Overview of Research on scintillator materials in Latvia: past, present, and future

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1st CERN Baltic Conference (CBC 2021) 28 June – 30 June, 2021

Introduction

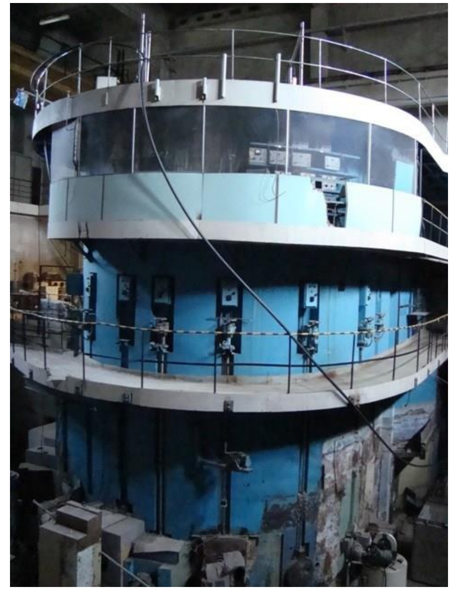
The research in solid state physics at the University of Latvia restarted after World War II.

In the beginning of 60^{th} - in the Institute of Physics, Latvian academy

The Institute of Solid State Physics (ISSP) of the University of Latvia was established on the basis of Laboratory of Semiconductor Research and Laboratory of Ferro- and Piezoelectric Research in 1978.

Since 1986 the ISSP has the status of an independent organization of the University and now is the main material science institute in Latvia.

Luminescence activities in Salaspils



1957 - 1990

Luminescence of alkali-halides, doped with ns^2 ions (NaI, CsI etc)

Luminescence of neutron irradiated halides and oxides (LiF, MgO, $MgAl_2O_4$ etc)

Time resolved optical absorption and luminescence studies under nano- and picosecond electron beam excitation (200-300 keV) (CsI, NaI, Al_2O_3, SiO_2

TLD dosimetry

Introduction

Four laboratories from the Institute of Physics of the Latvian Academy of Sciences joined our Institute in 1995.

Twenty scientists of the former Nuclear Research Centre joined the ISSP in 1999 and established Laboratory of Radiation Physics.

Type of Reactor: IRT pool-type reactor

The reactor was started up on September 26, 1961. Since 1979 thermal power was 5 MW.

10 horizontal and 17 vertical channels were employed in experimental research with using of neutron fluxes. Until, 1998.

Main fields of investigations:

- Nuclear spectroscopy;
- Solid state physics;
- Radiation materials science;
- Neutron-activation analysis.

History

Four laboratories from the Institute of Physics of the Latvian Academy of Sciences joined our Institute in 1995.

Twenty scientists of the former Nuclear Research Centre joined the ISSP in 1999 and established Laboratory of Radiation Physics.

In 2004 scientists from the Institute of Physical Energetics joined ISSP and established Laboratory of Organic Materials

Some Important milestones:

alkali halides

LUMINESCENCE OF BOUND EXCITONS IN ALKALI-HALIDE CRYSTALS WITH FIRST-GROUP CATION IMPURITIES.
YA **Valbis** - Opt. Spectros.(USSR)(Engl. Transl.), 21: 106-9, 1966

Thermally stimulated and tunneling luminescence and frenkel defect recombination in KCl and KBr at 4.2 to 77 K
DE Aboltin, VJ **Grabovskis**, AR Kangro, C Lushchik, AA O'Konnel-Bronin, IK **Vitol**, V.Zirap - physics status solidi (a), 1978

Tunneling recombination luminescence in KBr and KCl
VJ **Grabovskis**, IK Vitols - Journal of Luminescence, 1979 - Elsevier

Some Important milestones:
simple oxides

Luminescence of free and relaxed excitons in MgO
ZA Rachko, JA **Valbis** - physica status solidi (b), 1979

Recombination luminescence in single crystal Al2O3
PA **Kulis**, **MJ Springis, IA Tale**, JA **Valbis** - physica status solidi (a), 1979

Vacuum-ultraviolet luminescence of Be-Doped MgO epitaxial layers
IE Lacis, JA **Valbis** - physica status solidi (b), 1979

On the mechanism of the recombination luminescence of α-Al2O3 crystals with nonstoichiometric excess of aluminium
PA **Kulis**, MJ Springis, IA Tale, JA **Valbis** - physica status solidi (a) 1980

Some Important milestones:

other materials

Recombination luminescence mechanisms in $Ba_3(PO_4)_2$
I Tāle, P Kūlis, V Kronghauz - Journal of Luminescence, 1979

Tunneling recombination luminescence in $Na_2O \cdot 3SiO_2$ glass
AR Kangro, MN Tolstoy, IK Vitols - Journal of Luminescence, 1979

Optical Properties of the F-Centre in Beryllium Oxide
S.Gorbunov , AV Kruzhalov, MJ **Springis** - physica status solidi (b), 1987 -

Polarization of luminescence of colour centres in **YAG** crystals
M **Springis**, A Pujats, J Valbis - Journal of Physics: Condensed ..., 1991

Some Important milestones:

theory

Role of tunnelling recombination in radiation-induced F-centre creation in alkali halide crystals at liquid helium temperatures
I Tale, D **Millers**, E **Kotomin** - Journal of Physics C: Solid State, 1975
Temperature dependence of F-centre accumulation efficiency in doped alkali halides
E **Kotomin**, I Tale, I **Fabrikant** - Journal of Physics C: Solid State ..., 1977

