# SCINT 2017 - 14th Int. Conference on Scintillating Materials and their Applications

Monday, 18 September 2017 - Friday, 22 September 2017

Congress Centre "Le Majestic" **Programme** 

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# Monday 18 September 2017

# **Opening Session: Welcome Address (09:00-10:00)**

-Conveners: Christian Pedrini; Etiennette Auffray

time [id] title

09:30 [243] 25 years of Scint conferences

# Coffee Break (10:00-10:30)

## **Opening Session: Invited talks** (10:30-12:00)

## -Conveners: Etiennette Auffray; Christian Pedrini

#### time [id] title

# 10:30 [166] Advances in Scintillators for Nuclear Security

#### Presenter: MELCHER, Chuck

Increased concern about nuclear security in recent years has sparked a large effort to discover and develop new high performance radiation detectors for both gamma rays and neutrons. For a number of years, the primary options for gamma-ray detection have relied on: a) high purity germanium detectors with excellent performance but high cost and significant operational burden, b) cadmium zinc telluride semiconductors with very good energy resolution and room temperature operation but high cost, c) thallium-doped sodium iodide scintillators with reasonable cost but relatively poor energy resolution, and d) plastic scintillators, mainly polyvinyl toluene, with low cost but almost no spectroscopic capability. Recent research has focused on new inorganic scintillators with energy resolution that enables isotope identification previously attainable only with semiconductor materials. As a result, factors that impact energy resolution, such as nonproportionality and nonuniformity, are now much better understood than just a few years ago, and they continue to be active areas of investigation. Neutron detection with scintillators has also seen important advances as the shortage of 3He puts pressure on the development of alternative thermal neutron detection technology. For instance, inorganic crystals with high sensitivity for both gamma rays and neutrons have been developed, and organic crystals have been developed with gamma-neutron discrimination previously only attained by organic liquids. However, despite the discovery of numerous scintillating compounds that work well at the cubic millimeter scale of laboratory samples, most of them have defied cost-effective scaling up to the larger sizes required by security applications. Consequently, the development of inexpensive synthesis techniques will be critical to the successful widespread deployment of new radiation detection technologies.

# 11:00 [240] Studies of precision time-tagging of charged tracks with scintillating crystals for the phase-II upgrade of CMS

#### Presenter: TABARELLI DE FATIS, Tommaso

The projected proton beam intensity of the High Luminosity Large Hadron Collider (HL-LHC), planned to begin operation in 2026, will result in about 200 concurrent proton-proton interactions per 25 ns bunch crossing. The scientific program of the HL-LHC, which includes precision characterization of the Higgs boson, measurements of vector boson scattering, and searches for new heavy or exotic particles, will benefit greatly from the enormous HL-LHC dataset. However, particle reconstruction and correct assignment to primary interaction vertices present a formidable challenge to the LHC detectors that must be overcome in order to harvest that benefit. Time tagging of minimum ionizing particles (MIPs) produced in LHC collisions with a resolution of 30 ps provides further discrimination of interaction vertices in the same 25 ns bunch crossing beyond spatial tracking algorithms. The Compact Muon Solenoid (CMS) Collaboration is pursuing two technologies to provide MIP time tagging for the HL-LHC detector upgrade: scintillating crystals read out by silicon photomultipliers (SiPMs) for low radiation areas and silicon low gain avalanche detectors for high radiation areas. This talk will motivate the need for a dedicated timing layer in the CMS upgrade, and focus on the first technology. Test beam results and a reference design using small LYSO tiles will be presented. The requirements on the crystal and SiPMs properties and the R&D needed to optimize the performance within the constraints posed by the integration into the CMS experiment will be discussed.

# 11:30 [209] The 10ps Time-of-Flight PET challenge: Myth or reality?

Presenter: LECOQ, Paul Rene Michel

The future generation of radiation detectors is more and more demanding on timing performance for a wide range of applications, such as time of flight (TOF) techniques for PET cameras and particle identification in nuclear physics and high energy physics detectors, precise event time tagging in high luminosity accelerators and a number of photonic applications based on single photon detection.

There is a consensus for gathering Europe's multidisciplinary academic and industrial excellence around the ambitious challenge to develop a 10ps TOF PET scanner (TOFPET). The goal is to reduce the radiation dose (currently 5-25 mSv for whole-body PET/CT), scan time (currently > 10 minutes), and costs per patient (currently > 1000 € per scan), all by an order of magnitude, opening molecular imaging procedures to new categories of patients, including pediatric, neonatal and even prenatal examinations. Moreover such a time resolution will cause a paradigm shift in in-vivo molecular imaging, by enabling on-the-fly image formation and observation of bio-distribution and biochemistry in animals and patients, as well as an order-of-magnitude leap in molecular sensitivity and speed.

To achieve this goal it is essential to significantly improve the performance of each component of the detection chain: light production, light transport, photodetection, readout electronics.

This talk will concentrate on the light production and light transport. It will be shown that standard bulk scintillators are unlikely to achieve this very ambitious goal. On the other hand the introduction of a number of disruptive technologies, such as multifunctional heterostructures combining the high stopping power of well know scintillators with the ultrafast photon emission resulting from the 1D, 2D or 3D quantum confinement of the excitons in nanocrystals, as well as photonic crystals and photonic fibers, open the way to new radiation detector concepts with unprecedented performance.

Welcome Drink (12:00-13:00)

Lunch (13:00-14:30)

Applications: session 1 (14:30-16:30)

-Conveners: Paul Rene Michel Lecoq time [id] title

## 14:30 [212] Fiber-based Calorimeters for High Energy Physics

Presenter: PAUWELS, Kristof

Future high energy physics experiments will require major improvements in the performances of hadron and jet calorimetry. Because of the challenging conditions in which they will be operated, unprecedented levels of energy and timing resolutions, as well as efficient particle identification are required. An approach based on heavy inorganic crystal fibers to form a fully homogeneous calorimeter was proposed earlier. Designs based on assemblies of small elements of undoped and doped materials have to potential to combine excellent energy resolution and particle identification abilities with its dual readout and vertexing/tracking capabilities. Shaping the scintillators in elongated (fiber-like) geometries becomes a challenge when criteria on their performances are set so high.

The initial focus was set on LuAG since this garnet structure has enough density to allow for homogeneous designs. A careful analysis of the fiber geometries and the growth parameters led to an enhanced optical quality and light propagation. First demonstrators were then assembled and tested during multiple test beam campaigns, demonstrating the potential of crystal fibers in a set of calorimeter geometries (homogeneous and sampling both in pointing and transverse configurations). Because of cost considerations, more emphasize is being given to sampling geometries. As a consequence, prototypes with crystals of lower density (YAG) were later also assembled into calorimeter units and thoroughly tested. The flexibility of this innovative type of calorimetry was demonstrated and results were obtained with modules either with very fine granularity or rather loose sampling fraction. Based on Geant4 simulations, we also studied the best way to find a good compromise between cost and performances by smartly sacrificing the homogeneity of the calorimeters in specific regions.

Another line of work was directed to the improvement of the timing properties and of the radiation hardness of the fibers. Studies performed on both bulk and shaped materials demonstrated the crucial role of the raw material and impurities. Codopants were also used to balance compositions as an attempt to meet all the requirements. Because of the large quantities of fibers to be considered for the construction of a full calorimeter, extra care was taken to ensure the reproducibility of the growth processes. On this later point, as an alternative to crystal fibers, silica-based fibers were also considered. Their lower density is counter-balanced by growth processes more easily scalable to mass production.

This contribution will review the collective R&D effort which is on-going both on the bulk material, the fiber growth and the assembly of prototypes.

\*This work has been supported by the H2020 projects AIDA-2020 (GA no. 654168) and INTELUM (GA no. 644260).\*

# 15:00 [171] Applications of Very Fast Inorganic Crystal Scintillators for Future HEP Experiments

#### Presenter: ZHU, Ren-Yuan

Future HEP experiments at the energy and intensity frontiers require fast inorganic crystal scintillators with excellent radiation hardness to face the challenges of unprecedented event rate and severe radiation environment. This paper reports recent progress in application of fast inorganic scintillators for future HEP experiments, such as thin LYSO crystals for a shashlik sampling calorimeter proposed for the CMS upgrade at HL-LHC, undoped CsI crystals for the Mu2e experiment at Fermilab and a rare earth doped BaF2 crystals for Mu2e-II. Applications of very fast crystal scintillators for Gigahertz hard X-ray imaging for the proposed Marie project at LANL will also be discussed.

# 15:15 [90] Composite scintillators for high energy physics

#### Presenter: GEKTIN, Alexandr

High energy physics need radiation hard large area detectors. The composite base scintillators are an alternative to bulk scintillation detector due to ability to make the large area detectors at reasonable price.

This work is devoted to the development of thin-layer scintillation detectors development for high granularity calorimeters. Depending on radiation intensity, the use of detectors based on radiation-resistant single crystal or composite scintillators was proposed.

The design of thin-layer detectors based on silicate crystals with wavelength shifter (WLS) fiber based on garnet crystals were studied. The scintillators for detector and WLS fiber were selected on the base of overlapping of their excitation and luminescence spectra. The dimensions of single crystals and the position of the WLS fiber which providing high light output uniformity of detector were determined.

The granulometric composition and dimensions of thin-layer YSO:Ce composite detectors were optimized. YSO:Ce granules were obtained by sol-gel method, solid-phase synthesis or mechanical grinding of single crystals [1]. Radiation-resistant optical polysiloxane was used as a immersion binder for the granulas [2]. For light collection quartz or leukosapphirer light-conducting layer and WLS fiber Y-11 or YAG: Ce were used.

The radiation resistance tests of composites were carry out. YSO:Ce, YSO:Ce,Ca YAG:Ce single crystals, optical polysiloxanes, quartz glass and leucosapphire were irradiated with electrons (E0 = 8.3 MeV) at room temperature. The total integral dose reached  $300 \pm 0.5$  Mrad. Also degradation of optical polysiloxanes was investigated under exposure of 2 MeV protons, the fluence was 1014 protons•cm-2. Optical, luminescent and scintillation characteristics of tested materials before and after irradiation were measured. It is shown that up to 300 Mrad these material are radiation stable. The decrease of the light output is up to 2% for single crystal and up to 15% for composites. The transmittance decreasing for optical materials is within 5%. With increasing the proton fluence, the optical degradation of polysiloxanes occurs in the visible region.

1. Ukraine patent 111455 (2016).

2. Boyarintsev A.Yu., Galunov N.Z., Karavaeva N. L. et al. Func. Mat. 2013, 20, P.471-476.

15:30	[47] Design and status of the Mu2e crystal calorimeter
	Presenter: MISCEIII, Stefano
	The Mu2e experiment at Fermilab searches for the charged-lepton flavour violating
	neutrino-less conversion of a negative muon into an electron in the field of
	a aluminum nucleus. The dynamics of such a process is well
	modelled by a two-body decay, resulting in a mono-energetic electron with
	an energy slightly below the muon rest mass (104.967 MeV).
	If no events are observed in three years of running, Mu2e will set a limit on
	the ratio between the conversion rate and the capture rate \convrate of \$\leq
	6\ \times\ 10^{-17}\$ (@ 90\$\%\$ C.L.). This will improve the current
	limit by four orders of magnitude~\cite{MU2ETDR}.
	A very intense pulsed muon beam (\$\sim 10^{10} \mu/\$ sec) is stopped on
	a target inside a very long solenoid where the detector is located.
	The Mu2e detector is composed of a tracker and an electromagnetic
	calorimeter and an external veto for cosmic rays surrounding the
	solenoid. The calorimeter plays an important
	role in providing excellent particle identification capabilities, a fast online trigger filter
	while aiding the track reconstruction capabilities. It should be able to keep
	functionality in an environment where the n, p and photon
	background from muon capture processes and beam flash
	events deliver a dose of $\sim$ 120 Gy/year in the hottest area.
	It will also need to work in 1 T axial magnetic
	field and a \$10^{-4}\$ torr vacuum. The calorimeter requirements are
	to provide a large acceptance for 100 MeV electrons and reach:
	(a) a time resolution better than 0.5 ns @ 100 MeV;
	(b) an energy resolution {\it O(\$10\%\$)} @ 100 MeV and
	(c) a position resolution of 1 cm.
	The calorimeter consists of two disks, each one made of 674 pure Csl
	envetale read out by two large area array 2 <sup>s</sup> times <sup>2</sup> 2 of LIV extended
	SiDM $6$ times $6$ mm <sup>6</sup> /2 <sup>6</sup> . We report here all progresses done for the construction and test of the
	Signification of the second seco
	St Cobain, Siccas and America firms, Each cructal has been
	readout by two pro production Mu2o SiDMs colocted among the
	anos produced by Hamamateu. Sonsi or Advancid. Each photoconsor
	bas been amplified and regulated in bias voltage by means of
	nas been amplined and regulated in bias voltage by means of
	a FEE custom cmp. Find uigitization stage is also custom and telles $a_1 = 5$ as sampling. The module 0 will be expressed to an electron
	on a 5 ns sampling. The mounter-0 will be exposed to an electron
	Dean in the energy fange around 100 MeV at the BTF (Beam TeSt
	racinity) in riascau. Preinfindaty results of uninity and energy
	resolution at normal incluence will be shown as well as dependence
	or response and resolution as a function of the implinging angle.

# 15:45 [22] Development of a Crystal Calorimeter for the Electron Ion Collider

#### Presenter: WOODY, Craig

The Electron Ion Collider (EIC) is a new facility that has been proposed in the US to study the structure of nuclear matter with precision electromagnetic probes at sufficiently high energies and with sufficient luminosity to access the gluon dominated regime of QCD with high statistical precision, and with polarized beams to enable a complete picture of the spin structure of the nucleon. Two versions of this facility have been proposed, one at Brookhaven (eRHIC) and another at JLAB (JLEIC), which would have the capability of colliding beams of electrons in the energy range of 5-10 GeV (eventually up to 20 GeV) with heavy ions in the range of 10-110 GeV/A, and with polarized protons up to 275 GeV/c. These facilities will require new detector systems to measure the scattered electron with high precision in order to provide the kinematic constraints to reconstruct the overall event, as well as measure the hadronic fragments from both heavy ion and proton beams, including particle id.

In order to measure the scattered electron at small scattering angles, a high precision electromagnetic calorimeter will be required for the endcap region of the detector in the electron going direction that can provide an energy resolution ~ 1%//E + 0.5%. A crystal calorimeter would be the best choice to provide such a high resolution, and there has been considerable experience with such calorimeters over many years. Given the energy range of interest at EIC, lead tungstate (PWO) is one of leading candidates for this calorimeter. The calorimeter would consist of several thousand crystals, each ~ 20 cm long, similar to the PANDA endcap calorimeter. However, the light yield requirements would be somewhat less demanding than for PANDA due to the higher energy range, and the radiation damage requirements would be far less demanding than for the CMS crystals. The calorimeter would be located inside or just outside the solenoid spectrometer magnet and the readout would therefore have to work inside a magnetic field. Therefore, silicon photomultipliers or avalanche photodiodes would be used to read out the crystals.

A R&D program is being carried out by several groups interested in future experiments at EIC to investigate the requirements of this crystal calorimeter and to study the performance of its various components. We have been working with two principle suppliers of PWO crystals, the Shanghai Institute of Ceramics (SIC) in China and Crytur in the Czech Republic, to obtain high quality crystals with sufficient radiation damage tolerance to meet our requirements. We have also tested crystals in a test beam at Fermilab with a SiPM readout and measured their performance. Finally, we have carried out Monte Carlo simulations to study the requirements for the calorimeter that are needed to measure the scattered electron with sufficient precision in order to make the desired physics measurements. A summary and discussion of all of these topics will be presented at this conference.

## 16:00 [118] High-Quality Lead Tungstate Crystals for PANDA

#### Presenter: NOVOTNY, Rainer Willi

There is a strong interest and demand for high quality lead tungstate crystals (PbWO4, PWO) for electromagnetic (EM) calorimetry. PWO has been implemented into the EM calorimeter of the CMS-ECAL detector at LHC and is required for the completion of the PANDA-EMC in the target spectrometer. In spite of moderate radiation hardness in an environment of high hadron fluences, PWO represents an ideal material for EM-calorimetry on electron accelerators and/or experiments with primarily electromagnetic probes. The compactness, sufficient light yield and various recovery options combined with a moderate price require the availability of mass production. The Czochralski method has been proven to be the optimum growing technology. However, after bankruptcy of the Bogoroditsk Technical Chemical Plant in Russia as the major producer so far, a new manufacturer had to be found. The company CRYTUR (Turnov, Czech Republic) with good experience in the development and production of different types of inorganic oxide crystals has re-started end of 2014 in a common effort the development of lead tungstate for the mass production based on the Czochralski method. An impressive progress of the R&D was achieved since then. The growing technology was optimized to produce full size samples with the quality meeting the PANDA EMC specifications for PWO-II. The presentation will give a detailed progress report on the research program in collaboration with groups at Orsay and JLab. The full size crystals are characterized with respect to optical performance, light yield, kinetics and radiation hardness. The report will give a status on the ongoing pre-production of more than 120 tapered crystals for the barrel section of the PANDA-EMC and compare the achieved quality to the former production at BTCP and prototypes produced at SICCAS.

# 16:15 [196] Predicting the performance of the CMS precision PbWO4 electromagnetic calorimeter in the HL-LHC era from test beam results on irradiated crystals

#### Presenter: ZGHICHE, Amina

The harsh radiation environment in which detectors will have to operate during the High Luminosity phase of the LHC (HL-LHC) represents a crucial challenge for many calorimeter technologies. In the CMS forward calorimeters, ionizing doses and hadron fluences will reach up to 300 kGy (at a dose rate of 30 Gy/h) and 2E14 cm-2, respectively, at the pseudorapidity region of  $|\eta|=2.6$ .

To evaluate the evolution of the CMS ECAL performance in such conditions, a set of PbWO4 crystals, which had previously been exposed to 24 GeV protons up to integrated fluences between 2.1E13 cm-2 and 1.3E14 cm-2, has been studied in beam tests.

A degradation of the energy resolution and a non-linear response to electron showers are observed in damaged crystals. Direct measurements of the light output from the crystals show the amplitude decreasing and pulse becoming faster as the fluence increases. The evolution of the performance of the PbWO\_4 crystals has been well understood and parameterized in terms of increasing light absorption inside the crystal volume.

A double-sided readout configuration, in which two identical photodetectors are coupled to the opposite ends of each crystal, has also been tested. The separate and simultaneous readout of the light from the two sides of the crystal allows us to correct for longitudinal shower fluctuations and to mitigate the degradation of energy resolution in highly damaged crystals. The non-linear response to electromagnetic showers, arising from high non-uniformity of light collection efficiency along the longitudinal axis of irradiated crystals, can also be corrected by means of the double-sided readout technique.

## Coffee Break (16:30-17:00)

# Applications: Session 2 (17:00-18:45)

#### -Conveners: Ioan Dafinei

time [id] title

# 17:00 [83] Search for new Molybdenum based crystal scintillators for neutrino-less double beta decay search

#### Presenter: KIM, HongJoo

Absolute masses and Majorana nature of neutrinos can be revealed if neutrino-less double beta decays are observed. To achieve enough sensitivity for the extremely rare events, it is required to have detection techniques capable of distinguishing extremely rare signals over a significant radioactive background from both inside and outside of the detectors. One of the most promising techniques is a cryogenic

phonon-scintillation detector at a milli-Kelvin temperature using both photon and phonon signals by an event by event basis discrimination of the extremely rare signal from the huge backgrounds.

The AMoRE [1] and LUMINEU [2] collaborations are searching for the extremely rare event process of neutrino-less double beta decay ( $0v2\beta$ ) of 100 Mo isotopes using CaMoO4, ZnMoO4 and LiMoO4 crystals,respectively. Main advantages of the 100 Mo are its high transition energy (Q  $\beta\beta$  = 3034 keV) and a relative easiness to enrich. However, since the above crystals either have low light outputs, difficult to grow, or purification limitations, it is necessary to search for new Mo based crystals with better performances for the AMoRE-II and other next generation experiments.

We studied Li2O-MoO3, Cs2O-MoO3 and Na2O-MoO3 phases and have developed several new crystals grown by a Czochalski method. The syntheses of polycrystalline materials are discussed based on the TGA/DSC analysis and the crystal structures are reported based on the XRD analysis. Luminescence and scintillation properties such as emission spectrum, light yield and decay time of the crystals from a room temperature to 10 K were studied by exciting the crystal samples with a 280 nm pulsed LED or a beta source. Developed crystals are not luminescent at the room temperature but luminescent at the cryogenic temperatures and the decay time got longer. We studied Li4Mo5O17, Li2Mo4O13 (Li2O-MoO3), Na2Mo2O7, Na6Mo11O36 (Na2O-MoO3), and Cs2MoO4, Cs2Mo2O7, Cs2Mo3O10 (Cs2O-MoO3) crystals. Among those newly developed Mo-based crystals, the Na 2 Mo 2 O 7 crystal shows one of the most promising properties for the neutrino-less double beta decay search experiments.

[1] V. Alenkov et al., arXiv:1512.05957v1, 18 Dec 2015.

[2] http://lumineu.in2p3.fr/

# 17:15 [151] Optical and luminescent properties of 40Ca100MoO4 single crystals

#### Presenter: ANNA, Kozlova

Calcium molybdate based crystalline materials show good potential for laser physics and acousto-optics due to a combination of a wide range of functional properties [1]. Currently these crystals are efficiently used as humidity sensors and optical elements of stimulated Raman scattering lasers.

Over the last decades there has been a growing interest towards CaMoO4 (space group 4/m, scheelite structure) because of its applicability as a material for cryogenic scintillation detectors [2]. Calcium molybdate crystals contain the 100Mo molybdenum isotope for which the possibility of neutrinoless double beta-decay ( $0v2\beta$ ) has been predicted, i.e. it can be used in the physics of elementary particles. Authentic registration of neutrinoless double beta-decay could allow the scientists to determine the weight of the neutrino which is one of the most important tasks of advanced nuclear physics. Efficient search for neutrinoless beta-decay requires a sensitive calcium molybdate functional elements with high optical and sufficient scintillation properties, and the contents of radioactive isotope impurities of the U-238 and Th-232 series should be at an low level. The low-background plant should be installed deep underground with the aim of reducing the radiation background generated by space radiation and carefully screened with the use of radiation free materials [3]. In Russia, 40Ca100MoO4 single crystals for the functional elements of this type of detectors are only grown by Fomos-Materials OJSC.

The main requirements of crystalline elements of the detector are absence of color and the attenuation coefficient ( $\mu$ ) not higher than 0.01 cm-1 at 520 nm wavelength (maximum scintillation luminescence) [3]. The results showed that the quality of the crystals is insufficient for the stated objectives because 40Ca100MoO4 crystals grown in air acquire blue color. The color of the crystals is caused by the color centers formed during the crystal growth [1].

Therefore it is crucial to study the attenuation spectra of the material as a function of growth conditions and subsequent treatment and to choose the optimum 40Ca100MoO4 crystal growth conditions, which provide the required material parameters.

Optical and luminescent properties of 40Ca100MoO4 single crystals have been investigated. The influence of isothermal annealing on the attenuation spectra in the 350 to 700 nm wavelength range has been studied. A broad absorption band with a maximum at  $\lambda$ =460 nm is observed in the attenuation spectra of the samples. The dichroism phenomenon which is associated with anisotropy of the color centers in the crystals is observed along directions perpendicular to the optical axis. We calculated the degree of dichroism. The results showed that oxidative annealing of the 40Ca100MoO4 crystals at high temperature initially substantially reduces the 460 nm absorption band intensity and the attenuation.

[1] A.A. Blistanov, Kristally kvantovoi i nelineinoi optiki [Crystals of Quantum and Nonlinear Optics], MISiS, Moscow, 2007 (in Russian).

[2] V.B. Mikhailik, H. Kraus, Cryogenic scintillators in searches for extremely rare events, J. Phys. D: Appl. Phys. 39 (2006) 1181–1191.

[3] O.A. Busanov, R.A. Etezov, M. Gavriljuk Yu, A.M. Gezhaev, V.V. Kazalov, V.N. Kornoukhov, V.V. Kuzminov, P.S. Moseev, S.I. Panasenko, S.S. Ratkevich, S.P. Yakimenko, Background radioactivity of construction materials, raw substance and ready-made CaMoO4 crystals, EPJ Web Conf. 65 (2014) 4. http://dx.doi.org/10.1051/epjconf/20136503002.

## 17:30 [199] Scintillation Properties of (Zn, Mg) WO4 for Dark Matter Search

Presenter: KODAMA, Shohei Scintillation Properties of (Zn, Mg) WO4 for Dark Matter Search

Shunsuke Kurosawa\$^{1,2}\$, Hiroyuki Sekiya\$^3\$, Takahiko Horiai\$^4\$, Akihiro Yamaji\$^4\$, Shohei Kodama\$^4\$, Rikito Murakami\$^4\$, Yasuhiro Shoji\$^{4,5}\$, Yuji Ohashi\$^3\$, Yuui Yokota\$^1\$, Kei Kamada\$^{1,5}\$, Akira Yoshikawa\${1, 4, 5}\$, Akimasa Ohnishi\$^2\$, Mamoru Kitaura\$^2\$

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Dark Matter is one of the biggest issue in modern physics, and ones of the candidates for the Dark Matters are weakly interacting massive particles (WIMPs) which are expected to form a halo around our Galaxy. Our Solar System is rotating around the center of the Galaxy, and we expect that the Earth should experience a "wind" (named 'WIMP wind') against the direction of the rotation, where is direction to Cygnus. Thus, it is expected to be one of the evidence of Dark Mater to detect the WIMPs wind from Cygnus, and a direction sensitive detector is required.

Up to now, several groups have developed such detectors using gaseous detectors, while gaseous ones have low detection efficiency. In this study, we propose a new type Dark matter detector with single crystals with which is expected to have higher detection efficiency than gaseous ones; ZnWO\$\_4\$ and/or similar group can detect the direction of incident particles due to anisotropic [1]. However, the mechanism was not revealed.

We grew ZnWO\$\_4\$ and (Zn, Mg)WO\$\_4\$ single crystals with diameters of ~0.5 inch grown by the Czochralski process to reveal the mechanism. The bulk crystals were cut to cubic shape samples with a size of 10 mm x 10 mm x 10mm, and each sample had the surfaces with c-axis orientation. Moreover, we check the crystal structure using the powder X-ray diffraction. Even these samples had good uniformities of material composition and transmittance, anisotropic scintillation properties were observed.

Light outputs of the crystal irradiated with 5.5 MeV alpha rays and 59.5 keV X-rays were estimated for each direction (orientation) for ZnWO\$\_4\$ using a photo multiplier and an \$^{241}\$Am source. Here, we evaluated the light output ratio: Alpha-ray to X-ray. As a results, b-axis orientation had different ratio from other surface, and we confirmed the anisotropic for ZnWO\$\_4\$.

On the other hand, we found (Zn, Mg)WO\$\_4\$ had smaller anisotropic effect than ZnWO\$\_4\$. Moreover, the light output was smaller than ZnWO\$\_4\$ by ~25%. Here, lattice constant of b for (Zn, Mg)WO\$\_4\$ was smaller than that for ZnWO\$\_4\$ from X-ray diffraction pattern.

We discuss the mechanism of this anisotropic effect using also crystal structure data and other information in this presentation.

[1] F.A. Danevich et al., Nucl. Instrm. Meth. A544 552 (2005).

# 17:45 [35] Calibration and Performance of a 3D Imaging Calorimeter of DAMPE for Cosmic Ray Physics on Orbit *Presenter: WU, Libo*

The space experiment of DArk Matter Particle Explorer (DAMPE) developed in China is designed to find the evidence of dark matter particle by observing primary cosmic rays and gamma rays in energy range from 5 GeV to 10 TeV. Since its launch in December 2015, a large quantity of data has been recorded.

The BGO Electromagnetic Calorimeter (BGO ECAL) of the DAMPE is a total absorption calorimeter consisted of 308 BGO crystal bars that allows for a precise three-dimensional imaging of the shower shape. It provides a good energy resolution (<1%@200 GeV) and high electron/hadron discrimination (>10^5). The ECAL also provides a trigger capability for DAMPE. With the data set acquired during the first and a half years of operation in space, a precise time-dependent calibration for energy, shower topologies measured by the BGO calorimeter had been developed.

In this report, the instrumentation and development of the BGO ECAL is briefly described. The calibration on orbit, including the pedestal, minimum ionizing particle (MIP) peak, dynode ratio, and etc. is discussed, and more details about calibration methods and the performance in space are presented.

# **18:00 [160] Low-temperature studies of the scintillation of pure cesium iodide for cryogenic scintillator detectors** *Presenter: CLARK, Mike*

The search for particle dark matter is one of the most active fields in physics, with many experiments using different methods to search for possible dark matter candidates. Direct-detection experiments look for rare interactions between some detector mass and these dark matter particles. The DAMA/LIBRA experiment utilizes thallium-doped sodium iodide (NaI(TI)) crystals at room temperature to search for dark matter direct-detection, and have claimed an annual modulation signal for dark matter [1].

There has been recent interest in the use of cesium iodide (CsI) as a doped or undoped scintillator as a target material in cryogenic scintillator detectors [2]. Cryogenic scintillaton detectors compare light and phonon signals from particle interactions to discriminate between nuclear and electron recoils. Pure CsI is an interesting target because of its chemical similarity to NaI(TI) for comparison with the DAMA/LIBRA experiment.

Using an optical cryostat installed at Queen's University in Kingston, Ontario, Canada, we can observe cryogenic scintillators with room temperature photomultiplier tubes, providing a simple apparatus to measure light yield at low temperatures. Utilizing the multiple photon counting coincidence method [3], we can measure the light output of the CsI crystal to nanosecond precision over a large, millisecond timescale to completely capture individual scintillation events. We present the evolution of the light yield, scintillation time constants and alpha/gamma quenching factor of CsI from 300K to 3.4K. We observe a promising high light yield at low temperature, and an alpha/gamma quenching factor surprisingly greater than one for temperatures lower than 100K.

[1] DAMA Collaboration, R. Bernabei et al. Eur. Phys. J. C 73 (2013)

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# 18:15 [183] Scintillation properties of n-type GaAs at cryogenic temperatures

#### Presenter: DERENZO, Stephen

We describe the scintillation properties of n-type GaAs at cryogenic temperatures for the detection of sub-GeV dark matter particles. The density of dark matter in the galactic halo is about 0.6 GeV/c2/cm3, they have average velocities of about 0.001 c in random directions as they orbit the galaxy, and have not been detected by large-scale experiments designed for the GeV/c2 mass range. Dark matter particles in the unexplored 1-1000 Mev/c2 mass range only carry kinetic energies of 1-1000 eV and a scintillator with high efficiency and very low background will be required to detect them. Detection of single photons in the near infrared with high efficiency and low background at cryogenic temperatures is a challenge, but this technology is under active development using transition edge sensors and microwave kinetic inductance detectors. Anti-reflection coatings can be used reduce internal trapping of the scintillation light.

GaAs has a density of 5.32 gm/cm3, a refractive index of about 3.5 in the near infrared, and a direct band gap of 1.52 eV. When doped with silicon to provide a population of shallow donor electrons and boron to provide acceptor sites for ionization holes the luminosity is above 30,000 photons per MeV. Boron is naturally introduced during the crystal growth process. The silicon donor level is only a few meV below the conduction band minimum and the Mott transition concentration is about 2E16 per cm3. The boron acceptor level is about 0.19 eV above the valence band maximum and the donor-acceptor emission peaks at about 930 nm (1.33 eV). This emission is thermally quenched above 120K with a thermal barrier of about 12 meV. Single 1.33 eV photons can be produced with high quantum efficiency by excitation energies above the 1.52 eV band gap.

After prolonged exposure to a 50 keVp X-ray beam at 10 K we are unable to detect any thermally stimulated luminescence during an increase in temperature to 400 K. This is in contrast with Nal(Tl), which has more than six strong thermally stimulated emission peaks. The apparent absence of metastable radiative states in n-type GaAs can be explained by the efficient annihilation of metastable holes by the delocalized n-type donor electrons that fill the crystal when their concentration is above the Mott transition. An important consequence is the apparent absence of afterglow that produces single photon emission over long time spans. No other available scintillator allows the detection of dark matter particles at the single photon level.

## 18:30 [111] The liquid scintillator for JUNO experiment

#### Presenter: XILEI, Sun

The Jiangmen Underground Neutrino Observatory (JUNO) is a multi-purpose underground neutrino experiment. The center detector of JUNO consists of 20,000 tons of liquid scintillator (LS) contained in an acrylic sphere of 35.4 m in diameter and viewed by ~18,000 20-inch photomultiplier tubes. The energy resolution of JUNO is designed to be 3% at 1 MeV, corresponding to a light output of at least 1,200 photoelectrons per MeV. The light yield, optical transparency and low-level radioactive background are crucial for the liquid scintillator. This report will review the JUNO experiment and then focus on the LS R&D, including the LS composition, light yield, attenuation length, neutron gamma separation, radioactive background, and purification methods.

# Buffet Montagnard (19:30-20:30)

# History of Chamonix (20:30-21:30)

- Presenter: BURNET, Claire

# **Tuesday 19 September 2017**

Crystal growth: Crystal growth (08:30-10:00)

## -Conveners: Chuck Melcher

time [id] title

# 08:30 [220] Crystal Growth and Engineering of Inorganic Scintillators

Presenter: BOURRET-COURCHESNE, Edith

The pace of discovery of new inorganic scintillators has increased dramatically in the last decade with the demonstration of efficient scintillation in mixed and ternary halides1 and that of the power of engineering oxides compounds2. The ability to make new materials reproducibly is often key to major progress in fundamental physics and numerous applications. The field of scintillation is no exception and that single issue is impeding the rapid commercial development of these new materials. Development of a reliable crystal growth processes and engineering of the scintillation performance are linked as they affect each other. We will present recent results of research efforts in crystal growth achieved through simulations and imaging of the growth process, fundamental advances in physics of scintillation and specifics on use of co-doping.

E. D. Bourret-Courchesne, G. A. Bizarri, R. Borade, G. Gundiah, E. C. Samulon, Z. Yan, and S. E. Derenzo "Crystal growth and characterization of alkali-earth halide scintillators" Journal of Crystal Growth, vol. 352, pp.78-83, Aug 1 2012.
M. Nikl, A. Yoshikawa, "Recent R&D Trends in Inorganic Single-Crystal Scintillator Materials for Radiation Detection" Advanced Optical Materials Vol. 3, Issue: 4, Special Issue: SI, p. 463-481 (2015).

This work is supported by the Department of Energy, NNSA/DNN R&D and carried out at Lawrence Berkeley National Laboratory under contract #AC02-05CH11231.

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# 09:00 [37] Growth and characterization of eutectic scintillator taking the advantage of composite material

Presenter: YOSHIKAWA, Akira

As eutectic crystals consist of at least two different crystals, it can have two or more properties in the one body. The directionally solidified eutectic (DSE) systems have been discovered in various materials for many applications [1,2]. We have proposed mainly two approaches to develop the scintillators using the DSE system. One is the neutron scintillator using a eutectic body composed of lithium containing crystal and scintillation crystal [3,4]. Lithium containing crystal plays a role to react with neutron and generates alpha particle. The alpha particle reacts with the scintillation crystal and will give scintillation light. Another attempt is the submicron-diameter phase separated scintillator fibers (PSSFs). They possessed both the properties of an optical fiber and a radiation-to-light conversion. The PSSFs were fabricated using a DSE system. In PSSFs, the light emitted from the scintillator fibers is confined and transported along the fiber direction by a total reflection mode, so that high-resolution radiation imaging can be achieved. CsI/NaCI [5] and GAP/alpha-Al2O3[6] have been reported as PSSFs. The aim of this presentation is to review the growth and characterization of DSE system for scintillator application.

DSE systems were grown by the micro-pulling-down (u-PD) method with an RF heating system [1] at the eutectic composition. The eutectic phase structure was investigated by back scattered electron image (BEI). Radio-luminescence spectrum at room temperature was measured with the above spectrometer (EI FLS920) excited by 5.5MeV alpha rays from an 241Am source. To determine the light yield, we obtained the pulse height spectra of these crystals irradiated with gamma rays from a 137Cs (662 keV) source. Scintillation photons were detected with a photomultiplier tube (PMT, Hamamatsu, R7600U-200). The signals were amplified with a preamplifier (ORTEC, 113), shaped with a shaping time of 2 micro-sec (ORTEC, 572A) and read out with a multi-channel analyser (MCA, Amptek 8000A).

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# 09:15 [161] Growth and characterization of SrI2:Eu crystals grown by the Czochralski method

#### Presenter: GALENIN, Evgeny

Srl2:Eu2+ crystals are among the brightest discovered scintillation crystals with the light yield over 100 000 ph/MeV and the energy resolution up to 3% at 662 keV [1]. Despite many reports on Srl2:Eu growth by the Bridgman method, no significant progress has been achieved in development of large size crystal growth technologies targeted at reduction of production cost.

This report represents for the first time the Czochralski growth of SrI2:Eu crystals with diameters of up to 50 mm. Czochralski method is optimal from the point of scaling up of crystal growth technology. The development of Czochralski process for highly hygroscopic substances includes the optimized conditions of raw materials preparation and their loading into the growth chamber, as well as precise control over admixture content in the raw materials and growth atmosphere. pH of the raw material water solution is shown to be a key criterion of the raw material quality [2].

Compositions, as well as optical and scintillation parameters of SrI2:Eu crystals grown by the Bridgman and Czochralski methods are compared. The Czochralski process provides a uniform distribution of Eu2+ across the crystals within +/-5 %. The latter factor favors a high energy resolution within 3.6 -3.7 % at 662 keV obtained in detectors fabricated from different parts of Czochralski-grown crystals. Such values of the energy resolution are similar to those obtained with SrI2:Eu2+ grown by the Bridgman method both in this work, and in other laboratories. This certifies a high purity and a good quality of the Czochralski grown crystals and demonstrates a feasibility to successfully produce SrI2:Eu2+, as well as other highly-hygroscopic halide scintillation crystals by the Czochralski method. The growth of 50 mm dia. crystals in R&D scale industrial furnaces is the first step in adaptation to the large size SrI2:Eu growth at industrial equipment. Further advance will be based on the well-developed growth technology of large CsI, CsI(Na) and NaI(TI) alkali halide crystals with the diameter of up to 500 mm by the Modified Czochralski-Kyropoulos method [3].

\*The work is supported by the NATO multiyear Science for Peace Project NUKR.SFPP 984958 "New sensor materials and detectors for ionizing radiation detection"\*

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# 09:30 [148] Garnet scintillators, obtained by 3D printing

#### Presenter: DOSOVITSKIY, Georgy

Scintillation ceramics attracts attention for last two decades due to several potential advantages: lower production costs compared to single crystals, possibility to achieve high scintillation light yield and flexibility of composition. With a help of modern additive technology additional benefit arises – ceramic materials could be 3D-printed, which provide a new level of possibility to create a material with complex geometry. We report our latest results on 3D-printing of complex oxide garnet scintillators.

YAG:Ce scintillating material was obtained using a stereophotolithograpy approach for the first time. YAG:Ce nanopowder was synthesized by co-precipitation, then it was mixed with photocurable resin and surfactants to form a slip with volumetric bulk content ~25%. Then it was photocured layer by layer in stereophotolithograpy 3D printer to form a polymer-binded green body. Green body was carefully debinded and sintered at 1600 °C, which gave translucent ceramic objects with density ~98% of a single crystal.

Luminescence properties were found to be typical for Ce3+ doped YAG. Ceramics demonstrated challenging scintillation characteristics – average decay constant  $\tau$ sc under 60 ns and light yield measured under 5,5 MeV  $\alpha$ -particles excitation was found to be more than 60% higher compared to YAG:Ce single crystal. This is an ongoing research, and new results will be included into the talk.

The method developed may be useful to produce composite materials, sophisticated luminophores with a complex surface for LED lighting devices, complex shape scintillators for improved characteristics or special detector properties. Particularly, neutron detection materials could be produced by printing a complex permeable form and filling it with neutron moderator/absorber. 3D-printing allows obtaining virtually any geometrical form, including those, which could not be obtained by any other approach. The goal of this talk is to induce creativity to find applications to this new scintillator forming method.

Work is supported by grant № 14.W03.31.0004 of Russian Federation Government.

# 09:45 [106] In-situ diagnostics of phase separation and segregation during growth of Cs2LiLaBr6:Ce scintillator crystals by energy-resolved neutron imaging

Presenter: TREMSIN, Anton

Scintillators development is often limited by issues of crystal growth reproducibility especially for crystal grown by the Bridgman technique, an intrinsically "blind" technique not suited for in-situ monitoring. Recent progress in high resolution energy-resolved neutron imaging provides unique possibilities to perform in-situ measurements of process parameters, which currently can be obtained only indirectly.

Our proof-of-principle experiments demonstrate the possibility to measure the elemental distribution, shape and location of liquid/solid interface and structural defects in Cs2LiLaBr6:Ce scintillator crystals during growth of a cm-size crystal by a Bridgman process.

The concentration of several elements (most accurately for Li) is imaged with sub-mm spatial resolution during crystal growth, revealing the dynamics of elements segregation across the boundaries between the solid and liquid as well within the liquid phases. A distinct separation into two liquid phases is observed above the solid/liquid interface. In the lower liquid phase volume the concentration of Cs is increased above stoichiometric value accompanied by the decrease of Li concentration, with Li concentration increased in the upper one.

In combination with finite element modeling of thermal profiles during crystal growth these in-situ diagnostics can be used to optimize the growth parameters, such as thermal profile, growth and cooling rates, and some others in order to improve the quality and yield of resulting scintillator materials. Ultimately, optimization of growth parameters through a feedback control can be performed as information on the growth process can be obtained in real time (minutes to hours in crystal growth terms). This should allow quick path in the search for optimal growth parameters, thus greatly reducing timescale between the laboratory material discovery and upscaling to commercial/production.

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# Poster Session 1 (10:00-11:00)

# -Conveners: Remi Chipaux; Kristof Pauwels; Marco Pizzichemi [id] title

boar d

# [20] A scintillator detector for beam tuning of low energy single electron accelerator

#### Presenter: XIE, Yuguang

In general, the weak-current beam of accelerator can be measured by picoammeter, the Faraday cup and other devices. However, when the beam intensity is lower than picoampere, the methods above are not suitable. For some applications and researches, the quasi-single charged particle beam is a key and essential facility. Therefore, it is required to find a new solution to reach single particle sensitivity for beam tuning. In this paper, nine single particle sensitive beam detectors based on plastic scintillator and photomultiplier tube (PMT) were developed to measure the 100Hz pulsed electron beam in the range of 0.1 to 50 MeV. The beam was tuned from about 107 to quasi-single electron for each bunch by these detectors installed after slits along the beam pipe. In order to determine the number of electrons in a bunch, the gains of each PMT were calibrated by a precise single photon gain measurement system for PMTs; the signal response of each detector was calibrated with 207Bi radioactive source; the deposited energy in the plastic scintillator, and the electron detection efficiency and light collection efficiency of the probe, as a function of electron incident energy were also simulated in detail by Geant4. The commissioning result shows that the detector is an effective method to measure the weak-current beam lower than picoampere, instead of the Picoammeter and Faraday cup for beam tuning.

# [29] Application of a LaBr3(Ce) Scintillation Detector to an Environmental Radiation Monitor

#### Presenter: JI, Young-Yong

The importance of environmental radiation monitoring has increased since the nuclear accidents of the Fukushima nuclear power plant. Naturally occurring radioactive materials (NORM) also require radiation safety management to protect the public according to the protective guidelines against radiation in the natural environment developed by the Nuclear Safety and Security Commission (NSSC) in Korea. The spectrometric determination of the dose rate using scintillation detectors is a very useful method with respect estimating the ambient dose rate and the nuclide contribution to the ambient dose rate. The dose conversion from a measured energy spectrum for counts obtained using a gamma-ray spectrometer can be achieved from a G-factor, which means the theoretical detector response function, to make a spectrometric determination of the dose rate. The dose rate spectroscopy [1-3] was first introduced to make spectrometric determination more effective, that is, to get more information from one measurement, such as the ambient dose rate as well as an individual dose rate and its radioactivity for detected gamma nuclides. To expand its application to the environment, a 2"x2" LaBr3(Ce) scintillation detector within the environmental radiation monitor (ERM) was used to perform real-time monitoring of the dose rate and radioactivity for detected gamma nuclides around an ERM. After the preparation of dose rate spectroscopy of the used LaBr3(Ce) detector, which means the theoretical calculation of factors dedicated to the ERM to simultaneously estimate the dose rate and radioactivity from the measured energy spectrum, the ambient dose rate as well as the individual dose rate and its radioactivity for detected gamma nuclides were then calculated with only one measurement using the ERM during 15 minutes. The results were experimentally verified by an intercomparison of the in situ gamma-ray spectrometry results obtained by a portable HPGe detector and the analysis of samples taken around the ERM. The results showed that the dose rate spectroscopy using an ERM based on an 2"x2" LaBr3(Ce) scintillation detector could be applied to remotely monitor three variables in real-time around an ERM.

# [198] Coincidence Resolution Time Measurements of LaBr3 (Ce) Detectors with a Fully Digital Acquisition System.

## Presenter: SÁNCHEZ-TEMBLEQUE, Víctor

Fully digital acquisition systems are being increasingly used because of their high sampling rate, their more compact size and their ever-decreasing prize. The possibility of obtaining sampling rates on the order of 5 Gs/s at an affordable price make them a great choice for the study of ultrafast inorganic scintillators such as LaBr3(Ce), a key detector in various nuclear physics experiments, like for example the construction of the high-performance FAst-TIMing Array (FATIMA) for DESPEC. For this purpose relatively large (1"x1.5"x1") truncated cone LaBr3(Ce) crystals coupled to ultrafast photomultipliers (PMTs) were tested using traditional electronics, based upon constant fraction discriminators (CFD), time to digital converters (TDC) and multichannel analyzers, showing excellent results both in time and energy [1]. We compared these measurements with a fully digital acquisition chain (DDAQ), where coincidence measurements with Co-60 and Na-22 sources were acquired. Pulses from PMTs optimized for timing measurements were digitized to a switched capacitor array with a speed sampling of 5 Gs/s and a resolution of 16 bits. Different algorithms were applied to the raw data set obtained. Among them an in-silico version of the analog CFD was used. With this strategy we obtained coincidence resolving times below 150 ps FWHM for Co-60, outperforming the standard acquisition system. This result proves that the DDAQ can be a great substitute for analog processing signals in read out systems.

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#### [225] Scintillators in high-power laser driven experiments

# Presenter: TARISIEN, Medhi

Bunches of electrons, protons, ions and high energy photons produced during the interaction of Ultra High Intensity (UHI) laser pulses with matter, have received considerable attention throughout the last decade. The main characteristics of these new sources are their short durations (ns), their high fluxes (1012 particles) and their small dimensions (100µm). These bunches are already reaching kinetic energies of hundred MeV for protons and GeV for electrons and the associated Bremsstrahlung photons, which is amply enough to induce observable numbers of nuclear reactions in matter [1]. Unfortunately, such high-power lasers cannot shoot several times per hour. Nevertheless, new UHI lasers operating at high repetition rate like APOLLON in Paris, or ELI in Prague and Bucharest, will start shooting their first pulses in the very next years. This new generation of UHI lasers will be able to accelerate particles at repetition rates up to 10 shots per second, which will make possible to perform laser driven nuclear physics experiments accumulating statistics from shot to shot. Unfortunately, the flux of background particles generated during the laser matter interactions (mostly soft X rays) leads to an instantaneous huge energy deposit (µJ) in the scintillators usually outfitting nuclear physics experiments. It makes them blind during milliseconds after incident particles passed through the target. These extreme conditions for gamma spectroscopy require the use of fast scintillator and the study of their energy dissipation process (afterglow, etc...). The signals of scintillators cannot be treated as usual with standard photo-multiplier tubes associated with slow spectroscopic amplifiers and peak sensing analog-to-digital converters. We must use fast scintillators read with new kind of photo detectors (HPD) which present large signal dynamics, connected to a digitizer. With such system, we succeed to detect some 162 keV gammas rays emitted from radionuclide with 63µs half-life, created by one UHI laser shot [2]. After presenting the specificities of the background emission during the laser-plasma acceleration process, I will present the strategies of detection we have adopted to perform gamma spectroscopy in the future laser-driven nuclear physics experiments.

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# [34] Fast scintillation X-ray detector using proportional-mode Si-APD and a HfO2-nanoparticle-doped plastic scintillator

# Presenter: KISHIMOTO, Shunji

We have been developing a new fast scintillation detector using proportional-mode Si-APD for synchrotron radiation nuclear forward scattering (NFS) experiments in high-energy X-ray region of >30 keV. We fabricated a prototype detector with a 5wt% lead-loaded plastic scintillator (EJ-256, Eljen Technology) and a proportional-mode Si-APD (S8664-3796(X), Hamamatsu Photonics) as photodetector. The one-channel prototype detector was successfully tested for detection of 67.41 keV X-rays, where the X-ray energy is the same as the y radiation from Ni-61 (lifetime: 7.6 ns). The prototype detector could measure time spectra with a good time resolution of 0.50 ns by a nanosecond-width outputs. The intrinsic efficiency at 67.41 keV reached 6.9% for a beam path of 3 mm long [1]. We are now fabricating a four-channel detector using four 3×3×3 mm^3 EJ-256 scintillators and Si-APD arrays of 3×3 mm^2 pixel to increase detection efficiency. The detector has nanosecond response and a beam path of 12 mm long. At the conference, performance of the fast four-channel detector will be shown. We will also give a topic of a 10wt% HfO2-nanoparticle doped plastic scintillator (Hf-PLS), 3 mm in diameter and 1 mm thick. The new scintillator was examined with synchrotron X-ray beam for the prototype detector. Hafnium is one of heavy atoms, which has 72 of atomic number and the \*K\*-absorption edge of 65.351 keV. Its oxide nanoparticles can be incorporated in a polymer matrix. We previously tested a hafnium-doped organic-inorganic hybrid scintillator fabricated by a sol-gel method [2]. The present scintillator was improved in dispersion of hafnium-oxide particle. The pulse height distribution and time spectra for the prototype detector with Hf-PLS were measured at −34°C with an increasing APD gain of ~200 for 57.6 keV X-rays. Light yield of Hf-PLS was 1.2 times as that for EJ-256 of the same size and a good time resolution of 0.34 ns (full width of half maximum) were obtained, which were better than 0.54 ns for EJ-256. This would be due to difference in the scintillation solution, that is only b-PBD was added in Hf-PLS, on the other hand, binary solutions, like PPO and POPOP, were included in EJ-256.

