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; Etienne Auffray

- **09:30 [243]** 25 years of Scint conferences

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

**Opening Session: Invited talks (10:30-12:00)**
-Conveners: Etienne Auffray; Christian Pedrini

- **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.
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 known 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

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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 the 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 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 emphasis 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).*
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 leucosapphire 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 ± 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 this 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).
Design and status of the Mu2e crystal calorimeter
Presenter: MISCETTI, 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 \( \lambda_{\text{conv}} \) 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 ~ 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 \( \text{O}(10\%) \) @ 100 MeV and
- (c) a position resolution of 1 cm.

The calorimeter consists of two disks, each one made of 674 pure CsI crystals read out by two large area array 2$\times$3 of UV-extended SiPM $6\times6$ mm$^2$. We report here all progresses done for the construction and test of the Module-0 prototype that is an array of 51 pre-production crystals from St.Gobain, Siccas and Amcry's firms. Each crystal has been readout by two pre-production Mu2e SiPMs selected among the ones produced by Hamamatsu, Sensl or Advansid. Each photosensor has been amplified and regulated in bias voltage by means of a FEE custom chip. Final digitization stage is also custom and relies on a 5 ns sampling. The module-0 will be exposed to an electron beam in the energy range around 100 MeV at the BTF (Beam Test Facility) in Frascati. Preliminary results of timing and energy resolution at normal incidence will be shown as well as dependence of response and resolution as a function of the impinging angle.
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%/\sqrt{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.

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.
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 |η|=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 PbWO₄ 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

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 (0ν2β) of 100 Mo isotopes using CaMoO₄, ZnMoO₄ and LiMoO₄ crystals, respectively. Main advantages of the 100 Mo are its high transition energy (Qββ = 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.

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 (0ν2β) 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 (μ) 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 λ=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.

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

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Dark Matter is one of the biggest issue in modern physics, and one 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.


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.
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(Tl)) 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 scintillation 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(Tl) 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.


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 NaI(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.

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.

[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

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 halides and that of the power of engineering oxides compounds. 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.


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. Csl/NaCl [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).

09:15 [161] Growth and characterization of SrI2:Eu crystals grown by the Czochralski method
Presenter: GALENIN, Evgeny

SrI2: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 SrI2: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(Tl) alkali halide crystals with the diameter of up to 500 mm by the Modified Czochralski-Kypropolous 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"*


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 stereophotolithography 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 slp with volumetric bulk content ~25%. Then it was photocured layer by layer in stereophotolithography 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 tsc under 60 ns and light yield measured under 5,5 MeV α-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.
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.

References


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

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

Ionization Quenching Correction of Volumetric Organic Scintillators for use in Proton Therapy

Development of High Spatial Resolution Dosimeter for Medical Uses by Colorimetric Discrimination Method

Scintillation materials for PET/MRI coupled to digital SiPM

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

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

Development of a detector module suitable for Whole body PET with improved timing performance

Development of a detector module suitable for Whole body PET with improved timing performance

Temporal Imaging for PET: Coincidence Timing results on 20 mm LYSO crystals

Breast-dedicated PET system with a personalized gantry

Emerging Concepts in Organic Radiation Detection Materials

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

Presenter: KIM, HongJoo

Thallium-(Tl)-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 Tl2LiGdCl6/Br6, Tl2LiYCl6, Tl2LiLuCl6, Tl2LaCl5/Br5 [1-3], Tl2LiScCl6, and Tl2GdCl5. 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 Tl ion in the host lattice. Since Tl 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 (Ze$> 60$. 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 Tl-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 Tl-based scintillators and also an overview of the already developed scintillators.


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

Presenter: LAM, Stephanie

Cesium hafnium chloride (CHC, Cs,$\text{HfCl}_6$) is a recently-discovered scintillator with radiation detection properties superior to the incumbent detectors NaI(Tl) and CsI(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.*
[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 µm, 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.

[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+.


lunch (12:45-14:30)

Scintillation Mechanisms: session 1 (14:30-16:00)
-Conveners: Richard Williams

- time [id] title
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:Tl). 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 thermalized 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

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.
15:15 [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$_3$, 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$_3$: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.


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 (STEs) 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 hybrid density functional theory can accurately predict the different types of 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.
[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 Ti₂LaCl₅: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.