Temperature and impurity concentration dependences of the efficiency of Frenkel defect accumulation in alkali halide crystals
E **Kotomin** - Solid state communications, 1984 – Elsevier

Phenomenological kinetics of Frenkel defect recombination and accumulation in ionic solids
E **Kotomin**, V Kuzovkov - Reports on Progress in Physics, 1992 -

Some Important milestones: CROSSLUMINESCENCE

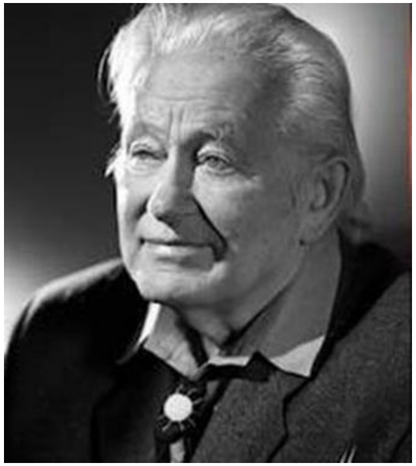
Luminescence due to radiative transitions between valence band and upper core band in ionic crystals (**crossluminescence**)
J.Jansons, VJ Krumins, ZA Rachko, JA Valbis - physica status solidi b, 1987

Photon yields and decay times of **cross luminescence** in ionic crystals
P.Dorenbos, R Visser, CWE Van Eijk, J **Valbis,** N Khaidukov - IEEE transactions on Nucl Sci , 1992 –

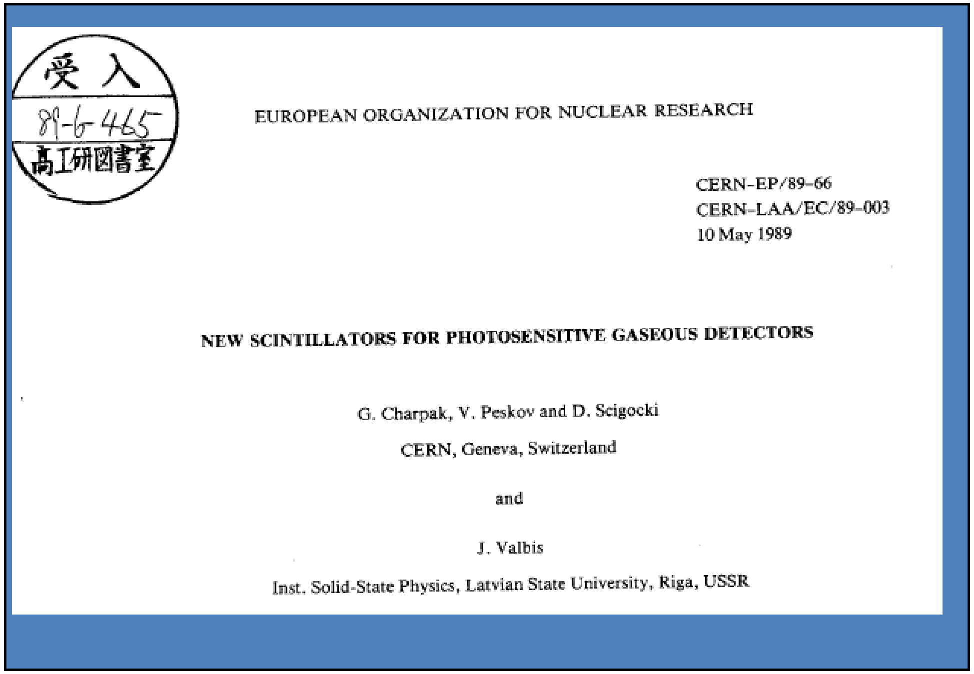
Report on the scintillation properties of the ternary inorganic crystals $KMgF_3$, KYF_4/: Rb, K_2YF_5, $KLuF_4$, $RbMgF_3$, $KZnF_3$, $BaTm_2F_8$, $LiYF_4$: Nd, and BaF_2: Rb.

About history of the collaboration between CERN and Latvia

Georges Charpak (1924-2010) was awarded the Nobel Prize in Physics in 1992 "for his invention and development of particle detectors, in particular the multiwire proportional chamber", with affiliations to both École supérieure de physique et de chimie industrielles (ESPCI) and CERN.



3 joint publication in CERN database, two of them are at WEB of Sci.



EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-EP/89-66
CERN-LAA/EC/89-003
10 May 1989

NEW SCINTILLATORS FOR PHOTOSENSITIVE GASEOUS DETECTORS

G. Charpak, V. Peskov and D. Scigocki
CERN, Geneva, Switzerland

and

J. Valbis
Inst. Solid-State Physics, Latvian State University, Riga, USSR

Janis Valbis (1936-2009)

About history of the collaboration between CERN and Latvia

Crystal Clear Collaboration

EC project (1995-1997)

phys. stat. sol. (b) 203, 585 (1997)

Subject classification: 78.55.Hx; S11.1

The Temperature Dependence of Scintillation Parameters in $PbWO_4$ Crystals

D. Millers (a), L. Grigorjeva (a), S. Chernov (a), A. Popov (a), P. Lecoq (b), and E. Auffray (b)

(a) Institute of Solid State Physics, University of Latvia, 8 Kengaraga St., LV-1063 Riga, Latvia

(b) CERN, Division PPE, Geneve, Switzerland

(Received January 10, 1997; in revised form April 14, 1997)

The luminescence spectra, decay kinetics and yield of luminescence in undoped $PbWO_4$ crystals were studied after pulsed electron beam irradiation. The rise time of luminescence pulses shows that two mechanisms – excitonic and recombination – were involved in luminescence center excited state formation. It is proposed that excited states of WO_3 and WO_4^{2-} luminescence centers were formed from some metastable state, possibly from Pb related excitation.