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Y. Sun, M. Koshimizu, N. Yahaba, F. Nishikido, S. Kishimoto, R. Haruki, and K. Asai, Appl. Phys. Lett. 104 (2014) 174104.

# [75] Development of SiPM based Scintillation Detector for Energy Selective X-ray Imaging

# Presenter: PARK, Chanwoo

A detector based on a scintillation crystal and a SiPM has been developed and tested for energy selective x-ray imaging. The detector consisted of 27 mm X 27 mm X 1 mm (and 2 mm) CsI(TI) and LYSO crystals coupled to an 8 X 8 array of 3 mm X 3 mm SiPM pixels with 3.36 mm pitch. The SiPM outputs were multiplexed to 4-channel position signals and the 2D image was constructed using simple anger logic. The Monte Carlo simulation tool, DETECT2000, was utilized for verifying the single x-ray photon counting capability. The number of scintillating photons incident on each SiPM pixel was calculated by varying crystal surface treatment. Then, the minimum detectable x-ray energy was characterized compared to the dark count rate(DCR) of the SiPM. Energy resolution, count rate, and 2D image were measured and evaluated using Co-57 source (122 keV) experimentally. The results showed that the detector can be used for energy selective x-ray imaging.

# [100] Tl2GdCl5 (Ce3+): A new efficient scintillator for X and y-rays detection

# Presenter: ROOH, Gul

Scintillators having a high effective atomic number (Zeff) and density is required for the efficient detection of X and  $\gamma$ -rays. Due to high Z-number and density of thallium ion in the host material enhances the density and Zeff of a scintillator [1-2]. Our group recently discovered new thallium based Tl2GdCl5 (TGC) scintillator which shows higher density and Zeff of 5.10 g/cm3 and 71, respectively. Luminescence and scintillation properties of TGC: x% Ce (x = 0, 1, 5 and 10 mole %) is presented. This material is grown by two zones vertical Bridgman technique. Luminescence under X-ray excitation for pure TGC crystal shows TI+ ion emission in the range of 350-600 nm peaking at 400 nm [3] while 1, 5 mole% Ce3+ doped TGC samples shows Ce3+ emission in the range of 350-500 nm peaking at 389 and 393 nm. Scintillation properties such as light yield, energy resolution and decay time are investigated under 662 keV  $\gamma$ -rays from a 137Cs source at room temperature. Best energy resolution and light yield are found to be  $\geq$ 5% (FWHM) and 53,000±5,300 ph/MeV, respectively. Three decay time components are obtained under  $\gamma$ -rays excitation for all the samples. The investigated results revealed that this scintillator could be a potential candidate for the radiation detection in different applications such as medical imaging etc.

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Y. Fujimoto et al., Jpn. J. Appl. Phys., 55 (2016) 090301–090303.

# [204] Characterizing some detection properties of Zinc Oxide nanowires in Anodic Aluminium Oxide membrane, as a novel high spatial resolution X-ray imager

#### Presenter: ESFANDI, Fatemeh

The novel idea of using scintillator nanowires in polycarbonate membrane as a high spatial resolution X-ray imager is proposed by our group at Amirkabir University of Technology [1-3]. In this work, a new generation of nano scintillator X-ray imagers based on ZnO nanowires in Anodized Aluminum Oxide (AAO) nanoporous template is characterized. This characterization consists of simulation, radiation test of the detector by soft X-ray and analyzing the experimental spectrums for extracting the detection properties of ZnO nanowires.

For this purpose, the optical response of crystalline ZnO nanowire arrays in porous AAO template irradiated by low energy X-ray is simulated by the Geant4 Monte Carlo code. The results show that for 10 keV X-ray photons, by considering the light guiding properties of zinc oxide inside the AAO template and suitable selection of detector thickness and pore diameter, the spatial resolution less than one micrometer and the detector detection efficiency of 66% are accessible. From experimental view, the AAO nanoporous template with average pore diameter of 240 nm and porosity of 30% was fabricated by anodizing the Aluminum. The ZnO was deposited in AAO nanopores by electro-deposition method. By using a new setup, some detection properties of ZnO nanowires electrodeposited in AAO nanoporous template were extracted. The results show that for 12 µm thickness of nano scintillator and the energy of 9.8keV near the K-edge of ZnO (9.65 keV), the detection efficiency of nano scintillator for direct X-rays is 24%. In addition, it is found that all the X-rays that are absorbed in 300 nm thickness of the gold layer (electrical contact for electrodeposition) on the top of zinc oxide nano wires can participate in scintillation process with efficiency of 6%. So, the scintillation detection efficiency of the whole detector for 9.8 keV X-ray energy is 30%. The extracted light yield of zinc oxide nano scintillator (9000). Much better spatial resolution of this radiation hard nano scintillator in comparison to bulk ones is an advantage which candidates this nano scintillator for imaging or even for particle tracking in high energy physics applications.

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# [216] High resolution Thick ZnO Nanowires in AAO Template for Hard X-Ray Imaging Applications

# Presenter: SARAMAD, Shahyar

High spatial resolution, fast decay time and high detection efficiency are the main parameters of a scintillator for X-Ray imaging applications. Recently, ZnO microrod scintillators with thickness of around 10 µm, electrodeposited in anodizing aluminum oxide (AAO) template [1] and polycarbonate [2] membranes have been used as a position sensitive soft X-ray imagers. In this structures, each microrod surrounded by a light reflective membrane acts as a light guide which conduct the generated photon in each array to photodetector separately. Although, the ZnO microrod fabricated by the electrodeposition method has spatial resolution less than 2 µm, but it's quantum efficiency isn't enough high for hard X-Ray applications. The small length of ZnO microrods in electrodeposition method is a limiting factor related to the properties of ZnO as nonconductive surface which decreases the electrical current during the electrodeposition. But, the methods based on vacuum system are appropriate to overcome this problem by filling the overall height of the channel [3]. In this work, a AAO template with 60 μm thickness, hole diameter 200 nm and inter pore distance of 283 nm (Whatman Company) was filled with ZnO microrod by vacuuming method. The AAO template impregnation was performed by immersing the template in the saturated Zn (NO3)2.6H2O under 150 mbar vacuum conditions for 30 min and then transferred into oven (60 oC for 24 hour). Finally, in order to crystallize the ZnO material, the samples were annealed in air at 500 oC for 3 h (heating rate of 10 oC min\_1). The samples were characterized by SEM, EDS, XRD and Photo Luminescence (PL) spectroscopy. The data analysis of SEM images and XRD pattern show that the AAO channels are filled with 40 µm ZnO. The PL spectrum of the sample excited by 250 nm laser reveals a strong and very weak PL emission band at 400 nm and 750 nm, respectively. Annealing of the defects at 500 oC is the source of strong PL related to excitonic emission.

The detection efficiency of 67 ZnO microarrays in AAO template for different thicknesses and X-ray energies was simulated by GEANT4. The results show that for the 15 keV and 20 keV X-rays, the detection efficiency of 30 µm ZnO microarray in 60 µm AAO membrane is 31.34% and 18.97%, respectively. The 60 µm thickness of ZnO microarray at the same energies has detection efficiency of 44.22% and 30.43%, respectively. The experimental and simulation results demonstrated that the vertically aligned ZnO microrod arrays in AAO template could be a good candidate for high spatial resolution hard x-ray medical imaging applications.

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# [13] Li2Se(Ag) A New Neutron Sensitive Scintillator

#### Presenter: RIEDEL, Richard

We report on the discovery of a new neutron sensitive scintillator. Single crystal Li2Se grown using a flux method and doped with silver was verified to scintillate with exposure to alpha particles (Am-241) and to neutrons (Cf-252). An estimate of the light yield is 2000 photons per neutron capture with a scintillation decay time constant of about 200nsec. Radioluminesce spectra show a strong peak centered near 410nm with a weak secondary peak near 560nm.

# [14] Evaluation of ZnS/6LiF and ZnO/6LiF scintillation neutron detectors read out with SiPMs Presenter: HILDEBRANDT, Malte

Helium-3 has been for several decades the most widely used converting material in detectors for neutron scattering experiments. The world-wide shortage of its supply stimulated the development of alternative detector technologies [1]. One of these alternatives is the scintillation technology based on 6Li (10B) loaded ZnS scintillators read out by wavelength-shifting (WLS) fibers. Currently all detectors of this kind utilize photomultiplier tubes (PMTs) as photosensors. Di□erent light-sharing schemes (channel coding) are used to reduce the number of PMTs. While in such parameters as the detection e□ciency and the gamma-sensitivity these detectors approach those based on 3He, the count rate up to which they can be operated without significant event losses is rather low, which is attributed to the slow emission of the ZnS scintillator. Here, however, one should clearly distinguish between two di□erent sources for the event losses, such as: a) finite dead time of a readout channel and b) coding errors.

At the Paul Scherrer Institut in Switzerland we are working on a novel approach [2,3] for the realization of ZnS-based scintillation detectors for neutron scattering applications. The absorption volume of the detector represents an (1D or 2D) array of individual sensitive elements (pixels), each equipped with its own signal conversion and processing chain. An efficient light collection from the scintillator is achieved by embedding WLS-fibers into its volume. The conversion of the collected light into electrical signals is done by a Silicon Photomultiplier (SiPM). This compact and inexpensive photosensor is well suited for application in multi-channel detection systems. As the channel coding is abandoned, significant improvement of the rate capability can be expected.

We demonstrated the potential of this approach by building a high efficiency, low gamma-sensitivity, high count rate capable 1D neutron detector aimed as a possible replacement of the 3He detector of the POLDI time-of-flight diffractometer [3]. The detector showed stable performance up to a counting rate of about 50 kHz/pixel with 10% dead-time conditioned event losses at 17 kHz/pixel [2]. Such count rate capability is already well above the requirements of most neutron powder diffraction instruments including those planned at bright neutron sources like ESS. However, to be of general applicability including also inelastic neutron scattering experiments, where spot counting rates in the order of 50 kHz/pixel are expected [1], the rate capability of our detector needs to be further improved. One way is to reduce the dead time, which is possible at the expense of the trigger efficiency [2]. Another way is to use a faster ZnO:6LiF neutron scintillator instead of the one based on ZnS. Such fast neutron detection screen has become recently available from Scintacor (UK). In this work we compare the performance of single channel neutron detectors built from ZnS/6LiF and ZnO/6LiF scintillators following the characterisation procedure developed in [2].

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## [16] Neutron detection using Li-loaded scintillators coupled to a custom-designed silicon photomultiplier array

#### Presenter: LIANG, Felix

Scintillators that are capable of detecting both neutron and gamma-ray have received much attention in recent years. Of particular interest is the use of silicon photomultipliers (SiPMs) as the photosensor for such scintillators in low-power and compact-geometry applications. Three types of Li-loaded scintillators, CLYC, CLLB, and NalL, have been tested with a custom-designed SiPM array for temperatures between -20 and 50 degrees C. The array consists of four 6x6 mm^2 SiPMs arranged in a 2x2 configuration. Pulse-shape discrimination is used for neutron and gamma identification. Because the pulse shape changes with temperature, the quality of neutron-gamma identification varies with temperature. Furthermore, the larger dark current in SiPMs at high temperatures results in poorer energy resolution and neutron-gamma separation. Comparisons of the energy resolution and the neutron-gamma pulse-shape discrimination for the three scintillators coupled the SiPM array will be discussed.

# [25] Novel scintillator screens for fast neutron detection with improved efficiency and spatial resolution *Presenter: WALFORT, Bernhard*

We report on the comparison of novel scintillator screens for fast neutron detection and neutron imaging with a commercially available screen with respect to efficiency and spatial resolution. The scintillator screens used for the comparison are a 480mm x 480 mm x 2.4 mm sized, 30% ZnS(Cu) loaded into PP matrix developed and produced by RC Tritec AG and other products available on the market.

For the characterization of the two screens fast neutron experiments have been performed at the NEUTRA beam line at the Swiss Spallation Source SINQ .We used a digital detector system based on a cooled CCD camera. For the comparison of both screens we characterized the (i) gamma sensitivity, (ii) the efficiency concerning light output and (iii) the image quality. For the image quality comparison we made radiographic images of a copper step wedge and hexagonal fuel assembly mockup.

We showed that novel ZnS+PP based scintillator is more efficient mainly due to its significantly higher light output. We compared our results with similar ZnS in PP-matrix screen to different plastic scintillators as reported in literature [1]. In [1], at about the same thickness (2.4mm ZnS vs. 3 mm plastic scintillator) the sensitivity of plastic scintillator was found only to be about 10% of that of the RC Tritec AG screen. For a 8 mm thick plastic scintillator in [1], the sensitivity is again only about 38% that of the RC Tritec AG screen.

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#### [194] Scintillation powders for neutron detection

#### Presenter: FIŠEROVÁ, Lucie

The work presents scintillation powders such as GdOBr:Ce, Srl2:Eu, Dy, BaMgAl10017:Eu, Mn, YAP:Ce, YAG:Ce and others for detection of neutrons of wide energy ranges in connection with suitable conversion isotope. The influence of fraction size was studied as well as the fraction size of conversion powder and choice of suitable mixture ratio. Special interest was devoted to the study of scintillation nanoparticles and nanoparticles for the use of neutron energy conversion. Several powders are commercially available, but not used as an object for neutron detection. The inaccessible powders were laboratory prepared by solid-state synthesis. The response for alpha, beta and gamma radiation was also performed. The results and properties of chosen scintillators were compared with zinc sulfide activated by silver in connection with lithium-6 fluoride. The scintillation powder was mixed with conversion isotope in an appropriate ratio and with a suitable optical binder. The mixture was then applied to the optical glass and coupled to photomultiplier. The binder selection is dependent on the physicochemical properties of the scintillator powder, 5% polyvinyl alcohol was used for water insoluble powders, transparent varnish and silicone fluid for water soluble powders. The shape of the output signal from the photomultiplier divider was generally studied in order to determine the possibility of pulses separation generated in the scintillator sensitive to alpha and gamma radiation. The study aims to take advantage of current knowledge in the field of scintillation materials synthesis and to find a fast scintillation material with properties similar to zinc sulfide activated with silver.

# [224] Newly developed flexible thermal neutrons detectors containing 6LiF nanocrystals

## Presenter: VESCO, Michela

During the last decade much effort has been devoted in the synthesis of radiation resistant siloxane based scintillators doped with primary dye and wavelength shifter to detect with good light output both y-rays and  $\alpha$  particles [1-3]. Moreover, liquid siloxane based scintillating cocktails have been prepared and recently tested as for fast neutrons detection and n-y discrimination [4]. The detection of thermal neutrons using boron loaded siloxane scintillator has been also pursued, by using soluble compounds of boron, such as carborane, dissolved in the precursor resins prior to the cross-linking reaction [5]. Quite recently, 6LiF nanoparticles have been synthesized and entrapped in siloxane based scintillators and thermal neutrons have been detected, owing to the capture reaction leading to α and triton ionizing particles [6]. Good performances have been recorded both using boron or lithium compounds dispersed into the doped polymer matrix, though the light output is much lower than ZnS:Ag based standard detector, one of the most popular thermal neutrons detector. The commercial plate for thermal neutrons is based on a mixture of the inorganic phosphor ZnS:Ag, which is known to display extremely high light output (around 50 photons/KeV), and micron-sized particles of 6LiF as thermal neutron sensitizer. The mixture is added with a small amount of polymeric binder and pressed on an acrylic disk acting both as a light guide and as a support, since the composite as a self-supporting film is very fragile. The mean free path of both the generated particles inside the 6LiF grain, alpha and triton, resulting from the capture reaction 6Li ( $n,\alpha$ ) 3H is in the order of some microns (around 30 µm and 6 µm for triton and alpha respectively). Therefore, the grain size of the 6LiF powder used as absorber is a crucial parameter and the entrapment of 6LiF nanoparticles into the composite could lead to greatly improved light output, owing to the increased interaction of ionizing particles with the surrounding light emitting phosphor. On the other hand, the entrapment of the mixture in elastomeric binders such as radiation hard polysiloxane allows the production of flexible thermal neutron detectors, with remarkable advantages on their application to different fields. In this work, the preparation of 6LiF nanocrystals has been pursued by co-precipitation technique, using a mixture of water and ethanol in different ratios. The as-prepared nanocrystals have been characterized as for crystal structure and crystallites size by X-Ray Diffraction (XRD), whereas the composition has been investigated by Energy Dispersive Spectroscopy (EDS). In this preliminary step towards the realization of a new detector, the commercial inorganic phosphor ZnS:Ag (EJ-600) has been used as scintillating medium. A fixed weight ratio of 6LiF nanocrystals and EJ600 phosphor has been chosen, namely 3:1 EJ-600:6LiF, whereas different amounts of the mixture have been added to the polysiloxane binder, prior to cross-linking. The composites have been benchmarked as for light output and thermal neutrons detection efficiency against a commercial sample of EJ-426 taken as a reference.

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# [159] Characterization of liquid scintillation detector BC501A

#### Presenter: RAWAT, Sheetal

Identification of different types of radiation e.g. alpha, gamma, neutron, etc. is very important in many areas such as nuclear non-proliferation, security, health, spectroscopy and nuclear reactors. For the detection and measurement of neutrons, in particular, liquid organic scintillators have been proved to be one of the best detectors because of their ability to detect both neutrons and gamma rays in the mixed field of radiation with the help of pulse shape discrimination techniques. Liquid scintillation detectors of type BC501A are widely used in neutron spectroscopy, neutron time-of-flight measurements etc. However, this detector suffers from very poor energy resolution of gamma rays. The present work aims to characterize BC501A detector in terms of energy calibration and pulse shape discrimination.

The energy calibration of BC501A detector was done using the technique of gamma-gamma coincidence [\*\*1\*\*]. This technique is used to find the exact position of Compton edge. We have used a 3"\$\times\$3" BC501A detector (supplied by Saint Gobain Inc.) as test detector and 2.18"\$\times\$2.36" n-type HPGe detector (supplied by Canberra industries) as reference detector.Three gamma sources, namely, \$^{137}\$Cs (661.6 keV), \$^{22}\$Na (511 & 1274.53 keV), \$^{60}\$Co (1173.24 & 1332.5 keV) were used in the present studies. A 4-channel digital acquisition system DGF PIXIE-4 (supplied by XIA LLC) was used for the acquisition of spectrum. In PIXIE-4, the incoming signal is digitized by 14-bit 75 MS/sec ADCs. Subsequently, the data obtained by PIXIE-4 was analyzed using Radware software \*\*[2]\*\*. The energy calibration plot was obtained from the positions of Compton edge peaks corresponding to gamma energies 340.66, 477, 963.43, and 1061.7 keV. The calibration plot was found to be linear represented by the equation y=0.29x + 28.02, where x is channel no. and y is energy of gamma ray.

The pulse shape discrimination was performed as an application of this detector. Cf-252 neutron source of activity 373 KBq was used. The study of neutron-gamma discrimination by analog zero-crossing method was carried out.Using this method, zero-crossing time difference between neutron and gamma peaks was found to be 124ns and figure of merit (FoM), which is the quality of discrimination between two radiations, was estimated to be 1.88. This FoM value is in good agreement with the value reported in the literature \*\*[3]\*\*. The details of experimental measurements will be discussed in the meeting. ## References ##

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- 3. Nakhostin M. and Walker P.M., Nuclear Instruments and Methods in Physics Research A 621,2010, p. 498–501.

# [124] Lithium di-silicate Li2O·2SiO:Tb bright scintillation glass for thermal neutron detection

# Presenter: TRATSIAK, Yauhen

Among the variety of scintillating inorganic materials glasses hold a unique position. They are transparent, easly handled, environmentally friendly, cheap to produce, and can be easily obtained in different forms in short times: from bulk to fibers. However, the disordered structure of the atoms in a glass and the presence of structural defects cause low efficiency transfer of electronic excitation to emitting centers preventing the achievement of a high scintillation yield. So far, encouraging results have been obtained with sol-gel SiO\$\_{2}\$ doped with Ce [1] and with other glasses with stoichiometric composition, in particular di-silicates [2].

Lithium di-silicate system, with Li\$\_{2}\$O.2SiO\$\_{2}\$ (DSL) composition and doped with Ce ions, can be obtained in the form of glass ceramics which have advantages with respect to glasses. Glass-ceramics, in fact, combine the luminescent properties of rare-earth ions in crystallites with the morphological ones of the remaining mother glass. Indeed, partially crystallized DSL glass, doped by Ce ions, and containing nano-crystallites of Li\$\_{2}\$Si\$\_{2}\$O\$\_{5}\$ displays a light yield of more than 7000 ph/neutron and provides energy resolution for thermal neutrons better than 8.5%.[2]

In the present work, photo- and radio-luminescence properties of DSL glass doped with Tb ions were studied. DSL glasses doped by Tb\$^{3+}\$ ions were obtained by heat treatment of a mixture of Li\$\_{2}\$CO\$\_{3}\$, SiO\$\_{2}\$ and Tb\$\_{4}\$O\$\_{7}\$ in a gas furnace (in CO atmosphere) at a maximum temperature of 1450 °C for 2 h. Tb concentration was set to 0.6 at. % with respect to Li ions. The obtained samples were annealed at 500 °C for 4 h in a muffle furnace to reduce stress. Bulk glass was found to be colorless. The samples for this study were cut from the synthesized blocks in 1mm thick plates and polished.

Both photo- and radio-luminescence spectra showed bright narrow bands corresponding to  $^{5}_{5}D_{4}^{+} + ^{7}_{5}F_{j}^{+} Tb^{3}_{5}^{+} + ectronic transitions (490, 545, 590 and 620 nm respectively for j = 6, 5, 4 and 3). The integrated radio-luminescence emission intensity of DSL:Tb glass was found to be two times higher than that of a BGO single crystal reference, measured at room temperature and in the same experimental conditions. So, the light yield was estimated to be 25000 ph/neutron. Although the scintillation kinetic is rather slow due to the forbidden nature of the Tb$^{3+} 4f-4f transitions, its high light yield still makes DSL:Tb a very promising material for applications in threshold neutron detectors, where the sensitivity to neutrons is particularly important.$ 

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# [72] A study of 40Ca{100}Mo4 scintillation crystals for the AMoRE-I experiment

#### Presenter: LEE, J.Y.

The AMoRE-I experiment is the first stage of the AMoRE (Advanced Molybdenum based Rare process Experiment) project intended to search for neutrinoless double beta (0\$\nu\$\$\beta\$\$\beta\$) decay of \$^{100}\$Mo with CaMoO\$ {4}\$ scintillation crystals at milli-Kelvin. The \$^{100}\$Mo has advantages of the relatively high released energy  $(Q_{\Delta}) = 3034 \text{ keV})$  and theoretically predicted decay probability. The calcium molybdate (CaMoO\$\_{4}\$) is the brightest one among molybdate scintillation crystals. The experimental sensitivity to the 0\$\nu\$\$\beta\$\$\beta\$ decay will be increased by an enrichment of \$^{100}\$Mo. The calcium depleted in \$^{48}\$Ca is used for the CaMoO\$ {4}\$ crystal production to decrease background caused by the double beta decay of \$^{48}\$Ca. \$^{48depl}\$Ca\$^{100}\$MoO\$ {4}\$ crystals grown by the FOMOS-Materials in Russia of weight around 5 kg (~2.5 kg of \$^{100}\$Mo) will be installed in the low radioactive cryostat at the Yang Yang underground laboratory (Y2L) in Korea and cooled to milli-Kelvin to enhance the detection performance of the AMoRE-I experiment. The Y2L is located at 700 m under the surface which is 2000 m-of-water-equivalent deep underground providing a shield against cosmic rays. A goal of AMoRE is an exploration of the inverted scheme of the neutrino mass hierarchy which corresponds to the half-lives of  $10^{20}$  –  $10^{27}$  years. Therefore, the experiment requires a very low radioactive background environment. We have set upper limits of internal radioactivity levels for \$^{228}\$Th, \$^{226}\$Ra, \$^{227}\$Ac (daughters of \$^{232}\$Th, \$^{238}\$U and \$^{235}\$U), and the total alpha activity of U/Th in the crystal scintillators via a GEANT4 Monte-Carlo simulation. As a preceding experiment of the AMoRE-I, we measured internal radioactive contamination of the \$^{48depl}\$Ca\$^{100}\$MoO\$ {4}\$ crystals by \$^{228}\$Th, \$^{226}\$Ra and \$^{227}\$Ac using a delayed-coincidence analysis. The approach allows us to separate the Bi-Po decay sequences in the chains thanks to the relatively short half-lives of polonium radionuclides in the decay chains (= 164 s, = 1.78 ms, = 145 ms). Two unexpected alpha peaks (at 660 keV and 790 keV in an energy scale calibrated with gamma guanta) observed in one of the crystals are identified by using the SRIM simulation code and compared with the previous result of \$^{48depl}\$Ca\$^{100}\$MoO\$\_{4}\$ scintillation crystal radioactivity. We also measured optical transmittances, relative light yields, and energy resolutions of the \$^{48depl}\$Ca\$^{100}\$MoO\$\_{4}\$ scintillators to be used for the AMoRE-I experiment.

# **[175] Phonon-scintillation properties of molybdate crystals for neutrino-less double beta decay experiment** *Presenter: KIM, Hyelim*

The AMoRE (Advanced Mo based Rare process Experiment) double beta project currently uses CaMoO4 crystals as the particle absorber of the low temperature phonon-scintillation detectors to search for neutrinoless double beta decay of 100Mo. However, an R&D of other molybdate crystals is in progress aiming to find higher performance molybdate crystal satisfying the AMoRE experiment requirements. We studied phonon-scintillation properties of several molybdate crystals (Na2Mo2O7, Li2MoO4). Simultaneous measurements of heat (phonon) and scintillation (photon) signals were carried out at milli-Kelvin temperatures using 1 x 1 x 1 cm3 crystal samples. The detector performances of each crystal are compared in terms of energy resolution, light yield, and particle discrimination capability using light/heat ratio and pulse shape of phonon signals.

# [180] Particle discrimination with low-temperature detectors based on CaMoO\_4 scintillating crystals

#### Presenter: JO, Hyon-Suk

Simultaneous measurements of heat and scintillation light signals from several \$^{100}\$Mo-enriched, \$^{48}\$Ca-depleted \$^{40}\$Ca\$^{100}\$MoO\$\_{4}\$ crystals have been performed at millikelvin temperatures using metallic magnetic calorimeters (MMCs). Each scintillating crystal was equipped with heat and light detectors, with the latter consisting of a Ge wafer placed near the crystal to collect the scintillation photons. The high energy resolution and fast response of the MMCs allowed us to discriminate \$\beta/\gamma\$- and \$\alpha\shnabeta-induced events in the \$^{40}\$Ca\$^{100}\$MoO\$\_{4}\$ crystals by using two discrimination methods: by estimation of the ratio of light and heat pulse heights, or by pulse-shape analysis of the heat signals (difference in rise and decay times). Measurements were performed at the 700-m deep Yangyang underground laboratory (Y2L) with the setup of AMORE-Pilot, that is the pilot phase of the AMORE (Advanced Mo-based Rare process Experiment) project aiming to search for neutrinoless double beta decay of \$^{100}\$Mo.

## [48] Pre-production and quality assurance of the Mu2e crystals

Presenter: ZHU, Renyuan

The Mu2e calorimeter is composed by two disks of 1348 un-doped parallelepiped CsI crystals of 34\$\times\$34\$\times\$200 mm\$^3\$ dimension, each one readout by two large area SiPM arrays. We translated the calorimeter requirements in a series of technical specifications for the crystals that are summarized by the following list:

(i) a well defined 3D crystal shape with \$\pm\$ 100 \$\mu\$m tolerance on the transversal dimension and 100 \$\mu\$m tolerance on each plane flatness and on the parallelism and perpendicularity between faces;

(ii) a light yield \$>\$ 100 p.e./MeV when readout with a UV extended 2" PMT;

(iii) a good longitudinal response uniformity (LRU) rms \$<\$ 5\% measured along the crystal axis;

(iv) a large ratio F/T \$>\$ 0.75 between the fast scintillating light component, integrated in 200 ns, and

the total light emitted within 3 \$\mu\$ sec;

(v) a good radiation resilience with a light output loss \$<\$ 65\% after a total ionization dose up to 100 krad and a neutron fluence up to

10\$^{12}\$ n/cm\$^2\$;

(vi) and a small contribution to the readout noise \$< 0.6\$ MeV related to the radiation induced fluorescence in the crystals.

A pre-production of 72 Mu2e crystals has been procured by three international firms (Amcrys, St. Gobain and Siccas). A detailed

quality assurance (QA) has been carried out on each crystal for the determination of its mechanical and optical parameters. The measurement of the radiation hardness have been carried out on 2 (1) random sample for each firm for irradiation wit ionization dose (neutrons). A summary of the techniques used and of the QA characterization of the crystals will be shown.

# [49] Pre-production and quality assurance of the Mu2e Silicon Photomultipliers

Presenter: MISCETTI, Stefano

The Mu2e calorimeter is composed by two disks of 1348 un-doped parallelepiped CsI crystals of 34\$\times\$34\$\times\$200 mm\$^3\$ dimension, each one readout by two large area SiPM arrays. We translated the calorimeter requirements in a series of technical

specifications for the SiPMs that are summarized by the following list:

(i) a high gain, above 10\$^6\$, for each monolithic (6\$\times\$ 6) mm\$^2\$ SiPM cell; (ii) a good photon detection efficiency, PDE, of above 20\% at 310 nm to well match the light emitted by the un-doped CsI crystals;

(iii) a large active area that, in combination with the PDE, could provide a light yield of above 20 p.e./MeV;

(iv) a fast rise time and a narrow signal width to improve time resolution and pileup rejection;

(v) a Mean to Time Failure (MTTF) of O(10\$^6\$) hours;

(vi) and a good resilience to neutrons for a total fluency up to 10\$^{12}\$ n(1 MeV\$ {\rm eq}\$)/cm\$^2\$.

A modular and custom SiPM layout has been chosen to satisfy these requirements. To well match the wavelength of the emitted light produced by the CsI crystals, the SiPM detection efficiency have been extended in the UV region. The configuration readout of 2 series of

three 6\$\times\$6 mm\$^2\$ monolithic SiPMs has been selected to overcome the issues related to the parallel connection that, due to the large

capacitance, could have spoiled the pileup rejection and the energy and time measurements.

A pre-production of 150 Mu2e SiPMs has been procured by three international firms (Hamamatsu, Sensl and Advansid). A detailed

quality assurance, QA, has been carried out on each SiPM for the determination of its own operation voltage, gain, quenching time, dark current

and PDE. The measurement of the MTTF for a small random sample of the pro-production group has been

also completed as well as the determination of the dark current increase as a function of the neutron fluency.

A summary of the techniques used and of the QA characterization of the sensors will be shown.

#### [54] A scintillator based charged particle veto system for the PADME experiment

Presenter: GEORGIEV, Georgi Stefanov

The PADME experiment will search for the  $e+e- \rightarrow vA'$  process in a positron-on-target experiment, assuming a decay of the A' into invisible particles of the hidden sector. The 550 MeV positron beam of the DAΦNE Beam-Test Facility, at Laboratori Nazionali di Frascati of INFN, will be used. The suppression of the background due to bremsstrahlung emission from the beam positrons requires high efficient charged particle detectors with optimized geometry. A fine-grained plastic scintillator veto composed of three stations in vacuum is foreseen. Two stations are placed inside a dipole magnet with 0.6 T magnetic field and will also provide momentum measurement at the percent level. Different prototypes for the design of the detector elements, the photo-sensor, and the front-end electronics were studied with single electron beam at the DAΦNE Beam-Test Facility to choose the optimal technologies and construction solutions. PADME is currently under construction and it is planned to take data in 2018. The design of the charged particle vetoes and the test beam performance of the prototypes will be reviewed.

# [105] Pulse Shape Discrimination with CsI(Tl) to Improve Hadron Particle Identification at High Energy Physics Experiments

### Presenter: LONGO, Savino

We will present studies evaluating the potential for the application of pulse shape discrimination (PSD) at high energy experiments using Thallium doped Cesium Iodide (CsI(TI)) electromagnetic calorimeters, such as Belle II, in order to improve neutral hadron vs photon separation. Using 30 cm long CsI(TI) crystals with PMT and PIN diode readout, the scintillation response for hadronic energy deposits is studied using proton test beams at TRIUMF with momenta ranging between 195-360 MeV/c and neutrons with kinetic energies up to 500 MeV. We show that in addition to the standard fast and slow scintillation components used to model photon pulses, the pulse shape variations observed for hadronic energy deposits in CsI(TI) can be modelled using an additional scintillation component with a decay time of approximately \$\tau\_\text{hadron} = 560\$ ns. By calculating the intensity of the hadronic scintillation component, neutron interactions containing combinations of secondary proton, deuteron and alpha particles are identified and separated from photon energy deposits with improved resolution compared to traditional charge ratio PSD methods. In addition, we have developed scintillation classes for GEANT4 to calculate the \$\tau\_\text{hadron}\$ scintillation intensity as a function of the ionization density of the interacting particles. These simulations reproduce the hadronic CsI(TI) response observed in the proton and neutron data. GEANT4 simulations are also used to study the expected improvement for neutral kaon long vs photon separation at CsI(TI) calorimeters using PSD.

## [179] PANDA Barrel Time-of-Flight Detector

#### Presenter: SUZUKI, Ken

The PANDA Experiment at FAIR is an in-ring fixed-target experiment, employing  $\overline{p}$  annihilation on H to nuclear target with  $\sqrt{s} = 2.3 - 5.5$  GeV, questing after unsolved problems in modern Hadron physics, e.g. light and charm exotics, multi strange baryons, hadrons in nuclei. The experiment is currently under construction. A detector installation is expected to begin in 2021.

PĀNDA Barrel Time-of-Flight detector is located in the target-spectrometer region in which a solenoid magnet is equipped. The barrel has a dimension of ~0.5 m diameter, ~2 m long, covering ~6 m^2 surface. The barrel is composed of 90x30x5 mm^3 scintillating tile, each end is read out by 4 SiPMs which are serially connected to act as a single sensor. The scintillator tile modules are mounted on a PCB backplane with coaxial MMCX connectors. High density signal transmission lines are embedded in the multilayer PCB backplane. The backplane serves as a mechanical frame as well as signal transmission lines, realising a simple and robust, cable-less design. In total, 2k tiles and 16k SiPM will be used. About 55 ps single tile time resolution (sigma) has been achieved.

The PANDA experiment adopts a free-running data acquisition concept in order to allow as much flexibility as possible which the complex and diverse physics objectives of the experiment require, and also to fully exploit the high interaction rate of up to 2×10<sup>7</sup> events/s. Each sub-detector system runs autonomously in a self-triggering mode, yet synchronised with a high-precision time distribution system, SODANET. Zero-suppressed and physically relevant signals are transmitted to a high-bandwidth computing network implementing a software trigger. Without a selection of data the whole data rate could be as high as 200 GB/s. The data acquisition system aims for an online data reduction of factor 100 – 1000. This demands a highly advanced online analysis. Another technical challenge that needs to be dealt with is a proper sorting of event data delivered from various sub-detector systems that would otherwise overlap at high interaction rates. Without a proper clustering of data from sub-detectors into an event packet, the events would be mixed up. One of the first steps in event sorting is the determination of the collision time of the event, t0, followed by a pattern-matching, online tracking and PID, which would iteratively increase accuracy. As the ToF system delivers the most precise timing information, the information will be an anchor to break into this challenge.

The detector R&D is in a matured phase. The technical design report is currently being reviewed by the FAIR. In this talk the detector concept and detailed design, expected performance validated by hardware tests as well as by software simulations, and an outlook towards mass production will be presented.

# [195] The CMS ECAL Upgrade for Precision Crystal Calorimetry at the HL-LHC

#### Presenter: JOFREHEI, Arash

The electromagnetic calorimeter (ECAL) of the Compact Muon Solenoid Experiment (CMS) is operating at the Large Hadron Collider (LHC) with proton-proton collisions at 13 TeV center-of-mass energy and at a bunch spacing of 25 ns. Challenging running conditions for CMS are expected after the High-Luminosity upgrade of the LHC (HL-LHC). We review the design and R&D studies for the CMS ECAL crystal calorimeter upgrade and present first test beam studies. Particular challenges at HL-LHC are the harsh radiation environment, the increasing data rates and the extreme level of pile-up events, with up to 200 simultaneous proton-proton collisions. Precision timing can be exploited to reduce the effect of the pile-up. We report about the timing resolution studies performed with test-beams. We also report on the R&D for the new readout and trigger electronics, which must be upgraded due to the increased trigger and latency requirements at the HL-LHC.

# [138] Radiation Damage of CMS HCAL Scintillator/WLS fiber readout during Run1 and Run2

We will summarize measurements of Radiation Damage of Scintillator/WLS fiber readout of CMS HCAL Endcap calorimeter during Run1 and Run2. Measurements were performed using Laser injection system as well as using collisions data, and then confirmed by in-situ Co60 calibrations.

Using these data we predict performance of HCAL Barrel and Endcap detectors for remainder of Run2, Run3 and beyond.

## [30] High Dynamic Range Front-End Circuit for SiPM-Based Readout of Large LaBr3 Crystals

#### Presenter: MONTAGANANI, Giovanni Ludovico

Lanthanium bromide is currently one of the best choices among scintillating crystals for high resolution gamma spectroscopy. Its fast output pulse (16 ns decay time) allows the minimization of parallel noise component in the detector readout chain, and its high output yield (63 ph/keV) contributes to the achievement of a better energy resolution (2.8% @662 keV ) than classic Nal(TI) scintillators (7% @662 keV [1]). Recent improvements in SiPMs have proven the possibility to implement the scintillator readout chain with these solid-state detectors, avoiding the use of high voltages or low temperature needed for respectively PMTs and SDDs, and obtaining similar energy resolution (3.7% @662 keV). High density SiPMs, exploiting a large number of microcells (~10000 cells/mm^2) together with a low DCR (30 kHz/mm^2), high PDE (30%) and a fast decay time (50 ns), suit well the need for the scintillator light output high dynamic range front-end. SiPMs could be also tiled in large area arrays, and so are becoming a feasible choice for the readout of large LaBr3 crystals. The target of the project here presented is the design of an analog front-end capable of capturing the full dynamic range of a 3" LaBr3 scintillator coupled to SiPM tiles, with negligible degradation of the overall resulting energy resolution. In this work we will demonstrate how the ENC of the front end circuit heavily contribute to the resulting resolution at lower energies (200 keV- 600 keV) if large optical signals are also to be detected (e.g. from a 15 MeV photon impinging on the scintillator) without manual gain adjustments. These energy resolution calculations, resulting in a need for almost 14-bit resolution, brought us to the design of an automatic gain control (AGC) circuit composed by a current conveyor input stage (6 Ohm input impedance) and a self-triggered gated integrator with automatic signal-dependent multiple gain adjustment. In this work we will show the topology of the AGC and of the integrator circuit. From circuit simulations the ENC of the resulting system is 0.3 pC and the saturation charge is 5 nC, thus reaching 84 dB of dynamic range, significantly improved with respect to the current state of the art. Based on GEANT crystal optical simulations and on field measurements, the resulting energy resolution is almost unaffected by the readout chain noise from 200 keV to 15 MeV and it is expected to be lower than 3% @662 keV. The overall designed front-end was implemented in a first 8-channel prototype ASIC in AMS C35B4C3 technology submitted in January and whose experimental characterization will be presented at the conference.

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A. Dawood Butt et al., "Comparison of SiPM and SDD based readouts of 1" LaBr3:Ce scintillator for nuclear physics applications," 2015 IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC), San Diego, CA, 2015, pp. 1-4.

# [135] Characterization of Cs2LiLaBr6:Ce (CLLB) Scintillator with Silicon Photomultiplier Arrays

#### Presenter: TUFF, Adam Garry

A Cs\$\_2\$LiLaBr\$\_6\$:Ce (CLLB) crystal has been characterised in terms of gamma energy resolution and neutron /gamma pulse shape discriminating (PSD) abilities, across the temperature range -20°C to 50°C. Performance was assessed through measurements taken with the crystal close coupled to a variety of SiPM devices and array configurations. One of these devices is a new Silicon Photomultiplier developed by Kromek. The SiPM devices have two outputs; a standard output that utilizes the full amplification potential of the device, ideal for making energy resolution measurements; a second output has low capacitance to provide a fast response signal, comparable to a PMT and is ideal for performing PSD. The PSD investigation examined two variants of the Charge Integration method; a 'prompt integration' method, that includes more of the prompt scintillation signal in the calculation; and a 'delayed integration' method, whose results are more dependent on the late portion of the scintillation signal. The measurements made with the Kromek-developed SiPM array yield a comparable energy resolution to that achieved with a super-bialkali PMT at 662 keV. The obtained figure-of-merit values as a function of temperature are presented, demonstrating the ability of the detector system to perform PSD.

# [145] Development of SiPM-based X-ray counting scintillation detector for security applications

# Presenter: PHILIPPOV, Dmitry

Conventional X-ray human inspection systems – security scanners – in airports, prisons, and other special guarded places are based on an integration mode scintillation detector. Usually, it consists of an array of PIN-photodiodes coupled with scintillators. The detector accumulates scintillation signals during the integration time. Thus, the output detector response represents an integral or average measure of all detected X-ray photons, i.e. all direct and scattered events with different energies. SiPM-based scintillation detectors have already been considered as potential candidates for X-ray imaging of integration and counting modalities [1], [2].

We assume that an ability to count X-ray photons with high efficiency and energy resolution is of high importance for precise and comprehensive imaging based on the spectral analysis in a number of security applications, especially with respect to the low dose human inspection.

Therefore, we proposed and developed the SiPM-based fast scintillation detector operateing in counting mode to increase the image quality and decrease the absorbed dose for the human body.

Prototyping the detector, we developed 1D 16 channel array of 3x3x5 mm2 GAGG scintillators coupled with 3x3 mm2 KETEK SiPMs. The array is connected to our homemade 18 channel front-end analog electronics board. It provides a voltage supply for SiPMs and the SiPM pulse shaping. Finally, the whole signal waveforms are digitized by the 4 channel Spectrum M2i.2031 analog-to-digital streaming PCI board [3] and analyzed by a specially developed software.

The first results of characterization, experimental studies, and measurements of SiPM-based X-ray spectral counting detector with a conventional X-ray tube and Am-241 source will be presented.

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[3] http://spectrum-instrumentation.com/en/m2i2031

# **[157] Investigation on electron beam radiation defects induced in KETEK PM3350 silicon photomultipliers** *Presenter: STANCALIE, Andrei*

In this work, the impact of electron irradiation on crucial SiPM parameters like dark current, dark count rate and PDE are investigated. Controlled radiation dose studies are important in order to provide precise quantitative information about SiPM radiation hardness [1]. The damage in silicon devices depends on the flux, type and energy of the particle.

The photodetectors, subjected to radiation, were KETEK "PM3350" type consisting of an active area of 3X3 mm2 with 50 µm micro pixel size. The photo detection efficiency is usually up to 38% (for ~406nm) for these devices. Experiments have been performed at the linear accelerator (LINAC) machine at the National Institute for Laser, Plasma and Radiation Physics, Romania while their characteristics were measured at KETEK GmbH laboratories, Munich. The samples were exposed gradually to different doses, so that the accumulated dose was linear dependent from one SiPM to another. Tests on IV and DCR characteristics have been performed before and after exposure with a low light level CCD using a probe station (sample was contacted with probe needles) and "Andor, Clara" type camera with "Mitutoyo, FS70" optical microscope. The exposures parameters are 28V (1kGy) and 26.50V (10kGy). The exposure time was set to 10min.

The electron beam irradiation has shown that a large increase in dark count rate and a partial loss of gain uniformity occur at relatively low doses. Before breakdown the dark current increases by ~1order of magnitude for the 1kGy sample with respect to the reference. Increasing the dose to 10kGy led to an additional increase of the dark current by 5 orders of magnitude (not a linear dependence). Currents before breakdown are attributed to the surface and Si/SiO2 interface. The dark current intensity after breakdown increases by ~2-3 orders of magnitude. Dark currents after breakdown are attributed to a superposition of contributions from the surface and the bulk. In order to investigate parameters like the the breakdown voltage, recovery time and PDE a lower dose or a smaller sample (for example 6x6µcell test structure) is required. These experiments are planned for the next stages of our investigations.

[1] S. Sanchez Majos, et. al. Nucl. Instr. & Meth. Phys. Res. A 602(2009) 506-510.

#### Acknowledgements.

This collaborative work is supported by the COST Action TD1401: Fast advanced Scintillator Timing (FAST). The Romanian authors acknowledge the financial support of the Romanian Executive Agency for Higher Education, Research, Development and Innovation Funding (UEFISCDI), under Grant 24PED/2017, project "Photonics devices under extreme operating conditions – PHOENIX".

# [207] Implementation of an analytical model of SiPM in GATE

#### Presenter: DUPONT, Mathieu

The aim of this work is to implement a model of SiPM that can be used to simulate scintillator-based spectrometers implementing SiPM like with PMT in the GATE simulation platform.

GATE [1] is a monte carlo simulation tool built on top of GEANT4. It has been developed in order to ease the simulation of complete systems used in medical imaging and radiotherapy applications like Computed Tomography (CT) scanner, Positron Emission Tomography (PET), radiotherapy devices, etc. To fully simulate an acquisition, GATE can manage time-dependent phenomena. Movements are synchronized with the evolution of the source activities by subdividing the simulated run into acquisition time frames with frozen geometries.

The use of SiPM, instead of PMT in PET field has increased for several reasons: it is cheaper than PMT, insensitive to magnetic field and provides potentially better timings. Improvements in timing benefits directly to PET imaging by the reduction the uncertainties on the localization of the source of emission. With the inclusion of an analytical model of the pair scintillation crystal/SiPM, GATE users will be able to predict the impact of SiPM over PMT in a PET system.

The first step was to develop this model. For this, we have started by implementing an analytical model of SiPM from Marano et al. [2] which describes the electrical response of a SiPM to a photoelectron. In this model, SiPM is treated like an electrical circuit and its characteristic times (rising time, quenching time, recovery time) are combinations of the component specifications of this circuit (number of cells, internal resistances and capacitances, etc.). Theoretically, experimental determination of these electrical parameters can provide a complete response scheme to one photoelectron. With the inclusion in this model of crosstalk, afterpulse, dark count rate, we intend to develop a comprehensive simulation tool for scintillator-based spectrometer coupled with SiPM.

The second step is to exploit these results that will be cross-validated against experimental data to assess a more generic model, identifying what quantities are fundamental to simulate SiPM with the GATE digitizer and to describe a systematic method to measure these quantities.

[1] Jan S. et al., "GATE a simulation toolkit for PET and SPECT". Phys. Med. Biol., 2004, 49, 4543

[2] Marano, D. et al. "New Improved Model and Accurate Analytical Response of SiPMs Coupled to Read-Out Electronics" IEEE Sens. J. , 2016, 16, 19-21

# [140] Measurements of Position and Depth of Interaction using Silicon Photostrip Sensors with a CsI(Tl) Crystal Scintillator

### Presenter: JEON, Hyebin

As a scintillation detector, silicon photo sensors have been widely used in variety of fields such as medical imaging system, high-energy/nuclear, astro-physics experiments and so on [1-2]. We developed AC-coupled single-sided photostrip sensors fabricated on 380 --thick n-type silicon wafers with high resistivity, and an anti-reflection coating process was applied to light entrance window of the sensors. They have a size of 3.8×2.8 cm2 and 128 strip channels. For an optical property, quantum efficiency was measured to be > 90% at the wavelength of visible light (450 – 700 nm) [3]. We optically combined the sensors with a CsI(TI) crystal scintillator to detect passing particles especially for gamma rays. Two photostrip sensors were orthogonally placed on the top and bottom sides of the crystal, so that 2-dimensional information of interaction position can be obtained by the two perpendicular sensors. In addition, depth of interaction inside the crystal can be also obtained from a signal ratio of the sensors. Analog signals from every second strips are serially read-out by a VATA ASIC chip and are converted to digitized signals by passing a flash analog-to-digital converter (FADC) on a data acquisition (DAQ) board, then all signals are stored in a PC as histograms of pulse height distributions. At first, the sensor with the crystal, exposed to an Am-241 radioactive source, was tested as the positions of the crystal and the radioactive source were varied together. Then different pulse height distributions were taken along the strip channels. The pulse height distribution appeared over about 10 readout channels which corresponded to a contact surface's area of the crystal to the sensor. Two sensors were also tested to get 2-dimensional position. The position was measured to be strip channel number of (55.7±1.4, 35.8±1.0) in range of 64 channels for each coordinate, and the signal ratio between two perpendicular sensors was obtained to be 0.46±0.05. Finally, a test of the detector consisting of two sensors combined with the crystal for gamma-ray detection is ongoing using the same experimental setup in the LED test, but a radioactive source instead of the LED. Besides, we are studying detector simulation using GEANT4 in order to compare experimental results and to give complementary information while changing the depth of interaction in the crystal with a size of 2.8 × 2.8 × 10 cm3. In preliminary results, the signal ratios as a function of distance from the sensor show 1.23×sigma discrepancy between the nearest and the farther interaction positions, 1.7 cm and 10.0 cm, respectively. We present measurement results of the energy resolution, 2-dimensional position and the depth of interaction from two perpendicular photostrip sensors with the CsI(TI) crystal scintillator, and GEANT4 simulation result for the depth of interaction in the scintillator.

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[3] D.H. Kah, et al., Nucl. Instr. And Meth. A628 (2011) 256.

# [149] Design of the fast radiation detector with 10-picosecond time resolution based on crossluminescence scintillator

# Presenter: KAMENSKIKH, Irina

For obtaining spatial resolution ~1.5 mm in TOF-PET devices, the coincidence time resolution ~10 ps is needed, which is beyond the current state of the art. Crossluminescence (CL) crystals having fast (nanosecond) decay and very short (subpicosecond) rise time of emission can be considered as very promising scintillators for ultrafast radiation detectors. Despite of relatively low light yield of CL (~\$10^3\$ photons/511 keV \$\gamma\$-quantum), estimations show that the limit time resolution of CL scintillators, determined as arrival time of the first scintillation photon, can be < 1 ps. CsF crystals are well-known scintillators which are produced by several manufacturers. Scintillation detectors based on CsF scintillators (with thickness ~4 cm) and standard PMT were tested in [1, 2] where time resolution of ~400 ps was obtained for the detection of 511 keV \$\gamma\$-quanta. However, the prospects of using such kind of scintillation detectors are underestimated. CsF is an exceptional example of CL crystals which emits under standard conditions only fast CL without the presence of any slow emissions [3]. Decay time of CL in CsF is 2.8 ns and the CL rise time is expected to be ~1 ps. The CL emission band of CsF is peaked at 390 nm which suits well the spectral sensitivity of standard SiPM detectors. The problem is its high hygroscopicity but there are well-known methods for the implementation of hygroscopic scintillators in radiation detectors. Besides that, some lower-hygroscopic multicomponent CL crystals based on CsF can be also considered.

