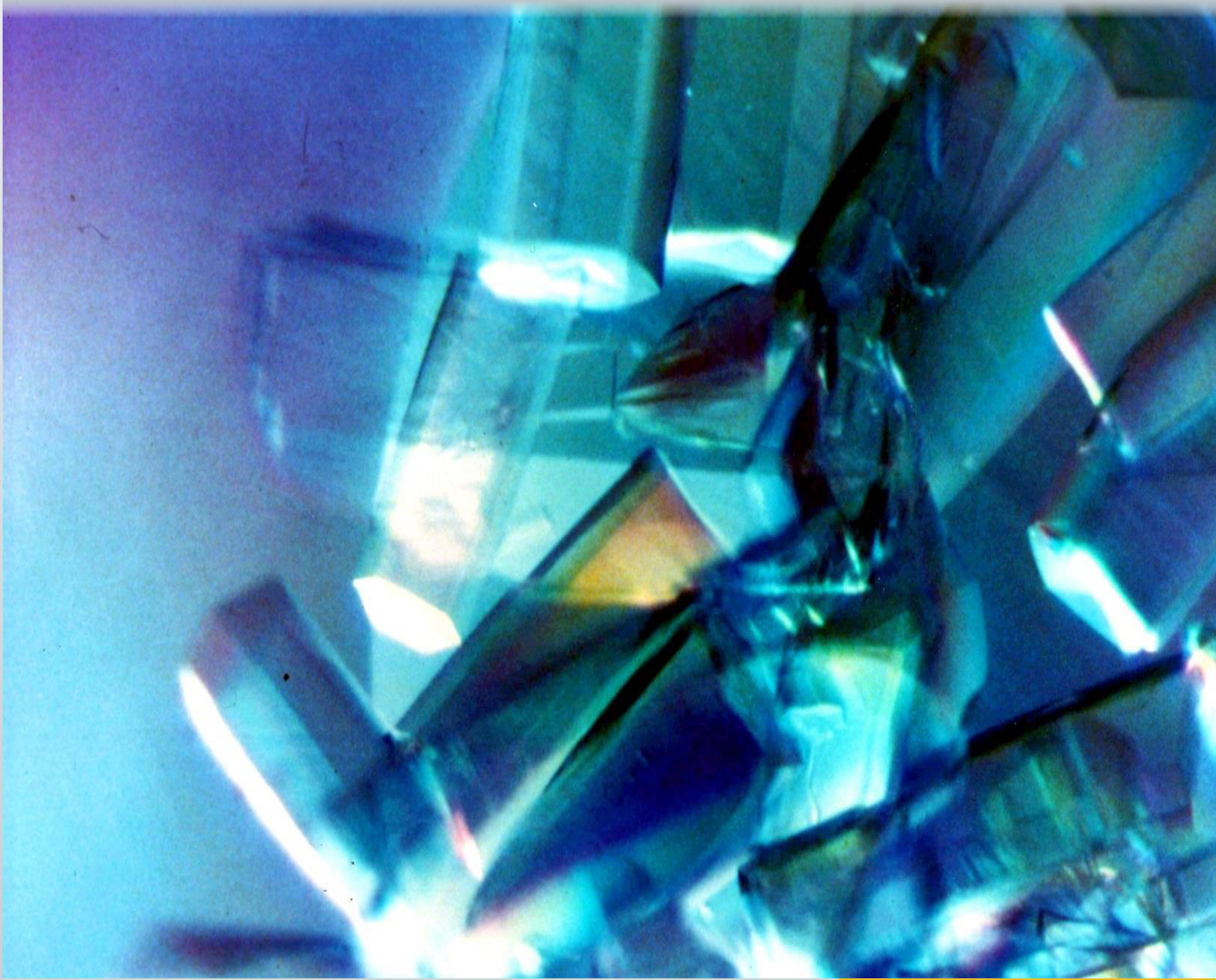
ANNUAL REPORT 2013

April 2013 - March 2014



Yoshikawa Lab

Since 2007

IMR, Tohoku University

ANNUAL REPORT 2013 Yoshikawa Lab., IMR, Tohoku Univ.

Contents

Preface

Dear Colleagues,

Thank you for downloading the Annual Report of the *Yoshikawa Laboratory* in the Institute for Materials Research (IMR) and *Yoshikawa Project* in the New Industry Creation Hatchery Center (NICHe), Tohoku University. Similarly to the previous year, we had decided to avoid publishing the Report as a hard copy to save couple of trees necessary for the paper production and to make it easier for you to access the copy of the Report at any time when you have your computer in your hands.

This is the second report of the laboratory that was originally established in April, 2007 as *Yoshikawa Group* at the Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University. It contains a summary of our research activities and selected papers published in FY2013. Current issue covers our progress within academic year from April 2013 to March 2014. Within this period we had continued development of our basic technologies considering both practical and fundamental points of view. Some of our achievements are summarized below:

- Further progress in studies of GAGG crystals for their application as gamma-ray scintillators. Food survey monitoring system and gamma camera for environment inspection are under development.
- Improvement of ^{6}LiCAF and ^{6}LiSAF growth yield. They are studied for neutron detection as an alternative to the detectors based on application of ^{3}He.
- Establishment of crystal growth technology of halide materials. As an example, Eu:SrI_2 crystals were grown by the micro-pulling down method including crystals of one inch in diameter.
- Development of La-substituted GPS crystal that has exceptionally high temperature stability.
- Bulk and shaped crystal growth of langasite type crystals. Al-substituted CNGG and CTGS crystals are studied for their application in oscillators, resonators, and combustion pressure sensors.

Our long term strategy is to develop chains of research capabilities that connect three areas of expertise including (1) materials production (crystal growth and solid-state synthesis), (2) materials physics and characterization, and (3) application of the materials in contemporary devices. Such chain was already built for the scintillating materials. Our current goal is to apply this strategy to piezoelectric materials.

Our projects are always supported by our colleagues from all over the world, and we note that this collaboration is in most cases very successful. The details of their contribution can be found in the photos and papers published within the above period and included into the Report.

Many laboratory members took part in preparation of this Report. I appreciate their efforts very much. I wish also thank all of our colleagues from Japan and overseas that had participated in our research projects and significantly contributed to their progress.

Akira YOSHIKAWA

Professor,
Institute for Materials Research (IMR), Tohoku University
New Industry Creation Hatchery Center (NICHe), Tohoku University
March, 2014

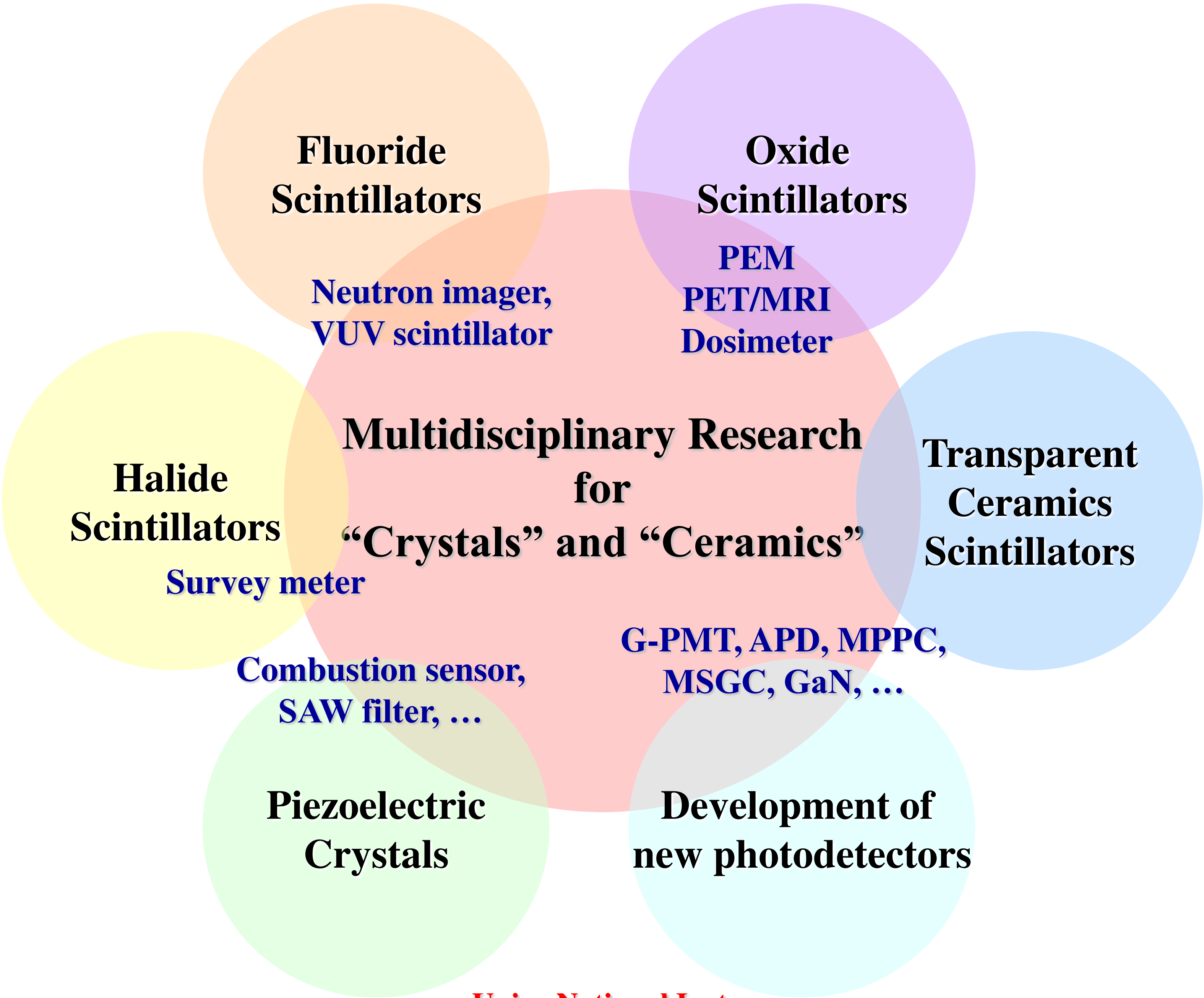
Research Digest

Research Activities in 2013

Development of scintillators, lasers, piezoelectric crystals and crystal growth technology

Int'l collaboration

Inst. Phys. (Czech), Pisa Univ. (Italy), Milan-Bicocca Univ. (Italy),
Ecole Polytechnique (France), Univ. Lyon 1 (France), General Physics Inst. (Russia)
Soltan Institute for Nuclear Studies (Poland), Delft Univ. (New Zealand)



Univ., National Inst.