N·H ELSEVIER

Journal of Luminescence 72–74 (1997) 693–695

JOURNAL OF LUMINESCENCE

Time-resolved luminescence and induced absorption in $PbWO_4$

D. Millers[a], S. Chernov[a], L. Grigorjeva[a,*], A. Popov[a], E. Auffray[b], I. Dafinei[b], P. Lecoq[b], M. Schneegans[c]

[a] *Institute of Solid State Physics, University of Latvia, 8 Kengaraga St., Riga, Latvia*
[b] *CERN, Geneva, Switzerland*
[c] *LAAP, Annecy, France*

Abstract

Luminescence and short-lived induced absorption are studied for two undoped $PbWO_4$ crystals. Luminescence decay at LNT is delayed relative to irradiation pulse. The delay observed is either due to reabsorption of luminescence or due to creation of luminescence center excited states via energy or/and charge transfer after irradiation pulse. Short-lived absorption is observed within 1.1–3.6 eV spectral region at LNT as well as at RT. It is proposed that the short-lived absorption band peaking at ~3.5 eV is intrinsic.

Keywords: $PbWO_4$; Time-resolved luminescence; Short-lived absorption

Since 2016 we are again member of CCC,
Anatoli Popov –CCC CERN board member

This was some history

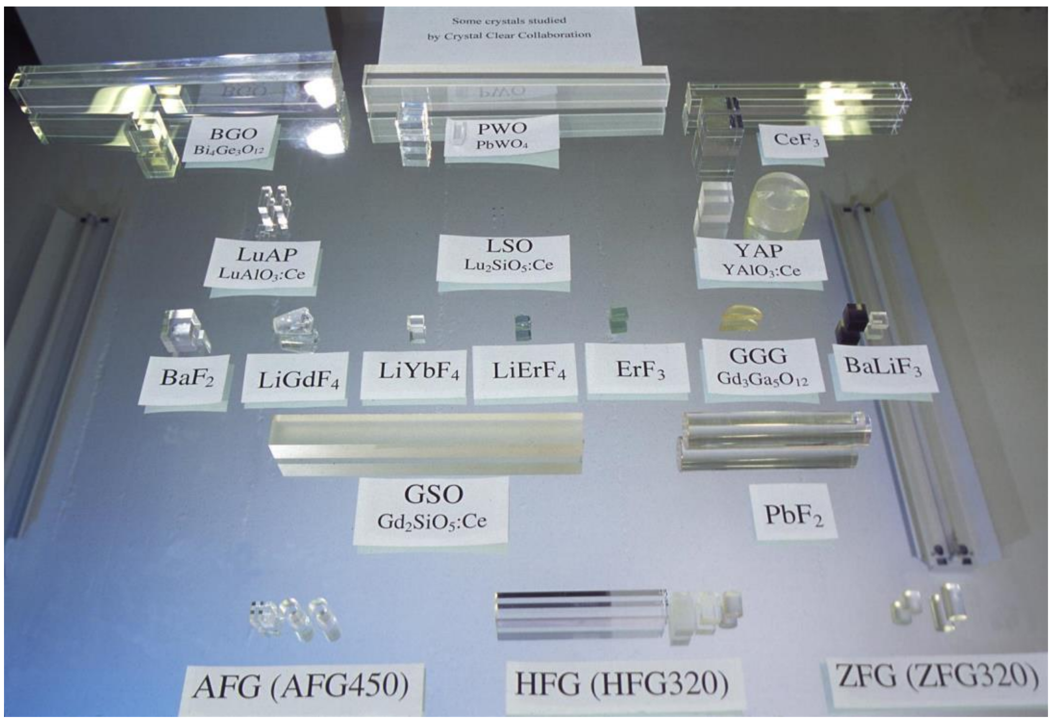
CRYSTAL CLEAR COLLABORATION

Since 2016 we are member CCC. Crystal Clear is involved in:

-Investigation of scintillator materials for high energy and nuclear physics, astrophysics, dark matter search, beam diagnostics, medical imaging and other industrial applications.

-- Development of new crystal production methods (micropulling down, ceramics etc.).

-- Development of ionising radiation detectors in particular high energy physics and medical and is working in close collaboration with industrial partners and experts in crystal growing, solid-state physics, defects in solids, optics, etc.



Materials of our interest:
$PbWO_4$
LSO, LYSO
YAP
$BaLiF_3$

One of the main achievements in 2020

135 Scopus papers published by the staff of Institute
(20% - luminescence)

Luminescence research – in house equipment, other universities and synchrotron light sources (DESY, Lund)

We have also an established access to ILL, ESRF, MAXIV, Soleil, GSI, GANIL large scale facilities.

Of course CERN !!!

Why Large Scale Facilities?

Synchrotron &Neutron radiation beamlines are high-performance instruments that allow to obtain multi-scale and multi-task researches on materials of industrial as well as fundamental interest.

- **ILL Grenoble, France**
- **LLB Saclay, France**
- **ESRF Grenoble, France**
- **FRM II Munich, Germany**
- **PSI Villigen, Switzerland**
- **DESY Hamburg, Germany**
- **XFEL Hamburg, Germany**
- **ELETTRA Trieste, Italy**
- **DAFNE Frascati, Italy**

etc ....