One more factor limiting time resolution when using such kind of CL scintillators is their relatively large absorption length. For the efficient absorption of 511 keV \$\gamma\$-quanta the thickness of CsF scintillator should be at least ~1 cm. Accordingly, in the standard scheme of scintillation detector with a single SiPM attached to the end face of the scintillation crystal there will be remarkable time difference for the detection of scintillation photons born near the front and end parts of the scintillator. In order to overcome this problem the SiPM array with the size of elements ~0.1 cm can be attached to the scintillator along the direction of \$\gamma\$-quanta propagation. The time resolution of such scintillation detector will be limited by time resolution of SiPM.

This work was supported by the Ministry of Education and Science of the Russian Federation (state contract no. RFMEFI61614X0006).

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# [99] Ionization Quenching Correction of Volumetric Organic Scintillators for use in Proton Therapy Presenter: ALSANEA, Fahed

Ionization quenching is a known phenomenon that causes non-linear scintillation response to heavy charged particles with high ionizing radiation density such as protons. In this work, we compare guenching correction models and their application to volumetric scintillators. We have investigated the feasibility of a volumetric organic liquid scintillator detector to verify and characterize clinical proton beams and intensity modulated proton therapy (IMPT) plans for cancer treatment [1], [2]. However, to fully achieve the potential of this concept, the ionization quenching in scintillators must be addressed to convert the 3D light distribution to its corresponding dose distribution. For the purpose of testing various semi-empirical quenching correction models, we have exposed our detector to five different proton beam energies produced by the synchrotron at M.D. Anderson Cancer Center (85.6, 100.9, 124, 144.9, and 161.6 MeV). We used Monte Carlo simulations to obtain the dose and linear energy transfer (LET) for these beams. The models we focused on are the Birks' formula and its variations, and energy density by secondary electrons (EDSE) model [3]. Birks' formula is a semi-empirical unimolecular model that relates the scintillation rate to specific energy loss. Other models include bimolecular quenching and a differentiation between singlet and triplet states. The EDSE model relates the light production inside a scintillator material to the energy distribution of secondary electrons produced along the ion tracks. Regions close to the particle track are of interest to understand ionization quenching. All models reasonably fit the measurements within 5%. We will discuss the level of agreement between the different models and Monte Carlo simulations. Furthermore, we will discuss guenching model modifications that are needed to describe the light distribution for a range of clinically significant proton energies (70-220 MeV). It will be important to understand the quenching mechanism and the limitations of semi-empirical models for our 3D scintillator detector to reach the clinically acceptable 3% dose accuracy.

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## **[134] Development of High Spatial Resolution Dosimeter for Medical Uses by Colorimetric Discrimination Method** *Presenter: SHIM, ChungBo*

It is of vital importance to measure accurately X-ray dose with high spatial resolution in medical radiation treatments. For that purpose, plastic scintillator detectors (PSDs) are often used, because good spatial resolutions can be achieved. The scintillation light from a PSD is, however, often contaminated by the Cherenkov light due to X-ray irradiation on the optical fiber particularly for high energy X-rays. One of the common methods of separating Cherenkov and scintillation light is a subtraction method, in which two identical optical fibers are tied together and a PSD is attached to only one of them. The difference in the light output from the two fibers can then be regarded as the scintillation light. This method requires two fibers tied together and works only with the assumption that the two fibers are exposed exactly to the same amount of irradiation. To overcome these difficulties, we have developed a PSD detector system by using a colorimetric discrimination method[1]. In our system, we attached a CMOS with a color filter array to one end of the optical fiber. The difference in the spectral shapes between Cherenkov and scintillation lights allows us to achieve the desired separation. Among the three given color components, red, green and blue, two of them are used for the estimation of the Cherenkov and scintillation lights, and the remaining information is used for the verification of our method. Since a CMOS has a large number of pixels, several optical fibers can be attached at the same time to a single CMOS, and we can measure simultaneously X-ray doses at different locations exposed to X-rays by using, for instance, 9 PSDs bundled together. Scintillators of 1 mm length and 1 mm diameter (Bicron BCF-60) are used to obtain high spatial resolution. We have performed demonstration experiments by using a Leksell Gamma Knife Perfexion and a Novalis Brain Lab.

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#### [12] Scintillation materials for PET/MRI coupled to digital SiPM

#### Presenter: SEITZ, Bjoern

Efficient PET scanners rely on inorganic scintillation crystals paired with efficient photon detectors. New PET machines are increasingly designed to co-register PET with MRI signals, which calls for a compact detector design insensitive to magnetic fields. The advent of SiPM, and digital SiPM (dSiPM) in particular promises a natural design route for an MRI compatible PET scanner.

The main detector requirements influencing the image reconstruction are the position resolution, energy resolution and coincidence time resolution. The use of SiPM technology mandates the study of the temperature dependence of the system.

We study the response of BGO, LYSO:Ce, GAGG:Ce and LuAG:Pr coupled to two modules of Philips DPC 3200 as a function of gamma energy between 100 keV and 1 MeV as well as in the ambient temperature range from 273K to 303K. The results are compared to the SensL J-series analog SiPMWe will present the characterisation of the dSiPM response in terms of breakdown voltage, dark count rate and duty factor. The energy resolution and coincidence time resolution for these four materials as a function of temperature will be shown. Last not least the influence of intrinsic background either from the photon sensor or the crystal will be discussed.

## [45] Development and Evaluation of PET-Compton imager based on Ce:Gd3Ga2.7Al2.3O12 and CeBr3 scintillators with SiPM arrays

#### Presenter: SHIMAZOE, Kenji

PET (Positron Emission Tomography) is currently an important tool in molecular imaging and medical diagnosis. Compton imager is a promising tool for future molecular imaging with multi-nuclides based on Compton scattering. Previously we have developed GAGG based Compton imager for environmental application [1][2] and GAGG based PET system[3]. Here we have developed PET-Compton hybrid imager based on two-layer structure using thin scatters and thick absorbers for multi-nuclide imaging. For achieving the good spatial resolution of Compton imager, the energy resolution of scintillators is one of the most important. CeBr3 is a promising scintillator because of its high light yield over 70000 photon/MeV, excellent energy resolution ~ 3% with no background radiation and fast decay time.

In this study, we present the development of PET-Compton hybrid detector which consists of 8 by 8 MPPC (Multi Pixel Photon Counter / SiPM) array individually coupled with 2.5 x 2.5 x 9mm3 Ce:Gd3Ga2.7Al2.3O12 scintillators (absorbers) and MPPC array individually coupled with 2.5 x 2.5 x 1.5mm3 CeBr3 scintillators (scatters). The pixel size of MPPC is 3mm by 3mm and operated at the voltage of 55V. CeBr3 scintillators are grown, packaged and mounted on the MPPC array. The signals from MPPC-scintillators are individually amplified and converted with dynamic time over threshold (dTOT) circuit to record the energy and timing information. In the experiment the image of 111In and 18F-FDG, which are used as SPECT and PET tracers, was acquired using the developed detector for Compton imaging and PET imaging.

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# [74] Development of a SiPM based DOI-PET detector module using depth-dependent reflector pattern within a single layer scintillator

#### Presenter: SONG, Hankyeol

A depth-of-interaction(DOI) PET detector has been developed based on a single layer scintillator coupled to the Silicon photomultiplier(SiPM) with depth-dependent reflector patterns. The DOI method uses different reflector patterns which varies distribution of lights according to the interaction depth. A crystal block consisted of 24×12 array of lutetium yttrium oxyorthosilicate(LYSO) crystals with a dimension of 2.1 mm × 2.1 mm × 20 mm, optically coupled to the 8×8 SiPM array with a size of 3 × 3 mm and pixel pitch of 3.36 mm. To evaluate the detector performance, a flood image was obtained with Na-22 gamma source. In the flood image, 24×12 of LYSO pixels are clearly distinguished and two layers in depth are well separated. The result proved that this DOI method can be applied to the preclinical PET for high resolution imaging.

#### **[108] Development of a detector module suitable for Whole body PET with improved timing performance** *Presenter: NIKNEJAD, Tahereh*

Most whole body PET scanners today are based on the use of PMT for detecting scintillating light. In recent years, a new solid-state light sensor, the Silicon photomultiplier (SiPM), become available. SiPMs with high gain are intrinsically faster, insensitive to magnetic field, thin, cost effective and the photosensitive area is subdivided in pixels. These characteristics make SiPM a great candidate for whole body PET scanners specially for time of flight applications. Previously, we have developed a detector module based on pixelated LYSO crystal and SiPM photosensor for small animal PET and organ specific PET scanners with depth of interaction (DOI) capabilities to improve the spatial resolution using an innovative method based on the light sharing technique with low complexity and a single side readout [1][2]. However, in whole body PET systems, the most important parameter is timing resolution and the improvement in spatial resolution due to the use of DOI is less important. Previous studies showed that time difference between the signal observed in two detectors for a given positron annihilation depends on DOI of the interaction in case of long crystal. The time difference between an interaction on the near side or the far side of the photodetector is around 200 ps for a 30 mm long crystal. This time difference is significant compared to the coincidence time resolution between two crystals in coincidence about 350 ps FWHM. The aim of this study is developing a detector module based on our newly developed technique in references [1] and [2] where the DOI information is used for improving the time resolution.

The detector module is a matrix of 4x4 or 8x8 LYSO scintillator pixels, each 3x3x20 mm3 and separated by reflective foils. The crystal pixels are in one-to-one coupling to SiPM pixels and readout by PETsys TOFPET2 ASIC with TDC capability (30 ps time binning) [3]. To have light sharing between the pixels, a glass light guide is optically coupled on top (i.e. the side opposite to the SiPM) of the LYSO array and out covered by a reflective foil. The lateral surfaces of the crystal pixels are optically depolished such that the amount of light arriving at the top side will depend on the position of the interaction point along the length of the crystal. Part of the light arriving on the top side will return to the SiPM through the crystals adjacent to the one where the interaction occurred.

Using an electronic collimation set-up and a single reference crystal pixel, the detector module is scanned along its depth. Having the fitting function between the DOI and the mean value of the time difference spectrum for coincidence events between the reference pixel and the module for each pixel of the module, the DOI correction is applied on timing measurements. Moreover, the light arriving on the adjacent SiPM pixels will contribute usefully in the timing measurement. This requires the development of a maximum likelihood based combination of the timing information from different pixels. We will present the results with this detector module.

References:

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3. A. Di Francesco, R. Bugalho, L. Oliveira et al., TOFPET2: a high-performance ASIC for time and amplitude measurements of SiPM signals in time-of-flight applications, Journal of Instrumentation, 11(03), 2016.

#### [139] Temporal Imaging for PET: Coincidence Timing results on 20 mm LYSO crystals

#### Presenter: HMISSI, Mohamed Zied

In this communication we are investigating the timing performance of an original time-based imaging concept within a monolithic scintillator: Temporal imaging. Acquisitions were done with a Phillips DPC digital Si-PM matrix with a delay-time correction map pixel/pixel. By combining the light distribution and the time distribution of the first detected photons, it was possible to recognize photo-electric events from those involving a Compton diffusion. Two criteria are used to make the selection: the symmetry of the light distribution and the presence or not of a secondary light maximum within the characteristic travel time of UV light within the crystal. Only photo-electric events are used for the present version of our software to reconstruct PET coincidence event.

The first step in our processing is to estimate the radius of the undiffused photon cone on the plane of detector by excluding zones where the first photon is detected >800 ps after the first one. The number of detected photons is then at least 100, which ensures the robustness of the statistical estimation of the critical disc filled by the un-scattered UV photons. This radius is proportional to the depth of interaction (Z).

The X-Y position is then estimated using light distribution barycenter. As we use thick high optical index crystals, the time jitter linked to the difference in propagation speed between gamma rays and UV scintillation light becomes important which introduces a big error in the measurement of the coincidence resolution time (CRT) in PET system.

Time of flight (TOF) was measured in PET system using the time based imaging concept. 32x32x20 mm3 monolithic LYSO crystal were used and the full width half maximum (FWHM) of the CRT obtained from raw data was 494 ps. Time correction was used in order to improve the previous result, it consisted of removing time delay introduced by slower propagation of UV light inside the crystal that contributes to deteriorate the CRT. Compensation is done using depth of interaction previously calculated. After this correction, the FWHM of the CRT reached 228 ps.

Taking into account the time information in the localization of the scintillation event is very important, it provides a good CRT measurement in PET system allowing an accurate location of the annihilation event along the line between the two detectors.

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2. Stefan Seifert, Herman T van Dam, Jan Huizenga, Ruud Vinke, Peter Dendooven, Herbert Lohner and Dennis R Schaart , Monolithic LaBr3:Ce crystals on silicon photomultiplier arrays for time-of-light positron emission tomography, Phys. Med. Biol 57 (2012) 2219-2233.

#### [80] Breast-dedicated PET system with a personalized gantry

#### Presenter: KANG, In Soo

We developed a breast-dedicated PET system with a personalized gantry for breast cancer diagnosis. The detector module consisted of 24 × 12 array of LSO and GAGG crystals with a pixel size of 2 mm × 2 mm × 10 mm. The scanner was composed of 4 rings and each ring was arranged in polygonal shapes. The ring shape can be changed from 12-square to 10-square, 9-square and 6-square shapes with a radius of 8.9 cm, 7.3 cm, 6.9 cm and 4.15 cm, respectively. To evaluate the effect of gantry size on the system sensitivity, total coincidence count rates were simulated with 5 MBq Na-22 source by GATE code. Sensitivity was measured as 6%, 6.9%, 7.2% and 10.3% for LSO and 2.7%, 3.1%, 3.2%, and 4.6% for GAGG for 12-square, 10-square, 9-suqare and 6-square, respectively. The simulation results indicate that high sensitivity can be achieved in breast PET by an applying transformable personalized gantry.

Novel Materials (11:00-12:45)

-Conveners: Aleksander Gektin

time [id] title

#### 11:00 [202] Emerging Concepts in Organic Radiation Detection Materials

#### Presenter: FENG, Patrick

There has been renewed recent interest in organic-based radiation detection materials, owing to their unrivaled fast neutron discrimination capabilities and ability to tailor the scintillation properties for different applications. Existing examples of crystalline, liquid, and plastic organic scintillators meet a multitude of radiation detection needs but are largely characterized by compromises between performance, cost, form factor, and mechanical/environmental stability. In this talk, we will describe our recent efforts to address these compromises via synthetic control over the materials chemistry and associated photophysical properties.

Particular emphasis will be placed on two classes of materials: (1) metal-loaded plastic scintillators for combined gamma-ray spectroscopy and neutron/gamma pulse-shape discrimination (PSD), and (2) melt-cast organic glass scintillators for high-efficiency PSD. These emerging materials have been shown to overcome several fundamental limitations of conventional scintillators such as NaI(TI) and trans-stilbene single crystals, respectively. These unique advantages will be discussed within the context of ongoing challenges and potential barriers to large-scale production.

#### 11:30 **[67] Novel all-solid-state hybrid film-crystal scintillators based on the epitaxial structures of garnet compounds** *Presenter: ZORENKO, Yuriy*

# The report presents our achievement in the developments of all-solid state hybrid scintillators based on single crystalline films (SCF) and single crystals (SC) of mixed garnet compounds for registration of different types of particles and quanta in the mixed fluxes of ionizing radiations. Hybrid scintillators have been made in the form epitaxial structures containing one or two scintillators in the form of SCFs grown by liquid-phase epitaxy (LPE) method onto substrates from SC scintillators. Film and crystal parts of hybrid scintillators were fabricated from effective scintillation materials with garnet structure with significantly different luminescent spectra or scintillation decay kinetics on the basis of different combinations of SCs and SCFs of the Ce3+, Pr3+ and Sc3+ doped Lu3Al5O12 garnets as well as the SC of Ga3Ga2.5Al2.5O12:Ce garnet and SCFs of (Lu,Gd)3(Al,Ga)5O12:Ce and (Tb,Gd)3(Al,Ga)5O12:Ce mixed garnets.

The report presents the original results on fabrication of scintillators based on the materials under study in the form of SC, SCF and hybrid scintillators as well as the results of investigation of their luminescent and scintillation characteristics. We provide also the comprehensive analyzes of the luminescent and scintillation properties of Ce3+, Pr3+, Sc3+ doped Lu3Al5O12 garnets, prepared in the SCF form in comparison with their bulk SC counterparts.

The results of testing the developed hybrid scintillators in the detectors for radiation monitoring of different components of mixed ionizing fluxes ( $\alpha$ -particles and  $\gamma$ -quanta) will be presented as well.

\*\*Acknowledgements:\*\* The work was performed in the framework of Polish NCN 2016/21/B/ST8/03200 project, Ministry of Education and Science of Ukraine SF-20 F project and Czech Science Foundation 16-15569S project.

#### 11:45 [57] Discovery, crystal growth and scintillation properties of Tl-based scintillators

#### Presenter: KIM, HongJoo

Thallium-(TI)-based inorganic halide single-crystals are the potential new scintillators and our group has initiated a pioneer work on the research and development of these scintillators. For example, we have recently discovered and reported new cerium-activated-scintillators that include TI2LiGdCl6/Br6, TI2LiYCl6, TI2LiLuCl6, TI2LaCl5/Br5 [1-3], TI2LiScCl6, and TI2GdCI5. These scintillators show excellent scintillation performance, including peak emission between 370 – 450 nm, good energy resolutions (less than 5% FWHM), high light yield (more than 50,000 ph/MeV) and fast decay-time (less than 40 ns), under gamma-ray excitation. Moreover, most of our grown pure crystals show strong luminescence and high light yields due to TI ion in the host lattice. Since TI possesses high Z-number (81) and density (11.85 g/cm3), therefore, our grown scintillators also show high density (\$\rho\$ > 4 g/cm3) and high effective Z-number (Zeff > 60). Like other halide scintillators, our reported single crystals are hygroscopic and, therefore, two-zones-vertical-Bridgman-method was used for their growth. Most of the commercial scintillators using in different applications are suffering due to their low effective Z-number, low light yield and slow scintillation decay constants, therefore we expect that our discovered scintillators will perform better than the commercially-available-scintillation-detectors. Preliminary results of the discovered TI-based scintillators are very promising from applications point of view while further investigations are under way for the enhancement of the present scintillators as well as for the discovery of new compounds of this family. This report will include the progress made in the discovery of our new TI-based scintillators and also an overview of the already developed scintillators.

[1] H. J. Kim, Gul Rooh, H.Park, Sunghwan Kim, J. Lumin., 164 (2015) 86 - 89.

[2] H. J. Kim, Gul Rooh, H. Park, Sunghwan Kim, IEEE Trans. Nucl. Sci., 63 (2016) 439 – 442.

[3] H. J. Kim, Gul Rooh, Arshad Khan, Sunghwan Kim, Nucl. Intrum. Method A, 849 (2017) 72–75

## 12:00 [26] Cesium Hafnium Chloride, a Non-Hygroscopic, High-Performance Scintillator

#### Presenter: LAM, Stephanie

Cesium hafnium chloride (CHC, Cs<sub>2</sub>HfCl<sub>6</sub>) is a recently-discovered scintillator with radiation detection properties superior to the incumbent detectors Nal(Tl) and Csl(Tl). Advantages of CHC include: an excellent energy resolution, no self-absorption, no self-activity, and non-hygroscopicity. Our first-grown crystals were measured to have a light yield of 30,000 ph/MeV and an energy resolution of 3.3%. With a decay time close to 4 microseconds, CHC is well-suited to most low-count scenarios encountered in homeland security applications. This presentation will address techniques for charge purification and preparation, crystal growth by the Bridgman technique, as well as the challenges associated with handling this line compound. Finally, we will discuss the use of alloying to reduce the decay time to 2 microseconds, and to enable dual-mode neutron/gamma detection.

\*This work has been supported by the US Department of Energy, under competitively awarded contract DE-SC0015733. This support does not constitute an express or implied endorsement on the part of the government.\*

#### 12:15 [52] Neutron detection and High resolution imaging using large area 6LixNa1-xI:Eu

Presenter: MARSHALL, Matthew S. J.

Here, we report on the synthesis and application of a novel scintillator material for neutron detection: a mixed halide compound: Eu-doped 6LixNa1-x (6LNI:Eu). 6LNI:Eu screens were made using physical vapor deposition. Additionally, we have grown single crystals of the same composition to compare with the thin films. In particular, a pseudo hot wall evaporation (HWE) approach was used to increase the efficiency of materials usage, which is import enter code here ant for controlling the cost. The ability to make 6LNI:Eu scintillators into a large format is particularly advantageous for detecting specular reflections in neutron scattering and diffraction experiments. The as-deposited material exhibit a microcolumnar structure that channels the scintillation light along the length of the column, thereby reducing the lateral spread of light and increasing the spatial resolution to as high as 50 um, vastly exceeding that of other solid (non-gaseous) neutron scintillators. While our primary application is detection of neutrons, this also makes them suitable for neutron radiography. Enriched 6Li was used to increase the neutron absorption cross section of the thin films, while preserving the brightness of the scintillation response. We performed a variety of tests on the 6LNI:Eu scintillator screens, including in house X-ray measurements to provide quick feedback on brightness and resolution, as well as neutron measurements at reactor facilities. For instance, the films were tested at the High Flux Isotope Reactor (HFIR) at ORNL. For a 650 µm thick film we have measured neutron detection efficiencies as high as 58% for 4.2 Å neutrons. Another 485 μm thick film demonstrated brightness 50% higher than commercial GS20 screens, and an FWHM resolution of 320 µm when coupled to an SiPM based detector, which is the highest reported resolution for a neutron sensitive Anger Camera.

#### 12:30 [43] Scintillation properties of advanced LuAG:Ce optical ceramic materials

#### Presenter: MARES, Jiri A.

Detectors of ionizing radiation are widely used in the medical (CT, PET or PEM), homeland security fields and in scientific applications as electromagnetic calorimeters at CERN [1,2]. After 2000 year, the fast and efficient LuAG:Ce single crystal of garnet structure was developed [1] and its latest generation reached L.Y. ~27000 ph/MeV [2]. LuAG:Ce crystals grown by the Czochralski method from high temperature melt (~2020 oC) do show deteriorated scintillation performance due to "antisite defects" (AD) constituted by the Al3+ ions at Lu3+ lattice sites [1]. On the other hand, transparent LuAG:Ce optical ceramics prepared by a solid state reaction method with the use of MgO sintering aid do not usually show these defects [2,3]. The sintering of ceramics is provided in vacuum on isostatically pressed samples at temperature around 1800 oC but no liquid phase arises during the sintering. These ceramics can reach (i) better homogeneity of dopants (or codopants) and also (ii) larger, tailored size resulting in less expensive technology [3]. The development of LuAG:Ce optical ceramic materials started in the first decade of 21st century. Nowadays, its highest prompt (shaping time less than 2 µs) L.Y. ~25000 ph/MeV was observed [2], but the highest values (shaping time 10 µs) of L.Y. ~ 30700 ph/MeV was observed on LuAG:Ce,Ba,Mg ceramic, ~29000 ph/MeV at LuAG:Ce,Li and LuAG:Ce,Ba ceramics and ~28000 ph/MeV at LuAG:Ce,Mg. The latter ceramic sample exhibits the best photopeak energy resolution ~5 % at 662 keV [2] while other ceramics exhibit resolution around 8 % comparable to that of LuAG:Ce crystal. Scintillation decays of the LuAG:Ce ceramic samples (with codopants and nonstoichiometric ones) consist of fast and slow decay component parts and the best nonproportionality (~a decrease to 80 % at 10 keV from 100 % at 662 keV) is found at LuAG:Ce nonstoichiometric ceramics. Results observed on various LuAG:Ce ceramics will be discussed under the model presented in [2] where the presence of Ce3+ and Ce4+ was verified from XANES spectra but various defects can be present due to Mg2+, Li+ and Ba2+ codopants.

We will present a review of scintillation properties (L.Y's, energy resolutions, non-proportionality and scintillation decays) of LuAG:Ce ceramics prepared from (i) stoichiometric or nonstoichiometric (both an excess or deficiency of Lu) compositions, (ii) with Mg2+codopant and (iii) with other codopants as Li+ and Ba2+.

[1] M. Nikl, A. Yoshikawa et al. Prog. Cryst. Growth Charact. Mater. 50 (2013) 41-72

[2] S. Liu, J.A. Mares, X. Feng et al. Adv. Opt. Mater. 4 (2016) 731-39.

[3] S. Liu, X. Feng, Y. Shi et al. Opt. Mat. 36 (2014) 1973-77.

#### lunch (12:45-14:30)

#### Scintillation Mechanisms: session 1 (14:30-16:00)

#### -Conveners: Richard Williams

#### time [id] title

#### 14:30 [188] The role of excitation distribution in the intrinsic resolution

#### Presenter: VASILYEV, Andrey

Non proportionality (NP) of scintillation response is usually used as an explanation of intrinsic resolution change, nevertheless there are no direct links between definitions of these scintillation parameters. At [1] the wide spread between LY and energy resolution (ER) was pointed for the same scintillator (NaI:TI). The literature data are between 5 and 12% resolution that is out of the LY spread! It is important to note that NP significantly depends on crystal purity, co-doping, integration time and so on [2, 3]. In other words it is necessary to assume that ER depends on the scintillator structure as well nevertheless there are no any "structure parameters" in ER definition (only statistical parameters).

These experimental data allows to assume that not all scintillation events have the same contribution to the PMT registered statistics, and it is necessary to find the core for such difference. Moreover, the decay kinetics also fluctuate significantly. These two factors approve the supposition that the main reason for such effects is the fluctuation of spatial distribution of electrons, holes and activators in the track region. In order to demonstrate this link we analyze electron-hole, electron-electron, hole-hole, electron-activator and hole-activator correlation functions just after the production of themalized excitations. This approach could also estimate the role of electric fields created due to the separation of electrons and holes. Fractal correlation dimensions of these functions shows the change of the track structure for different scales (e.g. 3D spherical for small distances where the clusters of excitations do not overlap, 1D cylindrical one for the case of overlapped clusters (intermediate distances) and 0D for distances comparable with the track length). The account for energetic Auger- and delta-electrons result in the change of this fractal correlation dimensions. The structure of these correlation functions determines different scintillation decay kinetics and corresponding yield at different stages of the scintillation and from different parts of the track.

It is important that "scale factor" based on the e-h separation and later thermalization for alkali halide scintillators is more significant than of alkali earth halides that explains the better ER for the latter materials. This is an additional (to the activator uniformity) factor at the predictable energy resolution description that has to looks like the new step to conventional NP and ER models.

#### References

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# 14:45 [88] On the impact of the nano-scale fluctuations of electronic structure in solid solutions on the scintillating properties

#### Presenter: BELSKY, Andrei

Many mixed crystal scintillators have been investigated during last years. It appears that tuning the composition of a mixed crystal allows to increase the scintillation yield, decrease the afterglow, and improve the energy resolution.

Some properties of mixed crystal scintillators exhibit a non-linear concentration dependence, which differs from the linear Vegard's law. We interpret these non linear behaviors as the effect of the modulation of electronic structure by the fluctuation of spatial distribution of the substitutional ions in the crystals along 3 typical behaviors.

The simulation of ionic crystal solid solution allows us to extract several conclusions concerning the influence of the affinity between the substitutional ions on the observed scintillation properties. Without any affinity between the substitutional ions, the distribution of them is non-correlated and the maximum of the spatial fluctuations of the energy gap (bottom of the conduction band in case of substitution in the cationic sublattice) is achieved at a concentration of 50%. Such fluctuations result in the decrease of the mean free path for elastic above-barrier scattering of electrons, and therefore thermalization length for electrons decreases. When electron energy becomes lower than the height of fluctuations of the bottom of the conduction band, the localization can occur. This increases the probability of geminate recombination and increase the fraction of fast luminescence.

On the opposite, the account of the affinity between the cations substantially alters their distribution and the properties of the mixed crystal. In presence of affinity AA and BB, nanophase separation effects are already observed at concentration about 10%. Nanoclusters enriched with one type of the substitutional ions are formed. The clusters have sizes about a few lattice constants. The fluctuations of the potential become deeper and wider, which facilitate the localization at lower concentration of one of the components, about 30% (70%). This allows suggesting that in scintillator solid solutions showing a maximum LY at these concentrations, the affinity plays a major role. The absorption coefficient in this case is characterized by rather long tail in the transparency region.

The third behavior occurs in presence of cross affinity between cations (AB). In this case, the cation distribution is almost uniform at each concentration. The spatial variation of the energy gap is small; it is much narrow than at a random distribution. In such crystals the energy gap varies in proportion to the concentration (Vegard's law). There are practically no new localization levels, due to the small change of the potential at the non-uniformities.

In this presentation, we discuss the combination of theoretical calculations and experimental evidences to confirm this interpretation.

#### 15:00 [184] Can self-trapped excitons deliver energy to impurities in tungstates?

#### Presenter: NAGIRNYI, Vitali

Tungstates are widely used as scintillator materials in security, medicine and particle physics devices (e.g., CaWO\$\_4\$, CdWO\$\_4\$, PbWO\$\_4\$). Their emission is represented by a broad band in the visible spectral region, which is usually ascribed to the radiative decay of self-trapped excitons (STEs) localized at oxyanion complexes. Due to the large energy of optical phonons and strong electron-phonon interaction in these systems hot electrons and holes created by high-energy quanta of ionizing radiation are believed to relax and form STEs very quickly, thus preventing energy from being transferred to defects and impurities. Despite tungstates of divalent metals have been popular research objects for decades, the basic physics features of their excitations, capable of confirming their excitonic nature, have not been revealed. No papers have been systematically dealing with this question except Ref. [1]. It is not known whether these excitations can be mobile or transfer energy to impurity ions. It is not even clear what is the reason of the thermal quenching of STE emission, i.e. is it thermal ionization, non-radiative recombination or hopping diffusion and energy transport. Here, we present the results of the studies on the STEs capability of transferring energy to impurity ions in tungstates.

The measurements were performed mainly on CdWO\$\_4\$:Sm, which seems to be especially suitable for such study due to the availability of bridging oxygens connecting neighboring oxyanions in the crystals structure. The experiments were conducted at the I3 beamline of the MAX III storage ring, Lund, Sweden and laboratory spectroscopy setups at the Institute of Physics, University of Tartu. The dependence of excitation spectra of the STE and impurity emissions on temperature was measured in a wide energy region from 3 to 40 eV. A special attention was paid to the energy regions of intra-centre impurity excitation, direct exciton creation in the Urbach tail and the creation of excitons via electron-hole recombination in the fundamental absorption region. The measurements of emission decay kinetics were performed in a wide range of 4-600 K in order to reveal the mechanisms responsible for the energy transfer to impurity ions.

It is shown that while the intensity of the Sm\$^{3+}\$ emission remains practically unchanged under the intra-centre excitation in the whole temperature range studied, it undergoes a remarkable evolution with temperature under excitation in the excitonic and fundamental absorption regions. In these energy regions, the intensity of the Sm\$^{3+}\$ emission remains modest at low temperatures due to the competitive process of STE formation, grows drastically in the region of thermal quenching of the STE emission and thereafter the impurity emission disappears almost completely. The former effect is ascribed to the enhanced mobility of STEs due to hopping diffusion at elevated temperatures, while the latter - to the ionization of excitons at temperature near 500 K, which exceeds the STE emission quenching temperature by 100 K. The temperature dependences are different in the excitonic and fundamental absorption regions. A detailed analysis of the mechanism of energy transfer by STEs will be presented based on the results of time-resolved spectroscopy studies.

[1] K.W. Meert et al., J. Lumin. 173 (2016) 263.

## 15:15 [191] A Theoretical First-principles Investigation of the Properties of Self-Trapped Excitons and Defects in Halide Scintillators

#### Presenter: CANNING, A.

The performance of new and improved materials for gamma ray scintillator detectors is dependent on multiple factors such as quantum efficiency, energy transport etc. In halide scintillator materials the energy transport is often impacted by both intrinsic hole and electron traps such as Vk centers and their associated self-trapped excitons (STE) as well as traps associated with defects and impurities. Recently there has been enormous progress in the development of quantum mechanical methods that allow us to investigate quantitatively these mechanisms. Here we present first principles calculations at the hybrid density functional theory level for the structure, mobility and optical properties of self-trapped excitons in three important families of scintillator materials, the alkali metal, lanthanum and barium halides. Alkali metal and lanthanum halides have been extensively characterized from an experimental point of view and serve in our studies as reference systems to assess the accuracy and reliability of our theoretical procedure. We show that hydrid density functional theory level for self-trapped excitons (on and off-center) found in these materials in agreement with EPR experiments. We present results of accelerated molecular dynamics to determine the migration pathways of excitons followed by nudged elastic band method calculations based on the migration pathways to determine the energy barriers to migration. This methodology was then used to perform studies of these defects in new scintillator materials including the barium mixed halides that we will compare to new experimental results.

This work is supported by the Department of Energy, National Nuclear Security Administration, Office of Defense Nuclear Nonproliferation Research and Development and carried out at Lawrence Berkeley National Laboratory under contract #AC02-05CH11231. This work does not constitute an express or implied endorsement on the part of the government

# **15:30 [141]** The electronic structure of Tl, Pb, and Bi based scintillators and how that relates to scintillator performance *Presenter: DORENBOS, Pieter*

The Sr co-doped LaBr3:Ce3+ scintillator is an almost ideal scintillator regarding energy resolution and speed. However, its relatively low density limits application in various fields. To develop scintillators with density higher than 8 g/cm3 one has to abandon the research field of the halides, and instead explore oxides containing high Z-cations like Hf, Ta, W, Tl, Pb, or Bi. The question then arises whether Ce3+, Tl+, Pb2+, or Bi3+ will scintillate in those type of compounds? To answer this, one needs to know the electronic structure. Where are the activator excited and ground state levels with respect to the host valence band and conduction band? The chemical shift model, that was developed in 2012 [1], enables to determine the electron binding energy in lanthanide impurity levels with respect to the vacuum. Recently we applied the model to obtain information on the vacuum referred binding energies (VRBE) of Tl+, Pb2+, and Bi3+ in luminescent phosphors [2]. An overview of the results will be presented, and we will explain why Ce3+ does not scintillate in Pb-, Bi-, Ta-, W-based compounds, and why it does scintillate in Tl2LaCl5:Ce [3] and BaHfO3:Ce. We will also address the possibility for Bi to luminesce in Pb- or Tl-based compounds or Pb2+ in Tl- or Bi-based compounds.

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[2] R.H.P. Awater, P. Dorenbos, J. of Lumin. 184 (2017) 221.

[3] H.J. Kim, Gil Rooh, Sunghwan Kim, J. of Lumin. 186 (2017) 219.

## 15:45 [102] Picosecond absorption spectroscopy of self-trapped holes, self-trapped excitons, and transient Ce states in LaBr\_3 and LaBr\_3:Ce

#### Presenter: LI, Peiyun

We report excitation-induced picosecond absorption over an extended spectral range from 320 nm to 2700 nm in LaBr\$\_{3}\$, LaBr\$\_{3}\$:Ce(4%) and LaBr\$\_{3}\$:Ce(20%). Preliminary identification of a V\$\_{k}\$ band near 3.6 eV, self-trapped exciton (STE) hole transition near the same energy, and STE bound-electron transitions spanning 0.46 eV to 1.1 eV have been made. Comparison to recent calculations of V\$\_{k}\$ and STE structure in LaBr\$\_{3}\$ by A. Canning and M. Del Ben [private communication] is helpful and in rough agreement on several points. We also observe transient induced absorption bands whose strength increases with Ce concentration, and they are tentatively attributed to carrier capture and/or excited states involving Ce. Strong Ce-correlated transient absorption bands are found at 2.8 eV, 2.2 eV, and with weaker Ce correlation at about 1.25 eV. We are working to establish identification with expected charge-transfer (CT) electron and hole transitions of the Ce\$^{3+}\$\* excited activator and CT transitions of Ce\$^{4+}\$ activator-trapped holes. Together with the identified STH and STE transitions noted above, these should constitute the main species in LaBrs\_{3}:Ce scintillation. The excitation in these experiments is two-photon absorption of 300 fs pulses producing total transition energies of 5.9 eV, only slightly above the band gap of LaBr\$\_{3}\$, and 8.86 eV, capable of creating hot electrons with almost 3 eV excess energy. Use of the corresponding two pump photon energies at 2.95 eV and 4.43 eV allows distinguishing effects of direct absorption of the pump photons by Ce dopant. Assembling information on the picosecond-scale sequential populations of trapped carriers and excited states that are main participants in scintillation, along with quantitative rates of capture, is necessary for a material engineering model of LaBr\$\_{3}:Ce. The present work follows our similar picosecond-measurement program in CsI, CsI:TI [1] which supplied a number of key rate coefficients for successful modeling of pulse shape, proportionality of decay components, and light yield in CsI:TI [2].

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#### **Coffee Break** (16:00-16:30)

#### Characterization: session 2 (16:30-18:15)

-Conveners: Pieter Dorenbos

time [id] title

## 16:30 [27] High Performance CLYC-PVT Composite Scintillators for Neutron/Gamma Detection

#### Presenter: LAM, Stephanie

While Srl<sub>2</sub>(Eu)'s excellent scintillation performance and CLYC(Ce)'s dual mode detection capability make these halides ideal candidates for nuclear radiation detection, these materials are expensive due to decreased crystal growth yield at large diameters (e.g. 3 inches). However, the cost of encapsulating small diameter crystals into a large plastic matrix such as polyvinyltoluene (PVT) is fairly small, and the plastic matrix can even shaped to improve light collection. Thus, composite scintillators hold the promise of providing high performance low-cost gamma/neutron detectors in sizes that would be otherwise prohibitively expensive or impossible to produce. This presentation will discuss the design, fabrication, and performance of our CLYC-PVT and Srl<sub>2</sub>(Eu)-PVT composite scintillators. Fabrication of these composites were guided by GEANT4 simulations to characterize the generation, transport, and collection of photons, and their dependence on surface conditions and index of refraction. Our first 2-inch diameter CLYC-PVT achieved an energy resolution of 4.4% at 662 keV and a PSD of 3.2. More recently, we demonstrated the feasibility for 5-inch diameter composite fabrication. Other important considerations for composite fabrication will be discussed.

\*This work has been supported by the US Department of Homeland Security, Nuclear Detection Office, under competitively awarded contract HSHQDN-15-C-00013. This support does not constitute an express or implied endorsement on the part of the government.\*

#### 16:45 [131] Scintillation properties of pure YAG crystals

#### Presenter: SHIRAN, N.

Last studies [1-3] demonstrated high efficiency of UV luminescence of undoped yttrium aluminum garnet (YAG). Radio-luminescence of pure crystals most probably related to excitons localized around defects, due to violation of the stoichiometric composition. At the same time there are different types of defects presence in the lattice. Pure YAG crystals emit a broad UV emission band under high-energy excitation, which is associated with the presence of anionic and cationic vacancies, their complexes and antisites. Which of them play the positive or negative role is still not clear that makes a problem with optimization of scintillation performance and potential application of pure YAG.

This study is directed to reveal the defects related to scintillator performance of pure YAG single crystals. The main goal is to evaluate the type of optimal defects presence for efficient scintillation.

The variable types and concentration of defects, which correspond to the absorption bands in the range of 190 - 400 nm, was estimated. It was found that the undoped crystals excited with X-ray demonstrate the broad complicated emission band in UV range with maximum at around 300 nm. The experimental results point out that some native structure defects and impurity' traces plays the negative role in scintillations. Iron and carbon ions presence is displayed by absorption, visible and NIR photoluminescence and high temperature TSL. Infrared absorption spectra provide the information about hydroxyl and carboxyl groups incorporation in the structure.

It was shown that the emission is enhanced in samples with the best optical transparency in UV and IR regions as well as the negligible contribution of additional luminescence. The intensification of intrinsic UV luminescence is only due to suppressing these trapping defects.

Factors affecting the scintillation efficiency in YAG are discussed. The best scintillation performance can be modified with purity and intrinsic defects change. The light output for such scintillators can reach the level of BGO or pure CsI scintillators and is about the same value for Ce-doped YAG crystal. Decay time consists of 7 and 460 ns components, which agrees the data resulted in [3]. The rate between these component and the optimal conditions are still under investigation.

The first data show that scintillation properties of pure YAG are reasonable for the use of this crystal for different applications. Modification of defects structure allows to reach better scintillation performance comparing to activated YAG.

[1] F.A.Selim, C.R.Varney et al., Rev.Sci.Instr. 83 (2012) 103112; AIMS Mat. Sci. 2 (2015) 560.
 [2] N.Shiran, A.Gektin, K.Gubenko et al., Funct. Mat. 23 (2) (2016) 191.
 [3] Y.Fujimoto, T.Yanagida, H.Yagi et al., Opt. Mat.36 (2014) 1926.

#### 17:00 [210] Comprehensive study on La-GPS scintillator

#### Presenter: HORIAI, Takahiko

Recently, Ce:(La, Gd) $$_2$Si$_2$O$_7$ (Ce:La-GPS) scintillator was reported the have a good energy resolution (FWHM) of ~5% at 662 keV, and its light output remained constant up to 150 $^{\circ}$C (423K) [1,2]. Moreover, we grew larger size Ce:La-GPS crystals by Czochralski process up to 2 inch diameter and studied their scintillation properties. Up to now, we have shown the scintillation properties of fixed La-concentration, and we have never revealed the La-concentration dependence of some properties such as bandgap energy, light output at high temperatures, rising time, etc. In this paper, we show the Comprehensive study on La-GPS scintillator with a La concentration of 20 – 50% in the Gd site. Ce-doped and pure Ce:(La$_x$, Gd$_{1-x}$)$_2$Si$_2$O$_7$ crystals were grown by the micro-pulling down method from the starting materials: 99.99% pure Gd$_2$O$_3$, La$_2$O$_3$, SiO$_2$ and CeO, where x = 0.2 to 0.5. The crystals were confirmed to have single phase using X-ray diffraction patterns.$ 

For Ce-doped samples, after cutting and polishing the samples, transmittance and photo-luminescence (PL) excitation and emission spectra were measured using a spectrophotometer (V-530, JASCO) and a spectrofluorometer (FLS920, Edingurgh Instrument: EI) with Xe lamp (EI, Xe-900), respectively. The pulse height spectra at the high temperature up to 200 \$^{\circ}\$C (473K) were measured with a ruggedized PMT (Hamamatsu R1288AH) in order to evaluate the temperature dependence. To evaluate the bandgap energy, transmittance spectra of pure La-GPS samples were measured at low temperature (7K) using a beam-line at a synchrotron faculty, Ultraviolet Synchrotron Radiation Facility (UVSOR), in Japan.

We found that both samples had almost the same emission and excitation spectra and good light output over 150\$^{\circ}\$C (423K). The band gap energies did not depend on the La-concentration, and the energies were estimated to be around 7.1 – 7.2 eV. We show the above results, rising time for the samples and the temperature dependence of light output or intensity from 7 to 500K in this presentation.

A. Suzuki, S. Kurosawa, and A. Yoshikawa et al., Appl. Phys. Express 5 (2012) 102601.
 S. Kurosawa, and A. Yoshikawa et al., NIMA 772 (2015) 72.

#### 17:15 [63] Li-Be-Si-Ce scintillation glass and glass ceramics with moderate properties

#### Presenter: KORJIK, Mikhail

A family of lithium silicate glasses and glass ceramics doped with Ce ions show high light yield under thermal neutrons [1]. There are several lithium containing scintillation glasses available on the market. The most widely applied scintillation glass is GS-20 type glass, which has a complex Si-Al-Li-Mg-Ce composition. This composition is hardly used to obtain glass ceramics, which has obvious advantages over an amorphous glass [2]. Glass ceramics combines the luminescent properties of rare-earth ions in crystallites and remaining mother glass. Efficiency of the neutron detection with lithium silicate glass is defined by 6Li neutron cross-section and its content in the glass. Due to this reason scintillation glass is an attractive material to detect thermal neutrons. Effective detection of epithermal and fast neutrons with 6Li glass requires application of a moderator, enriched with such nuclei as carbon, beryllium etc.

Here we report on scintillation properties of the light scintillation glass combining Li and Be ions. Be neutron cross-section predominantly is formed by scattering and is one of the largest among light nuclei, so epithermal and fast neutrons are effectively slowed down in such glass, which increases an efficiency of their capturing by neighbor 6Li nuclei. Li-Be-Si glass containing 20 mol.% of Li2O and BeO was prepared according to the technological approach described in [2]. In the present work, the photo- and radio-luminescence properties of the Li-Be-Si glass, activated by Ce ions, and with different Li/Be ratio were studied. The best achieved light yield at maximal Be content in the glass was measured to be 5000 photons/neutron. Results of the simulation of the neutron detection with the developed glass in a wide energy range are also discussed.

1. A.R. Spowart, Neutron scintillating glasses: part I. Activation by external charged particles and thermal neutrons. Nucl Instr Meth Phys Res 135(1976)441–453

2. P.Lecoq, A.Gektin, M.Korzhik, Inorganic Scintillators for Detector Systems, Springer, 2017, P.408

# 17:30 [97] Study of the glass and glass ceramic BaO\*2(SiO2):Ce (DSB: Ce) scintillation material for high energy physics application

#### Presenter: DORMENEV, Valery

The development of new crystalline materials for ionizing radiation detectors is still playing a significant role in applications in high energy physics (HEP). Further concepts of the detectors at HEP experiments will require an unique combination of the material features, particularly in case of collider experiments. A possible candidate can be the so called DSB: Ce glass ceramics obtained from the BaO\*2(SiO2). The transparent glass ceramics contains nano-sized particles of Ba2SiO5 which improve the scintillation properties of the resulted material. A systematic study of small volume (not more than 1 cm3) DSB: Ce material has been reported in [1,2]. On the other side, DSB: Ce glass ceramics heavily loaded with Gd can become a candidate for neutron detectors. Crucially important is a minimal level of radiation damage under the electromagnetic part of ionizing radiation and energetic hadrons as well: low deterioration of the optical transmission, low level of afterglow and low level of radio luminescence due to radio-nuclides being generated in nuclear reactions within the material of the detector. This report will focus on the investigations on scintillation properties of first bulk samples with 4 cm thickness made of DSB:Ce as well as DSB:Ce heavily loaded with Gd. We have measured the light yield and the optical transmittance of both DSB types before and after irradiations with 1.2 MeV gamma-quanta and 190 MeV protons, respectively.

1. K.-T. Brinkman, et al., Radiation Damage and Recovery of Medium Heavy and Light Inorganic Crystalline, Glass and Glass Ceramic Materials after Irradiation With 150 MeV Protons and 1.2MeV Gamma-Rays, presented at IEEE NSS 2014, Seattle, WA, USA, 8-15 November 2014.

2. A. Borisevich, et al., Optical transmission radiation damage and recovery stimulation of DSB: Ce3+ inorganic scintillation material, 2015, J. Phys.: Conf. Ser. 587, 012063.

#### 17:45 [65] Radiation hardness of Rare Earth doped sol-gel silica fibers for High Energy Physics Detectors

#### Presenter: COVA, Francesca

In recent years, the sol-gel technique was proven to allow a good control, at a relatively low densification temperature, of rare earth (RE) ions incorporation and of their dispersion inside the glass matrix [1]. The glass synthesis can be performed by using high purity precursors, reducing the level of unwanted impurities, which is an essential feature for the radiation hardness of such materials. Several studies demonstrated that RE-doped silica glasses prepared by sol-gel route are suitable materials for the realization of scintillating optical fibers [2], and have application perspectives as wavelength shifters for the collection and transport of scintillation light in High Energy Physics (HEP) experiments. Moreover, the use of RE-doped fibers as scintillators in HEP detectors, possibly in parallel with undoped fibers exploiting Cherenkov light [3], has been recently proposed. An extremely good radiation resistance and fast response are crucial properties for such application.

In this work we present a detailed study of the scintillation properties of SiO\$\_{2}: 0.05 mol% Ce glasses and the results of irradiation tests using gamma-rays from a \$^{60}\$Co source and X-rays up to an integrated dose of 1 kGy.

Radio-luminescence investigations have been combined with optical absorption and attenuation length measurements before and after irradiation with X-rays and with \$^{60}\$Co gamma-rays. Comparisons between bulk preforms and fibers have been carried out, in order to disclose the role of the fiber drawing process in the radiation hardness properties. Fibers with a lower (0.0125 mol%) Ce concentration have also been considered, pointing to a reduction of the radiation damage related to the decrease of the dopant concentration. Fibers with fluorinated glass or polymeric cladding have been compared, looking towards a future engineering of the fiber structure to improve the light propagation and the radiation resistance.

The evolution of the optical absorption spectra and of the attenuation length as a function of time after irradiation has been investigated in order to understand the room temperature stability of radiation-induced point defects acting as color centers. Moreover, the samples have been subjected to thermal annealing cycles, to check the temperature activated carrier release from radiation-induced defects and the possibility of a complete recovery of the damage.

Further analyses on the properties of sol-gel silica fibers have been carried out: the homogeneity of the Ce distribution along the fiber length has been tested by means of the X-ray fluorescence technique.

Eventually, we will report the results of the forthcoming test of Ce-doped fibers, assembled in a calorimeter prototype, with GeV electron beams at the CERN test beam facilities, to better investigate the application perspectives of such kind of material as scintillator in High Energy Physics detectors.

This work has been supported by the H2020 projects AIDA-2020 (GA no. 654168) and INTELUM (GA no. 644260).

[1] A. Vedda et al., Chem. Mater, 18, 6178, 2006

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[3] A. Penzo et al., Journal of Physics: Conference Series, 160, 012014, 2008.

IAC meeting (19:00-22:00)

## Wednesday 20 September 2017

Nanomaterials (08:30-10:00)

#### -Conveners: Christophe Dujardin

#### time [id] title

## 08:30 [189] Colloidal quantum dots design for scintillation applications

#### Presenter: MAHLER, Benoit

The synthesis of colloidal nanocrystals has greatly improved over the past 25 years. Semiconductor nanocrystals –the so-called quantum dots– can currently be synthesized in a wide range of sizes, shapes, crystal structure and compositions, allowing tuning their emission properties from the UV to the deep infrared. Furthermore, nanocrystal heterostructures of numerous geometries such as core/shell nanoparticles, dot-in-rod structures or core/crown nanoplatelets open up still new possibilities to tune the optical properties of these semiconductor nanoparticles.