Univ. of Tokyo (Kamiokande, Takahashi Lab.), Kyoto Univ. (Tanimori Lab),
Osaka Univ. (Sarukura Lab), Nagoya Univ. (Iijima Lab, Uritani Lab, Iguchi Lab),
Hiroshima Univ. (Fukazawa Lab), Kyushu Univ. (Ishibashi Lab), JAEA (Fukushima headquarters)

Company

Tokuyama, Furukawa, TDK, Hamamatsu Photonics, Canon, Mitsui Kinzoku,
Chiyoda Technol, Nihon Kessho Kogaku, Hitachi-Aloka Medical, Pony Industry, Oxide, GES
Tanaka Kikinzoku Kogyo, Furuya, Star seiki, TEP, Toei Scientific Industrial

Oxide Scintillators

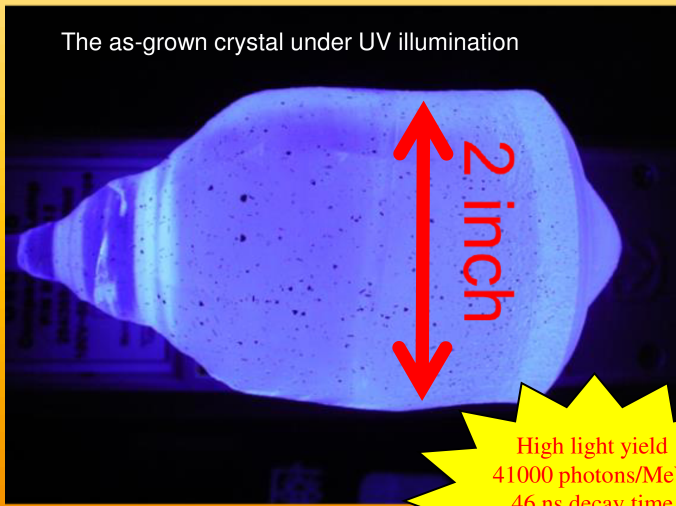
Gamma-ray detection

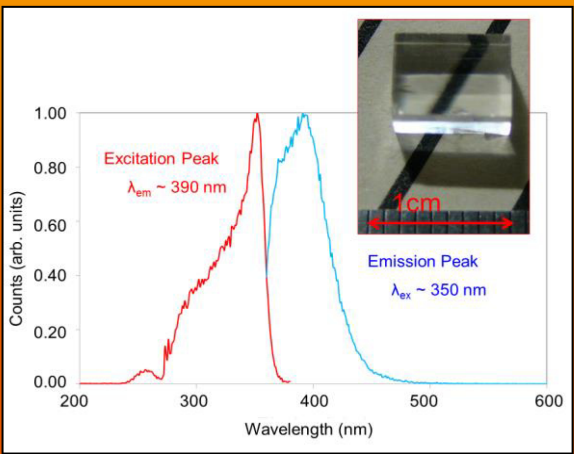
(PET, SPECT, Gamma-camera, oil logging)

Ce-doped and La-admixed gadolinium pyrosilicate
$(Ce_{0.01}Gd_{0.90}La_{0.09})_2Si_2O_7$

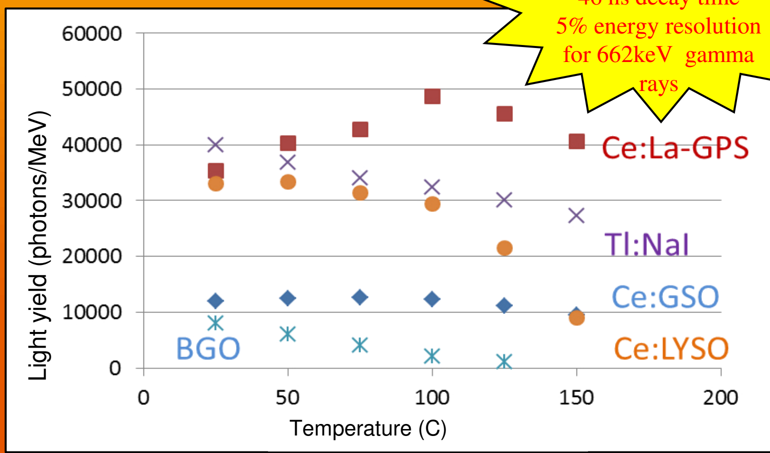
Ce:LaGPS

Ce:LaGPS





Emission at 390 nm

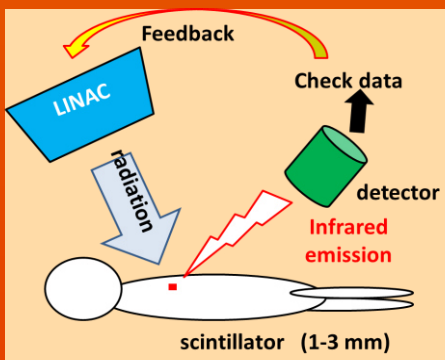


Good thermal stability until high temperature

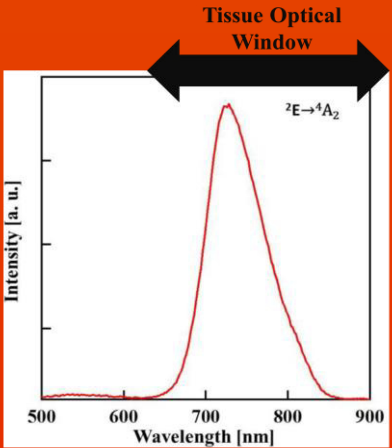
Infra-red scintillators

Cr-doped $Gd_3Ga_5O_{12}$

As-Grown Crystal Polished Samples



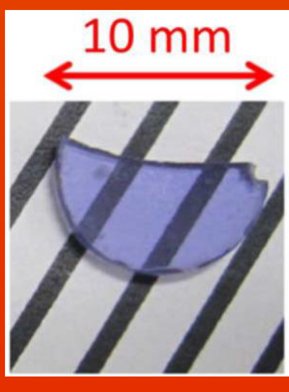
Real-time patient dosimetry system with infra-red scintillators for radiation therapy

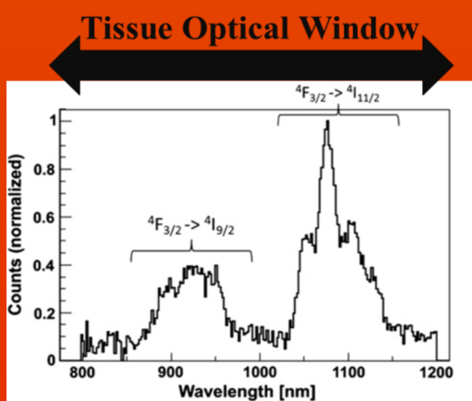


Cr^{3+} emission – matches tissue transparency window

A. Yamaji et al., Poster Presentation, SCINT2013 (2013)

Nd-doped Lu_2O_3

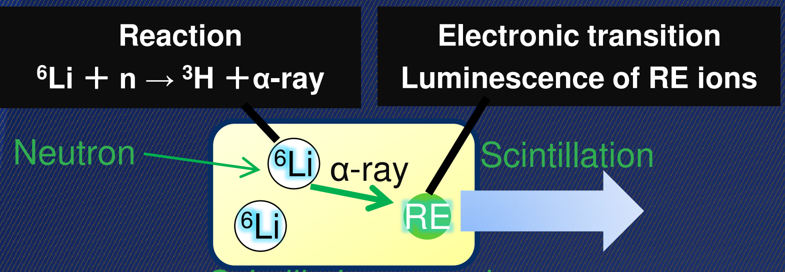




Kurosawa et al., IEEE TNS (2013).