Great opportunities for small countries

not only because LV is small country,

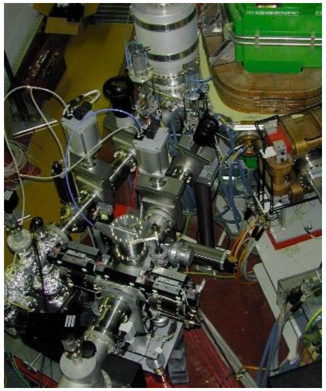
but also because of professional project evaluation etc)

ZnO	ESRF	2004	XEOL
ReO3	ESRF	2005	EXAFS
Ge (isotopes)	ESRF	2006	EXAFS
ZnO nano	ESRF	2006	XANES, XEOL
ZnWO4 nano	ESRF	2007	XANES, XEOL
ZnNiWO4	HASYLAB/DESY	2009	EXAFS
NiO	HASYLAB/DESY	2010	EXAFS
ReRAM	HASYLAB/DESY	2010	EXAFS
MnWO4	HASYLAB/DESY	2011	EXAFS
ReRAM	HASYLAB/DESY	2011	EXAFS
CuWO4	HASYLAB/DESY	2011	EXAFS
ReRAM	ESRF	2011	EXAFS/XANES
ScF3	ELETTRA	2011	EXAFS
SrTiO3	HASYLAB/DESY	2012	EXAFS
Cu3N	HASYLAB/DESY	2012	EXAFS
ReRAM	HASYLAB/DESY	2012	EXAFS
SrTiO3 (isotopes)	ESRF	2012	EXAFS
NiWO4	SOLEIL	2012	FTIR
ZnNiWO4	SOLEIL	2012	FTIR
SnWO4, CoCuWO4	SOLEIL	2013	FTIR
ODS steels	ELETTRA	2013	EXAFS/XANES
ODS steels	ELETTRA	2014	EXAFS/XANES
CuMoO4	ELETTRA	2015	EXAFS/XANES
SnWO4 HP	SOLEIL	2014	FTIR
SnWO4 HP	SOLEIL	2014	EXAFS/XANES
Cu3N HP	SOLEIL	2014	XANES
CuO HP	SOLEIL	2015	XANES
ODS steels	SOLEIL	2015	EXAFS/XANES
ODS steels	ESRF	2015	EXAFS/XANES
EuTiO3, CH3NH3PbX3	CLAES-ALBA	2015	EXAFS/XANES
CuO nano	HASYLAB/DESY	2016	EXAFS/XANES

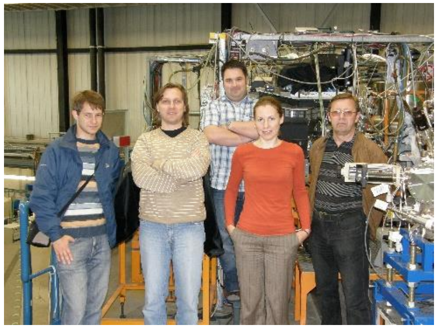
Number of projects before 2010: 6
Number of projects starting from 2010-2017: > 40

Since 2017 Number of project : 5-10 per year

AlN	LNF Frascati	2005	FTIR, XANES
AlN nano	LNF Frascati	2006	FTIR
AlN nano	ILL Grenoble	2006	INS, PD
CdI2	LNF Frascati	2007	FTIR
CsPbCl nano	LNF Frascati	2008	FTIR
SiC nano	LNF Frascati	2009	FTIR
Ag2CdI4	LNF Frascati	2009	FTIR
CdCoS	LNF Frascati	2010	FTIR, XANES
LaPO4 nano	HASYLAB/DESY	2010	VUV
NiWO4	HASYLAB/DESY	2010	VUV
LaCl3:Eu3+	HASYLAB/DESY	2010	VUV
SrI2:Eu	HASYLAB/DESY	2010	VUV
YVO4	HASYLAB/DESY	2011	VUV
PLZT	HASYLAB/DESY	2011	VUV
SrTiO3	HASYLAB/DESY	2011	VUV
ScF3	HASYLAB/DESY	2011	VUV
SrTiO3	HASYLAB/DESY	2012	VUV
CsBr	HASYLAB/DESY	2012	VUV
BaZrO3-Y	HASYLAB/DESY	2012	VUV
HAP	HASYLAB/DESY	2012	VUV
BaZrO3-Y	LNF Frascati	2015	FTIR
Ge_GaS_glasses	LNF Frascati	2015	FTIR

SINBAD facility (the synchrotron radiation IR beamline at DAFNE, **Frascati, Italy**)




VUV Superlumi beamline (DESY), Hamburg

New Project: Electronic structure of Mn, Ce or Ti-doped $YAlO_3$: Prediction from the first principles

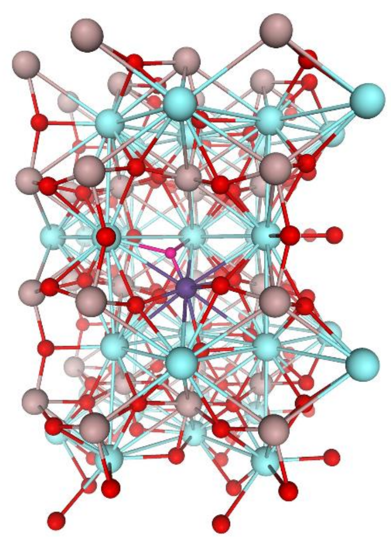
S. Piskunov, and A. I. Popov in collaboration with M.Brik (UT)