For scintillation applications, conversion yield as well as decay times are of particular importance. In the case of quantum dots scintillation, these parameters are strongly related to structural parameters such as the use of a core/shell structure, the band alignment between the core and the shell, and the size of both components.

In this presentation I will first present the usual strategies used in colloidal quantum dot syntheses to control the wavelength, fluorescence lifetime and quantum efficiency.

I will then discuss multi-charges phenomena arising in quantum dots: Auger assisted recombination and multiexciton generation. These effects are related to physical behaviors such as blinking and have been widely studied over the years in an attempt to suppress or control them.

Finally, different strategies to create quantum-dots in matrix systems will be analyzed, highlighting the potential uses of such composite materials, their advantages and drawbacks for scintillation applications.

#### 09:00 [114] Radiation synthesis of highly luminescent nanoscintillators with fast decay

#### Presenter: ČUBA, Václav

Direct radiation syntheses of nanoclusters of various metals or metal alloys have been studied for a few decades [1]. However, only recently the method was systematically used for preparation of simple or multicomponent metal oxides [2]. The procedure is based on irradiation of aqueous solutions containing soluble precursors by UV or ionizing radiation. Finely dispersed solid phase formed during irradiation is subsequently separated from solution, dried and either used directly, or further processed by annealing at higher temperatures under various atmospheres.

Nanopowder scintillators prepared via radiation method typically show good luminescent properties [3], as the processes initiated by radiation have some advantages over common chemical methods: they are mostly independent of temperature, they impart high level of interaction between individual components in precipitated precursor and they yield materials of high purity, with relatively narrow size distribution of particles and minimum of crystal defects.

This contribution summarizes various techniques for radiation or photochemical synthesis, that have been successfully used for preparation of numerous nanopowder scintillators, namely simple and multicomponent metal oxides, synthetic garnets, core-shell systems, quantum dots, heterostructures and nanocomposites. Such nanoscale powders can be further processed by ceramisation or by embedding into an optically transparent matrices.

Among prepared nanopowders, ZnO:Ga and synthetic garnets play prominent role, each representing a different group of materials. ZnO:Ga based scintillators show intense excitonic luminescence located at 390-400 nm with ultra-fast sub nanosecond decay. Their bandgap can be further modulated by introducing 5-20% of Cd or Mg ions into the crystal lattice. Defect related luminescence typical for ZnO can be completely suppressed by annealing in reducing atmosphere. Synthetic garnets, based on YAG or LuAG and doped with various ions also feature very high intensity of luminescence when compared to BGO standards and minimum defect related luminescence. Their high thermal and chemical stability and simple cubic structure make them good candidates for preparation of optically transparent ceramics or cores of PDTX drugs.

#### Acknowledgement

This research has been supported by the Czech Science Foundation grant GA 17-06479S.

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[2] V. Čuba, J. Bárta, V. Jarý, M. Nikl. Radiation-induced synthesis of oxide compounds. In: B.I. Kharisov, O.V. Kharissova, U.O. Mendez (eds). Radiation synthesis of materials and compounds. CRC Press, Boca Raton, 2013, pp 81-101.
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#### 09:15 [91] Spectroscopic Properties of Scintillating Hafnium Dioxide Nanocrystals

#### Presenter: VEDDA, Anna

In the last decade, many efforts have been devoted to the development of smart multifunctional materials. Among them, inorganic nanostructures have gained importance because of their outstanding luminescence properties and their potential applications as new building block materials for the next generation electronics and in a variety of lighting applications. In particular, many recent researches have been focused on the achievement of progress in the synthesis of nanosized metal oxides. In this field, a significant attention is paid to hafnium dioxide (hafnia or HfO2), which can be employed in optical protective and thermal barrier coatings. Thanks to its mechanical resistance, hafnia finds applications as ceramic, super hard materials and catalysts, or as component in gas sensors and fuel cell electrolytes. Lastly, HfO2 is now evaluated as potential alternative gate dielectric to replace SiO2 in the future generation of electronic nanodevices. Regarding the scintillating properties, the high atomic number Z=72 and the quite high density (9.6 g cm-3) make HfO2 nanocrystals good hosts for phosphor and scintillating applications where a large stopping power for ionizing radiation (X-rays, y-rays) is required (1).

Bulk hafnia is very difficult to grow due to its high melting point (2774 °C). Actually, HfO2 can be synthetized also in the nanocrystals form and studied to fabricate thin films, optical ceramics and nanocomposite materials (2). Indeed, besides the well-known luminescence of HfO2 nanocrystals activated by the incorporation of rare earth ions, some recent studies evidenced the occurrence of a blue fluorescence from undoped nanocrystals upon UV excitation; remarkably, a broad bluish luminescence appears upon X-ray illumination (RL). These findings suggest the potential of HfO2 nanocrystals as radiation detectors, but the lack of a detailed model that relates their structural and RL properties still hinders the development of efficient nanoscintillators with optimized structure and chemical composition.

We studied the RL features of undoped monoclinic HfO2 nanocrystals and their dependence on the structural properties of the material at the nanoscale in order to elucidate their origin. Upon X-ray irradiation, the nanocrystals show six emission bands in the near UV/visible spectral range, detectable between 10 K and 300 K. The visible luminescence bands at 2.2 eV, 2.5 eV and 2.8 eV are similar to those detected in our previous PL studies (3), while the UV emission at 4.2 eV and 4.6 eV have been observed for the first time. The excitonic behavior of the UV luminescence is evidenced. The strong increase of the 2.5 eV blue luminescence in annealed samples is likely related to the presence of titanium and it might be used for the design of highly efficient blue scintillating materials.

1. LeLuyer C. et al. (2008), HfO2:X (X = Eu3+, Ce3+, Y3+) Sol Gel Powders for Ultradense Scintillating Materials. J. Phys. Chem. A, 112, pp. 10152-10155.

2. Lange S. et al. (2006), Luminescence of Re-Ions in HfO2 Thin Films and Some Possible Applications. Opt. Mater., 28, pp. 1238-1242.

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#### 09:30 [41] Purcell-Enhanced scintillating properties in metal:ZnO nanostructures

#### Presenter: ZHU, Renyuan

Recent advances in nanotechnology enable novel nanomaterials for developing new scintillators. Outstanding scintillator materials require high absorption cross-section, fast response and robust radiation tolerance and good scalability. ZnO with fast decay time (<1ns) was proposed for ultrafast X-ray imaging. However, some applications require extremely high frame rate in the GHz range. We proposed to develop and perform some initial experiments on new types of ZnO based nanocomposites to reduce its response time. The nanocomposites consist of metallic nanoparticles such as Au, Ag embedded in ZnO films. To guide our experimental efforts, we first performed simulations on different metal/ZnO nanostructures. We have compared the effects of the metallic nanoparticle sizes, nanoparticle types and distances between the nanoparticles and ZnO structures. Simulation results show that Purcell effect, or shorter radiation time can be achieved by introducing of metal nanoparticles. We have fabricated Ag:ZnO nanocomposite films with Ag nanoparticles in ZnO film matrix have been grown on glass substrates by using pulsed laser deposition and spin-coating methods. To maximize the Purcell effect, we have systematically controlled the diameter and the density of Ag nanoparticles in nanocomposites by the growth temperature and thermal annealing. The structural properties have been characterized by X-ray diffraction and scanning transmission electron microscopy. We have investigated the scintillating properties including the decay time and photoluminescence using various charged particle and light sources.

# 09:45 [223] Growth and Characterization of InGaN/GaN Multiple Quantum Well Structures used for Scintillation Detectors

Presenter: HUBÁČEK, Tomáš

Growth and Characterization of InGaN/GaN Multiple Quantum Well Structures used for Scintillation Detectors

Tomáš Hubáček1, Alice Hospodková1, Jiří Oswald1, Jiří Pangrác1, Vítězslav Jarý1, Tomáš Parkman2, Dalibor Pánek2, Gilles Ledoux3, Christophe Dujardin3 and Martin Nikl1

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High-luminosity scintillators with fast decay time are crucial in many applications. These scintillators are a key component for improved time-of-flight mass spectrometers and could be used as detectors in synchrotrons or particle colliders. Fast scintillators are necessary in scanning electron microscopes (SEM) for industrial inspection of nowadays electronics. Generally, the nitrides are perspective candidates due to the strong exciton binding energy (short decay time in order of nanoseconds) and good radiation resistence. GaN and related ternary alloys with indium have been used widely in optoelectronics devices. On the other hand, GaN can be used as a scintillator material due to high light yields (105 Photons/MeV) and very short decay time (below 1 ns for its near-band-edge transitions) [1].

With the epitaxial growth, GaN can be prepared in high crystallographic quality. Perfect homogeneous epitaxial layers result in better emission homogeneity over large area, lower nonradiative losses and suppressed inhomogeneous broadening of the emission profile. Due to 1-D quantum confinement the GaN scintillation efficiency can be significantly enhanced by employing InGaN/GaN multiple quantum well structure. On the other hand, thin epitaxial layers have low planar light extraction coefficient. These structures are very similar to structures used for UV-visible light emitting diodes. There are two main differences, no p-n junction is necessary and much thicker active region (higher QW number), according to the electron penetration depth, is needed [2].

We optimized InGaN/GaN heterostructures (grown with MOVPE) with different growth parameters to get strong excitonic luminescence and fast response in the nanosecond range. We observed influence of the QW thickness on the decay time. Structures with thinner quantum wells had shorter decay time due to the higher electron hole wavefunction overlap. Photo-, radio- and cathodoluminescence were used for characterization of our structures as well as time resolved radioluminescence. We will discuss presence of the defect luminescence band, which is undesirable in scintillation detectors due to the very long decay time in microsecond range. The upper part of the active region in 30 QW-structure had the best luminescence properties in respect of ratio exciton/defect luminescence. The influence of Si doped layer under the active region on scintillation properties was studied as well.

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#### Poster Session 2 (10:00-11:00)

-Conveners: Remi Chipaux; Marco Pizzichemi; Kristof Pauwels [id] title

boar d

# [85] PHOTOELASTIC SPHENOSCOPIC OBSERVATIONS FOR RELIABLE AND FAST INSPECTION OF ANISOTROPIC CRYSTALS.

#### Presenter: NATALI, Pier Paolo

The development of new generation of scintillating crystals for High-Energy Physics, Medical Imaging and Security, asks for reliable and fast inspection techniques. Since the crystal production cost is a large part of instrumentation final expenses, it is mandatory an improvement of the production process in terms of production efficiency, i.e. reduced number of defected crystals. The residual stress is a signature of the crystal structural state and mechanical defects may be present, due to production process deviations, that negatively affect performances and compromise mechanical reliability. Inspection methods based on photoelasticty [1] allows to evaluate, in not invasive manner, the internal stress by means of the shape of the observed interference fringe patterns.

The traditional photoelasticity methodology works well along some specific directions of observation, but it lacks spatial resolution: the laser conoscopy [2] overcomes such a limit, but needs a great number of measurements and it is easy to apply only if the sample is cut in particular crystallographic directions. Polariscopes using conoscopic observation along the optical axis are devices used to evaluate crystals residual stresses in a precise, but time-consuming manner. In this work, we present a methodology for the photoelastic analysis of birefringent crystals, which is based on a modified polariscope. In order to reduce the time for crystal analysis, the light beam shape, that impinges on the crystal surface, has been changed from a solid cone (conoscopy) to a wedge (sphenoscopy [3]). Since the polarized and coherent light is focused on a line rather than on a spot, this allows for a faster analysis leading to the observation of stress distribution along a line. In conoscopy observations, the interference fringes can be modelled as quartic curves. The modelling is quite complex if the observation is performed out of the optic axis. In the sphenoscopy the modelling is guite easier and leads to a straight interpretation, independent on the observation direction. Instead of the punctual conoscopic observation, the sphenoscopy analyses a "stripe", reducing the observations number. For instance, to evaluate a crystal with a given resolution, the sphenoscopy needs at the most 2n observation instead of the n^2 observation necessary in conoscopy. In case of absence of stress or defects along a "stripe" the resulting sphenoscopic fringe pattern is composed by straight lines. Curvature or distance variations between the fringes are due to internal stress or defects. In this work, we show the application and validation of this innovative technique by means of a simple interpretation model.

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PHOTOELASTIC SPHENOSCOPIC ANALYSIS OF CRYSTALS
Review of Scientific Instruments, 87(1), 015113, 2016.

#### [186] Vacuum Ultraviolet Luminescence Spectroscopy Setup at Max IV Laboratory

#### Presenter: PANKRATOV, Vladimir

The MAX IV Laboratory (Lund, Sweden) is a new synchrotron radiation research center located in Lund Sweden. It was inaugurated in June 2016. Currently, seven beamlines have been installed at the MAX IV facility. With a circumference of 96 m and an electron energy of 1.5 GeV, the new storage ring at MAX IV Laboratory is a perfect synchrotron source for VUV and soft x-ray (XUV) photon generation. Time-resolved luminescence experiments will be performed in one of the branches of the FinEstBeaMS beamline, which is developed by a consortium of Finnish and Estonian Universities. The tuneability of synchrotron radiation and its inherent well-defined time structure makes it particularly well suited for time-resolved luminescence studies. The state of art of such investigations is summarized in the review of SUPERLUMI setup [1], which was a flagship for three decades. Particularly, it is proven that time-resolved vacuum ultraviolet (VUV) luminescence excitation spectroscopy under synchrotron radiation is a powerful tool for the study of electronic structure of any classes of wide band gap luminescent and scintillating materials.

The FinEstBeaMS will use an elliptically polarizing undulator light source, which produces soft x-rays of a variable polarizations with high flux 8x1013 ph/s – 1x1011 ph/s at photon energies between 4 and 1000 eV. Higher order excitation will be prohibited using either solid state (MgF2, SiO2, metal films) filters or gas phase filter in future. The duration of synchrotron pulses is 200 ps.

The luminescence endstation consisting of an ultra-high vacuum chamber with a closed-cycle helium cryogenic system was constructed providing sample temperatures T = 4 - 400 K. To analyze photoluminescence in the UV to near IR range a Shamrock 0.3 m spectrometer equipped with a CCD and several time-resolved photomultiplier detectors is coupled to an optical fiber, collecting emission from the sample. Time-resolved luminescence spectroscopy experiments can be implemented in single bunch mode of storage ring and/or utilize a chopper. The single bunch mode provides 320 ns time window to perform time-resolved experiments, while a chopper can extend a time window up to microseconds range. The possibility to use high-resolution excitation (0.01 nm at 100 nm) in scanning mode and the option of tuneable polarization of the incident light, not widely used so far, make this beamline very attractive for the field of luminescence and scintillating studies under VUV and XUV excitations.

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#### [36] Enhanced emission of plastic scintillators by plasmonic lattice resonance

#### Presenter: LIU, Bo

Plastic scintillators are extensively used in nuclear and particle physics experiments. However, because of the total internal reflection, the extraction efficiency of plastic scintillators with plane structures is very low. Additionally, the escaped light emission of the scintillator usually follows a Lambertian angular profile without specific directionality, leading to a low detection efficiency in practical detection systems. surface lattice resonances (SLRs) based on periodic arrays of metallic nanoparticles have been proposed to control the spontaneous radiation of luminescence materials. SLRs are collective lattice resonances originating from the radiative coupling among the localized surface plasmon resonances (LSPRs) of individual metallic nanoparticles, which can be significantly enhanced by diffractive orders in the plane of a periodic array. Such resonances have characteristically photonic-plasmonic natures.

We have demonstrated that periodical arrays of silver nanodiscs can enhance the light emission from a plastic scintillator layer on the surface of a silicon substrate. It is shown how the SLRs of periodic Ag nanodisc arrays enhance the light emission of a plastic scintillator layer on the surface of a silicon substrate. Because of the broad emission, a 1.81-fold emission enhancement for wavelength- and angle-integration has been obtained. The present study could be promising for fundamental and applied research into enhanced luminescent material layers on opaque substrates.

#### [61] Silica coating of scintillating nanoparticles

#### Presenter: TOMANOVÁ, Kateřina

Scintillating nanoparticles (NPs) may find various applications in medicine, one of them being X-ray induced photodynamic therapy (PDTX) [1]. In general, a material suitable for PDTX is a nanocomposite consisting of silica-coated scintillating core (e. g. Pr<sup>3+</sup>-doped lutetium aluminium garnet) subsequently functionalised with protoporphyrin IX (PpIX) [2] at its surface.

The first important step in the nanocomposite synthesis is ensuring that a uniform silica coating of the core particle was formed. The methods for silica surface modification include both the sol-gel and dense liquid processes as well as their combination. During the sol-gel process, controlled hydrolysis of tetraethoxysilane (TEOS) is performed in suspension of NPs in an organic solvent. The hydrolysed TEOS condenses on the surface of NPs forming a polysiloxane layer. The reaction proceeds at room temperature. However, the dense-liquid process requires careful control of reaction conditions such as the temperature and pH. The formation of silica layer on NPs is ensured by maintaining a solution of sodium silicate on supersaturation level (90 °C, pH = 9.5). The two step process combining the above mentioned methods was proposed by Liu et al. [3].

In this work we apply the above described silica coating methods on scintillating nanoparticles with three different particle sizes (up to 70 nm). We thoroughly characterise the materials prepared by several techniques, such as the X-ray diffraction, radioluminescence spectroscopy, transmission electron microscopy and surface area measurement. We compare and evaluate different coating methods to identify the one best suited for subsequent functionalization by PpIX.

#### Acknowledgement

This work has been supported by the Czech Science Foundation, project GA17-06479S, and by the Grant Agency of the Czech Technical University in Prague, grant No. SGS17/195/OHK4/3T/14.

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## [70] Composite scintillators with improved light yield, temporal and spatial resolutions

#### Presenter: POKIDOV, Anton

Our team developed composite scintillators from inorganic nanoparticles and organic phosphors. They possess unique combination of big capture ability for gammas, nanosecond decay time of light emission and high light yield comparable to CsI(TI). Super-fast transfer of the excitation energy from heavy inorganic particles absorbing X-Rays to organic phosphors emitting scintillations achieves these records. Usual organic scintillators have low radiation hardness resulting in decrease of light yield, optical transparency and mechanical strength during relatively short irradiation. But our composite scintillators demonstrate manifold increase of radiation hardness due to intermolecular fixation of polymers by nanoparticles. Earlier composites of this kind had disadvantage of non-homogeneous distribution of nanoparticles resulting in scattering of scintillation light. For better ordering, we applied fast solidification the composite solution by spinning on centrifuge disk. The centrifugal force aligned polymer molecules and provided better adjustment of benzene rings to nanoparticles. This adjustment increased the effectiveness of excitations transfer from nanoparticles absorbing gammas to polymers emitting scintillations. Due to this improvement the light yield of the oriented films was increased by 20 % in comparison with the films solidified spontaneously. Besides the centrifugal force ejected from the rotating substrate small drops of the composite solution which were transformed to microfibers during their flight. Diameters of these microfibers varied from 10 to 50 mcm. Measurements of X-Ray luminescence of these microfibers showed the enhancement of their light yield by 30% compared to non-oriented films. This enhancement on the one hand can be attributed to better adjustment between benzene rings and nanoparticles mentioned earlier. On the other hand optical microscopy of these microfibers revealed quasi-periodic modulation of their structure along the fiber with the periods of micrometer order. This kind of axial modulation can provide a sort of distributed feedback resulting in enhancement of the light emission due to optical resonances for the emitted wavelengths in analogy with photonic crystals. We observed propagation of the emitted light along the fiber beyond the borders of the region excited by X-Rays. This experimental fact supports the version about preferential emission of the light along the fibers. If so, future studies of the relation between the conditions of these microfibers preparation, their internal structure and light emission characteristics would result in further improvement of their scintillation parameters due to optimization of the technology. These fibers can be used for manufacturing of scintillation matrix detectors with high timing and spatial resolution. In this case the lateral surfaces of the fibers should be isolated optically from each other (e.g., by thin aluminum coating). Then the parallel to each other fibers can be assembled to dense bundles creating scintillation matrices with transverse spatial resolution corresponding to the fiber diameter and timing resolution of about 1 nsec taking place in the composite scintillators of this kind. Preferential propagation of the emitted light along the fiber axis will improve the light collection by photodetector matrix adjusted to the face of the scintillation matrix, as well as energy and timing resolutions due to minimization of light losses.

## [168] Impact of wrapping materials and bonding adhesives on light transfer efficiency (LTE) and light transfer time spread (LTTS) in scintillation detectors

#### Presenter: LOIGNON-HOULE, Francis

The light output from high-aspect ratio scintillators used in Positron Emission Tomography (PET) detectors is a critical factor to achieve good energy and time resolution. However, only a fraction of the light generated in the crystals is actually extracted from these scintillators, hence raising the need to identify the predominant causes of signal loss.

As a first step, in order to identify the key factors affecting the light transfer efficiency (LTE) in high-aspect ratio LYSO scintillators, various combinations of crystal geometry and wrapping conditions were investigated with Monte Carlo simulations through a full factorial design. It was concluded that the highly reflective material must be carefully selected along with the high-transmittance optical adhesive used to bond the reflector, since both factors were found to be strongly interrelated when optimizing LTE.

As a second step, the unexpectedly high light crosstalk (~30%) measured in crystal arrays assembled with the 3M-ESR reflector was investigated. Analytical modelling of the ESR reflector showed that the film becomes highly transparent to light impinging at large angles when surrounded on both sides by materials of refractive index (RI) higher than air. Monte Carlo simulations indicate that a large fraction (~25–35%) of scintillation photons is incident at these leaking angles in high-aspect ratio LYSO crystals. The film transparency was confirmed experimentally as significant light leakage, up to nearly 30%, measured through the reflector when coated on both sides with optical grease. The angular dependence of the reflector transparency was also confirmed experimentally for angles of incidence larger than ~60°. The major cause of light crosstalk in ESR-bonded arrays was thus elucidated.

Light crosstalk in bonded scintillator arrays is also dependent on the transmittance of the adhesive used to bond the reflector to the crystals. Simulations showed that high transmittance adhesives yield higher LTE, but results in higher light crosstalk (+15-20%). Moreover, the combined effects of high adhesive transmittance and ESR reflectivity quenching at large incident angles result in even higher light crosstalk (+25-30%). One thus has to seek low-RI adhesives approaching an air-coupling condition to eliminate the reflectivity quenching effect, in combination with high transmittance for optimum results.

The ultimate goal in scintillation detectors nowadays is to achieve ultra-fast time resolution, allowing accurate time-of-flight measurements. All aforementioned light collection factors enhancing LTE will contribute to improve time resolution, but the time fluctuation between production and detection of scintillation photons, the light transfer time spread (LTTS), must also be enhanced. The variance in optical path length becomes prominent in long crystal elements and must be taken into account in estimating the achievable time resolution. This was done by calculating a probability distribution function (PDF) representing the photons travel time in crystals. Wrapping conditions, including adhesive and reflector optical characteristics, were found to alter the scintillation light propagation. The influence on both LTE and LTTS, thus on the achievable timing resolution, will be discussed.

# [181] Improvement of light extraction from scintillators due to the surface modification in microscale using Xe-PFIB

#### Presenter: MODRZYŃSKI, Paweł

Positron Emission Tomography (PET) and detectors in High Energy Physics (HEP) both use scintillating crystals and face similar problems when it comes to light extraction. A large fraction of light produced in the crystals is trapped inside the crystal due to total internal reflection caused by their high refraction index. In previous works[1,2] we already showed the light extraction improvement of several LYSO crystals covered with Photonic Crystals structures (PhC) using Gallium Focused Ion Beam (Ga-FIB). However the use of Ga-FIB seems to be particularly problematic for structuration of materials for optical applications since it brings about amorphization of a sample's surface and the implantation of Ga ions to a depth of several dozen nanometers (depending on the acceleration voltage). The Ga implantation causes the metallization of implanted layer inducing surface conductivity and changing the surface reflection as well as introducing chemical Ga-compounds. Xenon Plasma Focused Ion Beam (Xe-PFIB) has many advantages over widely used Ga-FIB e.g. more than 20x higher milling speed, high-precision final cuts, no Ga implantation and others. Therefore we use Xe-PFIB for structuration of crystals without metallization of the surface.

In this work we show the enhancement of LYSO scintillator's surface by micro-structuration using Xe-PFIB which results in the improvement of light extraction. Together with created surface micro-pattern we show the results of spectroscopic measurements as well as the results of computer simulations of light extraction from LYSO crystal within Finite Difference Time Domain (FDTD) method.

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## [21] Facile Synthesis of High Purity Anhydrous Complex Rare Earth Halides by the Modified Mixed-Salts-Dehydration Method

#### Presenter: YU, Jinqiu

Various types of binary or ternary complex halides between alkaline and rare earth halides such as K2LaBr5:Ce, RbGd2Br7:Ce, Cs2LiYCl6:Ce and LiGdCl4:Ce have been developed as scintillators in the last two decades. Many of them, especially the elpasolite series, show excellent scintillation performance and good application prospect. Current crystal growth of complex rare earth halides usually uses high purity simple rare earth halides as starting materials. For example, in order to grow K2LaBr5:Ce crystal, high purity anhydrous LaBr3 and CeBr3 are needed as starting materials, which are very expensive due to their extremely hygroscopic nature and difficulty in preparation. As a result, K2LaBr5:Ce crystal is commercially unattractive because it costs similar but performs worse than LaBr3:Ce crystal, which is grown from merely the same rare earth starting materials. Therefore, new preparation technologies for low-cost complex rare earth halides must be developed.

Previous investigations of G. Meyer [1] on the ammonium-bromide route to anhydrous rare earth bromide revealed that when LaBr3 hydrates were dehydrated with NH4Br, (NH4)2LaBr5 formed as an intermediate product. This inspired us that if LaBr3 hydrates were dehydrated with KBr, K2LaBr5 might be easily synthesized as the product. We named the method mixed-salts-dehydration. Actually, G. Meyer had reported early that complex rare earth halides could be synthesized from the mixed solution of rare earths and other metal halides [2]. D.K. Ingole also used this method to prepare Ce3+-activated K2LaX5 (X = Cl, Br or I) phosphors before [3]. However, whether or not the purity of such products can meet the requirement of crystal growth has not been verified.

Our experiments revealed that the mixed-salts-dehydration method can yield K2LaBr5 with high phase purity. However, the oxygen content of the product is as high as 500 ppm, which is due to hydrolysis of the halide during the dehydration. Unfortunately, such purity can hardly meet the requirement of crystal growth, for which use, the oxygen content should be as low as possible and usually less than 100 ppm.

In order to restrain the hydrolysis and hence improve the purity of the product, the mixed-salts-dehydration method was slightly modified by adding some NH4Br into the solution at the first step. Correspondently, after the dehydration, the solids were further heated above 500 in order to remove NH4Br. High purity K2LaBr5 with oxygen content less than 50 ppm was successfully obtained.

The modified mixed-salts-dehydration method was further implied to prepare a lot of other complex halides including K2LaCl5, K2Lal5, Cs2LiYCl6, Cs2LiCeCl6, Cs2LiY0.995Ce0.005Cl6, RbGd2Br7 and LiGdCl4, and high purity samples were all successfully obtained with high phase purity and very low oxygen content (<100 ppm). These results indicate that the modified mixed-salts-dehydration method is a universal route to high purity anhydrous complex rare earth halides. Our experiments also revealed that it is much easier to obtain high purity complex rare earth halides by the modified mixed-salts-dehydration method than to obtain simple rare earth halides by the ammonium halide route, which may provide cost advantage to rare earth complex halides towards simple rare earth halides, and shed light on their commercial development and applications.

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#### [28] Growth and Scintillation properties of Cs2LiYCl6 crystals doped with different Ce3+ concentration

#### Presenter: REN, Guohao

Cs2LiYCl6 crystals doped with different Ce3+ concentration were grown with Vertical Bridgman method. Their scintillation properties including x-ray excited luminescence, photoluminescence excitation and emission spectra, energy resolution and scintillation decay time under the excitation of gamma-ray were also investigated. They all showd the intensity of CVL luminescence and the emission peaks of Ce3+ are dependent on the concentration of Ce ions in the crystals. The overlap between PLE and PL spectra demonstrated that self-absorption effect exsits in Cs2LiYCl6:Ce3+ single crystals. Of all samples doped with different Ce concentration, the best energy resolution was achieved at a concentration of 0.1mol%Ce3+ and the light yield was 18000±1000 ph/MeV . Three different luminescence decay component: CVL, direct electron-hole capture by Ce3+, binary Vk and electron diffusion could be identified. The scintillation decay time of direct electron-hole capture by Ce3+ decreaseing with increasing Ce3+ concentration proved Ce3+ concentration quenching effect in Cs2LiYCl6 crystals.

#### [51] Photo-induced preparation of band-gap-engineered garnet powders

#### Presenter: BÁRTA, Jan

Multicomponent oxide scintillators with the garnet structure (space group Ia-3d) have attracted significant attention in recent decade due to the possibility of alleviating structural defects and changing the luminescence properties by tuning the composition. Very often, the admixture of Ga into Ce-doped aluminate garnets is used to lower the conduction band minimum and thus to bury the anti-site defect traps in the conduction band [1], improving scintillation intensity. Moreover, the emission wavelength is slightly blue-shifted. At the same time, the thermal ionization of the Ce<sup>3+</sup> 5d state becomes more probable, necessitating further modifications of the composition. In a recent DFT simulation of  $A_2B_eO_{12}$  garnet structures (A = Ln, Y; B = Al, Ga, In, As, Sb), the substitution of the trivalent ion B was shown to have significant influence over the conduction band minimum and valence band maximum relative to LuAG (A = Lu; B = AI) [2]. Among these, the hypothesized Sb- and As-containing garnets exhibited a markedly raised valence band maximum, which would improve the transport of holes toward Ce<sup>3+</sup> while preserving the Ce<sup>3+</sup> 5d state position relative to conduction band. As both As<sub>2</sub>O<sub>2</sub> and Sb<sub>2</sub>O<sub>2</sub> are very volatile, the formation of such compounds precludes high calcination temperatures, let alone preparation using melting. In this study, we utilized a photo-induced formation of solid precursors to GGAG (A = Gd; B = Ga+Al) and Lu<sub>3</sub>(Al,Sb)<sub>5</sub>O<sub>12</sub> (LuSbAG) garnets in aqueous solutions of their salts and ammonium formate, followed by calcination in various atmospheres to form nanocrystalline garnets [3]. The resulting powder materials were examined by X-ray powder diffraction (XRD), X-ray fluorescence (XRF), differential thermal analysis (DTA) and luminescence measurements. The solid precursors to GGAG:Ce were prepared with almost 100% yield and contained identical concentrations of all elements as the initial solutions. During their calcination, a sharp exothermic DTA peak was detected between 800 and 900 °C (its temperature increased with decreasing Ga concentration), probably related to the crystallization of garnet phase. However, samples with high Ga concentration contained the garnet phase along with Gd<sub>2</sub>O<sub>2</sub>GaO<sub>4</sub> even when calcined at 1200 °C – this impurity disappeared only after further heating. Radioluminescence spectra featured an intense Ce emission band, a weak broad band at ~370 nm (caused by anti-site defects) with lower intensity in samples with high Ga concentration, and several emission lines of Eu impurity. The LuSbAG precursor was prepared by a two-step process due to fast Sb(III) hydrolysis into Sb(III) oxide - the precursor containing Lu and AI was formed in the solution, into which antimony acetate was then added. The produced solid phase with homogeneous distribution of elements was then calcined in different atmospheres. XRF and XRD confirmed that calcination in air somewhat prevents the loss of Sb from the material during heating at the cost of oxidation into Sb(V) oxides. Calcination in reducing or inert atmosphere led to significant losses of Sb, but a pure garnet phase with acceptable Sb content was formed after calcination at 700 °C for 3 hours in Ar. However, the additional heating required for high luminescence intensity causes further losses of Sb. This research has been supported by the Czech Science Foundation grant GA 17-06479S and EC H2020 project ASCIMAT no. 690599. [1] M. Fasoli, A. Vedda, M. Nikl, C. Jiang, B. P. Uberuaga, D. A. Andersson, K. J. McClellan, C. R. Stanek; Phys. Rev. B 84 (2011) 081102. [2] S. K. Yadav, B. P. Uberuaga, M. Nikl, C. Jiang, C. R. Stanek; Phys. Rev. Applied 4 (2015) 054012. [3] J. Bárta, V. Čuba, M. Pospíšil, V. Jarý, M. Nikl; J. Mater. Chem. 22 (2012) 16590-16597.

## [58] Optical band gap engineering of chemically synthesized PbS thin films by in situ Sn doping for photovolitic application

Presenter: HONE, Fekadu Gashaw \*\*Abstract:\*\*

Structural, morphological, and optical properties of undoped and Sn doped lead sulphide (PbS) thin films were systematically studied in this work moreover, efforts had made to tune the optical band gap of PbS thin films for PV application. The thin films were grown on silica glass substrates at a bathe temperature of 700 C by a chemical bath deposition (CBD) method. The concentration of Sn in the deposition bath represented by the ratio [Sn+2]/[Pb+2] and varied from 0% to 20 %. The X-ray diffraction analyses revealed that all the films were polycrystalline in nature with a face centered cubic crystal structure, however, the preferred orientations of the crystallites varied along the (111) and (200) planes with Sn doping concentration. The XRD results also verified that peak intensities and the crystalline size were decreased with increasing Sn concentration. Energy dispersive X-ray analysis confirmed that doping Sn plays a vital role on the surface morphology of the PbS thin films. The optical absorption spectroscopy study revealed that the optical band gap of the PbS thin films were engineered from NIR to visible region by incorporated Sn+2 ions by simple and cost effective route. This makes PbS:Sn thin films a good candidate for solar cell absorber layer.

\*\*Key words\*\*: Lead sulphide, Chemical bath deposition, Microstrain, Thin film

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## [62] LuAG:Pr3+ - BASED NANOHYBRID SYSTEMS FOR SINGLET OXYGEN GENERATION

#### Presenter: POPOVICH, Kseniya

With LuAG:RE<sup>3+</sup> (RE = rare-earth elements) single crystal and fiber applications in many fields of scintillator industry, including high-energy particle physics, medical imaging and security measures [1], there is a growing interest in the LuAG:Re<sup>3+</sup> powders. Due to the luminescence properties and very high chemical stability, such powders may be used as a light-emitting core of the nanocomposite material for X-ray induced photodynamic therapy (PDTX) [2]. PDTX uses tumor-destroying agents based on the nanoparticles (NP) conjugated with photosensitizer (PS) molecules. The agent accumulates preferentially in the target cells; subsequently, the external X-ray irradiation excites the scintillating NP, emitting secondary radiation, which activates the PS molecules. Their deexcitation via non-radiative energy transfer (ET) leads to the production of the reactive oxygen species, where the singlet oxygen is believed to be the most cytotoxic one [3].

Due to the suitable luminescent properties, especially the overlap of Pr<sup>3+</sup> emission and protoporpyrin IX (PpIX) absorption bands, LuAG:Pr<sup>3+</sup> may be a good candidate for a light-emitting core of the PDTX agent. In this work, we present a concept of preparation of the singlet oxygen producing

LuAG:Pr<sup>3+</sup>@SiO<sub>2</sub>-PpIX nanocomposites for PDTX application. LuAG:Pr<sup>3+</sup> nanoparticles with the average size of about 25 nm were prepared using photo-induced method [2]. Subsequently, the surface coating procedure with SiO<sub>2</sub> amorphous layer was performed via combined two-step sol-gel/dense liquid coating process. Finally, NPs were conjugated with a photosensitizer molecule (PpIX). Morphological characteristics of the materials were obtained from the X-ray diffraction analysis (XRD) and transmission electron microscopy (TEM). Energy transfer (ET) and luminescent properties of the nanocomposites were studied by radioluminescence (RL) and photoluminescence (PL) spectroscopies.

Acknowledgement:

This work has been supported by the Czech Science Foundation, project GA17-06479S, and by the Grant Agency of the Czech Technical University in Prague, grant No. SGS17/195/OHK4/3T/14.

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#### [76] Scintillation characteristics of liquid phase epitaxy grown GAGG:Ce single crystalline films

#### Presenter: CHEWPRADITKUL, Weerapong

The Gd3(Al,Ga)5O12:Ce single crystalline films were grown by liquid phase epitaxy (LPE) technique from BaO-B2O3-BaF2 flux. The scintillation characteristics were investigated and compared to the bulk Czochralski-grown single crystal of similar composition. The light yield (LY) and energy resolution were measured using an R6231 photomultiplier under excitation with  $\alpha$  - rays. At 5.155 MeV  $\square$  - rays, the LY value of 5980 photons/MeV obtained for the LPE sample is lower than that of 7050 photons/MeV for the bulk sample whereas an energy resolution of the LPE sample is better (6.0 % vs. 7.5 %). The LY dependence on integration time measurements show a lower contribution of slow components in the scintillation pulse of LPE sample with respect to bulk sample. The ratio of LY value under excitation with  $\square$ - and  $\square$ - rays ( $\square/\square$  ratio) is also determined.

Keywords: Energy resolution, Gd3(Al,Ga)5O12:Ce, Light yield, Liquid phase epitaxy, Scintillation

#### [98] Directionally Solidified Eu3+ and Y3+ Co-Doped HfO2/α-Al2O3 Eutectic Scintillators

## Presenter: YOSHIKAWA, Akira

Currently, CsI:TI and Gd2O2S:Tb (GOS) were recent commercially available scintillators for X-ray imaging. In this applications improvements of sensitivity and special resolution are required to obtain much fine morphological information. To improve sensitivity and special resolution, increase of thickness of scintillator plates or discover of new heavier scintillation materials were necessary. Up to now our group reported submicron-diameter phase-separated scintillator fibers (PSSFs) using directionally solidified eutectic (DSE) systems for higher resolution X-ray imaging. Csl/NaCl, GAP/α-Al2O3 [1] and LiF/ CaF2/LiBaF3 [2] have been already reported as PSSFs.

HfO2 doped with Eu has attracted attention due to its high density of 9.7 g/cm3, a high effective atomic number (Hf:72) and negligible intrinsic background. Light yield of Eu:HfO2 powders was reported to be 30000-31000 photons/MeV. Most recently fabrication of Eu:HfO2/α-Al2O3 eutectics and observation of its PSSFs structure were reported. HfO2 itself and HfO2 compounds have generally high melting temperatures of more than 2700 °C. HfO2 based DSE growth was firstly reported at the eutectic point blow 2000 °C. However, The Eu:HfO2/α-Al2O3 eutectics showed relatively week Eu3+ 4f4f emission comparing to the GOS standard[3].

In this study, Eu doped and Y substituted HfO2/ $\alpha$ -Al2O3 were investigated as a scintillator material for X-ray imaging application. Here, Y substitution effect on crystal structure and luminescence properties in HfO2 were investigated. Crystal growth was performed by the  $\mu$ -PD method for the eutectic composition, whose melting point is at significantly lower temperatures than that of HfO2 itself.

Eu doped (Hf,Y)O2/Al2O3 eutectics were fabricated with c-axis oriented a-Al2O3 seed by the □-PD method. Fiber-like eutectic structure were observed along the growth direction. Y substitution effect on crystal structure and luminescence properties in HfO2 were investigated.Y=20 and 30% subsuituted (Hf,Y)O2/Al2O3 showed cubic (Hf,Y)O2 and a-Al2O3 phases. Y=40% showed thirdly perovskite phase. Broad Eu2+ 4f5d emission peaking at 480nm was observed in the Y 3% sample. Photoluminescence decay time of the Eu2+ 4f5d transition was 8.93 ms 77%, 25.6 ms 16%, 326 ms 7%. References

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#### [109] Scaling and scintillation performance of TLYC:Ce

#### Presenter: KHODYUK, Ivan

Crystalline scintillators with high Z numbers, good energy resolution, and the ability to detect neutrons are highly attractive for threat discrimination and special nuclear materials identification. Traditional scintillators NaI and CsI have effective Z numbers of 50 and 54. However, they do not offer neutron detection. CLYC (Cs2LiYCl6:Ce) scintillator offers gamma energy resolution near 4%, thermal neutron detection when enriched with 6Li, and fast neutron detection when enriched with 7Li [1]. Being a cubic system, it is also relatively easy to grow, therefore its production has scaled rapidly over the last three years, both in terms of size and availability. But at Z=45, and relatively low density of 3.3 g/cc, more material is needed to absorb gamma radiation, as compared to NaI or CsI.

In this paper, we will discuss scaled growth and scintillation performance - of a relatively new scintillator, TLYC (Tl2LiYCl6:Ce), first presented by Kim et. al at SCINT 2015 [2] and later discussed by Hawrami et. al [3]. With cesium replaced by thallium, the material becomes 33% more dense (4.58 g/cc) and the Z number increases from 45 to 69. With higher density and effective Z, less TLYC is required to achieve the equivalent stopping power compare to CLYC. Our preliminary results with a small 10 mm in diameter and 20 mm long crystal show 4.1% energy resolution, a brightness of 22,500 ph/MeV at 662 keV, and FOM of 1.87 for thermal neutron detection. In the paper, we will present results of TLYC enriched with 6Li or 7Li to provide dual or tri-mode (y/n) detection through the well-known pulse shape discrimination technique.

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#### [113] Growth and scintillation characteristics of a Cs2Mo2O7 single crystal

#### Presenter: LEE, Moo Hyun

Recent results of neutrino oscillations have shown that neutrinos have masses. However, the results do not provide absolute masses and properties of neutrinos. To answer these questions, many experiments have been being carried out or prepared. One of the experiments is the Advanced Mo Based Rare Process Experiment (AMoRE) searching for the neutrino-less double beta decays ( $0v2\beta$ ) of 100Mo isotopes in 48deplCa100MoO4 scintillation crystals at mK temperatures [1]. Due to a background from  $2v2\beta$  decays from 48Ca and a limitation in calcium purification below an acceptable level [2], new molybdate-based scintillation crystals for the AMoRE phase-II are necessary. If a scintillation crystal containing the molybdenum emits a high enough light output, it can be a candidate scintillator for the neutrino-less double beta decay experiment. This study reports on a growth of a Cs2Mo2O7 single crystal and new results related to scintillation properties of the grown crystal.

We synthesized raw powders in a sintering process and confirmed the compound via an X-ray diffraction pattern. For the crystal growth, we also checked the phase transition of the compound via a Differential Scanning Calorimetry (DSC). After growing the single crystal by a Czochralski method, we prepared a sample for a luminescence property measurement. Temperature dependences of emission spectrum and decay time were measured from 10 K to the room temperature. We used a 280 nm light emitting diode (LED) as an excitation source for the emission spectrum and a 266 nm pulsed type laser for the decay time due to its high intensity. In addition, we measured the emission spectrum of a Li2MoO4 crystal to compare the relative light output with the Cs2Mo2O7. Since the Li2MoO4 crystal is well studied, we selected it as a reference. The new results of the Cs2Mo2O7 crystal show that Cs2Mo2O7 can be a candidate scintillator for the neutrino-less double beta decay experiment. We also measured transmittance of the crystal at a room temperature.

Keywords: Neutrino-less double beta decay, Scintillation properties, Czochralski technique, Cs2Mo2O7 crystal, Li2MoO4 crystal.

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# [121] Optimization of dopant and scintillation fibers diameter of GdAlO3/a-Al2O3 eutectic for X-ray phase imaging detector

#### Presenter: KAMADA, Kei

X-ray phase imaging techniques has been developing for the last decade because of its attractive potentials [1–7]. X-ray phase imaging provides three images such absorption, differential-phase, and visibility-contrast images. This future causes to higher resolution density variations in the sample than that conventional absorption-contrast X-ray imaging. Medical and biological imaging is the main target of X-ray phase imaging, and several trials using synchrotron radiation sources and laboratory sources have been made. In X-ray Talbot-Lau interferometry, single or mulch absorption gratings between a sample and X-ray generate detector differential-phase, and visibility-contrast images. The absorption gratings generate Moire fringes and differential-phase, and visibility-contrast images are obtained from analysis of the spatial frequency of the Moire fringes. However absorption gratings absorbed the transmitted X-ray and sensitivity is degraded. So exposure dose can become a problem in medical and biological imaging.

Recently submicron-diameter phase-separated scintillator fibers (PSSFs) were reported and they possessed both the properties of an optical fiber and a radiation-to-light conversion. The PSSFs were fabricated using a directionally solidified eutectic (DSE) system. Our group reported high-resolution X-ray imaging using Ce doped GdAlO3(GAP)/a-Al2O3 eutectic[9,10]. In this study, Tb and Eu doped GAP/a-Al2O3 eutectic scintillator was grown by the micro pulling down (□-PD) method with varius growth rate. Radiomuninescence intensity, decay time and scintillation fibers diameter were evaluated for X-ray imaging detecrtor.

1~15mol% Tb and Eu doped GAP/□□-Al2O3 eutectics were grown by the □-PD method. Tb3+ and Eu3+ 4f4f emission were observed in 470-700nm and 580-750nm ranges , respectively. Tb 8mol% and Eu 5mol% samples showed 5.5 and 4.4 times higher emission intensity than Ce doped one. Diameter pf scintillation fibers were 2.43-0.38mm for growth rate of 0.07-3.0 mm/min in the grown Tb 8mol% doped GAP/a-Al2O3 eutectics. The best contrast transfer function@122lp/mm of 0.28 was archived at the sample of growth rate at 0.3mm/min. The grown eutectic wafers were mounted on a CMOS sensor (SONY, 2.5µm pitch, 2080x1552pizels, 5.2x3.9mm sensitive area) with a fiber optic plate (FOP, Hamamatsu J5734, 3□m fiber diameter). By using this sensor, a prototype of X-ray Talbot-Lau imaging system was developed. Absorption, differential-phase, and visibility-contrast images were simultaneously obtained without absorption grating. Comparing to X-ray Talbot-Lau imaging system using CSI scintillator and a absorption grating, 2 time higher signal difference-to-noise ratio was achieved at the same radiation dose of 10mGy. This is the first result of direct X-ray phase imaging. Details on eutectic growth, detector design and X-ray phase imaging will be showed in the presentation.

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## [122] High proportional LaBr3:(Ce,Sr) scintillator at industrial scale, properties and characterization *Presenter: OUSPENSKI*, *Vladimir*

LaBr\_{3}:Ce is known as an efficient scintillation material with \$\gamma\$-ray spectroscopic properties for isotopes identification applications. Energy resolution of industrial detectors is on the order of 3.0%. Material engineering with \$Me^{2+}\$ co-doping demonstrated an efficient way of improving the proportionality and consequently the energy resolution of the co-doped scintillator [1,2]. Some properties of co-doped crystals are evaluated with respect to the reference: the decay time of the co-doped scintillator becomes more complex with additional 1 or 2 slower components; the light yield increases from the referenced 73000 ph/MeV to 78000 ph/MeV for co-doped material; the energy resolution is improved.

The level of co-doping in industrial crystals has been optimized by balancing the tradeoffs between increase of the principal decay time and improvement of the energy resolution.

The latest generation of industrial detectors with the crystal size 1.5"x1.5" consistently demonstrate an energy resolution of 2.2% at 662 keV.

The temperature dependence of the light yield of co-doped scintillator in the practically interesting range (20; 175)°C is becoming more flat.

The engineered material demonstrates enhanced PSD (Pulse-Shape Discrimination) for discrimination [3]. The PSD property enables the discrimination of the background from \$^{227}Ac\$ and its daughter products in the decay chain. The PSD makes it possible to actively suppress the contributions in spectroscopic measurements. The same PSD can also be applied in the use of \$LaBr\_{3}:Ce:Sr\$ material in composite neutron detection systems.

Saint-Gobain Crystals is launching the full-scale production of engineered \$LaBr\_{3}:Ce:Sr\$ scintillator in 2017 for the various customized applications. The maximum size of crystals in the detectors are limited to the diameter equals 4" and the length up to 10".

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# [128] Single crystalline film scintillators based on the Ce3+ doped Ca2RMgScSi3O12:Ce (R=Y, Lu) garnets *Presenter: ZORENKO*, *Yuriy*

In this work, we present for the first time the results on crystallization and investigation of the luminescent and scintillation properties of new prospective scintillators based on the single crystalline films (SCFs) of Ce3+ doped Ca2RMgScSi3O12 (R=Y, Lu) silicate garnet, grown by the liquid phase epitaxy (LPE) method onto Y3Al5O12 (YAG) substrates. The luminescent properties of Ca2RMgScSi3O12:Ce SCFs were compared with the properties of the reference YAG:Ce and LuAG:Ce SCF samples. The influence of the thermal annealing in 1000-1300oC range in N2/H2 reducing atmosphere on the optical and scintillation properties of Ca2RMgScSi3O12:Ce SCFs was investigated as well.

We have observed the formation of two types of Ce3+ centers in the Ca2RMgScSi3O12:Ce garnets in the emission and excitation spectra as well as in the decay kinetics of the Ce3+ luminescence in the SCFs (). These two types centers (labeled as Ce1 and Ce2) possess different local surroundings due to substitution by the Ce3+ ions of the different types of cations (correspondingly R3+ and Ca2+) in the dodecahedral positions of garnet host. Ce1 and Ce2 centers in Ca2RMgScSi3O12 garnets are characterized by the differing spectral behaviors (the positions of the emission and excitation bands, Stokes shift and decay time of Ce3+ photoluminescence) due to the different crystal field strength in the various dodecahedral sites for localization of Ce3+ ions.

We have observed also the formation of Ce4+ and Ce3+ valence states of Ce in the SCF of Ca2RMgScSiO12:Ce garnets due to the non-uniform distribution of the Ca2+, Mg2+ and Si4+ cations, and charge compensation requirement. The presence of Ce4+ ions in as grown SCF samples is confirmed by the presence of absorption due to the O2+-Ce4+ transitions in the UV range. The Ce4+ centers are also responsible for acceleration of the initial stage of the cerium photoluminescence decay and presence of the fast components with the lifetime in the few ns range in Ca2YMgScSi3O12 SCF. The Ce4+ --> Ce3+ recharging in these SCF is achieved by annealing of the samples in the reducing atmosphere at temperatures above 1000oC. Such thermal treatment leads also to more exponential-like decay kinetics of the Ce3+ luminescence in Ca2RMgScSi3O12:Ce SCF and enables the study of the energy transfer processes between the different Ce3+ multicenters in these garnets.

The results of this research can be suitable for the development of new generation of scintillators based on the epitaxial structures of Ca2+-Si4+ containing garnets, grown by LPE method onto undoped or doped substrates of garnet compounds for registration of the different components of mixed ionization fluxes as well as for the microimaging technique.

\*\*Acknowledgements:\*\* The work was performed in the framework of NCBR NANOLUX #286 project and partly also supported by Polish NCN 2016/21/B/ST8/03200 project and Ministry of Education and Science of the Ukraine SF-20 F project.