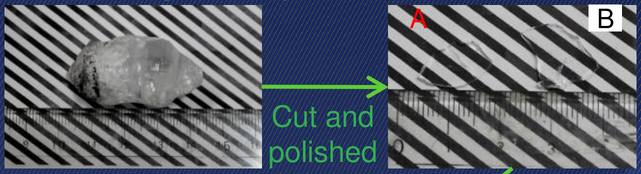
Fluoride scintillators

Neutron scintillators - alternative to ^3He

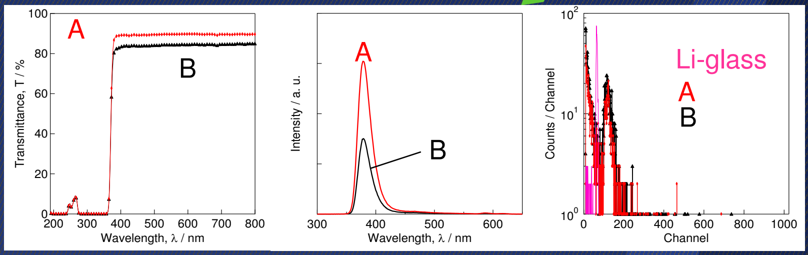


Scintillation crystal

Single crystal for neutron system grown by Cz method



Security applications employing neutron systems



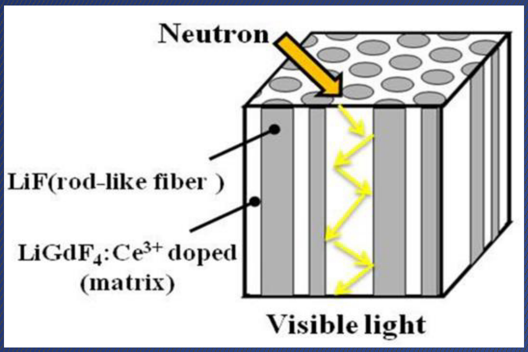
Transmittance spectra

Radioluminescence (^{241}Am)

Pulse height spectra (^{252}Cf)

Eutectic composite scintillators

Concept of eutectic composite scintillators for neutron detection



As-grown eutectic composite scintillators

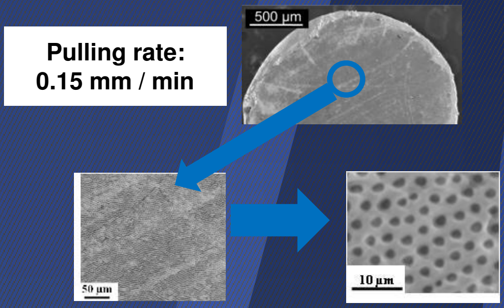
$LiGdF_4$-LiF undoped

1%Ce doped $LiGdF_4$-LiF

1%Ce and 0.5%Ca co-doped $LiGdF_4$-LiF

Backscattered Electron Images

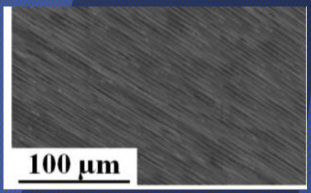
On transverse cross-section



Rods dispersed well.

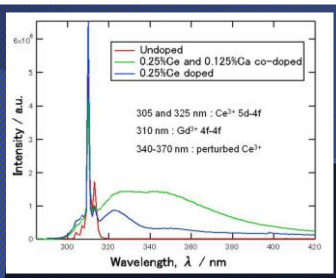
About 6600 rods / mm²

Along the growth direction

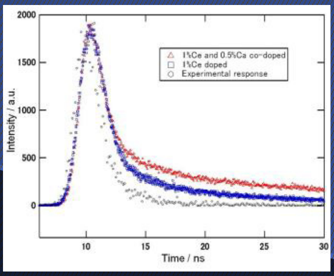


Rod fibers were grown well.

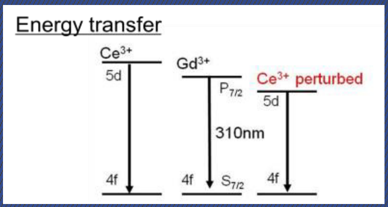
The length of longest rod : 800 μm



Photoluminescence spectra (ex.=270nm)

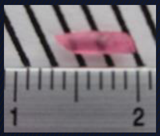


Photoluminescence decay curve (em.=340, ex.=320nm)



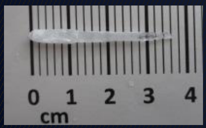
	Emission at 290 nm (Excitation at 270 nm)	Emission at 310 nm (Excitation at 270 nm)	Emission at 340 nm (Excitation at 320 nm)
Undoped	-	660 μs	-
Ce doped	2.10 ns	244 μs	16.78 ns
Ce and Ca co-doped	1.60 ns	137 ns	19.62 ns

Flux growth

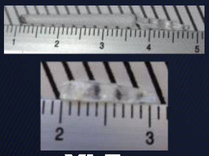


ErF_3

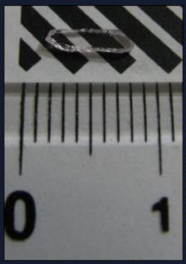
TmF_3



YF_3

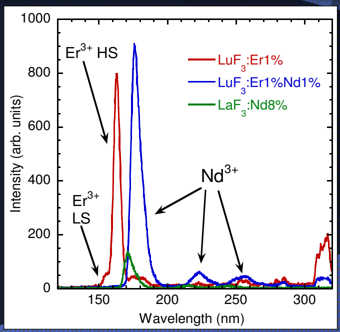


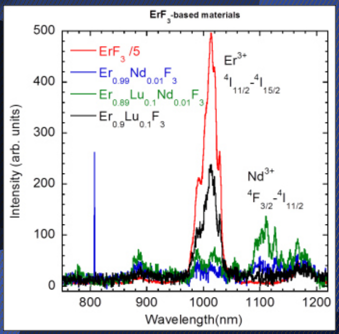
YbF_3



LuF_3:Er1%,Nd1%

VUV / IR scintillators





Radioluminescence spectra of VUV and IR scintillators

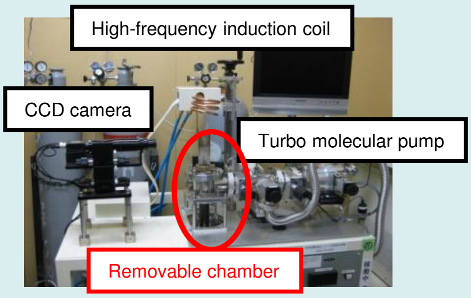
Halide scintillators

The aim of this work was to develop new halide scintillator materials for some applications for example gamma-ray survey meter, spectrometer, food radiation detector and so on. In this year, we have challenged to improve scintillation performance of Eu doped SrI_2 single crystals grown by the micro-pulling-down (m-PD) method. They are displayed below.

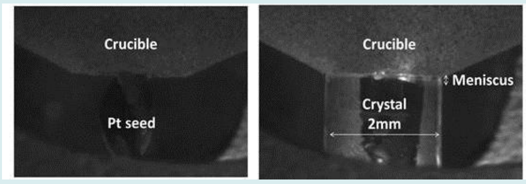
Experimental arrangement of halide m-PD machine.

Setting chamber in glove box

Crystal growth



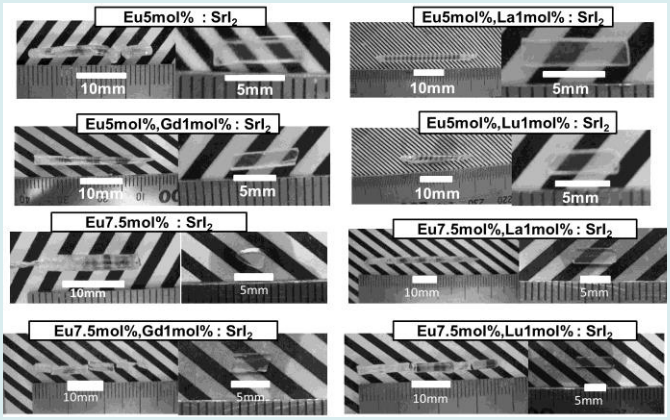
Stable crystal growth



Liquid-solid interface during crystal growth observed by the CCD camera. (left) initiation of growth with first seed contact with the melt and (right) stable growth.

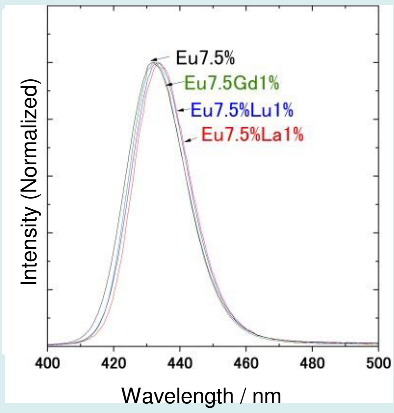
Halides are hygroscopic materials which easily react with air moisture to form oxy- and hydroxy- halides. Therefore, careful handling of starting materials under protective argon atmosphere in glove box is required.

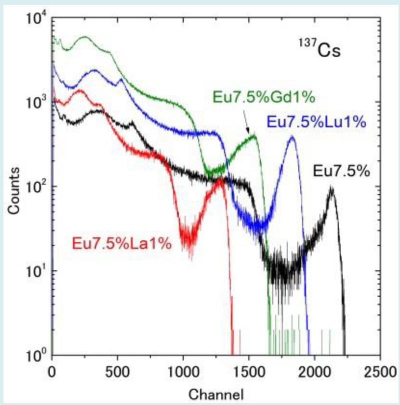
Grown rare-earth codoped Eu:SrI_2



As-grown and polished RE(La, Gd or Lu)1mol%, Eu5, 7.5mol doped SrI_2 crystals grown by the modified μ-PD method.

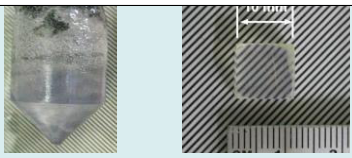
Scintillation properties

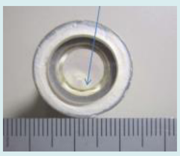




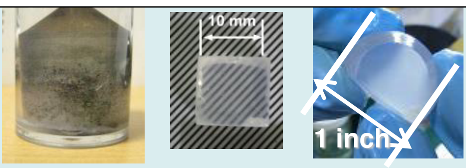
RE-codoped Eu:SrI_2 crystals were studied by X-ray radioluminescence measurements and showed Eu^{2+} emission peak around 430 nm. Under ^{137}Cs γ-ray irradiation light yield and energy resolution was decreased by approximately 20% and 5~7% respectively.