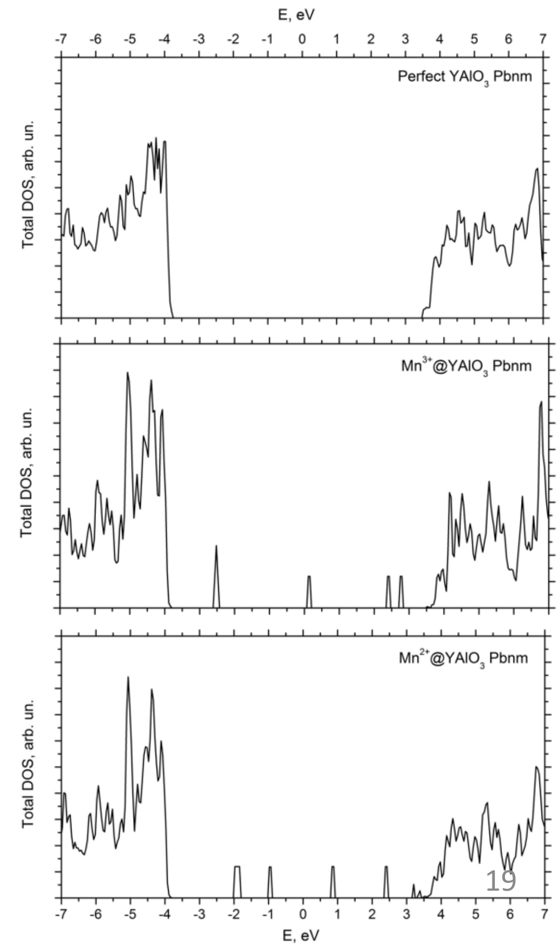
Motivation

The $YAlO_3$ crystals belong to a group of materials serving as the basic materials of the laser technique, scintillators, optical recording media etc Among them Mn-doped $YAlO_3$ is a TLD promising material for thermoluminescent dosimetry of ionizing radiation.

In this study - calculations obtained within the formalism of hybrid Density Functional Theory and Hartree-Fock method (using the HSE0 Hamiltonian as implemented in total energy LCAO-CO CRYSTAL14 computer code [http://www.crystal.unito.it/])

Schematic representation of 2x2x2 supercell of orthorhombic Pbnm $YAlO_3$ having an Mn^{2+} dopant designated as a violet ball and compensating F-center (small pink ball).





Total density of states for (from top to bottom) perfect orthorhombic $YAlO_3$ *Pbnm* bulk crystal,
trivalent Mn-doped,
and divalent Mn-doped stabilized
by the presence of the *F*-center.

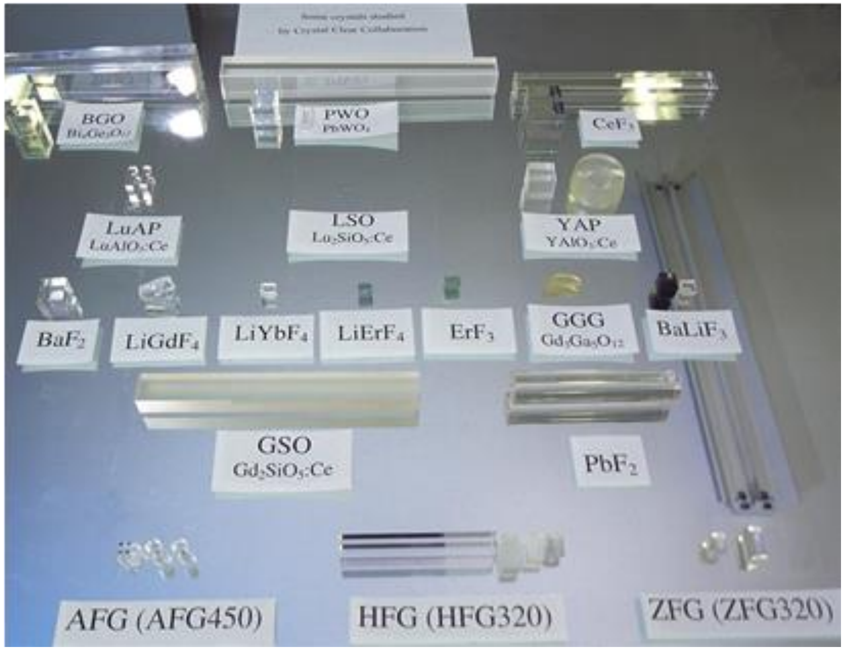
5 projects submitted for synchrotron at Lund and Hamburg were approved

EXAFS studies of cerium centers in GGAG crystals (BALDER)

Time-resolved luminescence and VUV-XUV excitation spectroscopy of ion irradiated (Lu,Y)2SiO5:Ce single crystals (FinEstBeAMS and DESY)

VUV excitation spectroscopy of ion and neutron-irradiated oxides (FinEstBeAMS and DESY)

3 projects for irradiation with swift-heavy ions at GSI, Darmstadt, Dubna and Kazakhstan were approved. Irradiated samples from Dubna and GSI were received



Samples of "Crystal Clear Collaboration" interest

Proposal for swift heavy irradiation of YAG, PbWO4 and PbF2
at GSI (Darmstadt) – contact person Prof. Dr. Christina Trautmann

Fluences for irradiation:
1e11, 5e11 and 1e12 of 8.6 MeV/u Au.