# [146] Scintillating screens based on the single crystalline films of Eu3+ doped mixed RAIO3 (R= Tb, Gd, Lu) perovskites

#### Presenter: ZORENKO, Yuriy

In this work, we continued our research directed on crystallization and investigation of the luminescent and scintillation properties of scintillating screens based on the single crystalline films (SCFs) of Eu3+ doped RAIO3 (R= Tb, Gd, Lu) mixed perovskites [1, 2]. SCFs of the solid solutions of (Tb1-xLux)AP:Ce and Gd1-xTbxAP:Ce, x=0-1.0 perovskites were grown by the liquid phase epitaxy (LPE) method onto YAIO3 (YAP) substrates using the traditional PbO-B2O3 flux. The absorbance, cathodoluminescence (CL), X-ray excited luminescence (RL), photoluminescence (PL) and scintillation properties of these SCFs under  $\alpha$ -particles excitation were investigated depending on the perovskite hosts. We have found that the shape of CL, RL and PL spectra, scintillation light yield (LY) and decay kinetics of Eu3+ ions in the RAIO3 (A= Tb, Gd, Lu) SCFs are strongly affected by the local surrounding of dopant depending on the cations-ligands distances in the perovskite hosts. Specifically, the orange emission band at 593 nm dominates in the CL/RL/PL spectra of (Tb,Lu)AP:Eu and (Tb,Gd)AP:Eu SCFs when the red luminescence band at 710 nm prevails in the spectra of LuAP:Eu and YAP:Eu SCFs. The effective Tb3+->Eu3+ and cascade Gd3+ --> Tb3+ --> Eu3+ energy transfer processes has been found in (Tb,Lu)AP and (Gd,Tb)AP lattices, respectively, giving reasons to increasing the efficiency of the Eu3+ luminescence in the perovskite host. We have found also that the highest LY of the CL and X-ray exited luminescence is observed in (Gd0.4-0.5Tb0.5-0.6)AP:Eu and TbAP:Eu SCFs. The LY of the RL in these SCFs is comparable with the LY of the best reference LuAG:Ce and TbAG:Ce SCF samples. For this reason, the SCFs of (Tb,Lu)AP:Eu and (Gd,Tb)AP:Eu perovskites can be considered as promising candidates for the scintillating screens in the microimaging detectors. For the last application, the X-ray absorption ability and LY of (Gd,Lu)AP SCF screens can be also improved by Tb3+ codoping due to the extension of "K-edge engineering" in such mixed perovskite compounds [1] and effective Tb3+ --> Eu3+ energy transfer.

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\*\*Acknowledgements:\*\* The work was performed in the frameworks of Polish NCN 2016/21/B/ST8/ 03200 project and Ministry of Education and Science of Ukraine in frame of SF-20F project.

## [156] Czochralski growth of YAG- and LuAG-based scintillators under reducing conditions.

#### Presenter: ARHIPOV, Pavlo

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Y3Al5O12 (YAG), and Lu3Al5O12 (LuAG) crystals doped with Ce3+ or Pr3+ ions, YAG, and LuAG possess attractive scintillation properties and, for instance, are candidates for application in new HEP experiments at colliders. The production cost is a basic criterion in the material choice for practical applications. In the present work, undoped, as well as Ce, Pr-doped YAG, and LuAG crystals were grown under the reducing Ar+CO conditions in W crucibles by a novel fabrication procedure enabling to avoid the usage of expensive Ir crucibles and ceramic heat insulation.

Optical and scintillation parameters, as well as phase and admixture content of crystals were analyzed. Effect of thermal annealing on YAG optical properties was studied on samples fabricated from the crystals grown in Ir crucible by the conventional technology under weakly oxidizing atmosphere, and in W crucible under Ar+CO reducing conditions. In parallel the admixture content in crystals, including carbon concentration was determined and the element composition of the ~50  $\mu$ m thick surface layer before/after the annealing were controlled. The important role of crystal surface interaction with the growth atmosphere was demonstrated [1]. The optimized post-growth annealing procedure of such crystals in reducing or oxidising atmosphere provides an irreversible discoloration of YAG crystals and a high transparency in the 200 – 1100 nm spectral range.

The growth procedure of YAG and LuAG doped with Ce3+, Pr3+ and Sc3+ was developed. Absorption, cathodoluminescence and photoluminescence spectra of LuAG:Pr and LuAG:Sc crystals grown under the reducing conditions, besides the activators bands, contain the strong bands corresponding to F+-centers. The procedure to suppress these parasitic absorption/luminescence properties of as-grown crystals was elaborated. As a result, the light yield and scintillation decay parameters of the doped garnets are similar to those obtained with the crystals grown by the conventional technology in Ir crucibles.

The work is supported by the H2020-MSCA-RISE-2014 Project No. 644260 (INTELUM).

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#### [163] EFG Growth of SrI2(Eu) and CLYC Scintillators

#### Presenter: SWIDER, Stacy

SrI2(Eu) is a promising scintillator having high brightness, 3% energy resolution at 662 keV, and no self-activity. The elpasolite Cs26LiYCl6(Ce) (CLYC) has also shown great promise for dual-gamma-neutron detection. However, high production costs have slowed their adoption into mainstream detection technologies. Costs related to precursors may be mitigated through continuing improvements, but features inherent to the Bridgman technique limit achieving high throughput. Interactions with the ampoule wall can create defects, and resulting stresses become aggravated by thermal gradients along the boule length. Therefore, growth rates are limited to less than 1 millimeter per hour. With edge-defined film-fed growth (EFG), crystal solidification proceeds from a free surface, which removes the possibility of defect formation at the ampoule wall. The lower density of surface defects in EFG-grown material allows the crystal to sustain higher levels of stress without cracking. Srl2 crystals have been grown by EFG at rates 10 times faster than the Bridgman-grown crystals. EFG also allows for near-net shape growth of cuboids, which reduces material loss to machining. We will share our latest results of EFG growth of Srl2(Eu) and CLYC cylinders and cuboids. We shall discuss considerations for seeding, material compatibility, die design, and overall equipment design. In addition, we will complement our discussion with 3D conjugate thermal models of the apparatus.

## [178] Crystal Growth and Scintillation Properties of Ce doped Barium Lanthanide Chlorides

#### Presenter: FURUYA, yuki

Recently, halide scintillators have been developed because of their high light yield which is originated from their narrow band gap. Thanks to great researchers' efforts, some excellent scintillators have been found, such as Eu:Srl2 for high light yield, CeBr3 and Ce:LaBr3 for high light yield and fast decay time. In addition to these promising scintillators, new halide scintillator screening have been carried out for long time. For example, Ce doped AnREmXn+3m (A : Alkali metal, RE : Lanthanide, X : Cl, Br, I) and Ce doped A2A'REX6 (elpasolite structure, A : Alkali metal (large), A' : Alkali metal (small), RE : Lanthanide, X : Cl, Br, I) and Eu doped AnBmXn+2m (A : Alkali metal, B : Alkali earth metal, X : Cl, Br, I) have been studied a lot. But, except for few exception like CsI, most of halide scintillators are hygroscopic, therefore, scintillator materials which have both usability and good scintillation properties are required.

In this work, we focus on Ba2RECI7 (RE : Lanthanide). According to past references, Ba2RECI7 series are expected to be both moisture stable [1] and high light output [2] (for 10%Ce doped Ba2GdCI7 powder, light yield was estimated to be 30000 ph/MeV). In spite of their potential as a promising scintillator, only few research have been taken for Ba2RECI7. Therefore, we tried to grow single crystal of Ce doped Ba2RECI7 (RE = La, Gd, Lu and their combination) by Bridgman–Stockbarger (BS) method, and evaluated its scintillation properties.

Mixed powders were prepared as starting materials, and put them into quartz tube in glove box filled with Ar gas. Then, the quartz tube was evacuated with 250 heating for several hours to remove moisture and other impurities. After sealing, the quartz tube was heated up to the melting temperature of the compound by a high-frequency induction coil and carbon heater. Then, the crystal was grown by pulling down the quartz tube at 1 mm/h.

The grown crystals were cut and polished in a dry room using synthesis oil.

The phases of obtained crystals were confirmed by the X-ray powder diffraction (XRD) analysis. The scintillation properties of grown crystals were measured in dry room. In radioluminesence spectra, Ce3+ 5d-4f emissions were clearly observed. The pulse height spectra and decay time irradiated by radiation were also measured using PMT. 5%Ce doped Ba2GdCl7 was grown and evaluated its scintillation properties, light yield was about one third of CeBr3, and decay time was 33ns. The phases of other grown crystals (Ce doped Ba2LuCl7, Ba2Lu0.5La0.5Cl7, Ba2Lu0.5Gd0.5Cl7) and difference of scintillation properties will be discussed.

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#### [187] Control of dopant segregation in colquiriite-type fluoride single crystal scintillators Presenter: YOKOTA, Yuui

Eu<sup>2+</sup> doped LiCaAIF<sub>6</sub> and LiSrAIF<sub>6</sub> [Eu:LiCAF, Eu:LiSAF] single crystals have been investigated as a neutron scintillator for homeland security. The Eu:LiCAF and Eu:LiSAF single crystals indicated high light yield, ~30,000 photons/neutron, compared to present neutron scintillators. However, the segregation coefficient, \*k\*<sub>eff</sub>, of Eu<sup>2+</sup> ion in the Eu:LiCAF and Eu:LiSAF single crystals is extremely small, \*k\*<sub>eff</sub> = 0.02~0.03, and the small segregation coefficient generated inhomogeneity in the bulk single crystals and decreased the yield rate of bulk single crystals. On these backgrounds, we grew Eu:LiCAF and Eu:LiSAF single crystals using AI metal as a starting material. Generally, EuF<sub>3</sub> powder was used for starting material to grow the Eu:LiCAF and Eu:LiSAF single crystals. However, Eu ion is doped in LiCAF as Eu<sup>2+</sup> ion, and Eu<sup>3+</sup> ion has to be reduced before doping to LiCAF and LiSAF. Therefore, we tried to reduce the valence of Eu<sup>3+</sup> ion in EuF<sub>3</sub> by AI metal.

Eu:LiCAF and Eu:LiSAF single crystals were grown using AI metal powder as a starting material by the micro-pulling-down ( $\mu$ -PD) method. Mixed powders with nominal compositions of

Li(Ca<sub>1-x</sub>Eu<sub>x</sub>)(AlF<sub>1-y</sub>Al<sup>M</sup><sub>y</sub>)F<sub>6</sub> and Li(Sr<sub>1-x</sub>Eu<sub>x</sub>)(AlF<sub>1-y</sub>Al<sup>M</sup><sub>y</sub>)F<sub>6</sub> with x = 0.005~0.03 and y = 0, 0.01 were prepared from LiF (7.5% <sup>6</sup>Li), CaF<sub>2</sub>, SrF<sub>2</sub>, AlF<sub>3</sub>, EuF<sub>3</sub> and Al metal powders (> 4N purity). In the chemical formula, Al<sup>M</sup> and AlF are Al elements derived from AlF<sub>3</sub> and Al metal, respectively. Rectangular specimens with the thickness of 1 mm were obtained from the grown crystals and they were polished for measurements of optical and scintillation properties.

Eu:LiCAF and Eu:LiSAF scintillator single crystals were grown using AI metal as a starting material to improve the segregation coefficient of Eu ion. All Eu:LiSAF single crystals using AI metal [Eu,AI<sup>M</sup>:LiSAF] indicated high transparency while Eu3%:LiSAF crystal without AI metal included milky parts in the crystal. The powder X-ray diffraction patterns indicated that all Eu,AI<sup>M</sup>:LiSAF single crystals were a single phase of colquiriite-type structure without impurity phase. Effective segregation coefficient, \*k\*<sub>eff</sub>, of Eu2%,AI<sup>M</sup>1%:LiSAF single crystal was significantly improved to the value of 0.986 using the AI metal. In the transmittance spectra, all

Eu,Al<sup>M</sup>:LiSAF single crystals indicated more than 70% transmittance and absorption peaks were observed around 200 and 300 nm. Light yield of Eu,Al<sup>M</sup>:LiSAF single crystals under thermal neutron irradiation increased with an increase of Eu concentration and the crystal with Eu2% indicated the maximum light yield, 10,000 photon/neutron. Details of crystal growth, optical and scintillation properties for Eu,Al<sup>M</sup>:LiCAF and Eu,Al<sup>M</sup>:LiSAF will be reported.``

#### **[192] Structure and luminescence of Li2O- xGeO2 glass-ceramics doped with some three charged ions** *Presenter: NEDILKO, Serhii*

Transparent oxide ceramics has attracted large attention of research due to their potential applications in solid-state lasers, medical and optoelectronic devices and scintillation technique too. In fact, transparent ceramics manufacturing has no of drawbacks such as long time and large sizes to grow of single crystals scintillators. Now, among the transparent ceramics produced for application as scintillation materials various glass-ceramics materials draw a close attention.

The low temperature (melting temperature of un-doped glass is 1277oC) glass-ceramics based on lithium- germanium oxides Li2O-x(GeO2) (LGO) as optical materials practically had not been studied before. The devitrification of the Li2O-x(GeO2) low temperature glass-ceramics start at 512 C. Heating leads to formation of ordered areas of the Li2Ge4O9 micro/nanocrystals in the volume of amorphous GeO2 body, in other words, the glass-ceramics is formed. The Li2Ge4O9 phase diminished when temperature riches T = 567 C, then the Li2Ge7O15 phase is formed.

The technologies of lithium germanate glass and nanostructured glass ceramics producing have been developed by us recently. The structure of the LGO crystals is of framework type that is formed by (rigid) germanium-oxygen tetrahedra and octahedra. Such structural elements are suitable for doping with ions of transition groups (Fe and La groups), that state Li2O-x(GeO2) compounds as attractive for new optical materials elaboration. So, we have shown after that multifunctional LGO glass-ceramics (e.g. capable to transit in ferroelectric state) is a promising optical material and particularly it can help to solve specific tasks of ionizing radiation transformation to light.

The LGO glass-ceramics doped with ions of Cr, Eu, and Nd ions were made and studied. In order to decrease the melting temperature to influence directly the absorption and luminescence spectral characteristics the addition of bismuth oxide, Bi2O3, to starting pre-cursor oxide composition will be applied.

Some correlation between morphology, conductivity and optical especially luminescent properties has been discussed. Selection of the conditions of the glass heat treatment makes possible to obtain high dispersive states of different composition, morphology and physical properties of which are determined by the chemical composition and phase's structure, by the relative volume and size of ordered regions, by the character of their spatial distribution. The subsequent cooling stabilizes such dispersion states and allows preparing materials of desired composition and luminescent properties.

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## [200] The effect of bandgap energy and electron trap of Ce-doped Y-admixed lanthanum gadolinium pyrosilicate scintillator

#### Presenter: HORIAI, Takahiko

Recently, oil well logging in the shale layer that exists at a depth greater than conventional oil resources have been required due to increase oil demand. Operating in a deep underground region at high temperature, the scintillator is required that a high light output can be maintained even at high temperature. In previous study, our group has been investigated Ce-doped (Gd,La,Y)2Si2O7, and the light output (LO) at high temperature was improved compared to before Y-admixed [1]. However, in that paper, we have not revealed the reason why LO at high temperature was improved. As the improvement factors, we considered (i) to extend the bandgap and energy gap between top of 5d1 level and bottom of the conduction band (CB) which contribute to suppress the thermal quenching, (ii) to generate new electron trap(s) under Ce3+ 5d1 level moderately, and contribute to be increased the LO at high temperature. In this study, to find the improvement factor of LO at high temperature, we evaluated band structure, the temperature dependence, traps for Y admixed Ce:La-GPS. UV absorption spectra of (Ce0.01 Gd0.59-x La0.4 Yx)2Si2O7 (x=0.00, 0.05, 0.10 and 0.15) grown by the micro-pulling-down method were measured to calculate the bandgap energy. Thereafter, we estimated the temperature dependence of photoluminescence (PL) intensity and LO excited by a Xe lamp and 137Cs as gamma-rays source, respectively, and evaluated electron trap. In addition, we measured thermoluminescence glow curve within 6-300 K. From the results of bandgap energies, the bandgap energies were almost constant regardless of Y concentration, and the effect of the bandgap energy on temperature dependence of LO was considered slightly. On the other hand, comparing the temperature dependence of PL intensity and scintillation LO, where all values were normalized to each value at 298 K, the scintillation normalized LO of Y 5% or 10% admixed samples at 448 K were higher than that of PL intensity. From those result, some electron traps were generated, and would contribute to the temperature dependence. The above results and the detail are shown in this presentation.

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## [205] Crystal growth and optical properties of benzoic acid crystals for neutron scintillator

#### Presenter: YAMAJI, Akihiro

Novel applications of neutron have been developed such as imaging method using a pulsed neutron source and neutron resonance absorption spectroscopy. These methods require position sensitive detectors for neutron with the Time-of-Flight (TOF), and one of the issues is development of fast response scintillator. Some of halide scintillators show high light yield and fast decay time, however, most of them are hygroscopic. Then, we focused on the organic scintillator crystals, which showed fast decay time in the nanosecond range and no hygroscopic nature. Their constituent elements include hydrogen which shows high reaction cross-section to thermal and fast neutron. However, conventional organic scintillator has low melting temperatures and would degrade with overheating. Therefore, we have developed organic crystals for neutron scintillators with high melting temperatures and fast decay times. In this work, we focused on benzoic acid crystals which include pi bonds in the molecular orbital. Thus, we grew benzoic acid crystals from the melt and evaluated their scintillation properties for neutron scintillator.

Benzoic acid crystal was grown by self-seeding vertical Bridgeman method using an enclosed chamber [1]. Raw material powder was charged into a double glass ampoule and the atmosphere in the chamber was replaced with high purity nitrogen. The ampoule was heated by a resistance heater and pulled down slower the rate of 0.03-0.06 mm/min. The grown crystals looked transparent. The scintillation decay were measured by using a 251Cf isotope as neutron source. The detailed results are presented on the presentation.

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#### Acknowledgment

This work was supported by JSPS KAKENHI Grant-in-Aid for Research Activity Start-up JP16H06633.

#### [222] Industrial development of fast scintillator materials

Presenter: SYKOROVA, Silvia

S. Sykorovaa, S.Valkovaa, R. Kucerkovab, J. Houzvickaa, M. Niklb

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In the recent years, the demand for faster scintillators increased significantly in many areas of research in nuclear and particle physics, in medicine and industry.

Crytur company is actively participating in R&D of novel faster single crystal scintillator materials for both the research and industrial applications.

For some of high energy physics applications the existing materials like PbWO4 (PWO) with very fast (less than 10ns) scintillation response, but very poor light yield, are still preferred. As industrial availability of this material disappeared, Crytur established after two years of intense R&D, a novel and reliable growth technology to prepare PWO single crystals for PANDA experiment at GSI in Germany.

The traditional materials like Yttrium/Lutetium Aluminum Garnets doped with Ce3+(Y3Al5O12:Ce, Lu3Al5O12:Ce) with the leading scintillation decay time of 60-80 ns are excellent candidates for high energy physics applications regarding the radiation hardness but their scintillation response is not fast enough.

For the development of novel faster materials there are several mechanisms which can be considered. One of them is based on nonradiative energy transfer from the excited luminescent centers. Such a process can be induced by co-doping of YAG/LuAG:Ce(Pr) with an element from the group of lanthanides (Nd, Sm, Eu, Tb, Dy, Ho, Er).

As a working example, Crytur has prepared recently a codoped LuAG:Pr single crystals: this material shows a faster response with 40% shorter decay time of the dominant component comparing with the non-codoped material. It is robust and temperature resistant, with a moderate light yield.

In this contribution, we will present the results of the industrial growth of PWO using the newly established growth technology in Crytur. Furthermore, the development of novel faster garnet scintillators based on co-doping with lanthanides and their scintillation properties in terms of light yield and decay time at room temperature will be presented as well.
# [23] Scintillation properties and radiation tolerance of Alkali Free Fluorophosphate Glasses with different dopant concentrations

### Presenter: LUCCHINI, Marco Toliman

One of the main challenges in the development of detectors for future collider experiments is finding materials that can operate in high radiation environments while maintaining their physical, chemical, and optical properties.

In this framework, scintillating materials and Cherenkov radiators, such as crystals and glasses, represent a powerful tool for the design of large area radiation detectors. In particular glasses are produced by using a relatively cheap melting process (with respect to crystal growth) which makes them attractive candidates for the instrumentation of large volumes as in the case of many High Energy Physics (HEP) experiments.

A set of heavy glasses of 1 cm<sup>3</sup> with density of ~4.1 g/cm<sup>3</sup> and different chemical compositions was produced by AFO Research Inc. and characterized at CERN. These glasses (FP2035) are multicomponent alkali free fluorophosphate glasses, based on Ba(PO3)2 -Al(PO3)3 - BaF2- MgF2 and doped and co-doped respectively with Pb and Ce oxides and fluorides [1]. Undoped glass samples have also been studied for comparison with doped ones.

The scintillation properties and timing performance of the samples have been investigated through laboratory measurements and irradiation studies have also been conducted to assess the effect of ionizing radiation on the optical properties of the samples.

The Ce-doped samples show an emission of light peaking around 370 nm, a decay time of about 40 ns and a light yield ranging from 100 to 600 ph/MeV, depending on the Ce-concentration. Measurements of coincidence time resolution were also performed using high energy pions and value of ot<35 ps were obtained with both doped and undoped samples read-out with SiPMs.

The irradiation campaign showed that Ce doped FP2035 glasses have a better radiation resistance, with respect to undoped and Pb-doped glasses, and maintain a better transparency after exposure to 1000 Gy of gamma-rays from a Co-60 source. A loss of transparency was noticed in the UV part of the spectrum in FP2035 Ce doped glasses between 360-450 nm, overlapping with the scintillation emission peak, and thus responsible for a decrease of light output after irradiation. Hence, further optimization of the chemical composition of these glasses is required

to allow their use as radiation resistant scintillators in radiation environments with Topic: Defects and Radiation Damage

ionizing doses above 1000 Gy. At the present time, these glasses may have a wide range of applications as radiation hard lenses, windows or optical fibers for light in the visible spectrum (above 450 nm) where the transparency is not degraded by radiation.

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[1] A.A. Margaryan. Ligands and Modifiers in Vitreous Materials, Spectroscopy of Condensed Systems (World Scientific Press, Singapore, New Jersey, London, Hong Kong, 1999).

### [50] Radiation hardness investigation of ZnO(Ga) and ZnO(In) with heavy ion beams.

### Presenter: BOUTACHKOV, Plamen

GSI [1] is a unique large scale facility for heavy ion research. Ion beams of many elements, including H and U, are produced with energies up to 4.5 GeV/u. For absolute beam intensity measurements a BC-400, organic scintilator, is used. Due to the low radiation hardness of this material, alternative inorganic scintilators like ZnO(Ga) and ZnO(In) [2] were investigated during the 2016 experimental campaign. The response of these ceramic detectors to C, Xe and U ion beams at 300 MeV/u will be reported.

1. www.gsi.de

2. P.A. Rodnyi et al., Novel Scintillation Material-ZnO Transparent Ceramics, IEEE 59, 2152 (2012)

### [94] Radiation tolerant YAGG:Ce scintillation material for collider experiments

### Presenter: DORMENEV, Valery

Engineering of garnet scintillators by co-doping with the divalent ions of the second group appears to be a powerful tool to improve the scintillation response. It leads to a strong suppression of phosphorescence in different garnets like Y3Al5O12 (YAG), Y3Al2Ga3O12, Gd3Al2Ga3O12 and diminishing the afterglow parameter correspondingly. However, crystal co-doping with Mg2+ or Ca2+ decreases the amount of Ce3+ in the crystal and deteriorates its light yield as well. Here we report on an alternative concept to control afterglow and timing parameters of garnets by the variation of the Al/Ga ratio in the crystal. It does not affect the valence state of the Ce3+ as activating ion, but acts via the control of the thermo-induced ionization rate of the excited state of Ce3+. We applied this approach to Y3(Alx-Ga1-x)5O12 (YAGG) [1], a medium heavy scintillation material with the emission spectrum pretty matched by the spectral sensitivity of SiPMs. YAGG can be produced in fiber or bulk geometry and, similar to YAG, is produced from inexpensive raw material. The only open question was the radiation hardness of this solid solution material to high energy protons, the main contributor to crystal damage effects at collider experiments. This report presents first results on the measurement of the radiation damage effect in the garnet solid solution crystal under 190 MeV protons. It was found that the crystals show a negligible loss of the optical transmission after irradiation with fluence beyond 5x1013 p/cm2. It opens an opportunity for an application of YAGG:Ce scintillation detectors in a combination with SiPM photosensors to construct a large area detectors to operate in collider experiments, particularly in the close to beam area.

### [1] O. Sidletskiy et al, Cryst Eng Comm, 2017, 19, 1001.

### [110] Radiation damage of LaF3 doped with rare-earth impurities

### Presenter: RADZHABOV, Evgeny

Lanthanum fluoride crystals are characterized as material with large density (5.94 g/cm\$^3\$) and the absence of hygroscopicity, which is promising for laser and scintillation applications . Lattice of LaF\$\_3\$ are perfectly isomorphic to the introduction of trivalent rare-earth ions and spectroscopy of all trivalent lanthanides was studied [1]. However, the radiation damage of rare-earth doped LaF\$\_3\$ crystals was poorly studied.

Crystals of lanthanum fluoride doped with fluorides of Y, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb with concentration 0.01, 0.1 and 0.3 mol. % were grown. The crystals were irradiated using X-ray tube (Pd-anode 40 kV, 20 mA) in our lab or by irradiation with fast electrons (10 MeV, dose 24 kGr) in Ural State University.

Several absorption bands were created by x-irradiation of doped LaF\$\_3\$. Strong band near 200 nm belongs to absorption of F\${^-\_3}\$ molecular ions [2]. The band near 330 nm belongs to V\$\_k\$-type centres. The bands at 500-550 nm belong to transitions of F centres splitted by crystal field. The induced absorption bands increased several times with increasing of impurity concentration from 0.01 to 0.3 mol. %.

Absorption spectra of all the crystals, induced by x-irradiation at room temperature, are divided into two groups. The first group includes crystals doped with Ce, Gd, Tb, Ho, Er. The absorption spectra of these crystals are characterized by bands at 520-550 (F-centers), 320 (V\$\_k\$-type) and 200 nm (F\$\_3^-\$). The relation between the absorption bands vary in different crystals. Due to the absorption bands at 500-550 nm crystals of this group become a pink colour.

The crystals of the second group (Nd, Sm, Yb, Tm) exhibit a strong absorption band at 200 nm (F\${\_3^-}\$ centres) and an order of magnitude less bands in the visible region of the spectrum. Due to the weak absorption in the visible region the crystals are almost colourless. The bands of Sm\$^{2+}\$-anion vacancy centres [3] and Yb\$^{2+}\$ bands at 310, 370 nm were recognized in LaF3-Sm and LaF3-Yb, respectively.

The absorption spectra of crystals irradiated at 77K were obtained and discussed also.

We conclude that the type of color centers created by x-irradiation of doped LaF\$\_3\$ depends on the electron affinities of Re\$^{3+}\$ and anion vacancy. The Re\$^{3+}\$ ions with higher electron affinities (deep traps) form the centers of RE\$^{2+}\$-anion vacancy. Other Re\$^{3+}\$ ions with lower electron affinities form the center Re\$^{3+}\$-F, which can be regarded as a perturbed F-centers. This conclusion is confirmed by preliminary quantum-chemical calculations and the estimation of the levels location in the energy-band diagram.

1. W. T. Carnall, G. L. Goodman, K. Rajnak, and R. S. Rana. J. Chem. Phys., 1989, 90, 3443-3457. 2. E.A.Radzhabov, Optics and Spectroscopy, 2016, 120, 294–299.

3. E. A. Radzhabov and V. A. Kozlovskiy. Bull. of the Russian Ac. of Sci. Physics, 2015, 79, 251–255

### **[154] Scintillation Efficiency and Position Sensitivity for Radiation Events in Plastic Scintillators** *Presenter: TRAN, Ngan N.T.*

A plastic scintillator (PLS) is a potential material for developing a dosimeter based on measurement of linear energy transfer (LET) for mixed radiation fields such as the accelerators' surroundings or in space because PLSs are composed mainly of hydrocarbon molecules, and have the effective atomic number and the density similar to those of water and human tissues. Additionally, there is a widespread application of PLSs in particle physics experiments due to their response to all types of radiation (photon, neutron, and charged particles). In order to employ PLSs in design and construction of LET spectrometer, some essential characteristics of PLSs are needed to be examined, such as the light yields as a function of energy deposited and radiation species, and the position sensitivity to observe tracks of incident radiation. Previously, we have performed the procedure for determining the energy deposited in a plastic scintillator (EJ-200) due to gamma rays from a Cs-137 radiation source using Compton Coincidence Technique (CCT). In the measurement, we found that the scintillation efficiency determined as a function of Compton electron energy approaches a saturation value at high energy region, and decreases in the region less than 100 keV. The decrease in the efficiency possibly pronounces the non-linearity between the light output and deposited energy. In this study, the scintillation efficiencies of PLSs (EJ-200, EJ-212, and EJ-252) are examined by focussing on the low energy region. In addition, the position sensitivity characteristic is examined by using four square-aligned PLS rods, multi-segmented photomultiplier tubes (PMTs) are attached to the both ends of the respective rods, and the positions of radiation incidence in the rod are measured by using signals from the PMTs. The experimental setups and results are given at the conference.

### [169] Proton-Induced Radiation Damage in BaF2, BGO, LYSO and PWO Crystal Scintillators

### Presenter: ZHANG, Liyuan

Because of their superb energy resolution and detection efficiency, inorganic crystal scintillators are widely used in HEP experiments. Future high energy physics experiments at the energy and intensity frontiers will face challenges of a severe radiation environment from both ionization dose and charged and neutral hadrons. The HL-LHC, for example, will present a very severe radiation environment, where up to 130 Mrad ionization dose, 3 × 1014 charged hadrons/cm2 and 5 × 1015 neutrons/cm2 are expected. In this paper, we report an investigation on charged hadron induced radiation damage in various crystal scintillators (BaF2, BGO, CeF3, LFS, LYSO and PWO) and a LFS/W/Quartz capillary-based Shashlik cell by using 800 MeV and 24 GeV protons at LANL and CERN respectively. Radiation-induced transmittance loss was measured in-situ before and immediately after radiation by 800 MeV proton beam at LANL. Degradations in both transmittance and light output measured after cooling-down of a few months are also reported. The results of these experiments provide important information for understanding proton-induced radiation damage in various fast crystal scintillators and their use in future HEP experiments at the energy and intensity frontiers.

### [170] Neutron-Induced Radiation Damage in BaF2, LYSO and PWO Crystals

### Presenter: ZHANG, Liyuan

One crucial issue for applications of scintillation crystals in future HEP calorimeters is radiation damage in severe radiation environment, such as at the HL-LHC. While radiation damage induced by ionization dose in inorganic crystal scintillators is well understood, investigations are on-going to understand radiation damage caused by hadrons, including both charged hadrons and neutrons. In this paper, we report an investigation on neutron induced radiation damage in BaF2, LYSO and PWO crystals irradiated at the Weapons Neutron Research facility of Los Alamos Neutron Science Center (WNR of LANSCE). Three groups of LYSO plates (6/each) of 14×14×1.5 mm3 were irradiated by fast neutron (>1 MeV) fluences of 2.2, 10 and 21 ×1014 n/cm2 respectively for 13.4, 54.5 and 118 days in 2015. The results show excellent radiation hardness of LYSO crystals against fast neutron. To address the issue of γ-ray background, three groups of 5 mm thick BaF2, LYSO and PWO plates with and without Pb shielding were irradiated to similar fluence. Degradations in both transmittance and light output are reported and compared to the damage induced by γ-rays and protons only.

### [229] Radiation damage tests of diamond and scintillation detector components for the ITER Radial Neutron Camera

*Presenter: CEMMI, Alessia* Radiation damage tests of diamond and scintillation detector components for the ITER Radial Neutron Camera

S. Baccaro, A. Cemmi, I. Di Sarcina, B. Esposito, G. Ferrara, A. Grossi, M. Montecchi, S. Podda, F. Pompili, L. Quintieri, M. Riva.

During the ITER reactor operation time, the plasma will give rise to high energy neutron and gamma flux and this intense radiation field will result in serious radiation damage and activation effects on various detectors components. In this work, neutron detector candidates for the ITER Radial Neutron Camera, i.e. diamonds and scintillator components (crystal and plastic scintillators, optical windows and PMTs), were investigated to establish their radiation hardness and stability under gamma ray and neutron irradiation.

Gamma radiation test were carried out at Calliope 60Co ENEA Plant in Rome. The facility is a pool-type irradiation plant equipped with a 60Co source (energy=1.25 MeV) with a current activity of 1.36 1014 Bq. Gamma radiation test were performed in the dark at room temperature for different total dose according to F4E requirements. Scintillators, PMTs and optical windows samples were irradiated up to 100 kGy absorbed dose while synthetic diamond samples up to 5 MGy. Scintillators and optical windows transmittance measurements were performed in the UV-VIS range (300-700 nm), paying particular attention to the behavior at 390 and 420 nm (scintillating emission wavelengths); samples were measured in the dark before and after irradiation and performance were monitored at room temperature for some weeks in order to study the damage recovery. For plastic scintillators photo-luminescence, optical bleaching and thermal annealing processes in air were made to reduce radiation damage. Quantum efficiency measurements were performed on the PMTs and the pulse height spectra and pulse shape capability of the scintillators were investigated by using gamma and neutron sources.

### Metamaterials (11:00-12:30)

### -Conveners: Martin Nikl

time [id] title

### **11:00 [203]** First step towards the design of metamaterials combining dense scintillator host with nanocrystals *Presenter: MARTINEZ TURTOS, Rosana*

In the search for prompt photon emission, colloidal semiconductor nanocrystals with supressed Auger recombination have proven to be a promising source of ultrafast light under ionizing radiation. Previous studies [\[1\]][1] have concluded in a sub-100ps effective decay time for CdSe 2D nanoplatelets and sub-1ns for CdSe/CdS giant shell quantum dots with an estimation of around 500 and 1000 photons per 511 keV emitted within the first 100 ps, respectively. However, the implementation of nanocrystal-based scintillators as a new generation of fast radiation detectors with sub-20ps time resolution requires that this amount of prompt photons is efficiently transported to the photodetector with improved SPTR and a minimum transfer time spread. In this work, we estimate the light coupling efficiency of drop-casted colloidal nanocrystal films deposited on the outer surface of optical fibers. Calculations are performed using the Wave Optics Module of COMSOL multiphysics software by implementing the high frequency homogenization method to reduce calculation time. Measurements of this quantity in terms of number of photons transported to the end of the waveguide are carried out under pulsed X-ray excitation up to 40 keV and SiPM readout. This study aims to maximize the amount of fast photons reaching the photodetector by correlating measurements and calculations in order to evaluate and conclude in an optimal nanocrystal-coupled-to-waveguide readout. It also constitutes the first step in the research path towards a design of an efficient radiation detector heterostructure combining fast nanocrystal photon-emission with dense materials suitable for electromagnetic calorimetry.

[1]: http://iopscience.iop.org/1748-0221/11/10/P10015/

### 11:15 [218] Perspectives On The Future Developments of Nano Scintillators

Presenter: SARAMAD, Shahyar

The spatial resolution of an indirect x-ray imaging detector is degraded by the light spreading phenomenon in scintillation layer. One way to improve the spatial resolution of these x-ray imaging systems with a thicker x-ray converter is preventing optical crosstalk between neighboring pixels by using segmented scintillators with optically isolated structures, which has applications in mammography, dental imaging, and micro-CT (computed tomography). By using microstructured scintillator layers in the form of thin and long ordered and densely packed needles of scintillator materials coupled to position-sensitive detectors, a higher spatial resolution can be achieved.

By using a new architecture based on the ZnO nanostructures proposed by our group at Amirkabir University of technology [1-3] a better spatial resolution in comparison to traditional imagers can be achieved. In the proposed imager, because of higher refractive index of the ZnO nanowires compared to their walls, each nanowire acts as a light guide (optical fiber) that prevents the generated optical photons to spread inside the imager. One of the advantages of ZnO nanowire scintillator is the simplicity of synthesized by template-assisted one-step electrodeposition technique. The results for ordered ZnO nanowire arrays in porous AAO template show that for 10 keV X-ray photons, by suitable selection of detector thickness and pore diameter, the spatial resolution less than one micrometer and detection efficiency of 66% are accessible. The simulation results also show that the conical frustum nanowires has better spatial resolution in comparison to cylindrical ones. The experimental light yield of ZnO nano scintillator is around 60% of the light yield of single crystal zinc oxide scintillator, which can be improved by suitable annealing or doping. According to the XRD results, the nanowires have a polycrystalline nature with a particle size around 20 nm. Concerning the scintillation spectrum, small but densely packed grains show dominant luminescence from the band gap transition in the UV and only minor contributions from defects. So, the X-ray excited optical luminescence (XEOL) of ZnO deposited by electrodeposition is due to band gap transition and has a peak at 390 nm.

In conclusion, better spatial resolution of this nano scintillator in comparison to bulk ones and the possibility of optimization its detection efficiency by increasing the porosity of the membrane and also it's thickness, are the advantages which candidate this nano scintillator for medical imaging or even high energy physics tracking in the future.

### References

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[3] A. Taheri, S. Saramad, S. Ghalenoi, S. Setayeshi, The European Physical Journal C 73 (2013) 1-7.

### 11:30 [190] Fast timing capabilities of the hybrid GRIFFIN array

Presenter: OLAIZOLA MAMPASO, Bruno

There is a growing interest in the nuclear physics community to develop hybrid detector arrays that combine the powerful energy resolution of the HPGe semiconductor detectors with the fast response of the LaBr\$\_3\$(Ce) inorganic scintillators. Examples of plans for such future arrays abound worldwide, like FATIMA for DESPEC, nu-ball at ALTO or the LaBr\$\_3\$-Gammasphere at Argonne and it is clear that, thanks to their versatility, they are going to play a key role in the following decades of experimental nuclear physics.

The Gamma-Ray Infrastructure For Fundamental Investigations of Nuclei (GRIFFIN) is a state-of-the-art \$\gamma\$-ray decay spectrometer facility at TRIUMF-ISAC in BC, Canada, used in \$\beta\$-decay experiments to study exotic nuclear structure. The core of GRIFFIN is an array of 16 large-volume HPGe clover detectors, but one of its strongest advantages is the variety of different ancillary detectors that can be coupled to the main array. SCEPTAR allows \$\beta\$-particle tagging with a maximum of \$80\%\$ solid angle coverage, while the 70 liquid scintillators of DESCANT allows for neutron-tagging and PACES has an array of 5 Si(Li) detectors with high-energy resolution for conversion electrons.

A new ancillary array of 8 LaBr\$\_3\$(Ce) detectors for \$\gamma\$-rays and a fast plastic-scintillator called Zero Degree for \$\beta\$-particles has been optimized for fast-timing experiments with GRIFFIN. The 51 mm x 51 mm cylindrical LaBr\$\_3\$(Ce) Saint Gobain BrilLanCe 380 crystals are coupled to Hamamatsu R2083 photomultipliers (PMT) with integrated pre-amplifier and amplifiers in the PMT base. After the signal amplification, the energy resolution is \$3.9\%\$ at \$^{137}\$Cs energy. After careful optimization of the CFD parameters, timing resolutions as good as FWHM\$\sim 200\$~ps and time-walks below \$\pm30\$~ps have been obtained for individual crystals using analog electronics.

The LaBr\$\_3\$(Ce) array has already been commissioned and it is foreseen to be present and operational for most of the experiments using GRIFFIN. Thanks to its good energy resolution and its exceptional timing properties, the LaBr\$\_3\$(Ce) array will be used to measure nuclear lifetimes of order of a few tens of picoseconds, using the General Centroid Shift Method. The near-future plans for the timing-array include the development of an active BGO shield for the LaBr\$\_3\$(Ce) crystals.

In this communication I will report on the careful optimization of individual detector and the excellent performance of the Zero Degree Scintillator and the LaBr\$\_3\$(Ce) array under online experiment conditions.

# 11:45 [193] Comparing BSO and BGO with different surface finishes as cost-effective, hybrid scintillation/Cherenkov detectors for TOF-PET

Presenter: BRUNNER, Stefan E

Before the introduction of L(Y)SO:Ce, BGO was used in many PET systems. Compared to L(Y)SO:Ce, BGO offers a number of superior properties, such as a higher stopping power, higher photoelectric-fraction, no intrinsic radiation, and cost-effectiveness. On the other hand, BGO was not thought to be a good scintillator for time-of-flight detection, due to its relatively low light yield and long scintillation decay time.

In recent years, TOF-PET detectors exploiting the fast nature of the Cherenkov effect have been proposed. For example, CRTs down to 70 ps FWHM have been achieved using PbF\$\_2\$ coupled to MCP-PMTs. One problem of this method is the low Cherenkov emission yield in the order of □10-20 photons per 511 keV annihilation photon, which complicates energy discrimination–a severe disadvantage in clinical PET.

We have recently observed a radioluminescence response in the 100 ps domain upon excitation of BGO with 511 keV photons, using time correlated single photon counting (TCSPC). This fast emission is likely to be connected with the Cherenkov effect. We have furthermore shown that this fast luminescence component can be used for improving the timing properties of BGO, while still using the scintillation signal for energy discrimination. This hybrid scintillation / Cherenkov scheme allows TOF detection while maintaining the previously mentioned advantages of BGO. For example, with BGO cubes of 3 mm x 3 mm x 3 mm we obtained coincidence timing spectra with FWHM values down to 200 ps FWHM. However, the timing histograms have non-Gaussian shapes with relatively high FWTM values, ranging from 1.3 ns to 3 ns for 3 mm to 20 mm long crystals, respectively.

Here, we show how the timing kernel of BGO can be improved for TOF-PET by optimizing the crystal surface roughness. We present an improvement of the FWTM by almost 50%, ranging from 0.6 ns to 1.65 ns for 3 mm to 20 mm long crystals, respectively.

A potential disadvantage when applying BGO in TOF-PET is a reduced rate capability due to the scintillation decay time of about 300 ns. We therefore investigate BSO (Bi\$\_4\$Si\$\_3\$O\$\_{12}\$) as a possible alternative for BGO. We show that with its 100 ns decay time and just 20% of the light yield, the FWHM can be improved by up to 20% compared to BGO. Moreover, while BGO can already be considered cost-effective compared to L(Y)SO:Ce, BSO allows substituting the relatively expensive GeO\$\_2\$ by SiO\$\_2\$ and, therefore, reducing the costs of material even further, while slightly improving the timing characteristics.

Coincidence timing results will be presented for BGO and BSO crystals with polished surfaces and with surfaces optimized for coincidence timing. Best results to date were achieved with BSO, viz. a FWHM of 192 ps and a FWTM of 725 ps (cubes with 3 mm edge-lengths). Moreover, temperature dependent TCSPC spectra will be presented for both materials. The results of this work show that the fast emission in BGO and BSO can be used to boost the timing performance of both materials.

### 12:00 [164] Light spread manipulation in scintillators using Laser Induced Optical Barriers Presenter: BLÄCKBERG, Lisa

As a diametric alternative to mechanical array fabrication, Laser Induced Optical Barriers (LIOB) is being explored for fabrication of high resolution and high sensitivity scintillator detectors for a number of imaging applications including Positron Emission Tomography (PET) [1], Single Photon Emission Tomography (SPECT) [2], and Computed Tomography (CT) [3].

With LIOB one can permanently change the crystal structure of a scintillator on a nano/micro scale using pulsed laser beam(s) tightly focused in a small focal volume. With this arrangement we can cause material optical breakdown which can be exploited to create crack(s), or lattice deformation and void formation in the focal point, depending on the crystal's thermo-mechanical properties and the nature of laser energy transfer to the crystal structure. The resulting so-called optical barriers will have a refractive index (RI) different from that of the crystal bulk, and their size, shape and RI can be engineered by carefully selecting laser parameters like pulse energy, pulse duration and repetition rate. Optical barriers incorporated inside the bulk of a monolithic detector block will redirect the scintillator light and can thus be used to manipulate and control the light spread in the detector.

We are exploring the capabilities of the LIOB technique for fabrication of thick LYSO:Ce detectors with depth of interaction (DOI) capability and single-side readout for PET. The behavior of these laser processed scintillators is between that of a monolithic block and a mechanically pixelated array. Given the flexibility in the optical barrier patterns that may be incorporated in the crystal, one important part of the detector development is the optical barrier pattern optimization. We are performing light transport simulations, using the Monte-Carlo code DETECT2000, to study the scintillation light spread in these laser processed crystals as a function of optical barrier pattern as well as characteristics of the barriers such as RI and roughness of the barrier-crystal interface.

Our results show that slab-shaped optical barriers, arranged in a 1 mm pitch pixel-like pattern all the way through a 20 mm thick crystal (resembling mechanically cut arrays) can provide up to 67% light confinement within individual pixels. The FWHM of the light response function (LRF) for a barrier RI of 1.0 and a smooth barrier-crystal interface was calculated at ~2.3 mm. A rougher interface increases the inter-pixel crosstalk but results in a depth-dependent width of the LRF that becomes wider further from the photodetector plane. The latter can be used for DOI extraction. We further studied the expected detector performance when the crystal is only processed partially through its thickness. Such barrier pattern shows improved transversal as well as DOI resolution compared to a monolithic detector especially close to the crystal edge.

Our experimental results show that the barriers can be placed at any depth within the crystal with barrier width as narrow as 2 microns. Through our preliminary results, we further demonstrate that the roughness of the barrier-crystal interface can be controlled.

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[2] H. Sabet, L. Bläckberg, D. Uzun-Ozsahin, and G. El-Fakhri, "Novel laser-processed CsI:Tl detector for SPECT," Med. Phys., vol. 43, no. 5, 2016.

[3] L. Bläckberg et al., "Scintillator-based Photon Counting Detector: is it feasible?," in 2016 IEEE Nuclear Science Symposium and Medical Imaging Conference, NSS/MIC 2016, 2016.

### 12:15 [221] Photonic crystals slabs applied to inorganic scintillators

Presenter: SALOMONI, Matteo

Photonic crystals slabs applied to inorganic scintillators

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The extraction of scintillation light from an inorganic scintillator is one of the major bottlenecks in Time-of-Flight Positron emission tomography (ToF-PET) as it directly affects the energy and time resolution of the gamma detector. The most widely used crystal in PET system is LYSO:Ce doped. Having a high index of refraction (IR = 1.82), the amount of light collected to the photo-detector is limited by total internal reflection (TIR): For crystals with high aspect ratios, as the ones used in PET scanners, up to 50% of the scintillation light will not be collected, even if Teflon wrapping and optical coupling are used. Photonic crystal slabs (PCS), defined as thin dielectric layers structured with a 2D or 3D periodic pattern, offer the possibility to increase this efficiency. A higher light output, combined with a reduction of the average path length of the photons in the crystal before their extraction, leads to a more precise evaluation of the particle time detection. This also implies a better coincidence time resolution (CTR). Together these will translate to ToF-PET reconstructed images with a better signal to noise ratio, which will lead to a better diagnosis, faster exams, and the possibility to reduce the patient dose. For our application, PCS need to be as well as an optical layer that is transparent for the emission spectrum of the LYSO. We have developed the nano-fabrication technology needed to realize the required large nanostructures needed to cover the scintillator readout face. This layer, when applied to the scintillator, helps overcome the TIR due to diffraction effects. Here we present our work focused on the simulations of the PCS parameters in order to optimize this effects. Using two different simulation tools, GEANT4 and CAMFR, we were able to take into account the geometry and configuration of the crystal to treat (wrapping, coupling, photo-detector, etc.). Optimization has been performed to find the best PCS pattern for a large variety of configurations. For the first time we produced centimeter size specimens patterned fully. We will present the results for LYSO that show a gain in light yield with respect to the same configuration without the PCS. Preliminary results shows also better performances from both the light yield and the timing point of view. In-depth analysis is currently underway. Details on the simulation steps, production process and characterization of the PCS will be presented. Acknowledgment:

This work was supported in part by the US Department of Homeland Security (DHS), Domestic Nuclear Detection Office (DNDO), under the competitively awarded contract(s) HSHQDC-13-C-B0040, and by European Research Council in the frame of the ERC Advanced Grant TICAL #338953. This support does not constitute an express or implied endorsement on the part of the Government. This work was also supported by the ERC Proof of Concept ULTIMA #680552, and was carried out in the frame of the Crystal Clear Collaboration (CERN).

<u>Conference Excursion: Martigny (Switzerland) through the scenic road. Visit of the city and of the Fondation Pierre</u> <u>Gianadda that will host a special exhibition with paintings from Cézanne.</u> (13:00-18:00)

<u>Conference Excursion: hike on the Balcon du Mont Blanc trail</u> (13:00-18:00)

<u>Conference Excursion: Train to Montenvers and the Mer de Glace (valley glacier) with a professional guide</u> (13:00-18:00)

### Thursday 21 September 2017

Scintillation Mechanisms: session 2 (08:30-10:00)

### -Conveners: Andrey Vasiliev

### time [id] title

### 08:30 [155] Free carrier absorption for study of fast excitation transfer in scintillation crystals

Presenter: TAMULAITIS, Gintautas

New requirements for timing of scintillator-based radiation detectors for future high energy physics experiments and medical imaging applications necessitate demand for novel tools to study the non-equilibrium carrier dynamics in scintillators on picosecond time scale. In this report, we summarize our current results on the application of free carrier absorption to study the carrier dynamics in several scintillators prospective for fast ionizing radiation detectors.

Our experiments have been carried out in pump and probe configuration. The non-equilibrium carriers were photoexcited by 200-fs-long laser pulses in visible or UV. The capability of photoexcitation to change the energy of excitation photons by using harmonics of the main laser radiation or gradually tune it by application of parametric oscillators was exploited for the resonant excitation of activator ions or structural units in the the host crystal. The change in optical absorbance induced by the excitation was probed in the infrared region. The change is caused by free carrier absorption. Its absolute value reflects the density of non-equilibrium carriers, while the spectral features might be exploited to characterize the density of states in the bands.

Both intrinsic (PWO) and Ce-doped scintillators (GAGG:Ce, YAGG:Ce) have been studied. The technique has been exploited to compare the excitation transfer in GAGG:Ce with and without codoping with divalent magnesium. In self-activated lead tungstate (PbWO4, PWO), the nonlinear optical response due to free carrier absorption occurs immediately after the short-pulse excitation. The possibility to use this fast response to provide precise timing capabilities in

novel radiation detectors is under discussion.