As-Grown 1 inch Eu5%:SrI_2 bulk crystal

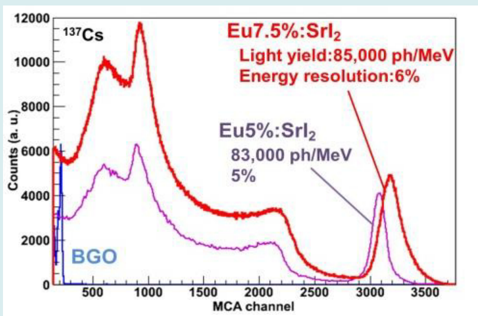




As-Grown 1 inch Eu7.5%:SrI_2 bulk crystal



Pulse-height spectra



Pulse-height spectra of 1 inch Eu:SrI_2 crystals were measured under ^{137}Cs γ-ray irradiation. Eu7.5mol%:SrI2 1 inch crystal showed high light yields and good energy resolutions: of 85,000ph/MeV and 6% respectively.

Transparent ceramic scintillators

Introduction

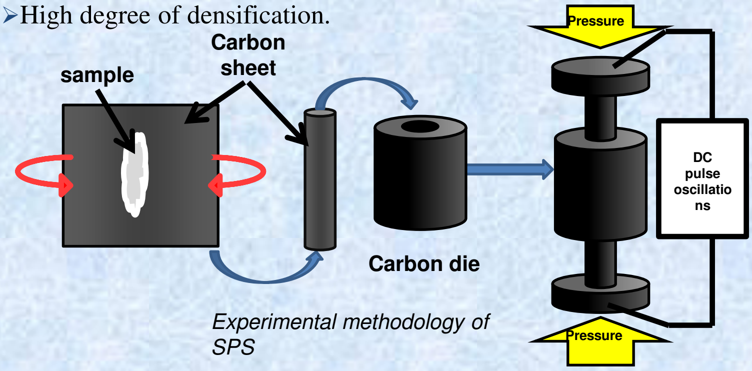
Advantages of ceramics

- Better chemical uniformity than single crystals.
- Can be produced with high dopant concentration.
- Economical especially for high-melting materials.
- Defects present in single crystals due to high melting temperature are often absent in ceramics → improvement of some properties (for example anti-site defects in $Lu_3Al_5O_2$:Ce are absent, which leads to absence of sub-microsecond slow components in scintillation decay)

Spark plasma sintering(SPS)

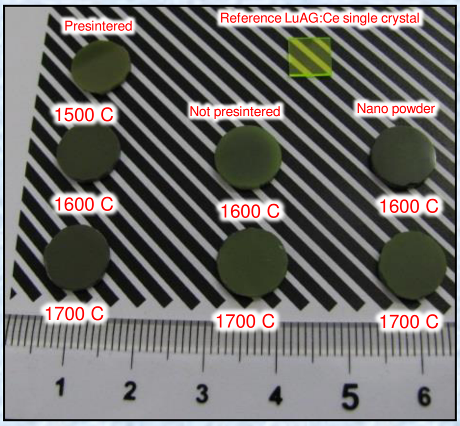
Advantages of SPS

- A rapid consolidation rate appropriate for densification of variety of ceramics.
- Highly reductive condition due to carbon die and punch and vacuum environment.
- High degree of densification.



Experimental methodology of SPS

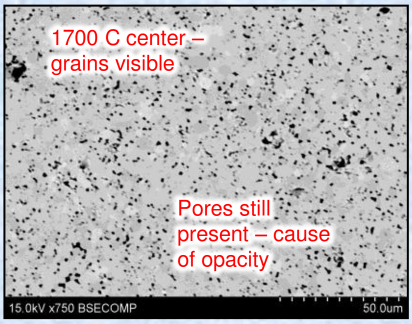
$Lu_3Al_5O_{12}$:Ce(LuAG:Ce)



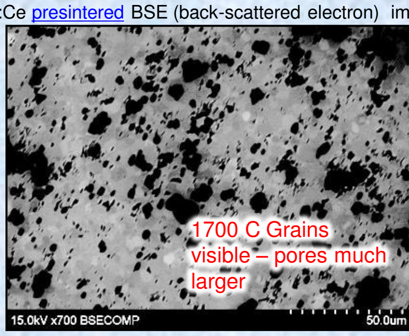
LuAG:Ce samples sintered by SPS method

99.99% Powders of Lu_2O_3, Al_2O_3 and CeO_2 mixed in mortar for 3 hours
1) Part of the mixture presintered at 1600C for 8 hours. Ce-concentration: 0.2 mol%
2) Non-presintered mixture Ce-concentration: 0.2 mol%
3) LuAG:Ce Nanopowder prepared by unique radiation-induced precipitation 20-60 nm size, Ce concentration around 1.5 mol%
(see J. Barta, V. Cuba et al.: J. Mater. Chem., 2012, 22, 16590 for details)

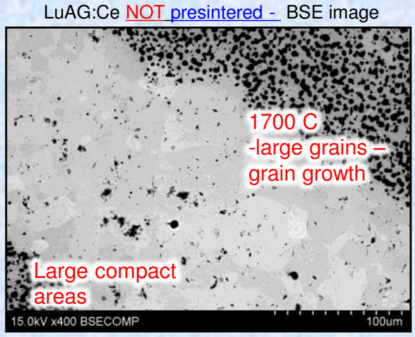
The above powders sintered by SPS at various sintering temperatures at 100MPa pressure for 45 minutes



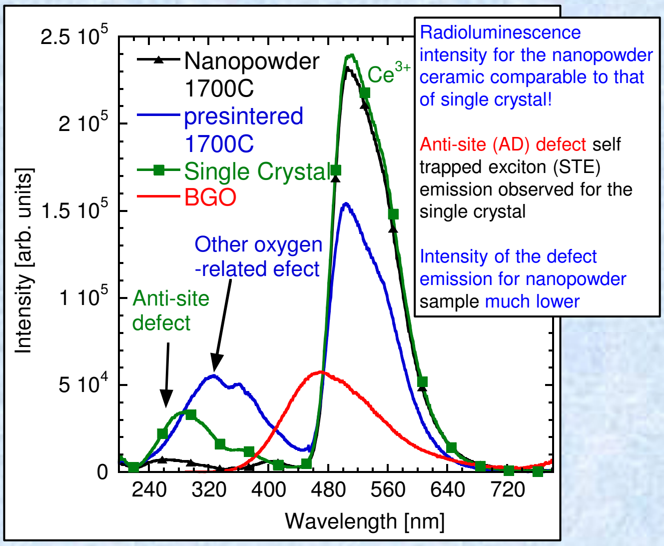
LuAG:Ce presintered BSE (back-scattered electron) image

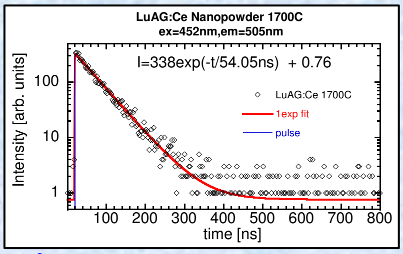


LuAG:Ce NOT presintered - BSE image

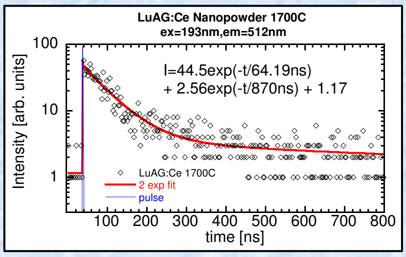


LuAG:Ce nanopowder - BSE image





Ce^{3+} emission under direct excitation (452nm)

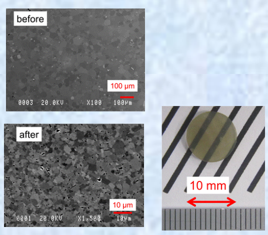


Ce^{3+} emission under band-to-band excitation (193nm) – slower components related to other complex defects

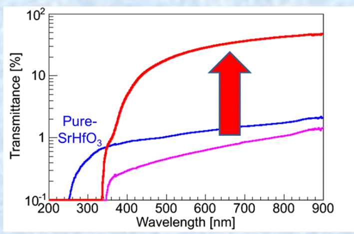
J. Pejchal et al., STAC7 2013

$Ce:SrHfO_3$

Hf : High Z, but high melting point.
→ Using SPS method.



Sintered $SrHfO_3$ sample and surface SEM images before and after annealing



Transmittance of $Ce:SrHfO_3$

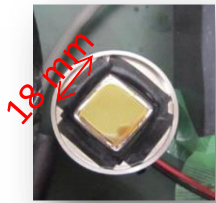
Photodetectors

We investigate radiation response of our scintillators using a photomultiplier tube and semi-conductor in a specially designed temperature chamber.

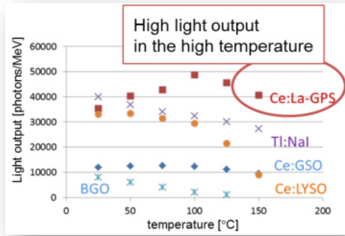
1, Pulse-height Measurement System

We use several photo-detectors for pulse height measurements.