15:45 [102] Picosecond absorption spectroscopy of self-trapped holes, self-trapped excitons, and transient Ce states in LaBr₃ and LaBr₃:Ce
Presenter: LI, Peiyun

We report excitation-induced picosecond absorption over an extended spectral range from 320 nm to 2700 nm in LaBr₃(3)%, LaBr₃(3)%:Ce(4%) and LaBr₃(3)%:Ce(20%). Preliminary identification of a Vₖ 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 Vₖ and STE structure in LaBr₃(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 Ce³⁺ excited activator and CT transitions of Ce⁴⁺ activator-trapped holes. Together with the identified STH and STE transitions noted above, these should constitute the main species in LaBr₃(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 LaBr₃(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 LaBr₃(3)%:Ce. The present work follows our similar picosecond-measurement program in CsI, CsI:Tl [1] which supplied a number of key rate coefficients for successful modeling of pulse shape, proportionality of decay components, and light yield in CsI:Tl [2].

References

Coffee Break (16:00-16:30)

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

-Conveners: Pieter Dorenbos

time  [id] title
[27] High Performance CLYC-PVT Composite Scintillators for Neutron/Gamma Detection

Presenter: LAM, Stephanie

While SrI\(_2\)(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 be shaped to improve light collection. Thus, composite scintillators hold the promise of providing high performance low-cost gamma/neutron detectors in sizes that would otherwise be prohibitively expensive or impossible to produce. This presentation will discuss the design, fabrication, and performance of our CLYC-PVT and Sr\(_2\)(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.*

[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 mostly 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.

[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 to have a good energy resolution (FWHM) of ~5% at 662 keV, and its light output remained constant up to 150 °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, Edingburgh Instrument: EI) with Xe lamp (EI, Xe-900), respectively. The pulse height spectra at the high temperature up to 200 °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 facility, 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 °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.


[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.

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 has 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 discard 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 INTELU (GA no. 644260).


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 concept 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 cm$^3$) 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.

IAC meeting (19:00-22:00)
Wednesday 20 September 2017

**Nanomaterials (08:30-10:00)**
- Conveners: Christophe Dujardin

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<td>08:30</td>
<td>[189] Colloidal quantum dots design for scintillation applications</td>
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<td>Presenter: MAHLER, Benoit</td>
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<td>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.</td>
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| 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.

References

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, γ-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.


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. 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 response time can be achieved by introducing of metal nanoparticles. To guide our experimental efforts, we have fabricated Ag:ZnO nanocomposite films and performed 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 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.
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 resistance. 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 (10^5 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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| [21] Facile Synthesis of High Purity Anhydrous Complex Rare Earth Halides by the Modified Mixed-Salts-Dehydration Method | YU, Jinqiu |
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Metamaterials (11:00-12:30)

- Conveners: Martin Nikl

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 suppressed Auger recombination have proven to be a promising source of ultrafast light under ionizing radiation. Previous studies [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/
[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


[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 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 $\gamma$(137)Cs energy. After careful optimization of the CFD parameters, timing resolutions as good as FWHM$\sim 200$ ps and time-walks below $30$ 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.
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 $\sim$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.
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.

References:


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).

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

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

Conference Excursion: Train to Montenvers and the Mer de Glace (valley glacier) with a professional guide (13:00-18:00)
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 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 an 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.
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.

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:Tl 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:Tl, the mapping of Tl* luminescence with 2 \( \mu \)m resolution in our experiment has indicated a granularity of response with feature size in the 20 \( \mu \)m range in standard CsI:Tl scintillators. This could indicate clustering or concentration of Tl 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:Tl scintillators.

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


Lead molybdate (PbMoO₄) crystal is isotruktral 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 β 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.
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:Tl 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:Tl 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:Tl 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

1. A. Syntfeld-Kazuch et al., Conference Record of the IEEE NSS-MIC, Seattle, 2014.
Excitation density distribution effects on fast ZnO excitonic emission

Presenter: MARTIN, Patrick

ZnO crystals and nanoparticles have promising scintillation properties as subnanosecond scintillators. There is a 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 depend 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 cases 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 results 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.