In GAGG:Ce, the signal is dominated by the absorption of free holes. The response peak occurs with a delay of few picoseconds due to hole delocalization from Gd3+ ground state to the valence band. The response in the garnet scintillator YAGG:Ce containing no Gd occurs immediately after the short-pulse excitation. The comparison of the results on free carrier absorption and those obtained by time-resolved photoluminescence spectroscopy enabled us to compile a electronic energy-level diagram of the main structural units of GAGG:Ce and GAGG:Ce,Mg to explain the influence of codoping with divalent Mg on the time characteristics and light yield of this scintillator.

### 08:45 [77] Some trends in the yield of the hot intraband luminescence

Presenter: OMELKOV, Sergey

Recent progress in various fields of scintillator applications has created a high demand for fast scintillators. In particular, the Time-Of-Flight Positron Emission Tomography (TOF-PET) technique requires coincidence time resolutions (CTRs) better than 100 ps FWHM in order to improve the image signal to noise ratio offering shorter scanning times, lower patient doses and better image quality. The CTR of 10 ps will potentially yield a breakthrough in PET allowing direct imaging without reconstruction. Developments in high energy physics also demand improving the timing capabilities of calorimeters down to 10 ps to distinguish several events per bunch crossing and make better use of high luminosity storage rings. Achieving such time resolution was set as an ultimate goal for European COST Action TD1401 "FAST".

Conventional Ce-doped scintillators were demonstrated to provide a CTR of about 73-120 ps FWHM when used with Silicon Photomultiplier (SiPM) detectors [1]. The time resolution in this case is limited by the photon time-density at the onset of the scintillation process and can be further improved only by using a different scintillation mechanism featuring a prompt response. It has been shown that already 40 prompt photons produced by 511 keV gamma quantum can significantly improve CTR below 70 ps, whereas for achieving 10 ps CTR about 500 are required together with corresponding advances in SiPM technology [1,2].

Hot intraband luminescence (IBL) is a good candidate for providing at least some of those prompt photons, alongside with Cherenkov radiation and quantum confinement driven luminescence [2]. IBL is a low yield ultrafast emission connected with the radiative transitions of hot electrons or hot holes between the sub-levels of the conduction or valence band of a crystal, respectively. The continuous and structureless spectrum of IBL covers the whole transparency region of a material, with increase of the intensity in NIR [3]. The IBL decay time and yield are defined by the competitive process of nonradiative transitions which are far more probable than the radiative ones. The decay time of IBL is expected to be below 1 ps, but the highest scintillation light yield (LY) measured so far is only about 30 ph/MeV (CsI at 120 keV electron beam excitation). Therefore, to achieve 10 ps CTR the LY has to be increased significantly. We demonstrate an inverse correlation between the LY and the energy of the highest phonon mode of the material, confirming the yield dependence on nonradiative transitions probability. However, the probabilities of radiative intraband transitions are not understood yet. Some materials, e.g. molybdates, display high spectral LY and simultaneously high-energy phonon modes, showing that the possibilities to increase LY are not yet exhausted. Extensive search of empirical trends in the LY as well as theoretical modeling are required to fully understand the IBL phenomenon. We have developed a technique which allows to compare the LY of solids in a reliable and reproducible way. The results of LY studies in various compounds will be reported, including alkali halides, binary oxides, silicates, garnets, tungstates, molybdates, simple and complex fluorides, oxyfluorides, etc.

[1] S. Gundacker, et al, J. Instrum. 11 (2016) P08008

[2] R. Turtos, et al, J. Instrum. 11 (2016) P10015

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### 09:00 **[103] Two-photon photoconductivity and luminescence in scintillators – measurement and mapping** *Presenter: ONKEN, Drew*

The radius of initial (femtosecond-scale) energy deposition around the local trajectory of a high-energy electron in a scintillator has been characterized as roughly 3 nm by several researchers, e.g. [1]. The resulting strong radial gradients of carrier density drive diffusion which can both dilute and separate carrier populations. This significantly influences the rate terms for capture, bimolecular recombination, and nonlinear quenching that govern the physical mechanisms of scintillation [2]. Particularly in halide scintillators, the holes self-trap rapidly and are initially confined near the track core, while electrons in the heavier halides persist as hot electrons for up to about 4 ps, during which time they outrun the self-trapped holes (STH) and thermalize or trap at a much larger radius than the STH. The subsequent evolution of the scintillation pulse starts from charge-separated carrier populations in a strong internal radial electric field. Thus carrier mobilities and diffusion coefficients are important characteristics of scintillator materials and essential input data for material engineering models. Inorganic scintillators most commonly are insulating crystals, and halide scintillators are often hygroscopic as well, so transport measurements can be challenging in this material class. Such data have been measured in some common alkali halides and a few oxide scintillator materials, but have yet to be measured in most of the recently developed high-performance scintillators, including rare-earth halides, alkaline earth halides, elpasolites, and materials with activator concentrations that can run to 10% or more.

This report describes experiments in which both photoconductivity and photoluminescence are measured under two-photon interband excitation of scintillator materials. Two-photon excitation enables generation of electron-hole pairs below the surface in the scintillator host. Use of sub-picosecond laser pulses allows resolving the very short carrier re-trapping times,  $\tau$ , expected in doped wide-gap scintillators in order to extract the mobility,  $\mu$ , from the measured  $\mu\tau$  product. Using the established principles of two-photon confocal microscopy, our experiment provides depth resolution and lateral resolution determined by the microscope objective. Therefore high-resolution 3-d mapping of luminescence response and somewhat lower resolution of photoconductivity is achieved.

We have previously reported simultaneous mapping of photoconductivity and two-photon luminescence with this system in the semiconducting scintillators LiInSe\$\_2\$ and ZnSe:Te [3]. The present report describes results on CsI and CsI:TI as a test of our experimental method applied to a well-characterized alkali halide scintillator preparatory to applying the technique in newer scintillator materials, including their modifications and effects of heavy doping. In addition to measurements of two-photon photoconductivity in CsI:TI, the mapping of TI\* luminescence with 2 µm resolution in our experiment has indicated a granularity of response with feature size in the 20 µm range in standard CsI:TI scintillators. This could indicate clustering or concentration of TI activator ions due to aggregation in the bulk or collection along low-angle grain boundaries. We are also using similar methods to investigate possible origins of double photopeaks in defective CsI:TI scintillators.

We acknowledge support from NNSA DNN-R&D under subcontract to LBNL in the Venture LB15-V-GammaDetMater-PD3Jf.

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[2] X. Lu et al., "Energy-Dependent Scintillation Pulse Shape and Proportionality of Decay Components for CsI:TI : Modeling with Transport and Rate Equations," Phys. Rev. Appl., vol 14007, 2017.

[3] K. B. Ucer et al., "Dual Detection Charge Collection and Light Emission in LiInSe\$\_2\$ and ZnSe," Oral Presentation R08-6, IEEE: RTSD Conference, 2016.

# 09:15 [123] Charge trapping processes and energy transfer in PbMoO4 studied by electron paramagnetic resonance and thermally stimulated luminescence

### Presenter: BURYI, Maksym

Lead molybdate (PbMoO4) crystal is isotructural to lead tungstate but it features a lower light yield. On the contrary, it is a promising candidate as scintillator for high energy physics and the neutrinoless double  $\beta$  decay experiment, due to the inevitable occurrence of the 100Mo isotope. PbMoO4 crystals were also suggested to find application in cryogenic dark matter experiments. Point defects (e.g., color centers, vacancies, accidental impurities etc.) responsible for trap states in a bandgap have a significant influence on the luminescence and scintillating properties of the crystal. The traps interfere with the charge transport process contributing to afterglow and degrading the light yield. Therefore, the present study is dedicated to an investigation of the charge trapping and energy transfer in lead molybdate.

Single crystals of undoped PbMoO4 irradiated with 420 nm light were studied by means of electron paramagnetic resonance (EPR) and wavelength resolved thermally stimulated luminescence (TSL). After light irradiation EPR spectra displayed two new. After a detailed analysis, both signals have been attributed to Mo5+ centers. Their spin Hamiltonian parameters have been determined and thermal decay characteristics studied. Both centers exhibit a relatively low thermal stability (limited to 90 K). The very good matching between EPR and TSL data allowed to correlate some of the observed TSL glow peaks within the 60-100 K temperature range to the EPR signals of the molybdenum centers. Additional TSL peaks were also detected in the 100-160 K temperature interval, possibly due to the depletion of non-paramagnetic traps. The traps are characterized by first order recombination kinetics: by using the partial cleaning procedure and initial rise method, the corresponding trap depths and frequency factors have been determined.

Remarkably, the temperature dependencies of TSL and RL emission spectra are drastically different: the former features a stepwise low energy shift of the maximum emission energy from 2.4 eV at 10 K to 2.15 eV at 300 K, whereas the latter displays an opposite trend, with a slightly lower energy shift of 0.08 eV in the whole temperature region. The

phenomenology is clearly due to the occurrence of several spectral components with different thermal stabilities in TSL and RL. The emission mechanisms in the two measurements are, therefore, different. The results are discussed by taking into account the origin of the different emission centers, their thermal stabilities, and the existence of different pathways of charge carrier recombination in case of TSL.

## 09:30 [104] Pulse shape analysis of individual gamma events - correlation to resolution and the possibility of its improvement

#### Presenter: GEKTIN, Aleksander

The recent measurement of non-proportionality of each of the decay components in CsI:Tl (fast, slow, and tail) [1] found opposite slopes of the fast and tail proportionality curves above about 10 keV. This suggests that combining both components could improve resolution by more than just the statistics of the additional photons. Indeed, Moszynski et al had shown this in studies of peaking-time and resolution in CsI:Tl several years earlier [2]. While modeling the results from [1], we considered that the relative amounts of the three decay components in the as-measured scintillation pulse probably do not represent the optimum linear combination to produce best total proportionality [3]. We presented an optimization analysis showing that a linear combination of the form "fast + 3.4 tail" produced a significantly flatter proportionality curve than the natural scintillation pulse "fast + slow + tail" [3]. However, to determine if an optimized linear combination could result in improvement of resolution requires applying a similar algorithm to each gamma event.

In this work we measured pulse shapes of individual gamma events in CsI:TI and other scintillators using an eMorpho multichannel analyzer as a digital oscilloscope. Decomposition of every scintillation pulse into three exponential decay components following [1] allowed to extract the weight of each component. It was found that the nominal energy resolution evaluated as FWHM over the position of the photopeak maximum in a reconstructed PH spectrum can be altered by scaling the weight of each decay component. The coefficients in the linear combination can be optimized to achieve an energy resolution which is better than the similarly reconstructed "natural" PH spectrum where the scaling factors are all unity, i.e. when the decay components preserve their as-measured amplitudes. The results are presently mixed due to a factor we believe is separate from the effect of an optimized linear combination of pulse shape components. Specifically, we measured the normal PH spectrum of CsI:TI without reconstruction in terms of decay components and found 5.8% resolution at 662 keV. Reconstruction in terms of three decay components lost resolution to 8.6%, which we believe is due to neglect of the rising part of the pulse in the present analysis scheme, losing both significant total counts and the proportionality information represented in that component. The important result of this study is that optimization of the weights of the three decay components improved the resolution of the reconstructed data from 8.6% to 6.6%. Thus while 6.6% is not a net improvement over 5.8%, it is a distinct improvement over 8.6% in the comparison of equivalent things, i.e. PH spectra represented as two different weighted sums of the decay components. If this direction continues to seem promising, such processing can be implemented in real time with available technology such as field-programmable gate arrays.

Some of the CsI:TI crystals we investigated exhibit a secondary peak in the PH spectrum. In the course of this study we used scanning absorption spectroscopy and spectrally resolved TSL measurements to understand the origin of the secondary peaks.

\*WFU acknowledges support of DNDO grant 2014-DN-077-ARI-077, with no express or implied endorsement on the part of the Government.\*

#### References

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2. M. Moszynski et al., "Energy Resolution of Scintillation Detectors - New Observations," IEEE TNS, vol. 55, no. 3, pp. 1062–1068, Jun. 2008.

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### 09:45 [153] Excitation density distribution effects on fast ZnO excitonic emission

#### Presenter: MARTIN, Patrick

ZnO crystals and nanoparticles have promising scintillation properties an subnanosecond scintillator. There is huge number of publications concerning laser excitation of ZnO. Nevertheless, it is still not clear how the properties of ZnO scintillation are connected with the physics of process in this crystal, which significantly differs from traditional inorganic scintillators being mostly ionic crystals.

The intensity and kinetics of ZnO emission strongly depends on the energy and fluence of excited photons both at ambient temperature and low temperatures. We perform systematic study of ZnO time-resolved luminescence: z-scan under intense interband femtosecond excitation 266 nm (3rd harmonic of Ti:Sa laser) and excitation by X-ray synchrotron radiation. Z-scan allows the systematic study of the luminescence intensity and decay characteristics in case when excitation density changes in space with characteristic scale of tens of microns. The results show non-monotonic behavior of luminescence intensity with excitation intensity. The possible reason of such behavior is the combination of (1) the increase of exciton production with the increase of electron and hole density and (2) the decrease of emission intensity and fastering the kinetics with the increase of exciton concentration.

The change of X-ray excitation energy result in nanometric changes in spatial distribution of excitations. The most prominent result is the excitation below (950 eV) and above (1100 eV) of Zn 2p core energy. We calculate the dramatic modification of the distribution of energy of Auger electrons created in cascade process, resulting in appearance of much higher number of Auger electrons with energy below 200 eV in case of 1100 eV excitation. Such electrons have small mean free paths (about and less than 1 nm) and therefore the spatial distribution of electrons and holes in track after 1100 eV absorption is characterized by higher number of dense regions in track structure. This analysis can be used to explain the increase of fast (about 100 ps) component in decay kinetics under 1100 eV excitation.

### Poster Session 3 (10:00-11:00)

### -Conveners: Marco Pizzichemi; Remi Chipaux; Kristof Pauwels

[id] title	boar d
[11] Light pulse decays and non-proportionality characteristics of CsI:Tl at temperatures down to -70°C	
Presenter: MIANOWSKA, Zuzanna	
Caesium iodides doped by thallium are one of the most popular scintillators used as gamma ray detectors whether in the scientific or application side. Despite such widespread use it is still ongoing work aimed at understanding the mechanism of light emission in these crystals. Some theoretical interpretation supported by experimental measurements have been already done and were presented in publications of A. Syntfeld-Kazuch et al., S.Gridin et al. or X.Lu et al. (see 1-3). But the theoretical models of light emission are still under development and experimental works are still needed. One of the crucial and unresolved questions are how light emission changed with:	f
- energy deposited,	
- temperature changing,	
- thallium concentration.	
For that reason light pulses of CsI:TI crystals straight from the PMT anode were collected and analysed. These pulses appears as a result of excitation of scintillators with gamma sources with energy from 17keV to 1274keV. Tested scintillators with variable thallium amount from 0.0008mole% till to 0.081mole% were checked. Additionally temperature dependence in a range from -70°C to +30°C was achieved. For quantitative description of light emission sum of exponentia functions were fitted to experimental data.	1
resolution equal to 5.15% and light output equal to 10000Phe/MeV (at 20°C and for 662keV). Decay time constants and a relationship between intensities of each of light component will be presented as temperature and gamma ray energy functions.	
References:	
[1] A. Syntfeld-Kazuch et al., IEEE Transactions on Nuclear Science 54, 1836 (2007).	

[2] S. Gridin et al., Journal of Physical Chemistry C 119, 20578 (2015).

### [18] Cerium-doped gadolinium fine aluminum gallate (Ce:GFAG) in scintillation spectrometry

### Presenter: IWANOWSKA-HANKE, Joanna

Scintillation crystals with high density and high atomic number coupled with photodetectors are commonly used in X-ray and gamma-ray spectrometry. There is continuous demand for new scintillation materials in such applications as industrial radiography, medical imaging techniques and nuclear and high-energy physics. In the case of modern scintillators, parameters such as high light yield, high gamma-ray absorption coefficient, good energy resolution, proportionality, and fast scintillation response, together with chemical and time stability and large sizes o the crystals, are of importance. These requirements were achieved by cerium-activated materials, such as Ce:LSO, Ce:LFS, Ce:GSO, and Ce:LaBr2, or Ce:LaCl3. The discovery of single crystal multicomponent garnet scintillators, based on YAG crystal with admixture of Ga and Gd, presented by Cherepy et al. [1] and Kamada et al. [2] provides new structures with high density and high atomic number. The emission spectrum of these crystals are about 520 nm, which makes them suitable for semiconductor photodetectors. The research on heavy ceramic scintillators led to the discovery of Ce:GAGG, which appeared to be one of the most attractive material for gamma-ray spectrometry, due to its high light output, fast decay time, good energy resolution and absence of intrinsic radioactivity [3]. The next step was the discovery of cerium-doped gadolinium fine aluminum gallate (Ce:GFAG), that has been reported by the manufacturer as faster decay time (about 50 ns of primary component of the light pulse) and better timing resolution than Ce:GAGG. High density of Ce:GFAG (Zeff = 52) and good energy resolution make it possible to apply in various nuclear technologies (PET, SPECT etc.). We have tested two samples of Ce:GFAG (both with dimension of 10x10x5 mm) in gamma-ray spectrometry with light readout through the photomultiplier (PMT) and the avalanche photodiode (APD). The parameters such as light output, energy resolution, non-proportionality, light pulses and timing resolution were measured.

The measurements of energy resolution obtained with Hamamatsu R-6231-100 PMT irradiated with 137Cs source gives the results of about 7 %, whereas for new samples of GAGG it was reported as about 6% [3]. The decay time measurements of Ce:GFAG gives the results significantly better that that of Ce:GAGG [3]: the fast component is at the level of 60 ns (54% intensity of the pulse) the medium one at about 120 ns (35% of the pulse). The third, slow component of the light pulse has a decay time of about 500 ns with only 10% intensity. A comparative discussion of GFAG performance in potential PET detectors to that of LSO, LFS, GSO and GAGG summarizes the study.

References:

[1] N. Cherepy et al Proceedings of SPIE 7079X, September 04, 2008

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### [19] Effects of lanthanum substitution on the band gap and luminescence properties of (Gd1-xLax)2Si2O7:Ce scintillator

### Presenter: WEI, qinhua

(Gd0.90, La0.09)2Si2O7:Ce0.01 crystals have been grown by different methods which had a high light output of 36,000 ph/MeV and excellent energy resolution (FWHM) of 5% at 662 keV. It is clear that the splitting of 5d levels of Ce3+, which play an important role on the luminescence properties of the scintillation materials. And the electronic structure perspective, the band gap and the location of lanthanide impurity levels both play important effects on the luminescence efficiency of scintillation materials.

Here, the (Gd1-xLax)2Si2O7:Ce (x = 0.1, 0.2, 0.3, 0.5, 0.7, 1.0) crystalline powders were synthesized by a sol-gel method. The thermoluminescence (TL), UV and X-ray excited luminescence spectra, photoluminescence decays as a function of wavelength and the temperature dependence of PL intensity were measured and discussed. Besides that, the influence of lanthanum on the electronic structure and band gap is calculated by first-principles method. The effects of lanthanum substitution on the band gap, luminescence properties and energy transfer mechanism are investigated precisely. The measurements and observations reveal that with the increase of La content, the entire glow curve shifts to lower temperatures, qualitatively indicating the lowering of conduction band edge, which has the same tendency with the calculated results by first-principles. Therefore, it is concluded that the scintillation efficiency deterioration with the increase of La content comes from the enhancing unwanted excited-state ionization of emission center. The activation energy for thermal quenching of samples is tunable from 0.4eV to 0.8eV with different La content. Besides, the energy transfer is occurred from Gd3+ to Ce3+, which can switch from a short-range exchange mechanism to a longer-range dipole–dipole mechanism.

### [31] Scintillation properties of Gd3(Al5-xGax)O12:Ce; x = 2.3, 2.6, 3.0 single crystals

### Presenter: CHEWPRADITKUL, Warut

Scintillation properties of Czochralski-grown Gd3(Al5-xGax)O12:Ce; x = 2.3, 2.6, 3.0 single crystals were investigated. The light yield (LY) and energy resolution were measured using an R6231 photomultiplier. LY non-proportionality and intrinsic resolution were evaluated. At 662 keV  $\gamma$ -rays, the LY value of 46,200, 65,800 and 59,400 ph/MeV was obtained, respectively, for Gd3(Al2.7Ga2.3)O12:Ce, Gd3(Al2.4Ga2.6)O12:Ce and Gd3(Al2Ga3)O12:Ce. In spite of a lower LY value, Gd3(Al2.7Ga2.3)O12:Ce shows superior energy resolution (5.4%) among the studied crystals due to its better proportionality of LY. The ratio of LY values under excitation with  $\alpha$ - and  $\gamma$ - rays ( $\alpha$ / $\gamma$  ratio) was also determined. Scintillation decay time and coincidence time resolution were measured in order to investigate the timing characteristics in the scintillation response. The dependence of LY on sample height was also measured and light loss coefficient was evaluated.

Keywords: Coincidence time resolution; Energy resolution; Gd3(Al,Ga)5 O12:Ce; Light yield; Nonproportionality of light yield; Scintillation decay

## [32] Comparative study of GdLu2Al2Ga3O12:Ce and GdY2Al2Ga3O12:Ce scintillation crystals for $\gamma$ - ray detection

### Presenter: SAKTHONG, Ongsa

The scintillation characteristics of Czochralski – grown GdLu2Al2Ga3O12:Ce and GdY2Al2Ga3O12:Ce single crystals were compared for y - ray detection. At 662 keV y - rays, light yield (LY) of 33,900 ph/MeV and energy resolution of 14.3% obtained for GdLu2Al2Ga3O12:Ce are inferior to those of 37,500 ph/MeV and 10.6% obtained for GdY2Al2Ga3O12:Ce. Scintillation decays were measured using the time-correlated single photon counting technique. A fast component decay time of 48 ns with relative intensity of 71% obtained for GdLu2Al2Ga3O12:Ce is superior to that of 106 ns (57%) for GdY2Al2Ga3O12:Ce. The coincidence time resolution were measured in reference to a fast BaF2 scintillator and discussed in terms of a number of photoelectrons and decay time of the fast component.

### [33] Intrinsic energy resolution in CeBr\_3 detector

### Presenter: RAWAT, Sheetal

CeBr\$\_{3}\$ is emerging as one of the best scintillation detectors having properties similar to Cerium doped lanthanum halide detectors. Due to its lower internal activity and stability at higher temperatures, CeBr\$\_{3}\$ is found to be suitable for many space applications and field explorations.

Energy resolution is one of the important parameters which greatly affects the productivity of the detector. Researchers have figured out that the major limitation to the resolution is the intrinsic resolution of the detector, which is proposed to arise due to non-proportionality of light output of the crystal, scattering of electrons ( $\delta$ -rays), landau fluctuations etc [\*\*1\*\*]. The actual origin of intrinsic resolution is yet to be fully understood. A Compton spectrometer based method to measure the intrinsic energy resolution and light yield non-proportionality of scintillation detectors was first proposed by John D. Valentine and Brian D. Rooney [\*\*2\*\*]. \*Swiderski et al.\* [3] have done a significant work in this field by using Compton Coincidence Technique (CCT) to determine the non-proportionality and intrinsic energy resolution of Compton electrons in LaBr\$\_{3}\$:Ce, LYSO:Ce, CsI:TI, etc. The basic principle of the CCT method is to register in coincidence the signals from scattering of gamma ray inside the tested scintillator followed by absorption of the scattered ray inside the reference detector. In the present work, we have used CCT to determine the intrinsic energy resolution of Compton electrons in CeBr\$\_{3}\$ using an n-type HPGe as the reference detector.

HPGe and CeBr3 detectors were kept face-to-face at a distance of 4 cm. A Cs-137 source was placed between them. The CeBr\$\_{3}\$ crystal was 1" in diameter and length, while the HPGe detector was a coaxial type reverse electrode detector of 2.18" diameter and 2.36" length. CeBr\$\_{3}\$ was biased with 680 V and was coupled to R6231 PMT. HPGe was given a negative voltage of 4500 V. We have used a PIXIE-4 module which is a multi-channel data acquisition system incorporating internal amplification system. In PIXIE-4, coincident data acquisition across channels is possible. Therefore, no external coincidence module was required. The signal from HPGe was directly fed to channel 0 of the PIXIE-4 module and that from CeBr\$\_{3}\$ to channel 1 of the module. The hit pattern of PIXIE-4 was so adjusted that only the signals detected in coincidence in channel 0 and channel 1 were recorded. The coincidence window was set at 1  $\mu$ s. By using offline gating and analysing the Compton edge peak we determined the intrinsic resolution using ref. [\*\*3\*\*]. In case of Cs-137, the intrinsic resolution was evaluated to be 4.48 ± 0.01% (at 477.6 keV). Work is in progress to measure the intrinsic resolution for different gamma energies.

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### [39] Study of the time response of a CLYC scintillator

Presenter: VEDIA, Victoria

The Cs\$\_2\$LiYCl\$\_6\$:Ce scintillator (CLYC) has become recently available in the market as a scintillating material with gamma neutron discrimination capabilities.

It is a viable replacement as neutron detector for \$^3\$He proportinal counters based on pulse shape discrimination. In addition, its moderately good resolution for gamma detection, below 4% FWHM at 662 keV, makes it a good candidate for gamma detection. It therefore possesses a very high potential for implementation as dual neutron and gamma detector for radiation monitoring, personal radiation detectors and other devices. Additionally, the Ce doping provides a fast decay constant of the order of 50 ns, which makes it a suitable candidate for fast timing measurements that could combine the detection of neutrons and gamma rays.

In this work we have measured the time response of a 1-inch cylindrical CLYC crystal, commercially available from RMD, and coupled to a fast photomultiplier and several Silicon photomultipliers. The time resolution was measured with time-delayed coincidence set-up based on analog electronics against a small reference BaF\$\_2\$ detector with ultra-fast time response. The parameters of the electronics were tuned to achieve the best time resolution. Alternatively a fully digital processing using a 5 Gs/s digitizer has also been implemented.

The energy resolution and energy linearity for gamma rays has been studied as well.

A good time resolution is obtained at \$^{60}\$Co energies and for 511-keV gamma rays, which makes CLYC a competitive crystal for timing measurements. We will describe the results as a function of the external parameters and the signal processing strategies.

### [42] Scintillation and timing characteristics of 1-inch diameter CeBr3 scintillator single crystal

### Presenter: YOSHINO, Masao

Recently, focus on halide scintillator crystals has increased due to the high light yield and energy resolution originated from the small band-gap. The relationship between light yield and band-gap of host material has been investigated for various scintillator materials, and previous reports indicate that light yield was inversely proportional to the bandgap [1], [2]. In addition, energy resolution is inversely proportional to the square root of photons detected by scintillators. In other words, the scintillator with the small band-gap of host material indicates the high light yield which results in high energy resolution. On the other hand, in many gamma-ray imaging applications such as Compton cameras, gamma cameras, positron emission tomography (PET) and single-photon emission tomography (SPECT), scintillators require such properties as good energy resolution, high stopping power, and high light yield. Especially for applying to PET detectors, the timing characteristic of the scintillators is one of the most important factors in determining the localization of the positron annihilation event. For these applications, the CeBr3 crystal is currently gathering worldwide attention due to great high light yield, good energy resolution and fast decay time[3].

In this study, CeBe3 bulk single crystals with 1-inch diameter were grown using the Bridgman-Stockbarger technique and special-shaped crucibles. We will report on the growth of the 1-inch size CeBr3 single crystal and the preliminary test of the same sample cut in the size of  $2 \times 3 \times 3$ mm3 with polishing. An energy resolution of 3.8 % (FWHM) and fast decay time of 20.2 ns was obtained for 137Cs. I also obtained the nonproportionality with a deviation of  $\pm 1.7\%$  in the 31-661 keV gamma-ray and X-ray energy range.

In addition, we measured the timing characteristics of the CeBr3 coupled with MPPC array. The signal from the MPPC array is first sent to newly developed 48 channel current comparing type time-over-threshold ASIC, then sent to the oscilloscope. In this presentation, we will report on growth, scintillation properties, and timing characteristics of CeBr3.

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### [53] Reduced Afterglow CsI:Tl,Sm for High Energy Imaging

### Presenter: NAGARKAR, Vivek

CsI activated with thallium is one of the most important scintillators in the radiation detection field. Long a standard for medical diagnostics, it has also become particularly attractive for radiographic scanning and active interrogation of cargo in transit via trucks or rail. CsI:Tl has one of the highest conversion efficiencies (60,000 photons/MeV) of all scintillators in current use, which along with its high density and effective atomic number, excellent light transport properties, broad commercial availability, and low cost, make it the detector of choice for a wide variety of applications. It does have one drawback, however, in its relatively long-lived afterglow, which restricts its use in high count-rate applications, causing reconstruction artifacts in CT, and reduced contrast and image blurring in high speed radiographic scanning and imaging. Addressing this issue, we report here on our development of an approach to reduce this limiting afterglow by a factor of ~50 without significantly degrading its other scintillation properties. This approach, which involves codoping of the material with small amounts of Sm2+ or Eu2+, has been demonstrated to be effective even at energies as high as the 7 MeV level relevant for use in typical cargo inspection systems. The improved CsI:Tl scintillator is a straightforward replacement to the standard material, increasing contrast and steel penetration while allowing dual-energy performance, thus enabling reduced scan times and increased speeds for the same performance level. This in turn leads to greater throughput efficiency at ports and borders, where imaging systems often create choke points in the workflow, as well as more efficient detection of contraband such as explosives, drugs, weapons, and special nuclear materials.

We have also produced CsI:TI,Sm in the form of microcolumnar films, where it exhibits the same reduction in afterglow as it does in the single crystal form. We currently have the capability to produce such films up to 800 microns thick and up to 45 x 45 cm2 in area, and expect yet further development in the future. This provides a combination of high-resolution, high-frame-rate, and large-area capabilities not hitherto available for imaging applications.

This work has been supported by the U.S. Department of Homeland Security, Domestic Nuclear Detection Office, under competitively awarded contract/IAA HSHQDC-15-C-B0032. This support does not constitute an express or implied endorsement on the part of the Government.

### [59] Optical and radioluminescent properties of ZnO, ZnO:Ga and ZnO:In ceramics

### Presenter: CHERNENKO, Kirill

Zinc oxide has a variety of practical applications due to its unique properties. In particular, high radiation hardness and the presence of an ultrafast emission component make this material prospective for scintillators [1]. As a rule, various forms of ZnO exhibit two luminescence bands. One of them is a narrow UV band located near the absorption edge of the crystal; therefore it is called near-band-edge (NBE) emission. Another one is a broad band, its maximum usually placed in the green spectral range, this luminescence with decay time in microsecond range is associated with deep-level (DL) emission. It is known that the introduction of elements of group III: In or Ga in ZnO leads to an increase in the intensity of fast NBE luminescence and reduction in the intensity of the slow component [1].

This work presents the results of the complex study of ZnO, ZnO:Ga and ZnO:In scintillation ceramics which were prepared by hot uniaxial pressing technique [2].

The obtained ZnO ceramics with thickness of 0.5 mm show the total transmittance of 55% at 550 nm wavelength, which is a rather high value for ceramics based on a material possessing a hexagonal structure. The transmittance of ZnO:In and ZnO:Ga ceramics reaches maximum near 500 nm and then begins to drop with wavelength. Increase in the dopant concentration leads to growth of absorption in long-wavelength spectral range due to creation of large amount of donor centers. Maximum value of transparency in total transmittance spectra is determined by average grain size: the finer the grain the smaller the total transmittance.

Reflectance spectra of the ceramics have been measured in the IR range to estimate concentration of free charge carriers. Minima of the spectral reflectance are located at 14.90, 3.75 and 6.88 um for ZnO, ZnO:Ga and ZnO:In, respectively. As expected, the free charge carriers concentration in ZnO:In(0.1 wt.%) (n=1.53×10<sup>19</sup> cm<sup>-3</sup>) and ZnO:Ga(0.1 wt.%) (n=3.439×10<sup>19</sup> cm<sup>-3</sup>) is much higher than that in ZnO ceramics (n=3.26×10<sup>18</sup> cm<sup>-3</sup>). Measurements of radioluminescence spectra show that introduction of indium or gallium suppresses the DL band and increases the intensity of the NBE band (peaking at the 388.3 and 389.6 nm for In and Ga, respectively). Maximum of NBE luminescence intensity is reached at dopant concentration 0.13 wt.% and 0.05 wt.% for ZnO:In and ZnO:Ga, respectively. Radioluminescence decay curves of the ceramics have been studied. The main decay constant of the ZnO:In(0.13 wt.%) ceramics is 0.7 ns. Annealing of the ZnO:Ga and ZnO:In ceramics in the Ar:H2 atmosphere allowed to increase the light yield of the fast component by ~ 6 times. Physical processes responsible for luminescence kinetic and spectral properties of the studied ceramics are discussed.

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P. Rodnyi, K. Chernenko, O. Klimova et al. Rad. Measurements. 90, 136 (2016).

### [60] CeBr3 - a well characterized new scintillator for gamma-ray spectrometry

### Presenter: WESTMEIER, Wolfram

Pure crystals of CeBr3 material were initially produced by Schott Lithotec (Mainz) who then transferred production of several types of pure crystals to the enterprise Hellma Materials GmbH (Jena). CeBr3 is a scintillating material which is very suitable for photon spectrometry in the X-ray and gamma-ray range, in particular because of its high quantum yield. When properly coupled with a photomultiplier the resulting detector unit can serve for X-ray and gamma-ray spectrometry in successful competition with new high-resolution crystals like e.g. the proprietary material LaBr3:Ce which has a resolution (i.e. FWHM at 662 keV) of around 3%. The resolution of peaks in spectra measured with CeBr3 is typically around 4%. The somewhat poorer resolution of CeBr3, however, is well compensated by the fact that the detector is free of intrinsic radioactive material, like the natural abundance of 139La in lanthanum, and its 227Ac contamination is significantly smaller than in LaBr3:Ce.

Having apparent advantages of CeBr3 detectors in mind, spectral properties of the material were surveyed [1] and applicability of CeBr3 detectors for spectrometric applications was tested.

As most users of scintillation spectrometry have in the past worked with Nal(TI) detectors, the comparisons are mostly focused on that material as a reference. Emphasis is laid onto the systematic display of properties of CeBr3 detectors, such as the full-energy peak efficiency function or the resolution function as function of photon energy, as these functions are relevant for quantitative peak analysis in measured spectra. To enable quantitative peak analysis the intrinsic peakshape of CeBr3 gamma-ray peaks was investigated and the resulting peakshape function was tested in analyses of spectra. For qualification of the CeBr3 material the internal contamination through 227Ac and its influence on the spectrum were experimentally determined.

Modern scintillation spectrometers often consist of a detector with PMT, plug-on multichannel analyzer and notebook PC which controls the unit and provides power to the MCA for over five hours of running time. Thus, portable scintillation spectrometry using a CeBr3 detector is well suitable for in-situ measurements such as for example:

- Environmental survey
- Survey and quantification in nuclear medicine
- Supervision in and around nuclear facilities
- Boarder transit controls
- Production water and scales in the oil and gas industry
- Prospecting

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### [78] Afterglow and quantum tunneling in Ce-doped lutetium aluminum garnet

### Presenter: MIHÓKOVÁ, Eva

Quantum tunneling between the activator and a nearby trap(s) is a phenomenon not infrequently encountered in scintillating materials. We confirmed its presence both theoretically and experimentally [1] (and references therein). Experimental evidence in a variety of Ce and Pr doped complex oxides was given by the delayed recombination technique, primarily developed to study the activator's excited state thermal ionization. In the delayed recombination measurement the slow tails of the luminescence decay after selective pulse excitation are monitored typically in the millisecond time window. The signal after successive pulses is accumulated for several minutes.

In this work we use an alternative technique to study quantum tunneling between the Ce3+ activator and host traps in Lu3Al5O12. We monitor an afterglow of several Ce3+ doped Lu3Al5O12 samples in the crystal and powder forms, as well as those Mg codoped, for several tens of minutes. Unlike in the usual afterglow measurement we do not use an excitation by ionizing radiation, we rather selectively excite by continuous light source directly into 5d1 Ce3+ excitation band. Such experiment (after initial emptying the traps) ensures that at low temperatures the traps cannot be filled by any other process than quantum tunneling from Ce activators. After excitation cut-off an afterglow signal is recorded and an inverse power-law function is fit to the data. Power-law exponent close to 1 confirms that the observed signal is due to quantum tunneling between the Ce activators and traps [2]. The experiment is performed in the extended temperature range within 8-400 K. The results obtained for different samples are compared and discussed.

\*\*Acknowledgement\*\*

This research has been supported by the Czech Science Foundation grant GA 17-06479S.

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### [82] Efficiency studies on Gd\_3Ga\_3Al\_2O\_12 scintillators: Simulations and measurements

### Presenter: RAWAT, Sheetal

For \$\gamma\$-ray spectroscopy, apart from having high light output, good energy and timing resolution, it is the high efficiency (both detection and photo-peak) of the detector which is of paramount importance. The detection efficiency is a measure of the percentage of radiation that a given detector detects from the overall yield emitted from the source. In the category of inorganic scintillators, recently developed cerium doped Gd\$\_{3}\$Ga\$\_{3}\$Al\$\_{2}\$O\$\_{12}\$ (GGAG) not only has high density (6.7 g/cm\$^{3}\$) and atomic number (55) but also has high light output (55,000 ph/MeV), nearly 6% (at 662 keV) energy resolution and 550 ps time resolution \*\*[1]\*\*. These properties make GGAG a suitable detector choice for many applications.

Several studies on efficiency measurements were made in the past with different types of scintillators. \*Melcher et al.\* \*\*[2]\*\* have measured photo peak efficiency of BGO and CdWO\$\_{4}\$ relative to NaI:TI as 3.3 and 3.2 respectively. \*G. Anil Kumar et al.\* \*\*[3]\*\* have carried out detailed energy dependent studies of total detection (TDE) and photo peak (PE) efficiency of 1"\$\times\$1" LaBr\$\_{3}\$ both experimentally and using GEANT4 simulation toolkit. In the present work, we have carried out detailed Monte Carlo simulations of absolute efficiency (both TDE and PE) of GGAG scintillators of different sizes for gamma rays up to energy of 5 MeV. Experimental measurements were also done to validate the simulation results.

A 18\$\times\$10 mm\$^{3}\$ GGAG:Ce crystal was mounted on a vertically setup Hamamatsu PMT. Sufficient layers of 0.1 mm thick teflon reflector was wrapped around the crystal and an optical grease was used to couple its polished surface to the PMT. We have used four gamma sources, namely, \$^{137}\$Cs (115 kBq), \$^{134}\$Cs (152 kBq), \$^{60}\$Co (104 kBq) and \$^{22}\$Na (60 kBq) for energy calibration and efficiency calibration. TDE and PE data was obtained by keeping \$^{137}\$Cs source above the detector assembly at various distances for the duration of 1000 s. After the background and dead time correction, TDE and PE at 662 keV for GGAG were measured to be (9.22±0.01)% and (3.87±0.01)% respectively. Whereas, simulated TDE and PE values were 10.53% and 3.92% respectively. These high efficiency values are in accordance to our estimation owing to crystal's high density and atomic number. ## References ##

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## [84] Response of alpha particles in GAGG:Ce scintillators and correlation with non-proportionality patterns down to 0.1 keV

### Presenter: SIBCZYNSKI, Pawel

The response of GAGG:Ce crystals with different Al/Ga ratio [1] to alpha particles, with the energies of 1.5 MeV up to 8.8 MeV, were studied in relation to that of gamma rays. It was done using ThC source and 241Am source with Si absorbers. The alpha-to-gamma (α/y) ratio, described as a ratio of the pulse amplitudes normalized to the energy unit, determined in relation to 662 keV gamma ray energy was also plotted versus a speed of the detected alpha particles and secondary electrons due to detected gamma rays. It allows to present a non-proportionality curve plotted vs velocity of detected particles covered response to alpha particles and gamma rays of a typical spectrometry range of 16 keV up to 1274.5 keV. Then, basing on alpha particle velocity, we calculated the energy of an electron, which would have the same velocity as the alpha particle. This technique was presented in [2], where scintillator was exposed to protons from cyclotron. In the presented study, use of alpha particles allows to get a non-proportionality characteristics of GAGG:Ce crystals down to about 0.1 keV of equivalent energy. The calculation provides new data and methods of scintillation non-proportionality characterization down to a very low energy with excellent precision, superior to these obtained with monochromatic synchrotron X-rays [3].

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# [86] Temperature Quenching of Radio- and Photoluminescence of Y3(Ga,Al)5O12:Ce3+ and Gd3(Ga,Al)5O12:Ce3+ Garnet Ceramics

### Presenter: VENEVTSEV, Ivan

Cerium doped mixed oxide garnets are promising luminescent materials to use as LED phosphors or scintillators. Their properties can be adjusted via composition engineering for use in a specific application [1]. The cubic crystal structure allows preparation of highly transparent scintillation ceramics, which is a more cost efficient process than crystal growth. Every change in the composition affects multiple parameters, meaning that the final composition is usually a compromise between several significant factors. One of these factors is the temperature stability of the scintillator. For many applications it is crucial to use a scintillator that shows stable signal in the range of operating temperatures.

The main goal of this study is the characterization of  $Ce^{3+}$  doped garnet temperature dependent properties and the analysis of mechanisms responsible for said properties. Luminescent properties of the following samples have been studied: Y<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce<sup>3+</sup> (x=1, 2, 3, 4) and Gd<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce<sup>3+</sup> (x=1, 2, 3). The temperature dependence of the Ce<sup>3+</sup> luminescence intensity has been measured using continuous X-ray excitation (40 kV, 10 mA). Measurements of the Ce<sup>3+</sup> luminescence lifetime have been performed at different temperatures using pulsed UV (445 nm) excitation. The samples show strong Ce<sup>3+</sup> 5d<sub>1</sub>  $\rightarrow$  4f band emission in the 500-600 nm range. The maximum of the Ce<sup>3+</sup> emission experiences a blue shift with increasing Ga content. For all samples the Ce<sup>3+</sup> luminescence decreases upon heating above

certain temperature. This process is usually called temperature quenching (TQ). According to modern concepts, TQ in Ce doped garnets is a result of thermal ionization of electrons from the  $Ce^{3^+}$  excited  $5d_1$  state [2,3]. Replacement of Y with Gd or admixture of Ga into the compositions causes TQ to start at lower temperature meaning that the energy gap between the  $Ce^{3^+} 5d_1$  excited state and the bottom of the conduction band decreases. Values of this energy gap have been determined for every composition by fitting the measured temperature dependence of the decay time with the Mott formula. Comparison of obtained values with literature is given.

While Y<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce<sup>3+</sup> samples have almost constant luminescence intensity at low temperature, Gd<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce<sup>3+</sup> show a significant decrease in intensity upon cooling from room temperature to liquid nitrogen temperature (negative TQ). An increase of the Ga content shifts the negative TQ region to even lower temperatures. Thermally stimulated luminescence has been measured for every composition. In the temperature region where the negative TQ takes place in Gd<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce<sup>3+</sup> samples, large thermoluminescence peaks have been observed which led to the conclusion that this behavior is most probably the result of significant localization of charge carriers on traps. References: K. Kamada, T. Endo, K. Tsutumi, Cryst. Growth Des. 11 (10), 4484–4490 (2011) G. Blasse, W. Schipper, J.J. Hamelink, Inorganica Chimica Acta, 189, 77-80 (1991) J. Ueda, P. Dorenbos, A.J.J. Bos et al., J. Phys. Chem. C., 119, 25003–25008 (2015)

### [87] Luminescence properties of rare earth ions in novel garnets and glasses

### Presenter: VAITKEVIČIUS, Augustas

Garnet structure is a very flexible crystalline platform which allows one to control the luminescence properties by engineering the excitation transfer processes. The ease of material engineering in garnet structures allows designing materials consisting of lighter ions that are preferable for applications in high energy physics experiments in a harsh radiation environment. Meanwhile, two component glasses might be fabricated by using inexpensive and unsophisticated production processes and, thus, are attractive, first of all, for rapid prototyping of photoluminescence (PL) and scintillation properties. The amorphous glass structure makes it possible to reach high rare earth doping concentration. This leads to more efficient excitation transfer and smaller emission decay lifetime.

In this study two sets of samples were investigated. The first set consists of modified cerium doped yttrium aluminum garnets. Yttrium atoms have been partially substituted by magnesium or calcium atoms, while aluminum atoms have been partially substituted by germanium: Y2MgAlGe(AlO4)3:Ce and Y2CaAlGe(AlO4)3:Ce. The second set of samples consists of two-component silicate glasses doped with different rare earth ions. Barium, lithium, calcium and strontium silicate glasses doped with cerium, terbium, dysprosium, and europium in different combinations and concentrations were studied. The change in optical properties after transformation from glass to glass ceramics was also investigated. The samples were studied using confocal PL spectroscopy. PL spectra were measured using a spectrometer with a thermally cooled CCD camera coupled via light fiber to a WITec Alpha 300 S microscope system equipped with an objective of high numerical aperture. This setup enabled us to perform spatial mapping of PL parameters with sub-micrometer in-plane resolution. A He-Cd laser emitting at 442 nm (2.8 eV) and CW laser diode emitting at 405 nm (3 eV) were exploited for excitation. Photoluminescence excitation spectra measurements and XRD crystal structure investigation were also performed.

The study of garnet samples showed that the partial substitution of yttrium by magnesium results in a red shift of cerium ion emission, which is by 7 mm larger than that in the garnet with partial yttrium substitution by calcium. The spatially-resolved study of different aggregates in the garnet samples revealed a high degree of homogeneity of emission spectra among the different aggregates. This is an indication of high structural quality of the garnets.

The PL spectra of different glasses and glass ceramics showed that the emission spectra are insensitive to the composition of the host matrix in cerium-doped glasses, but are substantially transformed in glasses doped with europium. The emission spectra of different combinations of rare earth ions in glass matrixes of different composition have also been studied and discussed.

### [89] Scintillation parameters improvement of LuAG:Ce epitaxial films by Mg co-doping

### Presenter: PRUSA, Petr

LuAG:Ce (Lu\$\_3\$Al\$\_5\$O\$\_1\$\$\_2\$:Ce) garnet scintillator is a scintillator of decent light yield and energy resolution, excellent chemical stability, very good mechanical properties, and fast scintillation decay. However, the LuAG:Ce response is negatively affected by traps, mainly related to Lu\$\_A\$\$\_1\$ antisite defects produced at high temperatures. Traps are responsible for very high intensity of slow scintillation response component and energy losses during the migration phase, therefore light yield decrease.

During the last decade, several approaches have been adopted to overcome the negative effects of traps. So-called band-gap engineering is based on targeted band positioning achieved by optimization of host matrix composition, resulting in trap inactivation. Alternatively, methods of growth at lower temperatures could be used to avoid emergence of antisite defects, e.g. liquid phase epitaxy or methods of ceramic scintillators production. Finally, co-dopants are applied as well, especially divalent Mg and Ca in case of LuAG:Ce.

Mg co-doping turned out to be particularly successful for LuAG:Ce [1,2] and YAG:Ce (Y\$\_3\$Al\$\_5\$O\$\_1\$\$\_2\$:Ce) [3]. Several researchers have reported increase of light yield, energy resolution improvement, and slow scintillation component intensity and afterglow reduction. Divalent ions presence changes Ce\$^3\$\$^+\$ ions to Ce\$^4\$\$^+\$ ions. It is widely accepted that Ce\$^4\$\$^+\$ center competes for electron with traps much more effectively than Ce\$^3\$\$^+\$ centers, which must at first capture a hole to be able to capture an electron. Therefore, less electrons are trapped in materials containing Ce\$^4\$\$^+\$ centers.

In this study, liquid phase epitaxy is for the first time combined with Mg co-doping of LuAG:Ce. 7 LuAG:Ce,Mg epitaxial films were prepared (Mg concentration: 0, 100, 300, 800, 1500, 1500, 3000 ppm). Light yield (LY) and photoelectron yield (PhY), respectively, exhibit strong dependence on Mg concentration. Using 1 µs amplifier shaping time, it reaches maximum for Mg concentration 800 Mg, 395 phels/MeV, i.e. 17 % higher than PhY of undoped sample. Decrease of slow scintillation component intensity with increasing Mg concentration is strongly evidenced by PhY(10 µs)/PhY(0,5 µs) ratio (monotonous decrease from 178 % down to 103 %), scintillation decay curves, and afterglow values. Results of the study are consistent with theory and previous results obtained on different LuAG:Ce,Mg systems (bulk crystals, ceramics).

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### **[92] Thermoluminescence evidence of grain boundary structural disorder in LuAG:Ce optical ceramics** *Presenter: MORETTI, F.*

Scintillating optical ceramics (OC) based on garnet structures have been the subject of intense R&D. The interest on this kind of scintillating optical materials spawned from the excellent results obtained on Nd:YAG laser OC, whose production route allowed better control on dopant concentration and spatial distribution in the final laser active element, substantially outperforming the single crystal counterparts.\$^{1}\$ However, the results obtained on scintillating garnet OC by using the same synthesis strategies employed for laser OC have not been as successful, and only recently OC with comparable or better performances with respect to those of single crystals have been reported, based on a divalent ion codoping strategy.\$^2\$ Indeed, sintering aids are used to improve the powder sinterability leading to a reduced amount of light scattering centres and thus to a better transparency. However, sintering aids can also give rise to point defects which strongly affect carrier migration and transfer to luminescence centres. The recent improvements are substantially related to a better understanding of the sintering aid role on defect formation\$^3\$ and the scintillation mechanisms, and the use of different sintering aids with respect to those used for the production of laser OCs.

In this contribution we will present results obtained on two different LuAG:Ce OC (with and without MgO as sintering aid) and on a Czochralski grown single crystal as a reference. Optical absorption (OA), Raman scattering, radio- (RL) and thermo-luminescence (TSL) results will be presented and discussed in view of the detected differences among the optical ceramics and the single crystal, with the aim to evaluate defect localization in the OC. OA measurements evidenced the typical Ce3+ transitions accompanied, in the case of Mg codoped ceramic, by the presence of Ce4+ charge transfer transition. RL spectra show only minor variations in the Ce3+ emission shape and intensity, related to the different cerium contents in the three samples. Raman spectra of all the samples are practically coincident, clearly demonstrating that the three samples are, from optical and vibrational points of view, indistinguishable.

TSL results, on the other hand, evidenced clear differences among the OC and the single crystal. The glow curves of the ceramics are in fact much broader. Trap energy evaluation indeed evidenced the presence of broad distributions that are usually encountered when structural (or compositional) disorder is present. This leads us to conclude, also taking into consideration the other results, that the detected broad TSL peaks are mostly related to defects at grain boundaries. In such regions, structural disorder occurs, giving rise to inhomogeneous broadening of defect levels.