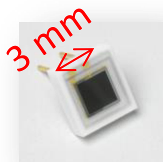
- **PMT (Photo multiplier tube)**
  - ✓ Ultra and super –bialkali (UBA and SBA)
  - ✓ Ruggedized photomultiplier for oil well logging



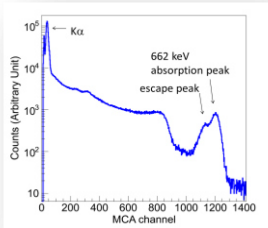
Ultra Bialkali (UBA)
R7600U-200



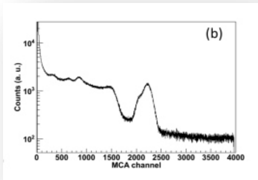
- **APD (avalanche photodiode)**
  - ✓ High quantum efficiency (up to ~80%)
  - ✓ Wide sensitive range (ex: 320 -1000 nm for S8664 HPK)



APD

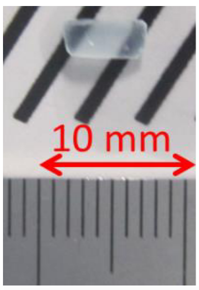


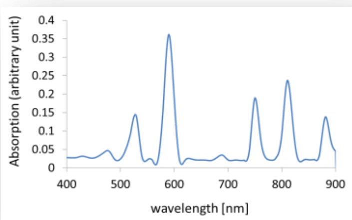
- **MPPC (Multi-Pixel Photon Counter)**
  - ✓ Photon counting regime

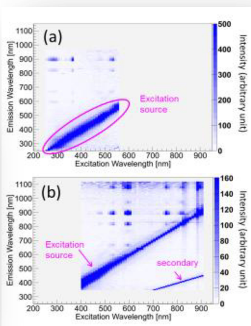


2, New Spectrometer

We have developed a new spectrometer for visible and near Infra-red (NIR) scintillators including halide crystals.

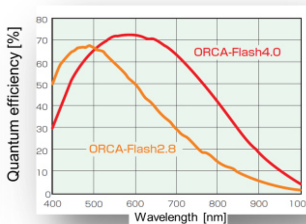


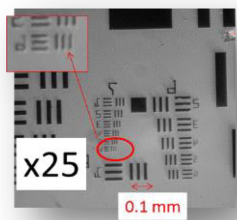


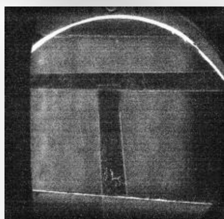


3, New Imaging system

We have developed a new imaging system for visible and near Infra-red (NIR) scintillators.







Development of Piezoelectric Single Crystals

Langasite-type crystals

Superior properties

- Electromechanical coupling factor is 2-3 times higher than that for quartz.
- The curie point not observed.
- High electrical resistivity at high temperature found for rare-earth-free langasite type crystals
- Small impedance at Low frequency

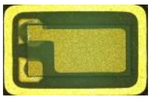
Applications

Combustion Sensors

Improvement of engine efficiency by pressure monitoring and combustion control to decrease amount of exhaust gases.

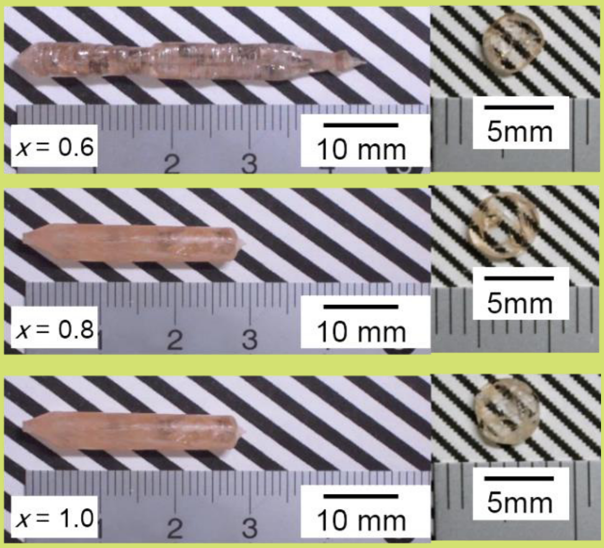
Stability up to 400℃ is required.

Oscillators

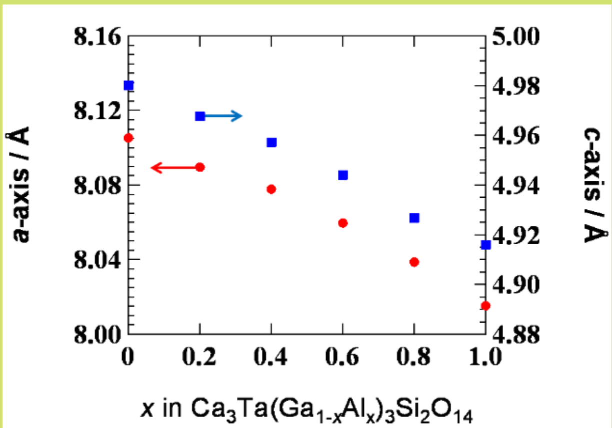


Development of next-generation smartphones and tablets employing smaller oscillators operating at low frequencies with low energy consumption.

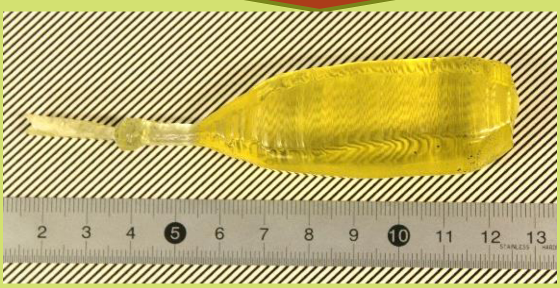
Growth of $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ crystals



Lattice constants



$Ca_3Ta(Ga_{0.5}Al_{0.5})_3Si_2O_{14}$crystal grown by Cz method



We successfully grew $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ crystals with various Al concentrations keeping the langasite-type structure.

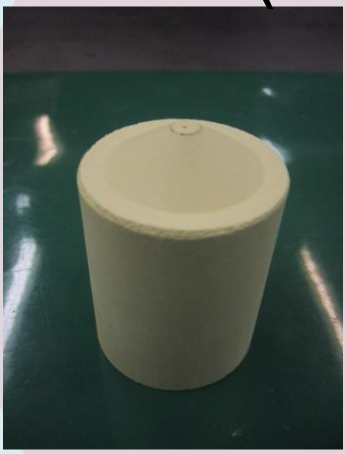
Development of shaped crystals

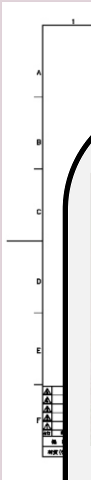
Development of fiber Ir alloy crystal with difficult workability (H23 Support Industry [Ministry of Economy, Trade and Industry] with TKK, Sutar Seiki ,TEP, Toei kagaku sangyo)

Necessity of development of low cost manufacturing technology of functional metal alloy with difficult workability

Crucible with high resistance

Heat-resistant crucible (> 2000ºC)