Similar proposal – Dubna (Russia)
YAG, PbWO4, PbF2 and GAGG
1e10 – 3.5e13
Xe ions (156 MeV) –Dr. Vladimir Skuratov
Bi ions (710 MeV)

MAIN CURRENT PROJECTS:

RADIATION DAMAGE STUDIES IN SCINTILLATOR MATERIALS FOR HIGH-ENERGY PHYSICS AND MEDICAL APPLICATIONS (2018 - 2021)

Principal Investigator: Dr. Phys. **Anatoli Popov**

Total funding: **300 000 EUR**

Project implementation period: **2018-2021**

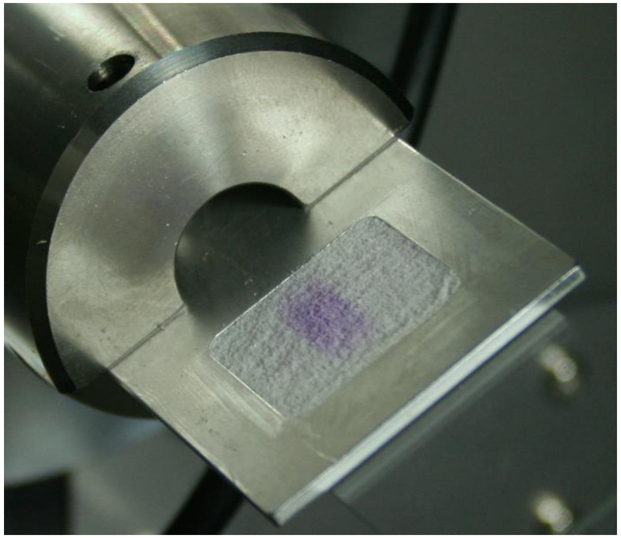
Latvian Council of Science Grant No. LZP-2018/1-0214

The experimental-theoretical project is devoted to systematic studies of radiation damage kinetics and basic processes in scintillating materials currently used for particle physics, neutron research and medical imaging. Radiation damage, together with the detection efficiency and time resolution parameters, as a very important factor determining long-term and stable operation is an important issue for all scintillator detectors operating in a hostile radiation environment. The main goal is prediction of long-time radiation stability of strongly irradiated scintillator materials based on careful analysis of the defect annealing kinetics.

Notes on radiation damage:

Radiation damage is very important and needs to be studied in details.

In many cases it is due to the formation of structural crystal lattice vacancies and interstitials, changing many functional properties, including optical absorption (change of the colour) and luminescence .



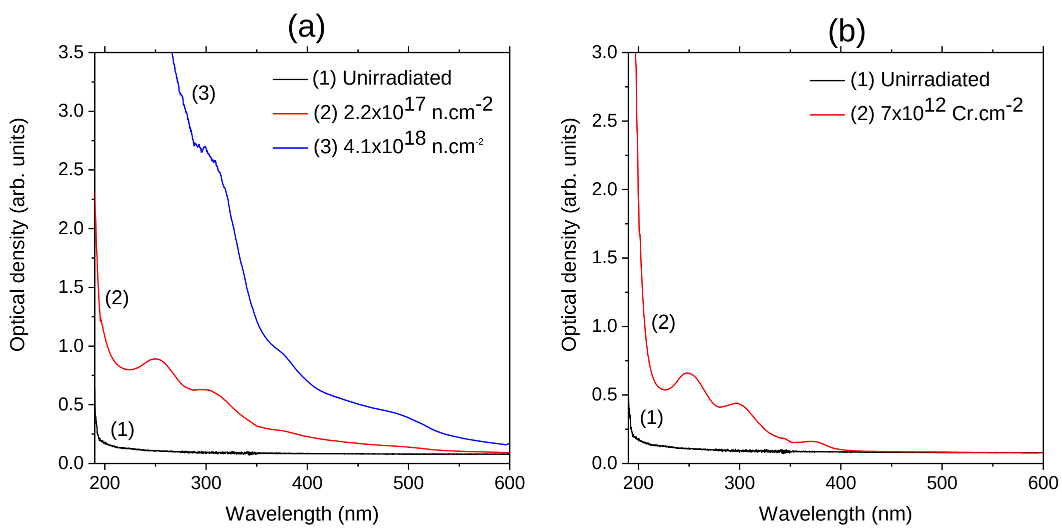
One of the most simple examples:
KCl single crystal.
Violet color of the part of the sample that was in the beam of the X-rays.

Defect recombination kinetics in irradiated $Y_3Al_5O_{12}$

Manucript in preparation: "**Peculiarities of defect recombination kinetics in irradiated yttrium aluminium garnet**" by

A.I. Popov , E.A. Kotomin, M. Izerrouken, V.N. Kuzovkov, R. Villa

(ISSP, Latvia; *Nuclear Research Center of Draria, Algiers* and *CIEMAT, Spain*)



Yttrium aluminium garnet
$Y_3Al_5O_{12}$(YAG)

Optical absorption spectra of YAG before and after irradiation. (a) YAG irradiated with neutrons and (b) YAG irradiated with 6.6 MeV/u Cr ions