Therefore, TSL appears as an extremely effective technique to evidence the presence of structural disorder in OC which would, otherwise, be practically undetectable by other more commonly used bulk characterization methods. This work has been supported by the H2020 project AIDA-2020, GA no. 654168.

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### [95] Role of defects in formation of the luminescence centers in ZnMoO4 crystals

### Presenter: HIZHNYI, Yuriy

Zinc molybdate crystals, ZnMoO4, are intensively studied at present as perspective scintillation materials. Elaboration of novel scintillators with characteristics suitable for particular experiments requires knowledge of origin of the scintillation material luminescence. Luminescence emission is the final stage of the scintillation process and in great measure determines scintillation light output and decay time characteristics of the scintillator. At present, origin of the ZnMoO4 complex luminescence is still under discussion, despite a number of studies carried out with use of various experimental and computational methods. The main subject of discussion is related to role of the lattice defects (impurities) in formation of the luminescence properties of zinc molybdate. In particular, an open question is, whether the components of luminescence emission of ZnMoO4 are connected to some structural defects, or they entirely originate from regular components of the crystal lattice. In this report we present a comprehensive study of this problem using both experimental and computational methods.

A sequence of ZnMoO4 samples was synthesized by solid state reaction method at different annealing temperatures to achieve a substantially different concentration of structural defects in different samples. Then these samples were the subject of extensive structural and elemental characterizations to determine the trends in their defect structure and composition. The luminescence properties of these samples were studied in 8 – 300 K temperature range with use of synchrotron radiation as an excitation source on SUPERLUMI station at HASYLAB (Hamburg, Germany). Obtained experimental data were analyzed together with results of the electronic structure calculations carried out by a DFT band-structure method [1]. A number of point defects which presumably can influence the luminescence properties of ZnMoO4 was considered in calculations, namely the oxygen vacancy Vo, compensated oxygen vacancy Vo+VZn, tungsten impurity WMo and the MoO3-deficient phase Zn3Mo2O9. Such a complex study has provided a solid base for explanation of the origin of the red and blue-green luminescence emission bands of ZnMoO4 as well as formation of their excitation spectra.

It is found that the blue-green luminescence emission components of zinc molybdate crystals peaking in 2.2 – 2.5 eV region originate from radiative annihilation of the excitons self-trapped on regular MoO4 groups of the crystal. The red components of ZnMoO4 emission peaking near 2.0 eV originate from MoO4 groups located close to point defects.

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# **[96] Luminescence, scintillation and energy transfer in the doubly doped LuAG:Pr,Dy single crystal** *Presenter: PÁTEREK, Juraj*

Ce and Pr doped Lu3Al5O12 (LuAG) single crystals started to be systematically studied around year 2000 and 2004, respectively, see review [1]. Among their advantages there is stable crystal growth, relatively high density, fast decay due to allowed 5d-4f transitions of Ce3+ and Pr3+, good light yield, mechanical and chemical robustness and radiation hardness. Degradation of figure of merit of these single crystals is due to a number of electron traps arising around the antisite defects (shallow ones) and oxygen vacancies (deep ones). The former were overcame by the concept of multicomponent garnets, namely by the pushing down the bottom of conduction band by the Ga admixture and even more restricted by the divalent alkali earth ion codoping [1,2].

In some applications, even faster decay than is defined by the inherent transition dipole moment of Ce3+ or Pr3+ is required. Shortening of the luminescence lifetime of Ce3+ can be achieved, e.g. by partial ionization of its 5d1 state by tuning the energy barrier between 5d1 and bottom of conduction band [3] or it can be achieved by embedding a suitable acceptor center, which will have its excited state in resonance with 5d1 level of Ce3+ (Pr3+) and thus enables a nonradiative energy transfer away from 5d1 state. Such an energy transfer will accelerate the measured decay time of Ce3+ (Pr3+) emission and this acceleration can be observed in both photoluminescence and scintillation decays.

In this contribution, we report the exploitation of the latter principle in the acceleration of the primary decay component of LuAG:Pr3+,Dy3+ luminescence. Such an energy transfer is realized due to the overlap of 5d1-4f emission of Pr3+ within 300-380 nm with the 4f-4f absorption transitions of Dy3+, resulting in shortening of the leading decay component by 30-40%. We will report the luminescence and scintillation characteristics of such a doubly doped LuAG and describe the energy transfer process in the terms of the Förster-Dexter model.

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### [112] Effects of Annealing and Mg-codoping on Luminescence and Scintillation Properties of Ce Doped Garnet-Based Scintillator Grown by Micro Pulling Down Method

### Presenter: KAMADA, Kei

Scintillators are widely used to detect high-energy photons and particles and applied in the medical imaging and other applications. Among the many kinds of scintillator materials, oxide scintillation materials based on garnet structure like Ce-doped Lu3Al5O12 (Ce:LuAG) are promising candidates for scintillator applications because of well mastered technology developed for laser hosts, and easy doping by rare-earth elements [1]. LuAG admixed with balanced Gd and Ga ratio was proved to be an excellent scintillator where the effect of shallow traps was suppressed; the spectrally corrected light yield value exceeded 50,000 photons/MeV [2]. Recently, cations (B, Ca, Ba) co-doping effects on scintillation mechanism and annealing effects on defect structures for Ce: Gd3Ga3Al2O12 (GGAG) has been reported [3], where further improvement of scintillation properties were also reported.

In this report, effects of annealing atmosphere and Mg co-doping on scintillation properties of Ce:GGAG were investigated. Mg 200ppm co-doped and non co-doped Ce:GGAG single crystals grown by micro pulling down method (m-PD) were prepared under N2 atmosphere. Temperature dependence of the light output and decay time were evaluated in the Mg/non co-doped samples before and after annealing under oxidizing (air) and reducing (Ar+2%H2) atmosphere. Temperature dependence of the light output was no change in the Mg co-doped sample. Temperature quenching of light output started at lower temperature in the case of non co-doped sample after annealing under reducing atmosphere. The 1st decay component of Mg/non co-doped samples changed to 50.0ns and 183ns after annealing under reducing atmosphere (Mg co-doped:54.1ns and non co-doped:61ns before annealing), respectively. Thease results indicates suppression of O2-defect by Mg co-doping. In our presentation, details of crystal growth and crystal structure as well as optical and scintillation properties will be reported.

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# [115] Luminescent ZnO:Ga nanopowder: Surface passivation and limiting the particle agglomeration *Presenter: PROCHAZKOVA, Lenka*

ZnO:Ga scintillation powder is a promising candidate for detectors with ultrafast timing, thanks to its extremely short decay and practically no rising time [1]. For practical use, the powder needs to be embedded in a suitable optically transparent matrix. However, heat treatment of ZnO:Ga particles is needed to increase the luminescence at the cost of particle agglomeration. This in turn results in low transparency of scintillating nanopowder embedded in a matrix and constitutes one of the biggest disadvantages, besides low light yield and low stopping power of ZnO:Ga powder. Limiting ZnO:Ga particle size is therefore a key step in order to prepare highly luminescent and transparent composites with high potential to be appropriate for PET TOF applications.

Therefore, the research presented has two goals – preventing the particles agglomeration during heat treatment to improve homogeneous embedding and the related transparency, and surface passivation of ZnO:Ga particles to preclude non-radiation loses on the grains boundary.

For surface passivation, the covering by either organic polymers or inorganic layers is acceptable, depending on target application [2, 3]. In this work, two methods for limiting growth of particles were studied.

The first method was tested for surface passivation and limiting the particle size;ZnO:Ga particles were annealed at 200 °C and coated by SiO2 amorphous layer via sol-gel method. Annealed powder was dispersed in the solution of absolute ethanol and TEOS (tetraethylorthosilicate) which was hydrolysed by dropwise adding ammonia. The amount of TEOS was calculated from the specific surface area of ZnO:Ga to cover the particles by two layers of SiO2. Product was dried and heat treated at 300, 600 and 1000 °C. XRD measurements show that linear crystallite size is constant up to 600 °C. At higher temperatures, increase of the crystallite size and formation of zinc silicate phase was observed. Particle size was also calculated from the evaluation of specific surface area and it does not exceed 14 nm, regardless of the calculation method. Parasitic Zn2SiO4 phase deteriorates luminescence properties and therefore this surface modification is suitable only for powders not requiring heat treatment.

In the second method for preventing particle agglomeration, ZnO:Ga was dispersed in water and the sufficient amount of PVA (polyvynilalcohol) was dissolved to form gel. This gelatinous suspension was dried at 50 °C and subsequently heat treated at 600, 800 and 1000 °C. TEM images show well separated particles with ~50 nm in diameter after heat treatment at 600 °C. Above 600 °C, the particle size increases with increasing temperature.

The combination of those two methods of post-treatment may allow for improvement of the optical and luminescence properties of ZnO:Ga-based composites.

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### [119] Scintillation Properties of TlGd2Cl7 (Ce3+) Single Crystal

### Presenter: KHAN, Arshad

Scintillation properties of new TIGd2Cl7 single crystal is presented. Different Ce-doped (0, 0.5, 1 and 5 mol %) single crystals of TIGd2Cl7 are grown by vertical Bridgman technique. All doped samples show typical Ce3+ emission under X-ray excitation between 350 nm and 500 nm. The emission peak positions slightly changed with the increase of Ce-concentration in the host matrix. Excellent light yield and good energy resolution is obtained under y-ray excitation at room temperature. Three exponential decay components are obtained for all Ce-doped samples at room temperature. Decay components changes with Ce-concentrations i.e. get faster with higher Ce-concentration. Effective Z-number is found to be 66 and therefore X- and y-ray detection will be detected efficiently with this scintillator [1, 2]. High light yield, high Z-number with moderate energy resolution and fast scintillation response suggest that this scintillator could be used in the medical imaging techniques. Further investigations are under way for the improvement of its scintillation properties with higher Ce-concentrations.

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# **[120] Luminescence and Scintillation properties of novel disodium dimolybdate (Na2Mo2O7) single crystal** *Presenter: PANDEY, Indra Raj*

The Advanced Mo Based Rare Process Experiment (AMoRE) collaboration is searching for the neutrino less double beta decays ( $0v2\beta$ ) of 100Mo isotopes using CaMoO4 scintillating crystals operating at milli-Kelvin temperatures [1]. Due to the  $2v2\beta$  background caused by 48Ca and the limitation of calcium purification within an acceptable level [2], new molybdate-based scintillation crystals for the AMoRE in the phase-II are necessary. In this work we studied luminescence and scintillation properties of the novel Na2Mo2O7 crystals grown by using two different techniques, the conventional Czochralski and the low-temperature-gradient Czochralski techniques. The transmittances of Na2Mo2O7 crystals were measured at the room temperature. The luminescence properties and decay times of the crystals at room and low temperatures were measured by using a light emitting diode (LED) source of 280 nm wavelength. The crystals showed a red emission peak of 663 nm at 10 K. The scintillator was measured by a photon counting technique using 90Sr as the excitation source in the temperature range from 300 K to 10 K. This work reports on the light yields of Na2Mo2O7 crystals grown at two different institutes at various temperatures and compared with a CaMoO4 crystal. Results for thermoluminescence measurements will be presented, which is going to let us understand whether the main constraint of the scintillator performance at low temperature is due to the self-trapping of charge carriers or the trap level in the crystals by defects or impurities [3].

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### [126] Temperature dependence of Ce3+ emission kinetics in Mg2+ co-doped GAGG:Ce epitaxial garnet films

### Presenter: KUCERA, Miroslav

The alkaline earth co-doping by Mg2+ or Ca2+ ions in LYSO:Ce and LuAG:Ce stabilizes tetravalent Ce4+ ions which then participate in the scintillation process. They act as electron traps and result in reduction of afterglow and slow components in the scintillation decay [1, 2]. Excellent scintillation properties have recently been reported in epitaxial garnet films GAGG:Ce,Mg grown by liquid phase epitaxy [3].

Here we study the effect of Mg2+ co-doping on timing properties, thermal ionization effects, and shallow electron traps in (Lu0.8Gd2.2)(Al2.5Ga2.5)O12:Ce (GAGG:Ce) multicomponent epitaxial garnet films with different Mg doping from 0 to 700 ppm. The films were grown by liquid phase epitaxy from BaO-B2O3-BaF2 flux. The kinetics of emission was studied under optical excitation in the temperature range from 10 to 500 K. The results are quantitatively compared with those obtained by e-beam excitation.

The photoluminescence decay kinetics allows to detect simultaneously both the prompt electron-hole 5d-4f(Ce3+) recombination and delayed components. It was found, that the effect of Mg2+ ions on the position of 5d state in the forbidden gap is small and the activation energy related to the ionization and/or quenching processes is not notably influenced. In particular, the activation energy was between 300 and 350 meV as obtained from the temperature dependencies of integrated decays and PL spectra in samples with Mg content between 0 and 700 ppm. The signal decreased to 50 % at ~370 K independently on Mg doping. Furthermore, significant improvements of timing performance of the Mg2+ co-doped garnets have been observed, namely a substantial acceleration of the scintillation decay and considerably reduced afterglow.

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### **[127] Effect of Co-doping on Pulse Shape Discrimination Properties of Gd\_3Ga\_3Al\_2O\_12:Ce Scintillators** *Presenters: TYAGI, Mohit, RAWAT, Sheetal*

Gadolinium gallium aluminum garnet (Gd\$\_{3}\$Ga\$\_{3}\$Al\$\_{2}\$O\$\_{12}\$, also called GGAG) doped with Cerium (Ce) has shown the best combination of the scintillation characteristics exhibiting high density (6.7 g/cm\$^{3}\$), effective atomic number (55), light yield of 54,000 ph/MeV and fast decay time of 55 ns \*\*[1]\*\*. The efficient and fast emission at 550 nm makes these scintillators very useful in the fabrication of compact detectors based on silicon photo-sensors. Single crystals of CsI:TI, also having 550 nm emission, are conventionally used with different photo-sensors for various detection applications. Therefore, the comparison of both the crystals has been studied in details by many researchers. However the pulse shape discrimination properties of GGAG scintillators have not been explored in much detail.

The scintillation decay time of CsI:Tl detector, for gamma excitation, consists of two component having 700 ns and 3500 ns time with relative contribution of 57% and 43% respectively. Therefore, the average time for gamma radiation is calculated to be 1200 ns. The alpha radiation causes the acceleration of decay time in comparison with gamma excitation. The average time for alpha radiation is calculated to be 600 ns only. However, this trend was found to be opposite in case of GGAG scintillators where alpha radiation slows down the decay time to 284 ns (average) in comparison with average decay time of 108 ns with gamma excitation. Thus, the quenching mechanism by the higher ionization density which successfully explains the dependence of decay in CsI:Tl crystals on the mode of excitation does not explain the behavior of scintillation properties \*\*[2]\*\*. The dependence of decay times on gamma and alpha radiations was studied in different co-doped GGAG crystals. The pulse shape discrimination capabilities were also observed to be dependent on the presence of co-doping, growth ambient and after growth annealing treatment. The results indicate the role of defect centers on charged particles discrimination characteristics of GGAG scintillators.

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### [129] LUMINESCENCE OF LiLa9(SiO4)6O2 SILICATE CRYSTALS DOPED WITH Ce3+ and Pr3+ IONS

### Presenter: PUSTOVAROV, Vladimir

Crystals doped with rare earth ions find numerous applications as scintillators in various fields of technology including nuclear physics, medical imaging, dosimetry, etc. In this paper, we report on luminescence and optical properties of a new class of LiLa9(SiO4)6O2 (LLSO) crystals doped with Ce3+ and Pr3+ ions which we found potentially attractive for scintillator applications.

The LLSO crystals belong to the nesosilicate family and are characterized by a partially disordered apatite structure that can be doped with optically active rare-earth ions substituting for two La3+ sites with different oxygen coordination and symmetry. The crystals for this study were synthesized and XRD verified in the Luminescent Materials Laboratory, University of Verona (Italy).

The spectroscopic study included measurements of photoluminescence (PL) and X-ray excited luminescence (XRL) spectra in the 1.5-5.5 eV energy range as well as UV-VUV excitation spectra in the 3.5-10 eV range at temperatures of 20, 90 and 300 K. Luminescence decay kinetics were recorded upon cathode and UV-pulse laser excitation. In addition, thermally stimulated luminescence of the crystals was studied by recording glow curves in the temperature range from 90 to 500 K.

PL spectra of LLSO:Ce3+ (1%) recorded upon UV-, VUV-, X-ray and e-beam excitation are composed of two emission bands centered near 2.4 and 2.94 eV with different Stokes shifts that correspond to interconfigurational  $5d \rightarrow 4f$  transitions in Ce3+ ions substituting for La3+ ions in two cationic sites with different coordination numbers. The Ce3+ sites demonstrate different thermal quenching that results in significant temperature dependence of PL quantum yield and shape of the PL spectra. The decay time of  $5d \rightarrow 4f$  transitions is 19 ns under UV laser excitation and  $\tau \approx 70$  ns under e-beam excitation at T=295 K. The latter is explained by delayed host-to-impurity energy transfer.

Emission of LLSO:Pr3+ is presented by intraconfigurational 4f  $\rightarrow$  4f transitions in the visible region and interconfigurational 4d  $\rightarrow$  5f transitions in the UV region (two broad bands) observed at 90 and 295 K. Positions of 4f  $\rightarrow$  4f bands in PL spectra are independent on excitation energy, while features related to 5d  $\rightarrow$  4f emission are dependent on excitation energy. As in the case of the Ce3+ crystal, this is explained by the existence of two Pr3+ sites with different symmetry. Two types of 4d  $\rightarrow$  5f emission are observed at T =295 K: type I is excited near 5.25 eV yielding a doublet emission feature peaked at 4.0 and 4.45 eV, while type II is excited near 5.0 eV PL yielding similar emission with maxima at 3.55 and 3.85 eV. The decay time of 5d  $\rightarrow$  4f emission is about 10 ns upon UV laser excitation in the region of 4f  $\rightarrow$  5d absorption at room temperature. It is worth noting that LLSO:Pr3+ excitation and emission spectra do not show any features which could indicate population of Pr3+ 1S0 level and enhancement of the 4f  $\rightarrow$  4f transitions. This suggests that the 1S0 level is above the lowest 4fn-15d state that prevents observation of cascade photon emission in the sample.

### [130] Suppression of the Slow Scintillation Component in BaF2 Crystals by Y3+ Doping

### Presenter: CHEN, Junfeng

It is well known that Barium fluoride (BaF2) crystals have a fast scintillation light peaked at 195 nm and 220 nm with a sub-ns decay time. This ultra-fast scintillation promises its wide application in future HEP experiments requiring extreme fast rate capability, Gigahertz hard X-ray imaging and TOF-PET etc. BaF2 crystals, however, have also a slow scintillation component peaked at 310 nm with a decay time of about 600 ns, which causes a pile-up problem. Two approaches have been proposed to handle the slow scintillation in BaF2: selected readout with optical band pass filters [1] or solar-blind photodetector [1] and selective doping in BaF2 with rare earth (RE), such as Ce, La and Y [2]. Our previous investigation shows a 20 cm long La/Ce doped BaF2 crystal grown in Beijing Glass Research Institute (BGRI) with effective slow component suppression [3]. In this study, we show that Y3+ ion doping has a great potential for slow component suppression in BaF2. Transmission and radio-luminescence spectra, light output, fast/slow ratio, scintillation decay kinetics and light response uniformity are measured for Y3+ doped BaF2 crystal grown in Shanghai Institute of Ceramics (SIC). The results show that the slow scintillation component in BaF2 crystal can be suppressed by up to a factor of 6 by Y3+ doping at 1at%, while the fast component remains almost unaffected. Development will continue along this line of research.

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### [133] Role of yttrium in thermoluminescence of LYSO:Ce crystals

### Presenter: DING, Dongzhou

This paper aims at an improved understanding of thermoluminescence of (Lu1-xYx)2SiO5:Ce (x=0 at.%, 9 at.%, 26 at.%, 45 at.%, 66 at.%, 88 at.% and 100 at.%). Wavelength-resolved thermally stimulated luminescence (TSL) spectra and vacuum ultraviolet excitation spectra as well as transmittance spectra of (Lu1-xYx)2SiO5:Ce were investigated as a function of yttrium (shortened as Y) content in it. Obtained thermal depth of electron traps Et indicates that: with the increase of Y content, Et corresponding to the higher temperature peak (~ 336 K) increases, while Et corresponding to the lower temperature peak (~ 100 K) presents a slight decrease and then increase trend. Besides, it was found that: with the increase of Y content, band gap presents an obvious increase trend. The kinetic process and recombination mechanism of de-trapped electrons are discussed.

### **[136] Improved cathodoluminescence performance of Mg-doped LuAG:Ce(GdGa) single crystalline films** *Presenter: LALINSKÝ*, *Ondřej*

Cerium activated lutetium aluminum garnet (LuAG:Ce) is a perspective material for applications in detection of X-rays, gamma radiation or high energy particles. However, the luminescence response of LuAG:Ce suffers from structural defects (mainly anti-site defects) that create unwanted slow microsecond-component in decay characteristics. Concentration of these defects decreases with decreasing growth temperature. Therefore, single-crystalline films grown by liquid phase epitaxy have recently attracted a lot of attention because their growth temperature is about half of that of the bulk single crystals grown by the Czochralski method. As shown earlier [1], the proper admixture of Gd and Ga into the garnet structure eliminates the effect of remaining anti-site defects and thus resulting in almost complete suppression of the slow decay component. In addition, such material has exceptionally high light yield (LY) exceeding 50 kph/MeV and its cathodoluminescence (CL) decay is dominated by a fast component with decay time of 50-80 ns. However, this decay time can be still too high, for example, for some special SEM applications where very fast e-beam scanning is required. Thus, new materials with faster decay have to be found. Recently, Mg\$^{2+}\$-doped garnet films have been intensively studied, primarily using photoluminescence and radioluminescence [1] but X-rays have high penetration depth in garnets. Consequently, unwanted substrate information is also recorded. The CL solves this problem because electrons having energy of 10 keV don't penetrate deeper than 1 µm under the surface. Therefore, CL was selected as the optimal tool for specimen characterization.

For the purpose of this work, set of Mg\$^{2+}\$-doped LuAG:Ce and set of Mg\$^{2+}\$-doped LuAG:CeGdGa multicomponent epitaxial films (thickness around 17 μm) with different Mg concentration (0-700 ppm) were grown from lead-free BaO-B\$\_{2}\$O\$\_{3}\$-BaF\$\_{2}\$ flux. The films were excited by an e-beam with energy of 10 keV using a specialized CL apparatus [2]. The CL spectra, CL decays under nanosecond and millisecond excitation and thermoluminescence glows in the temperature range between 100 and 490 K were obtained.

Firstly, it was shown for both sets of specimens that the CL decay time of the fast component related to the 5d-4f transition at Ce\$^{3+}\$ sharply decreased with increasing concentration of Mg\$^{2+}\$. For LuAG:CeGdGa with the highest concentration of Mg\$^{2+}\$, the decay time was only 28 ns. Secondly, considerable reduction of the afterglow (signal at 1 µs after e-beam cut off) for highly Mg\$^{2+}\$-doped specimens was observed. The afterglow of 3.1 % for Mg\$^{2+}\$-free LuAG:Ce was reduced down to 0.037 % in case of highly Mg\$^{2+}\$-doped LuAG:Ce, and even down to 0.015 % in case of highly Mg\$^{2+}\$-doped LuAG:CeGdGa. This is the best afterglow value ever reported for garnets so far. Finally, Mg\$^{2+}\$-doping has no influence on the start of thermal quenching, i.e. the quenching start is determined by the host composition, mainly by the Ga/AI ratio. Unfortunately, all these observations come at the expense of LY, however, the LY is still comparable to that of YAG:Ce.

Acknowledgement: The research was supported by Czech Science Foundation (GA16-05631S), by MEYS CR (LO1212), its infrastructure by MEYS CR, EC (CZ.1.05/2.1.00/01.0017) and by CAS (RVO:68081731).

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### **[143] Luminescent properties of Cesium Hafnium Chloride scintillators doped with alkaline earth metals** *Presenter: KODAMA, Shohei*

High-energy-resolution scintillators are demanded for food/environmental gamma-ray monitoring systems in Fukushima or for other applications. Generally, halide scintillators have high light output due to small band-gap energy, and therefore high energy resolutions are expected [1]. However, almost all halide materials have hygroscopic nature, which makes them difficult to handle.

In 2015,  $Cs_2HfCl_6$  (CHC) has been reported as non-hygroscopic halide scintillator [2]. CHC has a high light output of up to 54,000 photons/MeV, and its energy resolution is estimated to be 3.3%, from full width at half maximum (FWHM), at 662 keV. In order to improve the energy resolution, we focused on its non-proportional response. In the case of LaBr<sub>3</sub>, the non-proportional response improved by Sr<sup>2+</sup>-doping [3]. Therefore non-proportional response and energy resolution for CHC might be improved by doping alkaline earth metals as well. In this study, we report the effect of AE<sup>2+</sup>-doping (AE<sup>2+</sup> is alkaline earth metals; Mg<sup>2+</sup>, Ca<sup>2+</sup>, Sr<sup>2+</sup> and Ba<sup>2+</sup>) into Hf<sup>4+</sup> site on scintillation properties.

Non-doped and  $AE^{2+}$ -doped CHC crystals were synthesized from 99.9%-pure (Zr-free) HfCl<sub>4</sub>, 99.999%-pure CsCl, 99.999%-pure MgCl<sub>2</sub>, 99.99%-pure CaCl<sub>2</sub>, 99.998%-pure SrCl<sub>2</sub> and 99.99%-pure BaCl<sub>2</sub> from a nominal composition of Cs<sub>2</sub>(Hf<sub>0.995</sub>AE<sub>0.005</sub>)Cl<sub>5.99</sub> by the vertical Bridgman method. Crystal phases were identified by powder X-ray diffraction. Excitation/emission wavelengths were evaluated from photo- and X-ray excited radio-luminescence spectra. Light output, its non-proportionality, energy resolution and scintillation decay constant were evaluated using a <sup>137</sup>Cs gamma-ray source. Finally, we succeeded in growing non-doped and  $AE^{2+}$ -doped CHC single crystals. The crystal structure of all specimens was determined as Fm-3m. No other phase was observed. Non-doped CHC showed broad emission around 400 nm under X-ray excitation. The light output and energy resolution were estimated to be 42,000 photons/MeV and 5.2% at 662 keV (FWHM), respectively. The scintillation decay constant was estimated using double exponential fitting, and fast component and slow component were determined to be 0.27 µs (4.5%) and 5.52 µs (95.5%), respectively.

On the other hand, radio-luminescence emission spectrum of Mg:CHC was the same as for the non-doped CHC. Its light output and FWHM energy resolution were estimated to be 45,000 photons/MeV and 6.0% at 662 keV, respectively. The scintillation decay constant consisted of fast 0.69  $\mu$ s (7.5%) and slow 5.99  $\mu$ s (92.5%) components. In presentation, we show the results of other AE<sup>2+</sup>-doped CHC and discuss the relationship between their scintillation properties and co-doped elements.

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### [150] Characterizations and simulations of structured scintillators for synchrotron applications

### Presenter: DOUISSARD, Paul-Antoine

The European Synchrotron Radiation Facility builds a new ultra-bright synchrotron source, the ESRF-EBS [1] in the period 2015-2022, which will deliver an X-ray beam with performances multiplied by 100 in terms of brightness and coherence. In this context, diffraction and X-ray imaging experiments will benefit from X-ray detectors covering a large dynamic range (above 16 bits), large field of view (at least 10cmx10cm), while operating at high X-ray energies (between 30keV and 100keV) and high frame rates (above 100 frames per second). In the case of X-ray diffraction, indirect detection scheme using a scintillator to absorb and convert X-ray photons into light is important at high X-ray energies (starting from 30keV). Concerning X-ray imaging, indirect detection is the only scheme able to cope with the huge intensities of the X-ray beams while providing down to micrometer spatial resolution. Therefore, we plan to develop structured scintillators optimized for synchrotron radiation applications. The scintillating screen will be a key component, as it will determine the final X-ray absorption efficiency, speed, precision as well as stability of the detection system under high energy and intense X-ray flux.

In this context, we will present the evaluation of structured scintillators for synchrotron X-ray applications up to X-ray energies of 100keV. At these X-ray energies, the structuration of the scintillating screen is important to guide the emitted scintillation light and thus avoid the degradation of the Line Spread Function due to the spread of optical photons within the bulk of the scintillator. The properties of the scintillating screen will be focused on: spatial resolution below 50µm, low afterglow to solve at least 16bit dynamic range, high thermal and radiation stability, X-ray absorption >80% in the range 30keV-100keV, decay time <500µs, easily four-side buttable to cover large areas >10cmx10cm.

The presentation will be focused on experimental measurements of spatial resolution and afterglow. For the tests, we coupled the scintillating screens via a Fiber Optics face plate with 3µm diameter fibers to a 6.5µm pixel size sCMOS Andor Zyla 5.5 camera. The screens were exposed to a bright (flux >1010 ph/s/mm2) monochromatic X-ray beam at the ESRF BM05 beamline [2].

The spatial resolution measurements are compared with simulations using the Mantis-based simulation tool installed, modified and recompiled at the ESRF [ 3].

# [152] Luminescence and Scintillation properties of Ce-doped GdBO3 nanophosphor synthesized by aqueous sol-gel method

### Presenter: SERAICHE, Mourad

GdBO3:Ce3+ emitting phosphors as nanopowders were prepared using aqueous sol-gel method. The photoluminescence (PL) and scintillation properties were analyzed as function of the pH value of precursor suspension (pH=2, 5 and 8) and the nominal Ce3+ concentration (0.5%mol) at different annealing temperatures (600, 700, 800, 900, 1000, 1100 and 1200) °C. The crystal structures of the prepared materials were cheeked using several techniques such as: XRD, IR. All the samples of GdBO3:Ce3+ present pure phases which crystallize in the vaterite form. The higher PL, corresponding to the 5d-4f transition of Ce3+, as well as the scintillation light yield is obtained for the sample containing Ce3+ 0.5 mol.% prepared at pH=8 annealed at 800°C during 4 h. The scintillation yield has been deduced under X-ray excitation by comparison with the standard x-ray phosphor Gd2O2S:Tb3+ or Eu3+ (Gadox). These results, including scintillation decay and afterglow will be discussed as a function of the synthesis parameters.

### [158] Innovative LaBr\_3(Ce) geometries optimized for fast timing applications

### Presenter: VEDIA FERNANDEZ, Maria Victoria

LaBr\$\_3\$(Ce)-based detectors, which provide excellent time response together with high gamma detection efficiency and good energy resolution [1, 2], are optimal devices for a wide range of applications. Their scintillation properties are strongly influenced by their size and shape. Consequently, we have designed novel geometries of LaBr\$\_3\$(Ce) crystals aimed at enhancing the light collection, and thus their scintillation characteristics. Moreover the new geometries make it possible to construct rings of detectors surrounding implantation points (detectors or catcher foils) with higher packing factor than cylindrical geometries, increasing the  $\gamma$ -ray detection efficiency.

In this work we evaluate the overall performance of two new geometries of LaBr\$\_3\$(Ce) crystals with special attention to the time response. The crystals were coupled to Hamamatsu R9779 photomultiplier tubes (PMTs). The signals were processed with NIM analog electronics including ORTEC Constant Fraction Discriminator (CFD) 935 and Time to Amplitude Converter (TAC). Energy resolution and efficiency were measured using standard calibration sources and timing measurements were performed at \$^{60}\$Co and \$^{22}\$Na y-ray energies against a fast BaF\$\_2\$ reference detector. The time resolution was optimized by the choice of the PMT bias voltage and the fine-tuning of the CFD parameters. In addition to energy resolution and y-ray detection efficiency, we report the individual FWHM time resolutions for the two geometries, which are the best results reported to date for LaBr\$\_3\$(Ce) crystals of these sizes. This achievement has been possible thanks to the new geometry designs in combination with the use of the ORTEC 935 CFD at very short delays. Monte Carlo simulations using the Geant4 toolkit have been performed to gain a better understanding of the effect of the new geometries on the light transport, and thus on the performance of the crystals. We report on the results from the measurements and the simulations and on the best crystal choice for fast-timing applications.

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# [162] Influence of gallium content and position on thermally stimulated luminescence of multicomponent (Y,Lu,Gd)3(Ga,Al)5O12:Ce garnets

### Presenter: BABIN, Vladimir

Multicomponent (Y,Lu,Gd)<sub>3</sub>(Ga,Al)<sub>5</sub>O<sub>12</sub>:Ce garnets are efficient complex oxide scintillators, whose main advantages are achieved owing to the Ga-induced lowering of the conduction band (CB) bottom resulting in burying of shallow electron traps (see, e.g., review [1]). In this work, the

(Y,Lu,Gd)<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce single crystals with different Ga contents are studied by the photo- and thermally stimulated luminescence, NMR, and ESR methods. The results are compared with the data obtained in [2] for the (Lu,Gd)<sub>3</sub>Ga<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub>:Ce epitaxial films.

Under irradiation in the 4f - 5d absorption band of Ce<sup>3+</sup>, electrons can be thermally released from the 5d excited level of Ce<sup>3+</sup> into the CB, migrate along the CB and be trapped at different traps, thermally released from these traps, and finally recombine with the optically created Ce<sup>4+</sup> centers. As a result, the Ce<sup>3+</sup>-related thermally stimulated luminescence (TSL) appears. In case a TSL glow curve peak appears as a result of an electron release from the 5d<sub>1</sub> level of Ce<sup>3+</sup> into the CB, the activation energy E<sub>a</sub> for the TSL peak creation corresponds to the energy distance between the 5d<sub>1</sub> excited level of Ce<sup>3+</sup> and the bottom of the CB.

The increasing Ga content is found to result in the decreasing E<sub>a</sub> value, increasing TSL intensity, decreasing temperature of the luminescence quenching, low-temperature shift of the TSL peaks and decrease of the corresponding trap depths. It results also in the increasing concentration of Ga<sup>3+</sup> ions in the octahedral Al<sup>3+</sup> sites (N<sub>oct</sub>) as compared with the tetrahedral ones (N<sub>tetr</sub>) [3]. Both in the epitaxial films and single crystals of different composition and origin, having strongly different concentrations of non-compensated vacancies and antisite defects, the E<sub>a</sub> dependences on the Ga content practically coincide and are \*nonlinear\* (see, e.g., [2]). However, the dependences of the TSL peaks positions, the corresponding trap depths, and the N<sub>oct</sub>/N<sub>tetr</sub> ratio on the Ga content are close to \*linear\*. These data allow us to suggest that in the multicomponent garnets with x>1.4, E<sub>a</sub> is not equal to the 5d<sub>1</sub> - CB energy. Most probably, E<sub>a</sub> is the energy distance between the 5d<sub>1</sub> level of Ce<sup>3+</sup> and a defect level located between the 5d<sub>1</sub> level of Ce<sup>3+</sup> and a defect level located between the 5d<sub>1</sub> level of Ce<sup>3+</sup>

These data indicate that in the multicomponent garnets, Ga can play not only a positive but also a negative role (the appearance of additional defects, reduced activation energies for the photostimulated defects creation and luminescence thermal quenching) which increases with the increasing Ga content.

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### [165] Luminescence Properties of Mn^4+ Doped CaY\_2[MgM](AlSi\_2)O\_12 (M = Al, Sc, Ga) Garnets

### Presenter: VIELHAUER, Sebastian

Tetravalent Mn ions have a number of properties that makes them interesting candidates for use as activators in luminescent materials, such as phosphors for fluorescent lamps or white LEDs as well as for thermoluminescence dosimeters. They possess strong absorption bands in the blue and UV spectral region due to spin-allowed transitions \$-\ \$ \$^4\$A\$\_2 \rightarrow ^4\$T\$\_2\$ and \$^4\$A\$\_2 \rightarrow ^4\$T\$\_1 \$, respectively. A characteristic emission band due to the spin-forbidden \$^2\$E\$ \rightarrow ^4\$A\$\_2\$ transition is observed in the red spectral range. The energy levels of the Mn\$^{4+}\$ ions are sensitive to the crystal field, so that the spectral properties can be varied by incorporation of the Mn\$^{4+}\$ into different matrices. Using manganese as an optically active ion instead of rare earth elements can also be more cost efficient due to the much lower material cost.

In garnets, where Mn\$^{4+}\$ preferably occupies the octahedral sites, the distance to the neighboring O\$^{2-}\$ ions and their effective charges are important parameters to influence the energies of the absorption and emission bands [1,2]. To study the influence of these parameters, {CaY\$\_2\$}[\$MgM\$]\$(AISi\$\_2\$)O\$\_{12}\$ garnets with different metal ions AI, Sc and Ga were used. These metal ions also occupy the octahedral site in the garnet structure.

Single phase ceramic samples of garnets according to the composition and structure formula

{CaY\$\_2\$}\$[\$MgM\$]\$(AlSi\$\_2\$)O\$\_{12}\$ (M = Al, Sc, Ga) doped with Mn\$^{4+}\$ ions at different concentrations were obtained by high temperature solid-state reaction technique using precursors which were synthesized under hydrothermal conditions. The samples have been characterized by XRD, Raman and photoluminescence spectroscopy, including high resolution emission spectra in the temperature range of 8 \$-\$ 300 K.

The influence of the dopant concentration and modifications of the host composition on the luminescence properties will be discussed and compared with results of theoretical analysis for the Mn\$^{4+} levels in the studied hosts.

This research was performed within the ERA.Net RUS Plus program, project NANOLED # 361 (RFBR Grant 16-52-76028 ERA a).

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### [173] Effect of Au Co-Doping on the Scintillation Performance of BaBrCl:Eu Single Crystal

### Presenter: SHALAPSKA, Tetiana

Co-doping has been demonstrated to improve the light yield and/or energy resolution for compounds activated with Ce [1] and TI [2]. Limited efforts have been done for co-doping of Eu-activated halides. We report on the screening of co-dopants for the mixed halide BaBrCI:Eu. With a light output of 52,000 photon/MeV and energy resolution of 3.55%, theoretical performance of BaBrCI:Eu has not been achieved. Among a large number of aliovalent/isovalent co-dopants tested experimentally, Au, at low concentration, was found to give the best enhancement for the scintillator characteristics. We fount that co-doping with Au significantly improve the light output of BaBrCI:Eu for all Eu concentration in the range of 0.5%<Eu<8% with the optimum being at 3%. The results have a direct impact on the crystal growth process, as large concentration of Eu has been related to cracking of the crystals. The role of Au co-dopant in scintillation mechanisms of BaBrCI:Eu single crystals will be discussed considering the results of first-principal calculations and the defect structure of the compound.

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### [185] Optical properties of \mathrm{Eu^{2+} doped BaBrI, BaClI and SrBrI crystals

### Presenters: SHENDRIK, Roman, SHALAEV, Alexey

The rare-earth doped ternary alkali earth-halide systems are promising scintillators showing high efficiency and energy resolution. Some aspects of crystal growth and data on the structural and luminescence properties of BaBrI, BaCII and SrBrI crystals doped with  $\operatorname{Eu^{2+}}\$  ions are reported. The crystals are grown by the vertical Bridgman method in sealed quartz ampoule.

Emission, excitation and optical absorption spectra as well as luminescence decay kinetics are studied under excitation by X-ray, vacuum ultraviolet and ultraviolet radiation. The energies of first 4f-5d transition in \$\mathrm{Eu^{2+}} and band gaps of the crystals are obtained. The electronic band structure of the crystals are calculated using density functional theory as implemented in Vienna \*Ab Initio\* Simulation Package. Calculated band gap energies are in accordance with the experimental estimates. Vacuum referred binding energy diagrams of lanthanide levels are constructed using the chemical shift model.

### Acknowledgements:

This work was partially supported by a grant from the Russian Science Foundation, RScF project 17-72-10084 (Optical spectroscopy and calculations), and by a grant from Russian Foundation for Basic Research, RFBR project 15-02-06514 \_a (Crystal growth)

### [211] Scintillation properties of high-resolution La(BrxCl1-x)3:Ce and high-sensitivity CeBr3 crystals

### Presenter: PETRAK, Sibylle

Scintillation materials based on halide-lanthanide matrix elements are described in terms of material properties and applications. A new embodiment in this group of scintillators is presented, where the matrix material contains a mixture of two halides, namely lanthanum bromide and lanthanum chloride, that form a solid solution in the uranium tri-chloride lattice type. The matrix material also includes cerium as the activator. The scintillation properties of the new La(BrxCl1-x)3:Ce material are discussed and compared with those of CeBr3. We report on several aspects relevant to gamma-ray spectrometry, such as scintillation characteristics, energy resolution, intrinsic activity, decay time, non-proportionality of the response, and gamma-ray detection performance up to 3 MeV.

The unique advantages of each material are discussed in the context of specific detector requirements for high energy resolution and/or high detection sensitivity. The presence of 138La and 227Ac in these scintillators poses a limitation to their usage in low-intensity gamma-ray spectrometry, in particular for homeland security and environmental applications, which must rely on high detection sensitivity. Thanks to drastically reduced intrinsic activity, CeBr3 holds significant merit in ultra-high sensitivity detection when detection time is critical. The ability to detect signals and to distinguish peaks of nearly the same energy will be discussed for specific energy ranges and specific isotopes, including, for example, the detection of 40K and its interference with 138La.

### [217] Connection between TSL and afterglow in mixed oxide garnet ceramics

### Presenter: KHANIN, Vasilii

Oxide garnet materials are widely used as scintillators and phosphors and are being actively investigated. Composition versatility for this class of materials enables optimizing garnets' physical properties to specific application requirements in terms of density, light output or afterglow. Many medical and industrial scintillator applications have strict requirements to time performance of scintillators. E.g. medical computed tomography (CT) requires low (0.1%) afterglow levels in the ms-time range [1].

Relatively high light yields (> 50-60 ph/KeV) have been reported for the Ce-doped (Y,Gd)- and Gd<sub>3</sub>(Ga,Al)-mixed garnets [2]. However, many of the oxide garnets are characterized by a multi-exponential decay and severe afterglow [3]. The afterglow is due to trapping of electrons and holes. Trap centers are investigated using thermally stimulated luminescence (TSL) and isothermal decay (afterglow) measurements. Generally, TSL and afterglow curves of scintillators are studied separately, even though traps responsible for both processes can be the same.

This talk describes our research on the Ce-doped (Y,Gd)- and Gd<sub>3</sub>(Ga,Al)-garnet ceramics with the aim to correlate their TSL and afterglow measurements. The samples were either co-doped with 2–50 mole ppm of Cr, Yb or Eu, or left nominally pure. For all the samples TSL curves at 1-40 K/min heating rates and afterglow curves in the  $10^{-3}$ - $10^{4}$  sec time range and at several temperatures in the 300-430 K range have been measured.

We have developed procedures to determine trap parameters such as thermal trap depth and frequency factor in an unambiguous manner by connecting TSL and afterglow measurements. In order to accomplish that, we have devised a special method of extracting the lifetime of trapped carriers from afterglow measurements, independent of kinetic order. 1. Eijk van C. W. E. // Phys. Med. Biol. 47, R85 (2002)

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### [231] The emission centers in YAG crystals grown in various conditions

Presenter: VASYUKOV, Sergey

The emission centers in YAG crystals grown in various conditions S. Vasyukov 1, O. Sidletskiy 1, P. Arhipov 1, S. Tkachenko 1, A. Vedda 2, N. Shiran 1, V. Nesterkina 1

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The energy relaxation in the final stages of the scintillation process depends on the number of radiative and non-radiative relaxation centers, as also their properties. In many ways, the purity and quality of the crystals is crucial for minimizing energy losses at this stage. In the case of non-activated and doped YAG crystals, growing conditions and post-processing of the material are also important.

The work is aimed to research of YAG single crystals' luminescence at different excitations in the wide temperature range (from 77 to 520 K) and various conditions.

The optical properties of YAG crystals grown according to the "classical scheme" - using Iridium crucibles and crystals obtained by the "modified scheme" - as crucibles used Molybdenum or Tungsten will be compared and discussed. It is shown that special treatment and other conditions allows to reach high quality of YAG crystals.

### Characterization: session 2 (11:00-12:30)

-Conveners: William Moses

time [id] title

### 11:00 [208] Precise rise time measurements of inorganic scintillators using X-ray and 511 keV excitation

Presenter: GUNDACKER, Stefan

The emergence of new solid-state avalanche photodetectors, e.g. SiPMs, with unprecedented timing capabilities opens new ways to profit from ultrafast and prompt photonemission in scintillators. In time of flight positron emission tomography (TOF-PET) and high energy timing detectors based on scintillators the ultimate coincidence time resolution (CTR) achievable is proportional to the square root of the scintillation rise time, decay time and the reciprocal light yield, \$CTR \propto \sqrt{\tau\_r \tau\_d/LY}\$. Hence, the precise study of light emission in the very first tens of picoseconds is indispensable to understand time resolution limitations imposed by the scintillator. We developed a time correlated single photon counting setup having a Gaussian impulse response function (IRF) of 63ps sigma, allowing us to precisely measure the scintillation rise time of various materials with 511keV excitation. In L(Y)SO:Ce we found two rise time components, the first below the resolution of our setup \$<\$10ps and a second component being \$\sim\$300ps. Codoping with Ca\$^{2+}\$ almost completely suppresses the slow rise component leading to a very fast initial scintillation emission with a rise time of \$<\$10ps. A very similar behavior we observe in LGSO:Ce crystals. The results are further confirmed by complementary measurements using a streak-camera system with pulsed X-ray excitation. Additionally we will present the scintillation kinematics in Ca\$^{2+}\$ or Mg\$^{2+}\$ codoped LuAG:Ce, YAG:Ce and GAGG:Ce samples. Finally the merits of a very fast scintillation rise time will be discussed in view of prompt photonemission in the crystal (e.g. Cherenkov radiation) in order to achieve 10ps in TOF-PET.

### 11:15 [214] Significant improvement of GAGG based scintillation detector performance by control of the electronic excitation dynamics

### Presenter: KORJIK, Mikhail

Recently, we showed that the codoping of GAGG:Ce single crystal by Mg results in a strong acceleration of the rate of free carriers nonradiative recombination. This effect competes with the radiative recombination of free carriers via Ce3+ ions and, consequently, results in a decrease of the scintillator light yield. The nonradiative recombination occurs when a hole migrates to the vicinity of a Mg2+-related defect. Thus, a possible solution to recuperate the light yield loss is slowing down the hole migration rate. Due to a strong temperature (T) dependence of the migration rate w~exp(-E/kT), where E is the constant, defined by the nature of the compound, k is the Boltzmann constant, the migration can be inhibited by cooling the crystal or the whole detecting unit. This report presents the results on a significant improvement of the performance of GAGG-based scintillation detector with temperature decrease. When temperature of a PMT-based detector is lowered to -45oC, its amplitude response at registration of y-quanta is improved by 30%; 662 keV photo-absorption peak FHHM was found to be better by a factor of up to 0.85, whereas scintillation kinetics become even faster. All this opens an opportunity for a wide application of GAGG scintillation detectors, particularly in a combination with SiPM photo-sensors, the signal-to-noise ratio of which also improves with the temperature decrease.
# 11:30 [116] Timing performance of GAGG:Ce and LuAG:Ce epitaxial garnet films co-doped by divalent Mg2+ ions *Presenter: KUCERA, Miroslav*

The Mg co-doping in Ce-activated garnet scintillators has shown several highly beneficial effects, such as acceleration of the scintillation decay or improved afterglow, while the LY remains sufficiently high [1]. Excellent scintillation properties have recently been reported in garnet epitaxial films GAGG:Ce,Mg grown by liquid phase epitaxy [2].

In this work, the effect of Mg2+ co-doping on timing characteristics is studied in (Lu0.8Gd2.2)(Al2.5Ga2.5)O12:Ce and Lu3Al5O12:Ce garnet films. The films were grown by liquid phase epitaxy from the lead-free BaO-B2O3-BaF2 flux. The kinetics of scintillation emission was studied under X-ray excitation pulse with FWHM of 620 ps. The results are quantitatively compared with those obtained by e-beam and optical excitations. Significant improvements of timing performance of the Mg2+ co-doped garnet scintillators have been observed, namely a substantial shortening of the rise time and acceleration of the scintillation decay. In particular, the rise time in GAGG:Ce,Mg decreased from 400 ps in Mg-free sample to 70 ps in the sample with 700 ppm Mg (values refer to the rise of the signal from 10 to 90 %). Furthermore, the signal decreases to only 0.07 % just at 1 microsecond after the X-ray pulse excitation and the scintillation decay to 1/e value accelerated from 70 to 15 ns in Mg co-doped samples. Any notable delay was not observed under optical excitation into the 5d1 absorption band. Similar improvements of the timing properties were also observed in LuAG:Ce,Mg.

Such excellent timing properties makes these garnet films competitive candidates for high rate imaging techniques or for the electron detectors in e beam devices.

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### 11:45 [137] Scintillation properties of LuAG-based scintillators: Influence of Ga-admixture, non-stoichiometry and Mg-codoping

### Presenter: PEJCHAL, Jan

Scintillators based on lutetium-aluminum garnet LuAG:Ce have been studied in past decades. Besides many favorable properties, slow decay components related to retrapping of electrons on the shallow traps related to anti-site defects in caused deterioration of scintillation parameters. These defects, where Lu resides at Al sites, are created at high temperatures during crystal growth from the melt. One of the strategies of improvement was Ga and Gd admixture leading to highly-efficient GAGG scintillators. Ga admixture lowered the edge of the conduction band burying the trapping levels. Gd admixture increased the separation of the Ce<sup>3+</sup> 5d1 level from the conduction band decreasing the probability of 5d-state ionization. Another strategy was Mg-codoping, which stabilized the tetravalent Ce<sup>4+</sup>, which can directly compete for electrons in the early stage of scintillation mechanism, suppressing the slow components and increasing the light yield.

Another way to improve the scintillation properties of LuAG:Ce can be the shift of the crystal stoichiometry towards the Al-rich compositions. There is also a question if combination of such stoichiometry and Mg codoping or Ga admixture can bring further improvement.

Ce, Mg-codoped samples grown from Al-rich melt (1at% and 3at% excess) were prepared by micro-pulling-down method to study the influence of non-stoichiometry. Ga admixed LuGAG crystals with the same Ce and Mg concentrations were grown as well together with the Al-rich analogs.