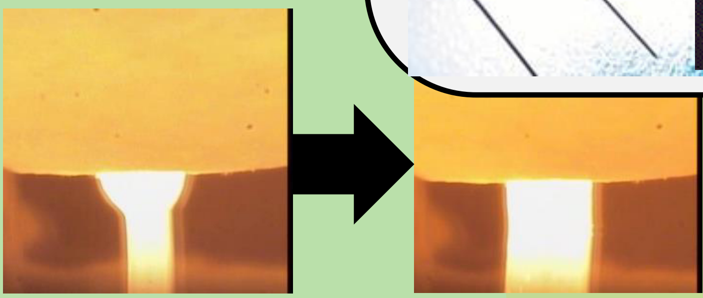


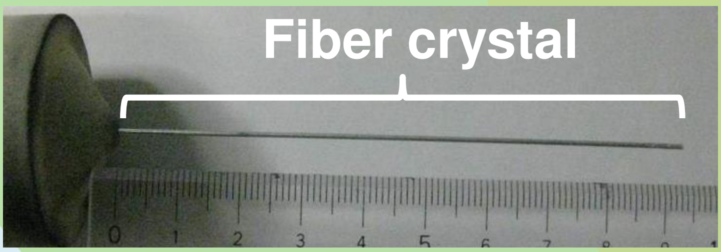
Manufacturing technique

Manufacturing of ceramics

Applications

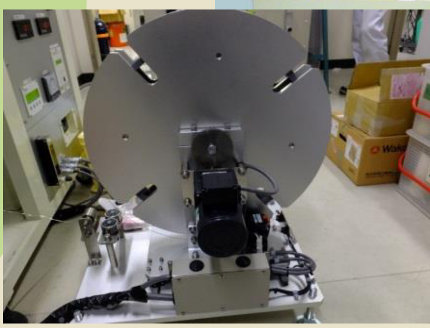
Ir alloy fiber





Growth method of shaped crystal

μ-PD furnace for shaped crystal



Growth furnace

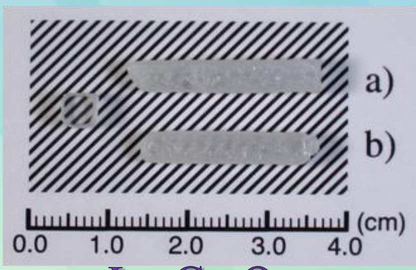
CRYSTAL GALLERY

OXIDES

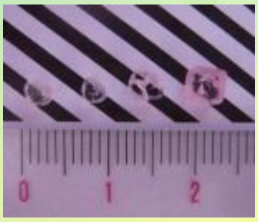
< Garnet type >

$Y_3Al_5O_{12}$

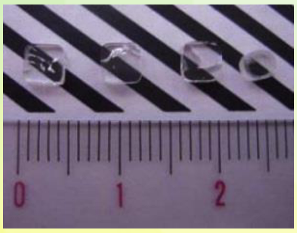
$Yb:Y_3Al_5O_{12}$



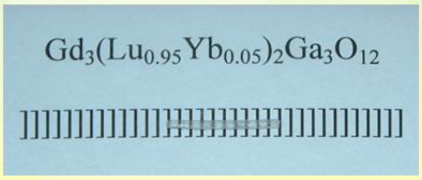
$Lu_3Ga_5O_{12}$



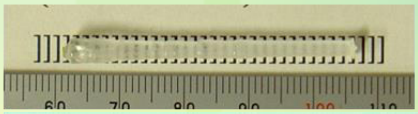
$Ho:Lu_3Al_5O_{12}$



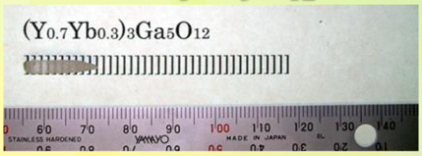
$Tm:Lu_3Al_5O_{12}$



$Gd_3Lu_2Ga_3O_{12}$

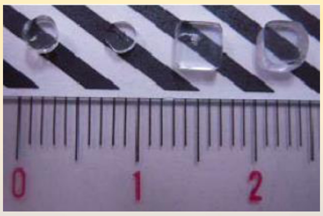


$Lu_3Al_5O_{12}$



$Y_3Ga_5O_{12}$

$Ce:Lu_3Al_5O_{12}$

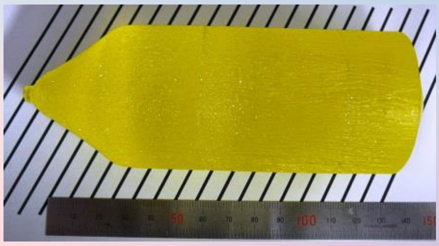


$Nd:Lu_3Al_5O_{12}$

$Pr:Lu_3Al_5O_{12}$

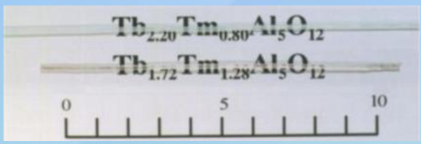
$Gd_3Ga_5O_{12}$

$Gd_3Al_2Ga_3O_{12}$

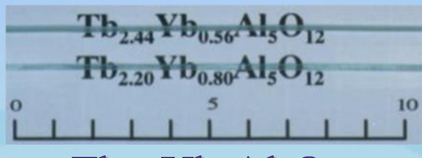


$Ce:Gd_3Al_2Ga_3O_{12}$

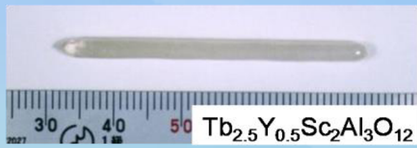
< *Garnet type* >



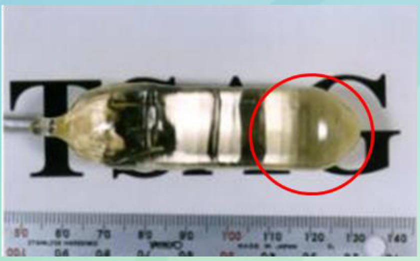
$Tb_{3-x}Tm_xAl_5O_{12}$



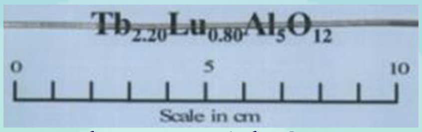
$Tb_{3-x}Yb_xAl_5O_{12}$



$Tb_{2.5}Yb_{0.5}Sc_2Al_3O_{12}$



$Tb_{2.2}Sc_{2.8}Al_3O_{12}$



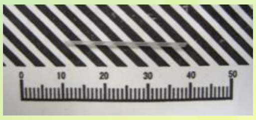
$Tb_{3-x}Lu_xAl_5O_{12}$

$Tb_{3-x}Sc_{2+x}Al_3O_{12}$

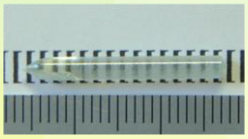
< *Ilmenite type* >

$LiNbO_3$

< *Perovskite type* >

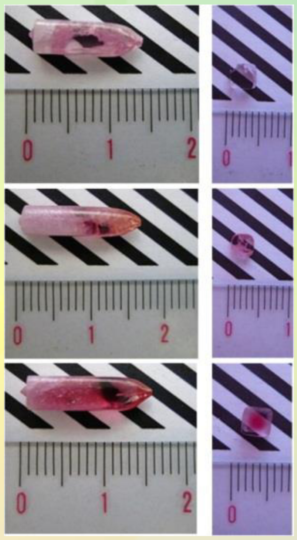


$YAlO_3$

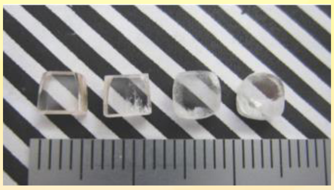


$(Lu, Y)AlO_3$

$KNbO_3$



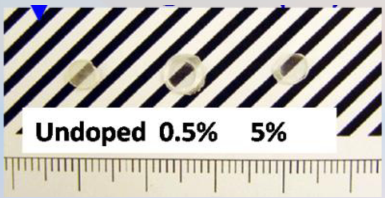
$TbAlO_3$



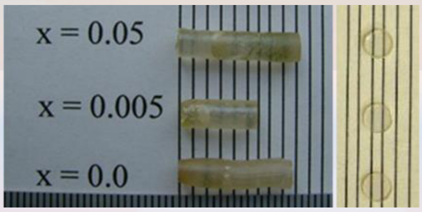
$Tm:YAlO_3$

$Cr:YAlO_3$

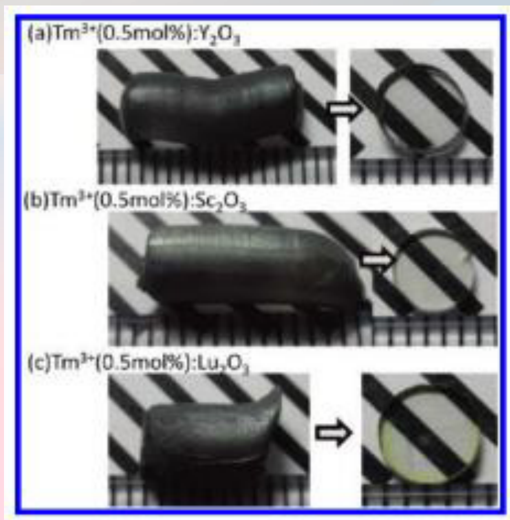
< *Sesquioxide type* >

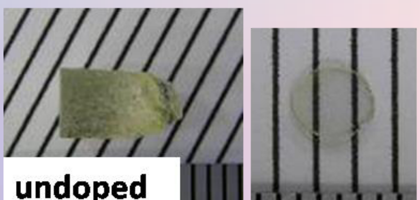


Sc_2O_3

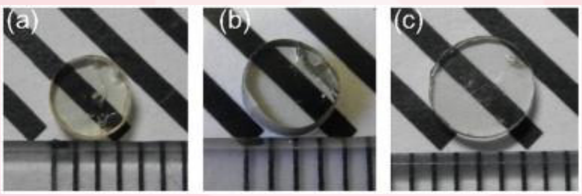


Y_2O_3





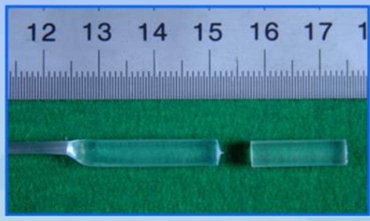
Lu_2O_3



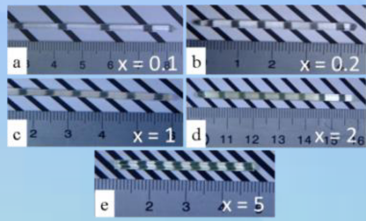
< Apatite type >

$Ca_8La_2(PO_4)_6O_2$

< Spinel type >

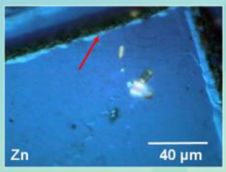


$Ti{:}MgAl_2O_4$



$Mn{:}MgAl_2O_4$

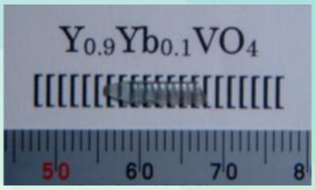
< ZnO type >

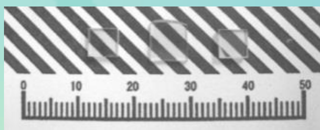


LPE ZnO Single Crystalline film

ZnO Single Crystal

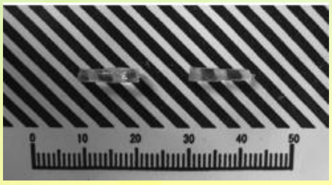
< Vanadate type >





YVO_4, $(Y,Lu)VO_4$, $LuVO_4$

< Aluminate type >



$LiAlO_2$

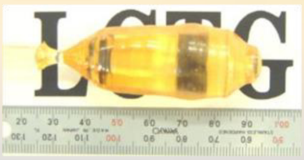
< Corundum type >

Al_2O_3

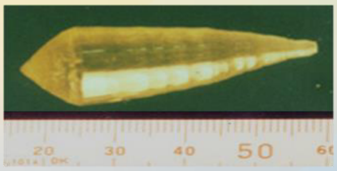
$Cr{:}Al_2O_3$

< Langasite type >

$La_3Nb_{0.5}Ga_{5.3}Al_{0.2}O_{14}$

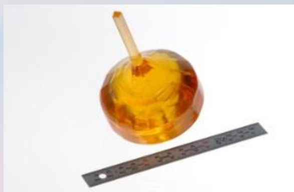


$La_{2.95}Ca_{0.05}Ta_{0.525}Ga_{5.475}O_{14}$



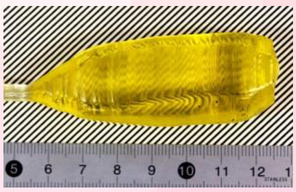
$Sr_3TaGa_3Si_2O_{14}$

$La_3Ga_{5.3}Al_{0.2}SiO_{14}$



$La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$

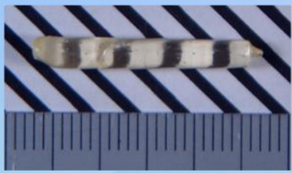
$La_{2.95}Ba_{0.05}Ta_{0.525}Ga_{5.475}O_{14}$