Defect recombination kinetics in irradiated $Y_3Al_5O_{12}$

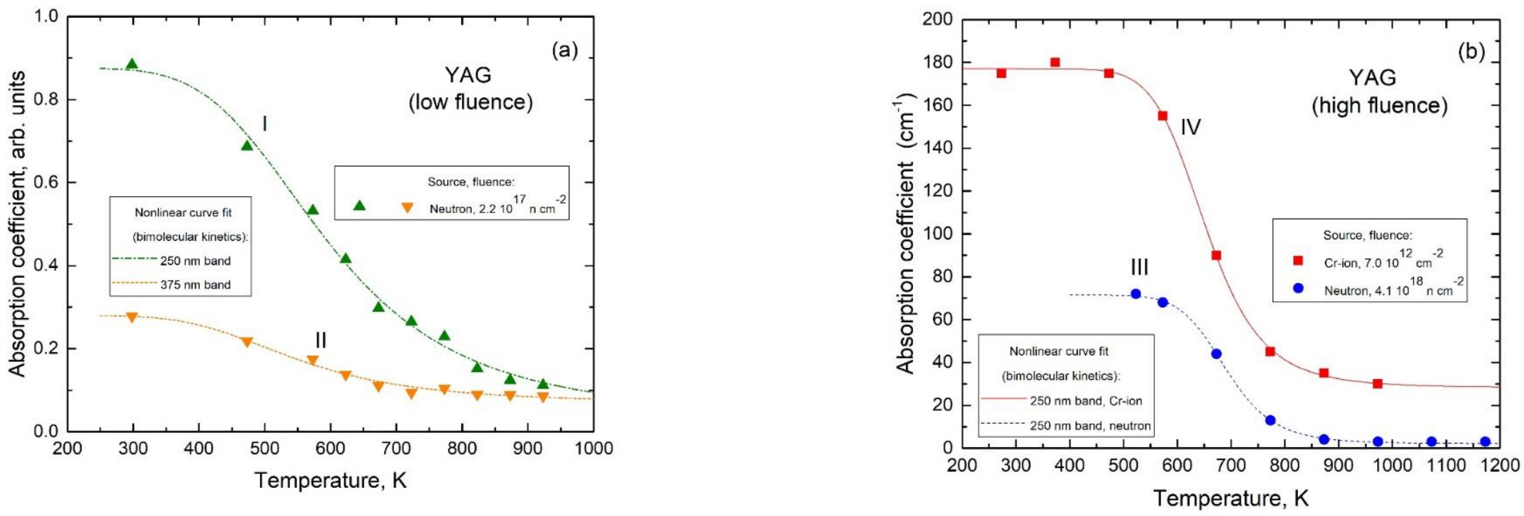
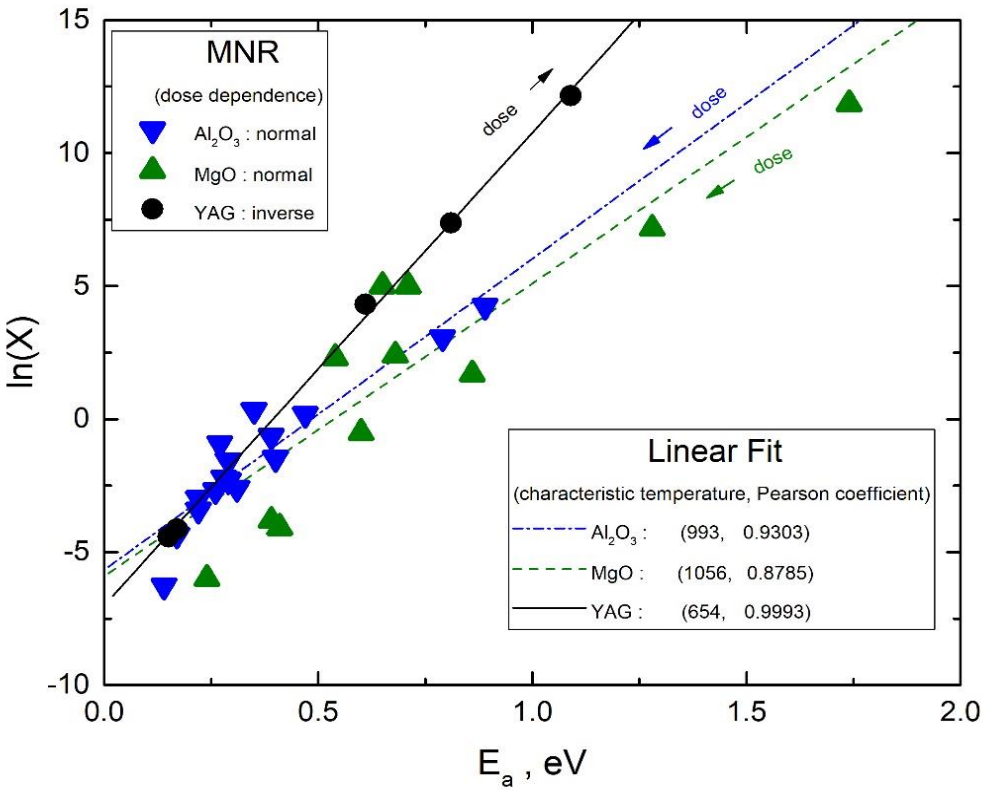


Table . The calculated migration energies E_a and pre-exponential factors X obtained from analysis of experimental data. The experimental and theoretical kinetics (points and full lines I-IV) for the four typical cases are shown in Figure.

Nr.	Type	E_a (eV)	X (K^{-1})	Legend
1	F^+(375 nm)	1.09	$1.9x10^5$	Fast neutrons (>1.2 MeV), fluence 4.1 x 10^{18} n cm^{-2}
2 (I)	F (250 nm)	0.17	$1.6x10^{-2}$	Fast neutrons (>1.2 MeV), fluence 2.2 x 10^{17} n cm^{-2}
3 (II)	F^+(375 nm)	0.15	$1.2x10^{-2}$	Fast neutrons (>1.2 MeV), fluence 2.2 x 10^{17} n cm^{-2}
4 (III)	F (250 nm)	0.81	$1.6x10^3$	Fast neutrons (>1.2 MeV), fluence 4.1 x 10^{18} n cm^{-2}
5 (IV)	F (250 nm)	0.61	$7.5x10^1$	Cr ions, fluence 7.0 x 10^{12} cm^{-2}

Meyer-Neldel rule in $Y_3Al_5O_{12}$



YAG shows the same MNR

but in opposite dose direction

The correlation between the migration energy and pre-exponential factor (the Meyer-Neldel rule) for YAG, as well as Al_2O_3 and MgO.