Codoping of Mg at a concentration around 300 ppm led to improvement of overall scintillation efficiency, which is attributed to the alternative scintillation mechanism involving Ce<sup>4+</sup> stabilized by Mg<sup>2+</sup>. Non stoichiometry 3 at% in the LuAG matrix for the Ce0.2% Mg 300 ppm sample led to significant improvement of the overall scintillation efficiency (almost a factor of 2) when compared to the stoichiometric samples. Exceptionally high light yield (17000 photons/MeV) under 667 MeV gamma-ray excitation (6 µs amplifier shaping time) was observed when compared to the standard stoichiometric LuAG:Ce crystals (5000 photons/MeV). Lower light yield values are due to lower quality crystals grown by the micro-pulling-down method. As was shown by the measurement of the scintillation decay under pulsed soft X-ray excitation, the highest Mg concentration brings additional slow components to the scintillation decay and did not bring any other improvement in general.

Very similar trends were observed for the Ga-admixed LuGAG, but the anti-site defect-related luminescence was strongly suppressed, which led to further improvement of the overall scintillation efficiency.

Thermally-stimulated luminescence (TSL) and temperature dependences of photoluminescence characteristics were measured for selected samples. Interestingly, tunneling of electrons towards Ce luminescence center from the adjacent defects was observed after excitation to the lowest Ce<sup>3+</sup> 5d state. For the non-stoichiometric sample, decrease of the steady-state luminescence intensity was observed at low temperatures around 150K, while for the stoichiometric sample the quenching starts above 450K. On the other hand, the photoluminescence decay time starts to decrease above 600K for all the samples, which points to the fact that a non-relaxed excited state of Ce<sup>3+</sup> can be involved. Luminescence and scintillation mechanism in relation with composition and stoichiometry will be presented and discussed.

### 12:00 [93] Consequences of Ca co-doping in YAIO\_3:Ce single crystals

### Presenter: MORETTI, Federico

Point defects play a relevant role in the scintillation process since they can act as traps for free carriers, created by the interaction of ionizing radiation with the scintillator, slowing down the carriers migration toward the radiative recombination centres. The competition which arises between charge trapping and recombination is ultimately responsible for the degradation of the scintillator performance with a reduction of light yield, the presence of rise time and long scintillation time decays - or even afterglow - and luminescence sensitization.[1] The defect impact on the scintillation process is well established and several strategies have been proposed to reduce the concentration of traps or their relevance upon the scintillation economy.[2] Co-doping with Ca or Mg in cerium doped garnets and silicates has proven itself as a very effective strategy to improve scintillation timing and light yield performances of these matrices. The detected improvements have been related to the partial oxidation of cerium ions to their tetravalent form induced by the presence of the co-dopant. Although Ce\$^{4+}\$ is not photoluminescent by itself, it can still take part in the scintillation process by temporarily capturing an electron, thus becoming trivalent and able to give rise to the typical Ce\$^{3+}\$ luminescence. In this contribution we will discuss the applicability of Ca co-doping in order to improve the scintillation characteristics of YAIO\$\_3\$:Ce (YAP:Ce) single crystal grown by Czochralski method. Four different Ca concentrations (from 0 to 500 ppm) combined with two cerium contents differing by one order of magnitude have been considered.[3] The optical properties of these samples have been studied by means of optical absorption (OA), steady state and sub-nanosecond time resolved photo- (PL) and radio-luminescence (RL). OA data showed a clear increase in absorption in the UV region as a function of Ca content and related to the Ca-induced Ce\$^{4+}\$ ions presence alongside regular 3+ ones. Steady state PL and RL data evidenced a rather clear reduction in the luminescence intensity related to reabsorption of the emitted light as well as energy transfer phenomena occurring from Ce\$^{3+}\$ to Ce\$^{4+}\$ ions. Time resolved RL profiles showed an acceleration of the main Ce\$^{3+}\$ luminescence decay coupled with a reduction in the contribution of long scintillation tails by increasing the Ca content; in weakly Ce doped samples, the observed scintillation decay improvements are also accompanied by a clear acceleration in the luminescence rise time. TSL results evidenced a reduction in carrier capture probability at defect sites. All these improvement are, however, accompanied by a substantial reduction in scintillation light yield. The obtained data clearly suggest that Ca co-doping is not a useful strategy to improve YAP:Ce crystal scintillation properties, but they also give valuable information on the applicability of this strategy on other matrices and activator ions. Moreover they also provide further insights in the understanding of charge carrier migration and trapping role on the scintillation time response.

This work has been supported by the H2020 project INTELUM (GA no.644260)

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[3] Moretti et al. ChemPhysChem 18(2017)493

### 12:15 [132] Pulse shape studies of various scintillators with waveform digitizing techniques

#### Presenter: WOLSZCZAK, Weronika

In this study we used a waveform digitizer to record, store and analyse individual scintillation pulses from the following scintillators: CeBr3, LaB3:Ce, LaBr3:Ce,Sr, NaI:TI, CsI:TI, and BaF2. With off-line analysis software we sorted the pulses according to their charge integrals. Then we obtained the pulse shape as a function of energy deposited within a scintillator. The result on pulse shape change with density of excitation can be used to verify currently investigated scintillation mechanisms and the postulated excitation track structure [1]. By exploiting a time correlation of nuclear decay events we separated 219-Rn and 215-Po alpha peaks from an internal alpha contamination spectrum in LaBr3:Ce, CeBr3, and LaBr3:Ce,Sr. We found that the alpha peaks are not symmetric in lanthanum-based scintillators. The origin of this phenomenon is unknown, but it may indicate a presence of domains in the crystal structure or a directional anisotropy [2]. This type of analysis can be used for collecting data on an intrinsic alpha excitation in scintillators, while avoiding surface effects [3]. We will show that by using a waveform digitizer all experimental data can be recorded and then analysed off-line, giving not only high flexibility, but also exceeding what is possible with classical nuclear electronics. For example, it is possible with a 137-Cs source to collect within one hour multiple scintillation decay curves for different deposited energies ranging from 10 to 662 keV. The same may take a few weeks using a classical start-stop method.

[1] X. Lu et al. "Energy-Dependent Scintillation Pulse Shape and Proportionality of Decay Components for CsI:TI: Modeling with Transport and Rate Equations", Phys. Rev. Applied, Vol. 7, Iss. 1, 2017.

[2] W. Wolszczak, P. Dorenbos, "Shape of intrinsic alpha pulse height spectra in lanthanide halide scintillators", NIM A, http://dx.doi.org/10.1016/j.nima.2017.02.041, 2017.

[3] W. Wolszczak, P. Dorenbos, "Non-proportional response of scintillators to alpha particle excitation", submited to IEEE TNS, 2017.

### COST ACTION TD1401 FAST: industrial Event: Industrial Presentations (14:30-17:00)

This year we launched the 3rd Industrial Workshop on FAST (Fast Advanced Scintillator Timing http://www.cost.eu/COST\_Actions/TDP/Actions/TD1401) at the European Trans Domain COST Action in Chamonix. This workshop brought together scholars, industry leaders and visionaries from across the world to discuss how academia and industry can partner to address the challenges and the opportunities that scintillator-based detectors with time precision better than 100ps presents.

A round-table discussion between the representatives from the industry and science should demonstrate what the industrial side need from the academia and vice versa at the end of the workshop. The success of the Research and Industrial Workshop and the dialogue that we started should continue in the following years. contact: k.ziemons@fh-aachen.de,

-Conveners: Karl Ziemons

time [id] title

### 14:30 [176] Scintillator materials and phenomena studied for fast timing

Presenter: NIKL , Martin

Fast and efficient scintillators are required by a number of applications, namely in high energy physics and medical imaging (Positrom Emission Tomography – PET) where so call time-of-light (TOF) techniques are used and timing coincidence resolution (TCR) is of critical importance. Today, with LSO-type single crystals, TCR values closed to 100 ps have been demonstrated, but it seems improbable to reach values below 70 ps using a bulk scintillator of this kind. The goal of COST FAST project is to seek the concept(s) to advance TCR up to by one order of magnitude, down to 10 ps which would give an unprecedented improvement in the applications using TOF techniques, e.g. a direct image reconstruction could be used in TOF-PET imaging.

To reach such a goal sufficiently high number of scintillation photons must be detected within several tens of ps after the absorption of high energy photon (particle) by the scintillator. Such a scintillator should posses high light yield, as short as possible decay time and negligible rising components in scintillation response. Direct band gap semiconductors as ZnO, GaN, PbI2, CdSe or CsPbCl3 [1] could be used as their luminescence based on Wannier exciton shows the radiative lifetime below 1 ns and theoretical light yield could exceed 100 000 phot/MeV due to smaller value of band gap. However, small Stokes shift, which is a consequence of Wannier exciton nature, practically disable to use these materials in a bulk form due to huge reabsorption losess. Moreover, due to the fact that such excitation is not localized, nonradiative quenching e.g. at the surface or interface of these materials becomes a critical problem. The transparent composite materials, in which the scintillation nanophase is embedded in an optically transparent host, e.g. ZnO:Ga nanocrystals in a polystyrene matrix, became intensively studied [2]. Moreover, excitonic emission in GaN-GalnN multiple quantum wells shows also very promising timing characteristics in this respect [3]. Finally, other ultrafast phenomena as crossluminescence, Cherenkov and intraband luminescence are also considered for this purpose though their yield seems to be too low at the materials studied so far.

In the presentation we will discuss in the detail some of the above mentioned materials and phenomena and their practical potential for fast timing.

Acknowledgement. This work is based upon work from COST Action (TD1401, FAST), supported by COST (European Cooperation in Science and Technology).

1. S.E.Derenzo et al, Nucl. Instr. Meth. Phys. Research A 486 (2002) 214.

2. H. Buresova et al, Optics Express 24 (2016) 15289.

3. A. Hospodkova et al, Nanotechnology 25 (2014) 455501.

### 15:00 [213] From Academic Research to Scintillator Crystal Industry

#### Presenter: CHAI, Bruce

Changing from Academic to Entrepreneurship is certainly a glamorous dream for everyone. But this is a very hard road in reality. Research in crystal growth and application has a lot of fun and we all enjoyed to do it. There seems to have endless number of materials that can be studied in research. However, when to put it in real industrial application, the usable material list drops significantly with only a few survivors.

Scintillator crystal industry is no difference. It is totally technology driven. There are hundreds of known scintillating crystals. But only a few of them are useful. Nevertheless, the actual demand of crystal quantity is much higher than people would have expected. The reason is simple. We need finite volume of crystals in order to capture high energy rays and particles. We are very fortunate that single crystals still provide the best scintillating properties over ceramics, plastics and other composites. This is the only reason why scintillator crystal industry still can exist.

Current known scintillator crystals can serve the demand adequately. But there is always the need of research for better materials. So academic research and industrial production are in a beautiful balance between them.

### 15:30 [230] Industrial Scale R&D of Fast Scintillators

Presenter: HOUZVICKA, Jindrich Industrial Scale R&D of Fast Scintillators

Silvia Sykorová1, Martin Nikl2, Jindřich Houžvička1

1) CRYTUR, spol. s r.o., Palackého 175, 511 01 Turnov, Czech Republic,, www.crytur.com

2) Institute of Physics, Academy of Sciences, Na Slovance, 180 00 Praha, www.fzu.cz

Large infrastructure projects e.g. in CERN or FAIR require typically large volume scintillators of the unique technical parameters, but manufactured with affordable cost and production capacity corresponding to the size of the detector and available time frame. Example of such scintillator development can be documented on the recent re-start of the PWO production, as well as on the new development for the considered calorimeter up-grades in CERN using the modified FastYAG crystals.

The new patented crystal growth method (CRIG - CRystal Improved Growth) was developed to grow large, core-free garnet crystals (YAG, LuAG, etc) recently. The maximum diameter reached so far is 140mm, and the maximum crystal weight is up to 12 kg. Crystals of unmatched quality are produced using this method – with their unique size, homogeneity and the low material stress.

Garnet materials (especially YAG:Ce) show excellent radiation hardness. Hence, they can be used, for example, even for very large multipixel detectors in very strong radiation fields. Around 8.000 fibres, 1x1x200mm can principally be made out from one such a big size crystal, thus meeting the requirements both from the perspective of the cost and manufacturing capacity. Fibre optical parameters still remain very impressive, significantly better than those of fibres produced by micro-pulling down technology so far reported.

Size of the mentioned crystals and their homogeneity matter also for many other applications. YAG:Ce crystal is frequently used as a screen for imaging of various objects. Enlarged size of the screen brings obvious advantages. The screens can be made in such a quality that submicron resolution can be achieved when inspecting various material, even those with low Z-contrast like living tissues or carbon fibre/carbon resin composites.

The key scintillation parameters like light yield and decay time can be modified to meet the parameters specific and critical in certain detectors. Example of the new FastYAG crystal, doped with cerium, will be shown. Decay time of this material was tuned down to 40 ns only, and afterglow minimized as well, to reduce pile up in the considered up-grade of HL-LHC calorimeter. Still, the crystal can be made of such size, that its application seems to be the competitive solution also from the cost point of view.

15:45	[242] Fomos-Materials experience to control crystal properties Presenter: BUZANOV, Oleg O.Buzanov1, V.Alenkov1
	1- JSC "Fomos-Materials" Moscow, Russia
	Here we report some results for the scintillation crystal growth and their performance. JSC "Fomos-Materials" is a Company which is involved in the crystal growth by different methods of the crystal pulling from the melt. We produce not only crystal ingots but also elements and more complicated assembly from the crystalline elements. One of the branches where our Company demonstrated a high level of the technology is a production of radio-pure scintillation materials with preselected isotopes. Molybdate single crystals are perspective for the cryogenic scintillating bolometers that operate in ultra-low temperature conditions and used to search for neutrinoless double beta decay ( $0v\beta\beta$ ) of 100Mo. Among tens of possible candidates, the isotope 100Mo is one of the most promising one. Furthermore, scintillating bolometers possess potentially very low background counting rate. Recently, we showed that performance of GAGG:Ce single crystal can be sufficiently improved at multi-doping. Each doping, which play specific role at the creation of scintillation, promotes fast transfer of electronic excitations. Some aspects of the production of the mixed garnets with multi-doping will be discussed.
16:00	[235] C&A Presenter: KAMADA , Kei
16:15	[236] scionix Presenter: SCHOTANUS, Paul
16:30	[237] Scintacor Presenter: DESTEFANIS, Carlos
16:45	[244] St Gobain Presenter: OUSPENSKI, Vladimir

### Coffee Break (17:00-17:30)

### COST ACTION TD1401 FAST: industrial Event: Round Table (17:30-18:30)

This year we launched the 3rd Industrial Workshop on FAST (Fast Advanced Scintillator Timing http://www.cost.eu/COST\_Actions/TDP/Actions/TD1401) at the European Trans Domain COST Action in Chamonix. This workshop brought together scholars, industry leaders and visionaries from across the world to discuss how academia and industry can partner to address the challenges and the opportunities that scintillator-based detectors with time precision better than 100ps presents.

A round-table discussion between the representatives from the industry and science should demonstrate what the industrial side need from the academia and vice versa at the end of the workshop. The success of the Research and Industrial Workshop and the dialogue that we started should continue in the following years. contact: k.ziemons@fh-aachen.de,

-Conveners: Karl Ziemons

Conference Banquet (19:30-00:30)

### Friday 22 September 2017

Crystal growth: session 2 (08:30-10:00)

### -Conveners: EDITH BOURRET

### time [id] title

### 08:30 [101] Ultra radio-pure scintillators for Rare Events Physics

### Presenter: DAFINEI, Ioan

Rare event search experiments, like the experiments searching for a direct evidence of dark matter and those searching for the neutrinoless double beta decay (0uDBD), are among the most exciting challenges of the modern physics. The sensitivity of such experiments is driven by the background, which depends substantially on the radio-purity of the materials used for the construction of the experimental apparatus. When scintillating crystals are used as detectors, dedicated production lines need to be conceived for the entire manufacturing process from the synthesis and conditioning of the powder used for crystal growth to the cutting and surface processing of the finished crystal, compliant with radio-purity constraints specific to rare event physics application. Besides routine check of impurities concentration, high sensitivity measurements are made for radio-isotope concentrations in raw materials, reactants, consumables, ancillaries and intermediary products.

When very expensive isotopically enriched materials are used for 0uDBD experiments, special precautions are taken for acquiring the maximum yield in the mass balance of all production stages.

The current work will make a review of scientific and technological aspects related to the crystal production for 0uDBD and dark matter search applications. Details will be given on the production of enriched zinc selenide (Zn82Se) and tellurium dioxide (130TeO2) crystals for 0uDBD experiments together with the performance of these crystals used as scintillating and Cerenkov bolometers respectively. The synthesis of ultrapure Nal powder and the growth and processing of Nal:TI crystals for possible use in experiments dedicated to the direct detection of dark matter will also be described. In this case, given the chemical affinity between sodium and potassium, dedicated measures will be discussed for the reduction of potassium content below 10-8 g/g limit, imposed by radio-purity constraints. The work will also describe improvements brought to cutting edge measurement techniques like mass spectrometry (MS) for a fast, sensitive and efficient screening of the materials used for the production of crystals and the certification of the whole crystal production process in a reliable and reproducible way.

### Acknowledgements

This work was made with the contribution of the Ministry of Education, Universities and Research of the Italian Republic, MAECI-PGR04136

# <sup>08:45</sup> [44] Epitaxial Growth of Ce-doped (Pb,Gd)3(Al,Ga)5O12 Films and Their Optical, Photoluminescence and Scintillation Properties

### Presenter: VASIL'EV, Dmitrii

The epitaxial films grown by liquid-phase epitaxy (LPE) have found an application as scintillation screens with the development of the technique of microimages obtaining using hard X-rays. Lu2SiO5:Tb, Gd3Ga5O12:Tb and Gd3Ga5O12:Eu epitaxial films are already used for such screens [1]. Recently, studies of the stimulated scintillation emission depletion (STED) properties of Lu3Al5O12:Ce (0.07% at.), Lu2SiO5:Ce, Gd2.96Ce0.03Al3.14Ga1.86O12, Lu2SiO5:Tb (12% at.), Gd3Ga5O12:Eu (2.5% at.) single-crystalline films were carried out [2]. It was shown, that Lu2SiO5:Tb scintillating film and a 628 nm cw STED-laser is the most promising combination for stimulated scintillation emission depletion X-ray imaging [2,3].

Here we report the results of the study of the epitaxial growth, optical, photoluminescence and scintillation properties of (Pb,Gd)3(AlGa)5O12:Ce films. The goal of the study was to determine the composition of the melt solution, which allows to grow films with maximal photoluminescence and light output.

The garnet films were grown on (111)-oriented single crystal Gd3Ga5O12 substrates by LPE from a supercooled PbO–B2O3 based melt solutions with gadolinium oxide (C(Gd2O3)) concentrations between 0.2 and 0.5 mol% in the mixture, C(CeO2) concentrations 0.2 and 0.3 mol% and C(Al2O3) concentrations between 2.1 and 4.5 mol%. Studies of the optical absorption of the grown films have shown that the decrease of the intensity of the absorption band of 5d2 level and the increase of the absorption in the region up to 360 nm occurs for films which were grown from a melt solution with C(Gd2O3)=0.5 mol%, C(CeO2)=0.2 mol% and C(Al2O3)= 4.5 mol%. The effect is attributed to the formation of Ce4+ centers, which energy levels are situated near the bottom of the conduction band. Photoluminescent studies have shown that the most intensive photoluminescence was observed in the Pb0.01Ce0.03Gd2.96Al3.14Ga1.86O12 film which was grown from the melt solution with C(CeO2)=0.2 mol%, C(Gd2O3)=0.4 mol% and C(Al2O3)= 4.5 mol%. The cathodoluminescence decay curve for this film was fitted by triple-exponential decay law with parameters t1=2.1 ns (2%), t2 =24.9 ns (30%) and t3=61.0 (68%). The mean scintillation decay time in this film was 43 ns. The cathodoluminescence light output was estimated relatively to that of the well-known scintillator LYSO:Ce. The values were obtained: of ~ 51500 photon/MeV under electron excitation at 80-120 keV and ~20000 photon/MeV under the Radio Isotope source excitation at

32 keV.

1. Douissard P.-A., Cecilia A., Martin Th. Chevalier V., Couchaud M., Baumbach T., Dupre K., Kuehbacher M., Rack A. J. Synchrotron Rad., 17, 571 (2010).

2. Alekhin M.S., Renger J., Kasperczyk M., Douissard P.-A., Martin T., Zorenko Y., Vasil'ev D. A., Stiefel M., Novotny L., Stampanoni M. J. Optics express, 25, 1251 (2017).

3. Alekhin M.S., Patton G., Dujardin C., Douissard P.-A., Lebugle M., Novotny L., Stampanoni M. J. Optics express, 25, 654 (2017).

09:00 [197] Scintillation Properties of La\_2Hf\_2O\_7 Transparent Ceramics by the Spark Plasm Sintering Method Presenter: KUROSAWA, Shunsuke Shunsuke Kurosawa\$^{1,2\*}\$, Koichi Harata\$^3\$, Shohei Kodama\$^3\$, Shinosuke Yamato\$^3\$, Akihiro Yamaji\$^3\$, Jan Pejchal\$^4\$, Yuji Ohashi\$^3\$, Kei Kamada\$^{1, 5}\$, Yuui Yokota\$^{1}\$, Akira Yoshikawa\$^{1, 3, 5}\$ \$^1\$New Industry Creation Hatchery Center (NICHe) 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai, Miyagi 980-8579, Japan \$^2\$Faculty of Science, Yamagata University, Kojirakawa-machi 1-4-12, Yamagata 990-8560, Japan \$^3\$Institute for Materials Research (IMR), Tohoku University 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan \$^4\$Institute of Physics, AS CR, Cukrovarnická 10, 162 53 Prague, Czech Republic. \$^5\$ C&A Corporation, 6-6-40 Aoba, Aramaki, Aoba-ku, Sendai, Miyagi 980-8577, Japan. Scintillators are used in medical imaging and high-energy physics as radiation detectors, and high effective atomic number materials are required in order to obtain the high gamma-ray detection efficiency. Recently, pyrochlore structure type materials have been investigated as scintillation materials, because these materials had good light outputs [1]. Since La\$\_{2}\$Hf\$\_{2}\$O\$\_{7}\$, a member of the pyrochlore group, has high an effective atomic number of approximately 62, this material can be high gamma-ray detection efficiency. On the other hand, the melting point of this material is over 2,500 \$^\circ\$C. Since almost crucibles cannot be operated under the such high temperature, and the common single-crystal-growth method is hard to apply to this material. Thus, we investigate the luminescent properties of La\$\_{2}\$Hf\$\_{2}\$O\$\_{7}\$ as ceramics. Here, the spark plasma sintering (SPS) method is one of the methods to fabricate the ceramics, and it takes shorter time to obtain the ceramics compared with other methods such as HIP. First, pure RE-doped La\$ {2}\$Hf\$ {2}\$O\$ {7}\$ ceramics powers were prepared by the solid state reaction, where is Eu, Tb, and transparent ceramics were obtained by the SPS method. All samples were confirmed to be the single phase, and these samples had transparency. Both Eu and Tb-doped Las\_{2}\$Hfs\_{2}\$O\$\_{7}\$ samples had some sharp emission peaks around 550-700 and 350-650 nm, respectively, in the radio luminescence spectra excited by X-rays or 5.5 MeV alpha rays from an \$^{241}\$Am source. Each peak was originated from Eu\$^{3+}\$ and Tb\$^{3+}\$ 4f-4f transition. In this paper, we show the optical and scintillation

[1] A. Suzuki, S. Kruosawa, A. Yoshikawa et al., Applied Physics Express 5 102601(2012)

properties of these materials including temperature dependence and time profile in this presentation.

# 09:15 **[40]** Comparative study of rare-earth aluminate scintillation crystals fabricated under different conditions *Presenter: SIDLETSKIY, Oleg*

Currently, Ce- or Pr-doped rare-earth aluminates with the garnet and perovskite structures, Y3Al5O12 (YAG), Lu3Al5O12 (LuAG), YAlO3 (YAP), as well as Al/Ga substituted garnets are the candidates for a range of scintillation applications, including future HEP experiments at colliders. High melting temperatures of garnets and the need for expensive Ir crucibles and shields stimulates the development of less costly crystal fabrication technologies. For instance, garnets and perovskites have been successfully grown in Mo and W crucibles under the reducing Ar+H2 atmosphere conditions by the Bridgman [1] and Czochralski [2] methods. The reducing atmosphere is necessary to avoid Mo(W) oxidation and melt contamination by its products.

This report represents a novel fabrication process of undoped, and Ce3+, Pr3+, Sc3+-doped YAG and LuAG, as well as perovskite crystals (YAIO3, CeAIO3) by the Czochralski and EFG methods in Mo and W crucibles under the reducing Ar+CO atmosphere. This method has potential advantages over the growth under H2, namely, avoiding the use of explosive H2, as well as substitution of expensive ZrO2 and corundum heat insulation with graphite.

Optical and scintillation properties of the crystals fabricated by the different methods are discussed. Creation/elimination of point defects, or change of valence state of admixtures, which act as electron or hole traps, are the cause of garnet crystals coloration. Therefore, the reversible coloration/discoloration of YAG grown under neutral atmosphere (Ar) is controlled by post-growth thermal annealing in reducing/oxidizing atmosphere, correspondingly. Meanwhile, the coloration of garnet crystals grown under Ar+CO can be eliminated irreversibly by the oxidizing or reducing high-temperature annealing. This is an evidence of more complex mechanisms of defect formation in YAG grown under the Ar+CO involving carbon and/or Mo(W) admixtures. Carbon is introduced into garnets due to interactions of melt and crystal with Ar+CO atmosphere. The carbon concentration in as-grown YAG determined by the element analysis is ~10-2 wt%. As carbon oxidation state may vary from -4 to +4, it may act as an active electron trap and compete with color centers for electron capture. This explains why an expected increase of concentrations of oxygen and Al vacancies in YAG:C under the reducing annealing not results in formation of color centers. A similar mechanism of trap decoration by hydrogen and elimination of trapping at cation vacancies in YAG crystal grown under H2 atmosphere was suggested [3].

From the practical point, such features of crystals grown under Ar+CO open new possibilities to optimize the scintillation properties of rare-earth aluminates. The presence of active electron traps makes it possible to transfer activators (Ce, Pr, or Nd) into the optically active lower valence state by the reducing annealing thereby increasing a quantity of luminescence centers without a loss of crystal transparency.

\*The work is supported by the H2020-MSCA-RISE-2014 Project No. 644260 (INTELUM).\*

[1] A. Petrosyan, J.Cryst.Growth139 (1994) 372.

[2] J. Houžvička, K. Bartoš, Patent US 9,499,923 B2 (2016).

[3] F. Selim, C. Varney, M. Tarun, et al, Phys. Rev. B88 (2013)174102.

# 09:30 **[201] Shaped crystal growth of novel oxide scintillators by the edge defined film fed growth method** *Presenter: KAMADA, Kei*

Scintillator materials are used for radiation detection applications such as medical imaging techniques, high energy, homeland security, well loging, nuclear physics detectors, etc. In the last two decades, great R&D effort brought several novel scintillator material systems[1-4], namely the Ce-doped orthosilicates as Lu2SiO5 (LSO), Y2SiO5 (YSO), pyrosilicates based on(La,Gd)2Si2O7 (La-GPS), aluminum perovskites as LuAIO3 (LuAP), YAIO3 (YAP) and garnets as Lu3AI5O12 (Ce:LuAG), Y3AI5O12 (Ce:YAG), Ce:Gd3(AI,Ga)5O12(GAGG). These scintillator single crystals are commercially produced by the Czochralski method using Ir crucibles because of their high melting point around 1900-2130 °C. At the production higher material costs of Ir crucibles and their repairing costs occupy most of the crystals costs. Scintillator crystals are used mainly as rectangular shaped pixels which are processed from 3-4 inch diameter bulk single crystals. These processing costs are substantially an economic burden, too.

Up to now, shaped crystal growth of sapphire single crystal with shapes of tube, plate, fiber, etc was commercially developed by Edge defined Film Fed Growth (EFG) method [2]. Recently a few companies are producing shaped sapphire single crystal by EFG method using Mo crucible and die. Mo is several hundreds times lower cost material than Ir. Shaped growth by the EFG method using the low cost Mo crucible and die is a today's factor of cost reduction of sapphire. In this study, possibility of mass production of above mentioned oxide scintillators by the EFG method using Mo crucible and die is investigated.

At the beginning of this study, reactivity and contact angles of these oxides melts and Mo were investigated. A EFG furnace equipped with a graphite resistive heating unit and shields was used for these investigation under Ar atmosphere. Each oxides powder are melted in Mo crucibles. Radioluminescence, Mo contamination measurements by ICP and powder XRD are performed to check the reactivity. Mo dies are designed according to the contact angles. For example, Ce doped LuAG and YAG were grown using 1 x 30 mm Mo dies at a growth rate of 0.3mm/min under Ar atmosphere using <100> LuAG seeds. 1 x 30 x 50mm plates of Ce doped LuAG was successfully grown by the EFG method. Ce3+ 4f5d emission was observed 500 nm. Light yield was comparable to a Cz standard sample and around 18000 photon/MeV. Mo contamination was around 52 ppm. There is no harmful effect from the usage of Mo for the growth of Ce:LuAG scintillator. Furthermore 10 x 10 x 30mm Ce:YAP, Ce:La-GPS, Ce:YSO, Ce:LSO crystals were grown by the EFG method. In our presentation, details on reactivity, contact angles, Crusible and die designs, growth conditions, Mo contamination, chemical composition analysis, optical and scintillation properties of the grown crystals will be discussed.

[1] C. L. Melcher et al, IEEE Trans. Nucl. Sci., vol. 39, no. 4, pp.502–505, Aug. 1992.

[2] P. Lecoq et al, IEEE Trans. Nucl. Sci., vol. 49, no. 4, pp. 1651–1654, 2002.

[3] M. Nik et al, Meas. Sci. Technol. Vol.17, pp.R37-R54, Feb. 2006

[4] K. Kamada et al, Cryst. Growth Des., vol.11 part10, pp 4484–4490 Aug. 2011,

[5] H.E. LaBelle Jr., J. Cry. Growth 50(1980) 8-17

00.45	
09.45	[46] Growth of Ce-doped garnets with additional monovalent impurities and related effects
	The vertical Bridgman method is widely used in research of various scintillator crystals including garnets in order to
	improve their performance [1] Conversion of a part of Ce3+ ions to Ce4+ by divalent co-doning is presently considered as
	one of the most efficient ways to improve the performance of Ce-doped garnet scintillators [2]. While cations with unit
	charge difference and nearly the same size are easily tolerated in garnet hosts [2.3], the case of monovalent cations is less
	evident and insufficient information is available to consider their substitution preferences and functional role. Most of
	reported studies were performed on crystals grown by Czochralski and micro-pulling techniques.
	In this presentation we shall discuss incorporation of monovalent impurities (Li+ and Na+) and related effects in YAG:Ce
	and LuAG:Ce single crystals grown by Bridgman. Measurements of lattice constant (ao), optical absorption,
	radioluminescence and pulsed X-ray decays were performed to reveal the tendencies of impurity incorporation and
	localization within the lattice, charge balance maintenance and impact of impurities on optical properties and performance.
	Annealing and gamma-ray irradiation effects have been examined in comparison to those in crystals with divalent co-doping
	and without any co-doping.
	Measurements of ao in series of ceramic samples prepared by solid phase reactions did not give clear evidence for
	substitution at any cation site within the lattice. In most cases the slopes of measured and calculated dependences of ao on
	Li or Na concentration (assuming incorporation in dodecahedral sites) are very different suggesting, among others,
	substitutions at interstitial positions. In contrast to crystals with divalent co-doping, which show an increase of absorption in
	the UV range due to conversion of a part of Ce3+ to Ce4+, there is no such effect in most of the studied crystals with Li or
	Na. Presence of impunities is nowever evidenced in acceleration of the fast decay component and decrease of radialuminascences intensity. Rediction bordances of envetals is high showing as induced absorption bonds in the range of
	adioidininescence intensity. Radiation maturess of crystals is high showing no induced absorption bands in the range of
	prepared by other techniques. The advantages and limitations of the co-doning approach with various species will be
	discussed basing on observed tendencies of incorporation and crystal growth and perfection
	This work was performed in the frame of the International Associated Laboratory (CNRS–France & SCS–Armenia) IRMAS
	and European Union Horizon 2020 Program under grant agreement no. 644260 (Intelum).
	[1] A.G. Petrosyan, K.L. Ovanesyan, R.V. Sargsyan, et al, J. Crystal Growth 312 (2010) 3136.
	[2] M. Nikl, K. Kamada, V. Babin, et al, Cryst. Growth Des., 14 (2014) 4827.
	[3] A.G. Petrosyan, K.L. Ovanesyan, M.V. Derdzyan, et al, J. Crystal Growth 430 (2015) 46.

### coffee break (10:00-10:30)

Applications: session 3 (10:30-12:15)

-Conveners: Christian Morel time [id] title

### 10:30 [38] Light Yield Enhancement of the 157-Gadolinium Oxysulfide Scintillator Screens for the High-resolution Neutron Imaging

Presenter: CRHA, Jan

LIGHT YIELD ENHANCEMENT OF THE 157-GADOLINIUM OXYSULFIDE SCINTILLATOR SCREENS FOR THE HIGH-RESOLUTION NEUTRON IMAGING

Jan Crha1,2, Joan Vila-Comamala3, Eberhard Lehmann1, Christian David4, Pavel Trtik1,2

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The work presented here follows upon the recent enhancement of Gadox (Gd2O2S:Tb3+) scintillator screen within the Neutron Microscope project [1] at Neutron Imaging and Activation Group (NIAG) of the Paul Scherrer Insitut (PSI). Recently, highly enriched gadolinium (enriched in Gd-157 isotope) of Gadox phosphor powder [2] was utilized for the enhancement of the neutron absorption. This allowed for a significant reduction of scintillator layer thickness and thus led to the corresponding improvement of the spatial resolution of the neutron imaging setup [3].

Here we present the first attempts on micro/nanostructuring of the high-resolution neutron-sensitive scintillator substrates using atomic layer deposition (ALD). For this purpose, iridium layers of different thicknesses were deposited by ALD onto silicon substrates. The application of the iridium layers resulted in an increase of the light output up to 60 % in comparison with the uncoated silicon substrates. This increase in the light yield can be attributed to both the higher reflectance of the substrate and the enhanced back-scatter of the conversion electrons off the high density layer of the deposited iridium. The decrease in the spatial resolution of the Ir-deposited scintillator screen was found to be rather minor. The results are supported by Monte Carlo simulations. The outlook of the future steps regarding micro/nanostructuring of the neutron-sensitive scintillators will be presented.

### References

[1] P. Trtik, J. Hovind, C. Grünzweig, A. Bollhalder, V. Thominet, C. David, A. Kaestner and E. H. Lehmann, "Improving the Spatial Resolution of Neutron Imaging at Paul Scherrer Institut - The Neutron Microscope Project," Phys. Proc., vol. 69, 2015.

[2] P. Trtik and E. H. Lehmann, "Isotopically-enriched gadolinium-157 oxysulfide scintillator for the high-resolution neutron imaging," Nucl. Instr. Meth. Phys. Res., vol. 788, 2015.

[3] P. Trtik and E. H. Lehmann, "Progress in High-resolution Neutron Imaging at the Paul Scherrer Institut - The Neutron Microscope Project", J. Phys. - Conf. Series, vol. 746, 2017

### 10:45 [69] Ultrafast Scintillator for Dynamic Compression Studies

Presenter: NAGARKAR, Vivek

The Dynamic Compression Sector (DCS) at the Advanced Photon Source (APS) links a high-energy, tunable X-ray beamline to state-of-the-art dynamic compression plat-forms. This novel development will permit real-time X-ray measurements (diffraction, imaging, scattering) in dynamically compressed materials (peak stresses to over 350 GPa, and time duration ranging from ~5 ns to microseconds). Thus, the advent of the DCS will enable understanding of the dynamics of materials response to intense shock waves, geomaterials and other materials under extreme stress. High-speed X-ray imaging, however, is inherently photon starved. Even though the beam flux at synchrotron facilities is extremely high (~10^20 X-ray photons/mm^2/s) the number of X-rays available for imaging (100 ps pulse duration) may be limited due to weak reflections and/or attenuation in the material under test. Consequently the burden of maintaining high signal-to-noise ratio (SNR) in the image is shifted to the scintillator used in high speed imaging systems. We manufactured scintillator screens of cerium-doped lutetium iodide (LuI3:Ce). This material is well-suited for large-area coatings using vacuum-based physical vapor deposition methods. With its high density (~5.6 g/cm^3`), high effective Z (59.7), bright (exceeding 115,000 ph/MeV) green emission (540 nm range) well matched to commercial optics and CCD sensors, and rapid, afterglow-free decay (~28 ns in crystals), this scintillation material is arguably the best suited material candidate for the dynamic compression and alternative high-speed imaging applications. In its film format, a primary decay time component of 12.6 ns has been demonstrated. This, combined with the low afterglow, made it possible to resolve the 153 ns synchrotron bunch structure demonstrating the high speed imaging ability of this materials.

Our lutetium-iodide film provides us with about 8 µm resolution at 70 keV which energy is necessary to penetrate metallic structural materials. Also, superior performance in imaging and diffraction mode at DCS have been demonstrated. Lul3:Ce is highly hygroscopic. Given the deleterious and effect of moisture, it is of critical importance that the coating and our novel dust-free, hermetic packaging steps be carried out in a climate controlled, dry environment. In order to address this challenge, and realizing the tremendous commercial potential of the ultrafast Lul3:Ce, we have acquired a unique tool called the "Glove-box-enabled Evaporator" (GBE). The GBE features a vacuum evaporator fully integrated in a glovebox and enables commercial grade manufacturing of high efficiency but moisture sensitive scintillating materials, such as Lul3:Ce scintillators while preventing exposure to moisture during the process.

Here, we report on the scintillation properties of films and those for corresponding crystalline material. The vapor grown films were integrated into a high-speed CMOS imager to demonstrate high-speed radiography capability. The films were also tested at the DCS of the Advanced Photon Source at Argonne National Laboratory under X-ray irradiation. The data show a high image quality and the sufficient temporal resolution for the recent bunch mode.

### 11:00 [66] Optimization of micro columnar CsI:Tl scintillators for X-ray medical imaging applications

#### Presenter: DOREL, Marc

X-ray detector market faces an annual growth of about 5% due to technological advancement, ageing of the population and the increasing use of medical imaging. Trixell, founded in 1997, was one of the first producers to propose digital flat panels for medical radiology. In its detectors, the X-ray detection is indirect and the scintillating function is given by a microcolumnar CsI:Tl layer. In this paper, the radiological context is first described. Trixell was able to keep up with the fast growth of the market and to meet the requirements of the market and the customer needs; the corresponding capacity increase enables Trixell to be among the world leaders in its domain. An important effort of research and development is still maintained by the company, in particular for the scintillator material, to facilitate the design of products of higher performance and consolidate its leader place. Depending on the application of the digital flat panel developed, the required characteristics of the scintillator can be very different in terms of light output, X-ray absorption and resolution (characterized by Modulation Transfer Function MTF). A specific bench was developed in Trixell to measure the performance of these scintillators independently of the photodiode array which converts visible light in electronic signal in the detector. The research and development work is illustrated in a second part with a presentation of a study on the compromise between the MTF and the sensitivity of CsI:Tl layers deposited on optical fibers. The thickness of the scintillating layers produced in Trixell for this study varies between 250µm and 1 mm, and their properties are measured with three X-ray different spectra: RQA1, RQA5 and RQA9, which correspond to the range of applications of detectors in X-ray imaging applications.

# 11:15 **[172]** Miniature inorganic scintillation detectors for on-line treatment verification during brachytherapy *Presenter: KERTZSCHER, Gustavo*

Scintillation based point detectors have been used in radiation therapy since the early 1990s [1] primarily for pre-treatment quality assurance of patient plans and radiation sources. One common treatment modality is brachytherapy (BT) in which a sealed radioisotope is guided inside pre-inserted catheters in the tumor volume to deliver high radiation doses to the tumor with steep dose gradients to spare adjacent organs and normal tissue. The scintillation detectors could have an important role as on-line verification during BT treatments to detect errors that can lead to harmful consequences for the patient [2]. However, on-line treatment verification is presently not performed during BT, partially because commercial technology does not exhibit adequate signal intensities over the entire range of absorbed dose rates during BT treatments, which spans >2 orders of magnitude. The limited use of treatment verification is problematic because errors can occur unnoticed during single treatments or systematically over longer time periods [3].

We have developed miniature inorganic scintillation detectors (ISDs) for BT that are based on the scintillators Al<sub>2</sub>O<sub>3</sub>:Cr, Y<sub>2</sub>O<sub>3</sub>:Eu, YVO<sub>4</sub>:Eu, Y<sub>2</sub>O<sub>2</sub>S:Eu, Gd<sub>2</sub>O<sub>3</sub>:Eu, ZnSe:O or Csl(Tl). The ISDs consist of a 1 mm-size scintillator that is optically coupled to a 1 mm-diameter and 15 m-long fiber-optic cable made of poly(methyl methacrylate). The fiber-optic cable transmits the scintillation to the photodetector system which consists of a charge-coupled device camera or a spectrometer spectrograph. We have tested the ISDs under BT treatment irradiation conditions using a 10 Ci (370 GBq) <sup>192</sup>Ir source, and compared their performance with organic scintillators BCF-12 and BCF-60 which are the current standard for scintillation detectors in radiotherapy.

We will discuss the characteristics of our scintillation detectors and read out systems and their suitability for on-line verification of BT treatments, based on radiation exposures to the BT photon energy in water (average 300 keV) and the clinical range of dose rates (1-500 mGy/s). Our measurements show that the scintillation intensity of the inorganic materials are up to 3 orders of magnitude larger than those of the organic scintillators, and that the miniature ISDs exhibit the required dynamic range for precise dose rate measurements in the steep dose gradients near BT sources. The accuracy of the ISDs are furthermore not significantly affected by the stem signal, which is the contaminating Cerenkov and radioluminescence induced in the fiber-optic cable. Some of the inorganic scintillators exhibit unstable scintillation intensities for the larger dose rates and afterglow components with order of 1 s time constants, and we will describe how these luminescence properties can introduce inaccurate dose measurements during BT. Finally, we will discuss the promising potential for some of the inorganic scintillator materials for on-line treatment verification during BT, and describe the required detector characteristics.

[1] Beddar AS, Mackie TR, Attix FH. Water-equivalent plastic scintillation detectors for high-energy beam dosimetry. Phys Med Biol 1992;37:1883-1900.

[2] Tanderup K, Beddar S, Andersen CE, Kertzscher G, Cygler JE. In vivo dosimetry in brachytherapy. Med Phys 2013;40:1-15.

[3] IAEA Safety Report Series 17. Lessons learned from accidental exposures in radiotherapy. Vienna, Austria: IAEA; 2000.

### 11:30 [17] Real-time 3D scintillation dosimetry using organic liquid scintillators for proton therapy *Presenter: BEDDAR, Sam*

We have recently developed a novel application for organic liquid scintillator detectors in Radiation Oncology to measure or image the radiation absorbed dose from external radiation therapy beams in 3D. Initial feasibility studies<sup>1</sup> using one charge-coupled device (CCD) were investigated by Beddar et al in 2009. The present study will be focused only on scanning proton beams used for patient treatments. The basic concept is to use a 3D volume of a liquid scintillator material to measure or image<sup>2</sup> the dose distributions from proton beams in three dimensions. In this configuration, the scintillator material fulfills the dual role of being the detector and the phantom material (mimicking a patient) in which the measurements are being performed. In this case, dose perturbations caused by the introduction of a detector within a phantom will not be at issue. A larger liquid scintillator (LS) detector system was recently developed and consists of a transparent acrylic tank (20x20x20 cm3) filled with a water equivalent, commercially available liquid scintillator that when irradiated with protons generates scintillation light. To track rapid spatial and dose variations in spot scanning proton beams we use three high speed scientific-complementary metal-oxide semiconductor (sCMOS) imagers (2560x2160 pixels) that collect the scintillation light signals from three orthogonal projections in cine mode at up to 30 frames per second. The system that will be presented has been fully developed and characterized at the Proton Therapy Center at MD Anderson Cancer Center in Houston, Texas. The various optical artefacts that arise as the light propagates from the scintillator through the optical chain will be briefly presented<sup>3</sup>. The presentation will show that such systems can provide fast and accurate measurements of the range, lateral profile, and lateral position for scanned proton beams with higher spatial resolution (~ 2.5 mm) than other commercially available detectors. We will also show the ability of such detectors to rapidly measure or image proton beam characteristics and intensities at multiple energies which makes them particularly promising as a tool for scanned proton beam quality assurance as well as the verification of patient treatment delivery (i.e. prostate cancer).

<sup>1</sup>Beddar S, Archambault L, Sahoo N, Poenisch F, Chen GT, Gillin MT, Mohan R. Exploration of the potential of liquid scintillators for real-time 3D dosimetry of intensity modulated proton beams. Med Phys 36(5):1736-1743, 5/2009.

<sup>2</sup>Hui C, Robertson D, Beddar S. 3D reconstruction of scintillation light emission from proton pencil beams using limited viewing angles - a simulation study. Phys Med Biol 59(16):4477-4492, 8/2014.

<sup>3</sup>Robertson D, Hui C, Archambault L, Mohan R, Beddar S. Optical artifact characterization and correction in volumetric scintillation dosimetry. Phys Med Biol 59(1):23-42, 1/2014.

### 11:45 [167] Scintillating fibers devices for Particle Therapy applications

Presenter: MATTEI, Ilaria

Particle Therapy (PT) is an increasingly widespread kind of radiation therapy in which solid tumors are treated with charged light ions beams to exploit the highly localized dose delivery that can be achieved, allowing to spare the healthy tissues surrounding the organs at risk. During the irradiation a large amount of secondary particles is produced as a consequence of the interactions between the beam particles and the patient tissues. Secondary charged fragments and photons, namely annihilation and excited nuclei de-excitation photons, show an emission spectrum correlated to the released dose distribution that can be used to monitor the beam range during the treatment. The development of a range monitoring technique capable of reaching a sub-millimetrical precision is considered one of the key steps in optimizing the PT efficacy and assuring the treatment quality. Besides charged fragments and photons there is also the secondary neutron component that contributes to an undesired and not negligible dose deposition far away from the tumor region, enhancing the risk of secondary malignant neoplasies development after the treatment. An accurate neutron production characterisation (flux, energy and emission profile) is hence needed to significantly improve the evaluation of possible long-term complications.

In this contribution two tracker detectors, that employ layers of scintillating fibres as active mean, are presented. The first one, named Dose Profiler (DP), is designed for secondary charged fragments measurements and is planned to be used as a beam range monitor in PT treatments with Carbon ions beam. The second one is dedicated to the fast and ultrafast neutron measurements for the characterisation of the secondary neutron component, in the frame of the MONDO (MOnitor for Neutron Dose in hadrOntherapy) project. The DP is currently under development within of the INSIDE collaboration (Innovative Solutions for In-beam Dosimetry in hadrontherapy)[1]. It is composed by six layers (20 \$\times\$ 20 cm\$^2\$) of BCF-12 square scintillating fibres (500 \$\mu\$m) coupled to Silicon Photo-Multipliers, followed by two plastic scintillator layers of ~6 mm thickness. The detector characterisation with cosmic rays is currently undergoing and a data taking campaign with protons will take place in May 2017. The DP design and the performances measured with using MIPs and protons beam will be reviewed. The MONDO detector[2], that exploits the tracking of the recoil protons produced in double-elastic scattering neutron interaction to measure the neutron kinetic energy and incoming direction, is a matrix of scintillating fibres, arranged in x-y oriented layers (total active volume 10\$\times\$10\$\times\$20 cm\$^3\$ filled with squared 250\$\mu\$m fibres BCF-12), that are read-out by a dedicated SPAD sensor produced by FBK (Fondazione Bruno Kessler). The detector is currently under development and its full completion and assembly is expected before the end of the year. The expected performances computed using a MonteCarlo simulation and the preliminary measurements obtained using MIPs and a tracker prototype will be presented.

[1] G. Traini et al., Physica Medica 34 (2017), pp. 18-27, doi: 10.1016/j.ejmp.2017.01.004.

[2] M. Marafini at al., Phys. Med. Biol. 62 (2017), pp. 3299–3312 , https://doi.org/10.1088/1361-6560/aa623a

## 12:00 [215] Development of a high resolution module for PET scanners with DOI capabilities *Presenter: POLESEL, Andrea*

In order to ensure early stage detection of cancer, PET scanners have to achieve high performances in terms of spatial resolution and sensitivity.

An innovative PET detector module able to provide high spatial resolution was developed [1]. The high spatial resolution can be achieved thanks to a new method to extract Depth Of Interaction (DOI) information while keeping the complexity and the cost of the module low. This result is possible by means of light sharing and recirculation: the ratio between the light read by the MPPC directly coupled to the crystal hit, and the total light that hit the photodetectors array, collected using a reflector on the back side of the matrix, is correlated to the DOI information.

This method was calibrated and validated using an external tagging crystal. The procedure however is slow and infeasible on a full size detector. For this reason, an innovative calibration method was also developed [2]. This method consists in deriving a relation between the distribution of the coordinate reconstructed and the DOI, and showed a very good agreement with experimental data obtained using the tagging bench.

The possibility to improve the timing resolution taking advantage of the DOI information was investigated as well. The idea is to correct the timestamps measured by each detector in the MPPC array using the DOI information, and then combine for each scintillation event the timing information provided by the detector directly coupled to the scintillation crystal and by the surrounding detectors in order to obtain the best possible timing estimator time of interaction between the incident gamma and the crystal. This approach was preliminarily studied by means of computer simulations using the Geant4 Monte Carlo toolkit. The results are encouraging and suggest a substantial improvement in timing resolution can be achieved with this method.

This work has been performed in the frame of the Crystal Clear Collaboration and funded by the Knowledge Transfer department of CERN.

 M Pizzichemi, G Stringhini, T Niknejad, Z Liu, P Lecoq, S Tavernier, J Varela, M Paganoni, E Auffray, A new method for depth of interaction determination in PET detectors. 2016 Physics in Medicine and Biology, 61 (12) 4679.
G Stringhini, M Pizzichemi, A Ghezzi, A Stojkovic, M Paganoni, E Auffray, Development and evaluation of a practical method to measure the Depth of Interaction function for a single side readout PET detector. 2016 JINST 11 P11014.

### Conference Summary (12:15-12:35)

- Presenters: DUJARDIN, Christophe; DUJARDIN, Christophe

### presentation site SCINT2019 (12:35-12:50)

### Conference closure (12:50-13:00)

- Presenter: AUFFRAY, Etiennette