$Ca_3TaGa_3Si_2O_{14}$

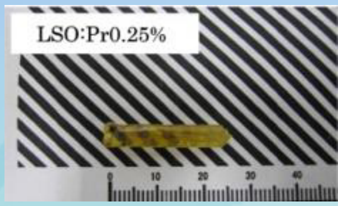
$Ca_3NbGa_3Si_2O_{14}$

< Silicate type >



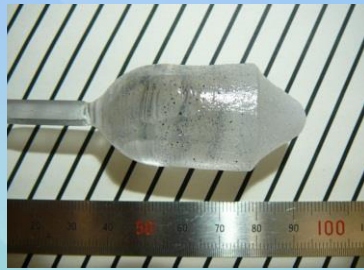
Lu_2SiO_5

$Pr:Y_2SiO_5$



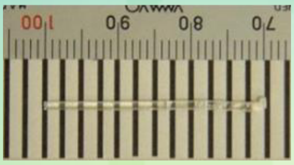
$Pr:Lu_2SiO_5$

$Ce:Gd_2SiO_5$

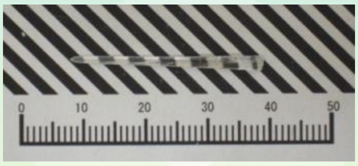


$Ce:(Gd_{1-x}La_xSi)_2O_7$

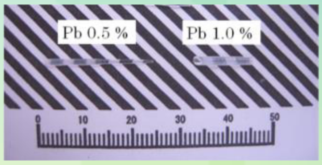
< Borate type >



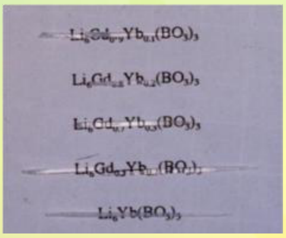
$Li_6Y(BO_3)_3$



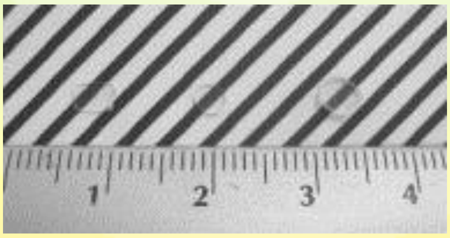
$YCa_4O(BO_3)_3$



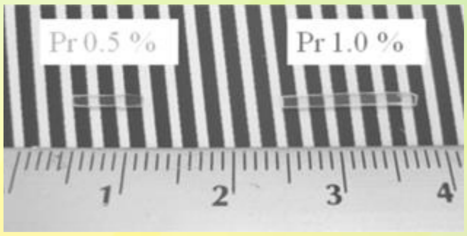
$Pb:YCa_4O(BO_3)_3$



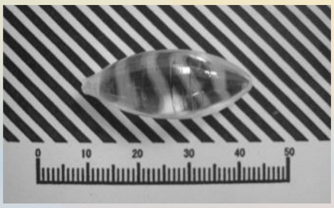
$Li_6Yb(BO_3)_3$,
$Li_6Gd(BO_3)_3$



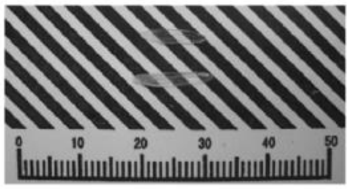
$Tm:Ca_3(BO_3)_2$



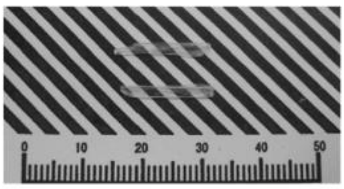
$Pr:Ca_3(BO_3)_2$



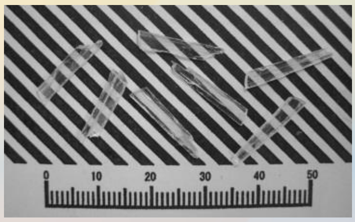
$Ce:Ca_3(BO_3)_2$



SrB_2O_4

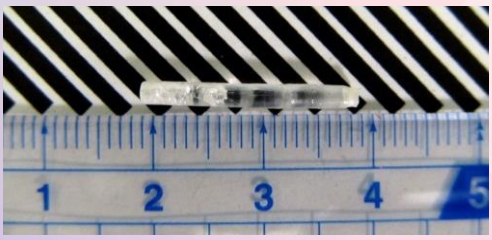


$Ce:SrB_2O_4$

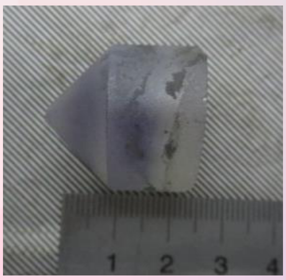


CaB_2O_4

Halides

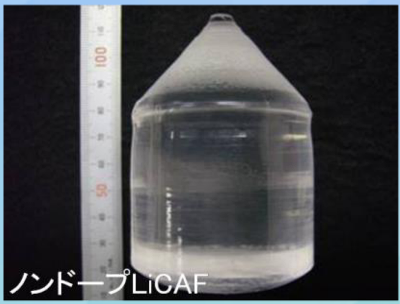


$Yb:RbPdCl_5$

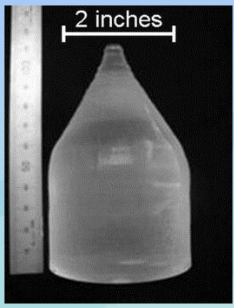


$Eu:SrI_2$

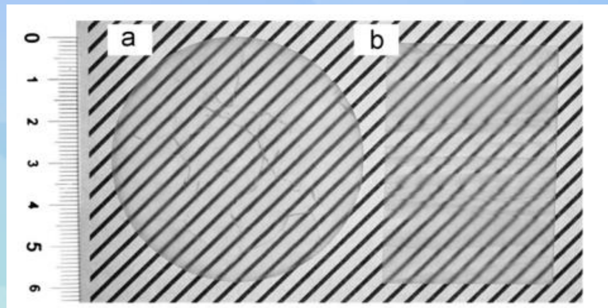
Fluorides



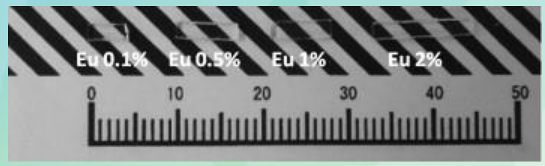
$LiCaAlF_6$



$Ce:LiCaAlF_6$



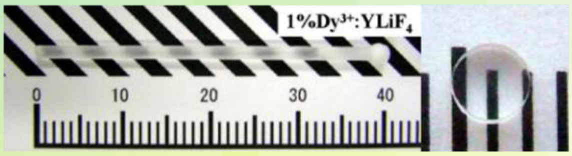
$Eu:LiF/CaF_2$



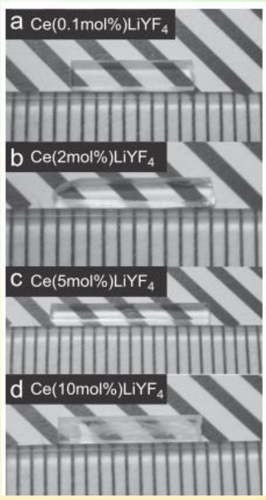
$Eu:LiCaAlF_6$

$Ce:LiSrAlF_6$

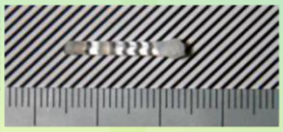

$Eu:LiSrAlF_6$

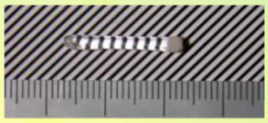


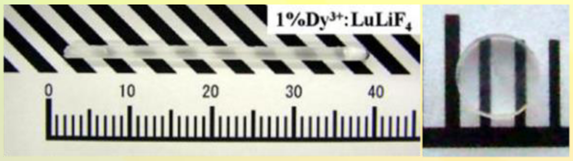
$Dy:LiYF_4$



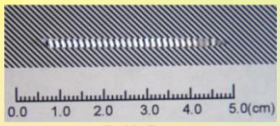
$Ce:LiYF_4$


$SrMgF_4$

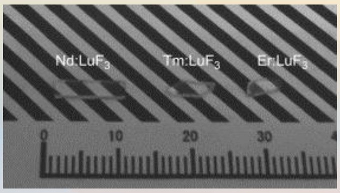

$Ba_{0.2}Sr_{0.8}MgF_4$



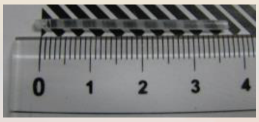
$Dy:LuYF_4$

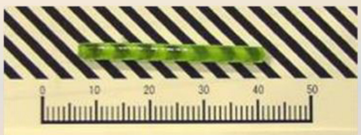


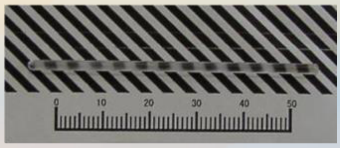
$BaMgF_4$

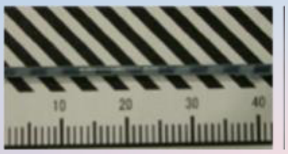


$Nd:Tm:Er:LuF_3$


$Tm:Nd:BaYLuF_8$


$Ce:PrF_3$

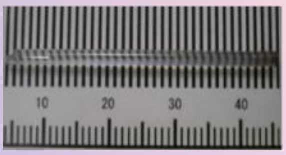

$K(Y_{0.99}Pr_{0.01})_3F_{10}$

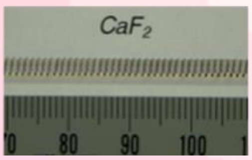

NdF_3

BaF_2

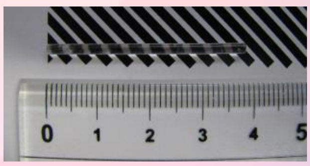

ErF_3

$LiGdF_4$-LiF


CeF_3

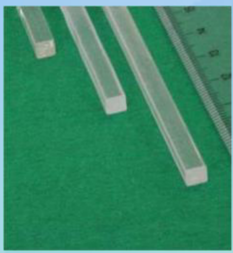


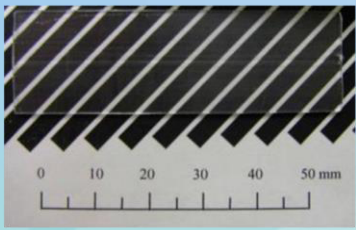
CaF_2

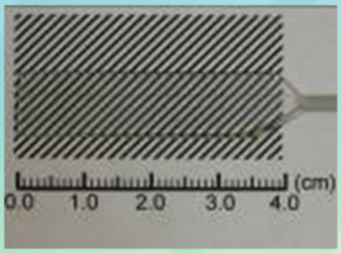

KYF_4

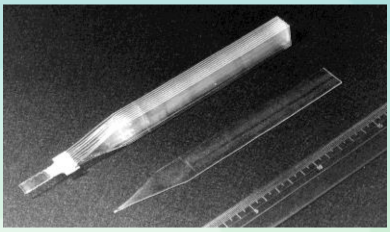