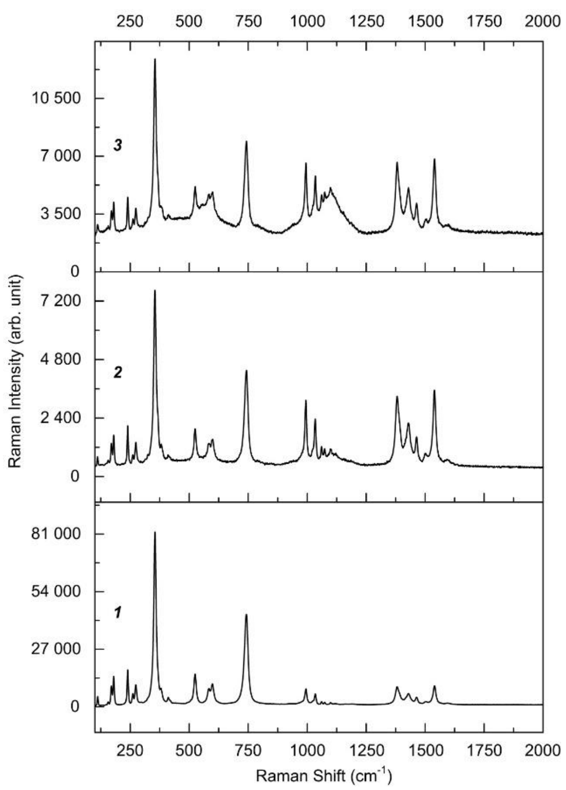
Directions of the radiation fluence (dose) increase are shown by arrows. YAG shows the same MNR but in opposite dose direction

: OPTICAL ABSORPTION AND RAMAN STUDIES OF NEUTRON- IRRADIATED $Gd_3Ga_5O_{12}$ SINGLE CRYSTALS

N. Mironova-Ulmane, I. Sildos, E. Vasil'chenko, G. Chikvaidze, V. Skvortsova, A.Kareiva, R. Pareja, E. Elsts , A. I. Popov **NIMB 2020**

The samples were prepared in the form of a flat wafer 0.4 to 0.8 mm thick oriented in the (111) plane with polished surfaces.The neutron irradiation was performed at Latvian 5 MW water-water research reactor. The fluence of fast neutrons with energy E> 0.1 MeV was in the range of $10^{14} – 10^{18}$ cm^{-2}. The appropriate irradiation temperature was not exceeded 350 K. A cadmium filter was used for absorption of thermal neutrons.

F^+ neutron-induced optical absorption bands are also observed. Their thermal annealing, which is actually similar to MgO and other oxides, is also measured.



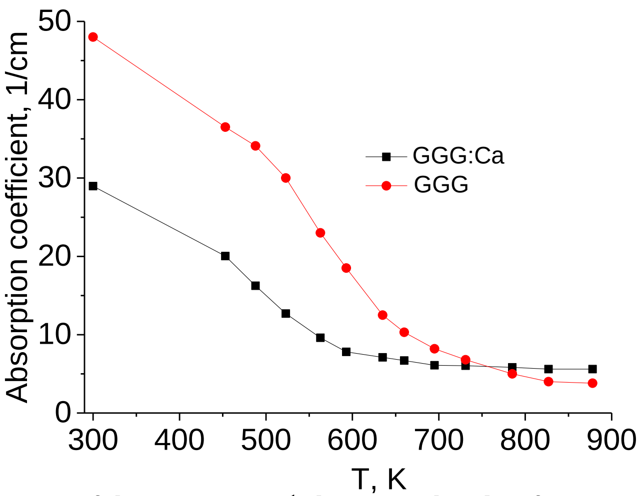
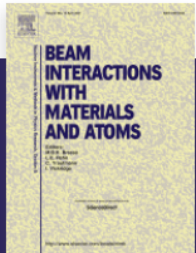


Fig.5. Intensity of the ~34000 cm^{-1} absorption band as function of thermal annealing temperature for irradiated with fast neutrons $Gd_3Ga_5O_{12}$ and $Gd_3Ga_5O_{12}$:Ca

Fig.6. Raman spectra of the irradiated by fast neutron $Gd_3Ga_5O_{12}$ single crystals: 1. before radiated, 2. fluence 10^{16} cm^{-2}, 3. fluence 10^{18} cm^{-2}



Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms

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What is the Next

We were inform that on 03.03 2021 , following recommendations from the Scientific Board and the Programme Manager,

the EUROfusion General Assembly has endorsed the funding for Enabling Research project proposals, including our joint proposal (CfP-FSD-AWP21-ENR-02-UT-01):

"**Investigation of defects and disorder in non-irradiated and irradiated Doped Diamond and Related Materials for fusion diagnostic applications (DDRM) – Theoretical and Experimental analysis**"

Anatoli POPOV (ISSP LU) with Institute of Physics, University of Tartu (PI- Prof. Aleksandr Lushchik) and KIT Karlsruhe Institute of Technology (Prof. Theo Scherer).

This is our second project in a row this time will be for 3 years !

Diamond is also important detector material for CERN

Thank You

Contact: Popov@latent.lv, a.popov@cern.ch