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

-Conveners: Marco Pizzichemi; Remi Chipaux; Kristof Pauwels

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

- Conveners: William Moses

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<td>11:00</td>
<td>[208] Precise rise time measurements of inorganic scintillators using X-ray and 511 keV excitation</td>
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  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, \( \text{CTR} \propto \sqrt{\tau_r \tau_d / L Y} \). 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 \(\sim300ps\). 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 Ce\(^{3+}\) 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 Mg\(^{2+}\)-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 \(\gamma\)-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 (Lu0.8Gd0.2)(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.


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 Ce5+3+ level from the conduction band decreasing the probability of 5d-state ionization. Another strategy was Mg-codoping, which stabilized the tetravalent Ce4+…, 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 Ce4+… stabilized by Mg2+2+…, 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.

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 Ce4+… level. 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+3 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 YAlO$_3$: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$^{3+}$ emission.[3]

In this contribution we will discuss the applicability of Ca co-doping in order to improve the scintillation characteristics of YAlO$_3$: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$^{4+}$ 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)

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:Tl, CsI:Tl, 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 quality 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.


Lunch (12:30-14:30)
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

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<td>14:30</td>
<td>[176] Scintillator materials and phenomena studied for fast timing</td>
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Presenter: NIKL, Martin

Fast and efficient scintillators are required by a number of applications, namely in high energy physics and medical imaging (Positron Emission Tomography – PET) where so call time-of-flight (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 loseness. 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 embeded in an optically transparent host, e.g. ZnO:Ga nanocrystals in a polystyrene matrix, became intensively studied [2]. Moreover, excitonic emission in GaN-GaInN 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).

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 different. 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.
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 scintillation bolometers that operate in ultra-low temperature conditions and used to search for neutrinoless double beta decay ($0\nu\beta\beta$) of $^{100}\text{Mo}$. Among tens of possible candidates, the isotope $^{100}\text{Mo}$ 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.

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 $100\text{ps}$ 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)
### Crystal growth: session 2 (08:30-10:00)

- **Conveners:** EDITH BOURRET

#### 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 (0υDBD), 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 0υDBD 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 0υDBD and dark matter search applications. Details will be given on the production of enriched zinc selenide (Zn$^{82}$Se) and tellurium dioxide (130TeO$_2$) crystals for 0υDBD experiments together with the performance of these crystals used as scintillating and Cerenkov bolometers respectively. The synthesis of ultrapure NaI powder and the growth and processing of NaI:Tl 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**

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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, Gd₃Ga₅O₁₄: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, Gd₂.96Ce₀.03Al₁.14Ga₁.86O₁₂, Lu₂SiO₅:Tb (12% at.), Gd₃Ga₅O₁₄: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(Al,Ga)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 Gd₃Ga₅O₁₄ 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 Pb₀.₀₁Ce₀.₀₃Gd₂.₉₆Al₁.₁₄Ga₁.₈₆O₁₂ 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.

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 La$_{2}$Hf$_{2}$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 properties of these materials including temperature dependence and time profile in this presentation.

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 (YAlO3, CeAlO3) 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.

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[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 logging, 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 LuAlO3 (LuAP), YAlO3 (YAP) and garnets as Lu3Al5O12 (Ce:LuAG), Y3Al5O12 (Ce:YAG), Ce:Gd3(Al,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, Crucible and die designs, growth conditions, Mo contamination, chemical composition analysis, optical and scintillation properties of the grown crystals will be discussed.

Growth of Ce-doped garnets with additional monovalent impurities and related effects

Presenter: PETROSYAN, Ashot

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-doping 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 (a₀), 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 a₀ 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 a₀ 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 impurities is however evidenced in acceleration of the fast decay component and decrease of radioluminescence intensity. Radiation hardness of crystals is high showing no induced absorption bands in the range of emission after gamma-ray irradiation with 1 kGy. The results will be compared with available data reported for crystals prepared by other techniques. The advantages and limitations of the co-doping 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).

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 Institut (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

[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 platforms. 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, geomatics 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 (LuI$_3$: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. LuI$_3$: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 LuI$_3$: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 LuI$_3$: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.

[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.
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, Gd<sub>2</sub>O<sub>2</sub>S:Eu, ZnSe:O or CsI(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.


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 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 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. 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).


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 the INSIDE collaboration (Innovative Solutions for In-beam Dosimetry in hadrontherapy)[1]. It is composed by six layers (20 cm$^2$) of BCF-12 square scintillating fibres (500 µm) 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 10×10×20 cm$^3$ filled with squared 250 µm 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.


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.