$LiAlF_6$-CaF_2

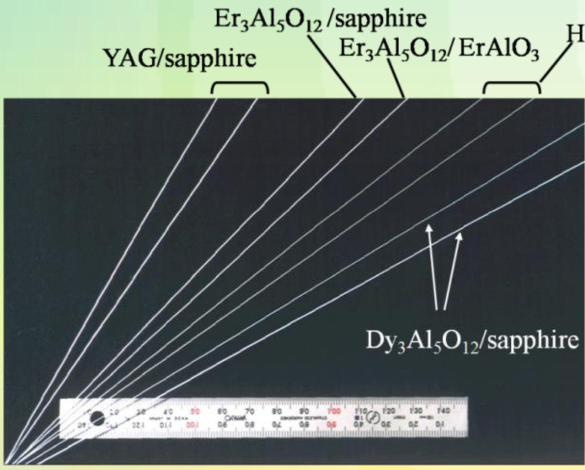
Shaped Crystals

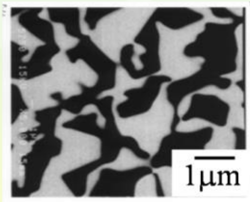


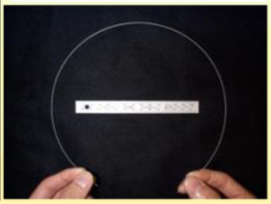


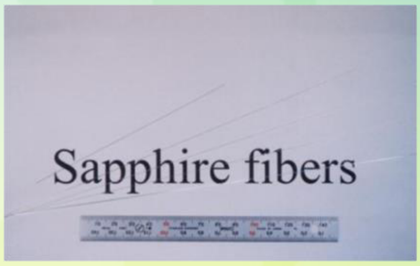




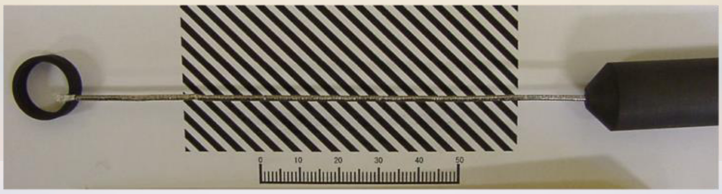




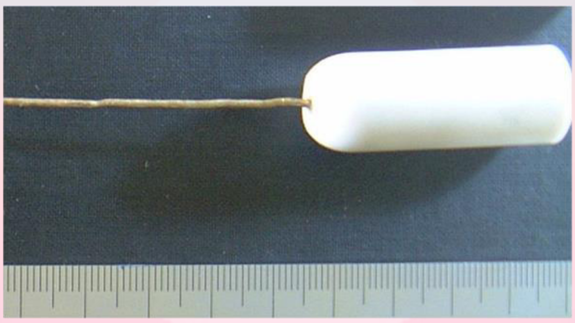




Others



Fe-Ni-Ga alloy fiber produced from carbon crucible (right) with conically shaped bottom (Scale in mm)



$Cu_{72}Al_{17}Mn_{11}$ alloy fiber produced from Al_2O_3 ceramic crucible (right) with spherically shaped bottom (Scale in mm)

Instruments in 2013

Growth Equipment (μ-PD Method)

Fluorides (RF-Heated μ-PD System)

TDK, MPD-HT
Anti-vibration system
Temperature: ～ 2500°C
Atmosphere: Ar,N_2,H_2,CF_4,O_2

TDK, MPD-HT
Anti-vibration system
Temperature: ～ 2500°C
Atmosphere: Ar,N_2,H_2,CF_4,O_2

Halides (RF Heated μ-PD System)

Toei scientific industrial co., Ltd.
Temperature: ～ 1200°C
Atmosphere: Ar,N_2,H_2,CF_4,O_2

Oxides (RF Heated μ-PD System)

SAPOIN,
CNMPD
Anti-vibration system
Atmosphere: Ar,N_2,H_2,O_2

TACHIBANA RIKO,
SCF-600M
Anti-vibration system
Temperature: ~ 2000°C
Atmosphere: Ar,N_2,H_2,O_2

TDK, M-PD2-A
Anti-vibration system
Temperature: ~ 2500°C
Atmosphere: Ar,N_2,H_2,O_2

Sapoin 2
Anti-vibration system
Atmosphere: Ar,N_2,H_2,O_2

Growth Equipments (Cz Method)

Fluorides (RF Heated Cz System)

Nisshin Giken
Temperature: ～ 1600°C
Atmosphere: Ar, N_2, H_2, CF_4, O_2

Oxides (RF Heated Cz System)

CYBERSTAR, OXYPULLER 05-03
Heating system: RF
Vacuum: 30Pa
Atmosphere: Air, Ar, N_2, O_2
Temperature: ～2200°C

30kW-Cz
Heating system: RF
Atmosphere: Air, Ar, N_2, O_2
Temperature: ～ 2000°C

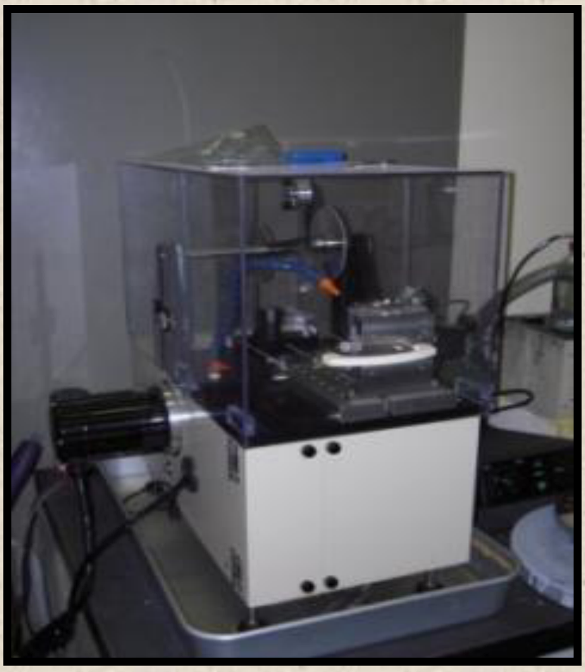
Sample Preparation

Fine cutting machine
HeiwaTechnica, HS-25A

Glove box

Automatic lapping polishing machine

Electric balance
Shimadzu



Diamond Wire Cutting Machine
New Metals & Chemical corporation

Measurement & Analysis Equipment

Nomarski-Type Differential Interference Contrast Microscope

NIKON, Eclipse ME600,TYPE 120
Light Source: White light
DIC Microscope System
Magnification: ×1000

Polarized microscope

NIKON, Eclipse, E600POL
Light Source: White Light
DIC Microscope System
Magnification: ×500

NIKON, SMZ-U
Light Source: Halogen lamp

Horiba, EDX, EMAX X-act

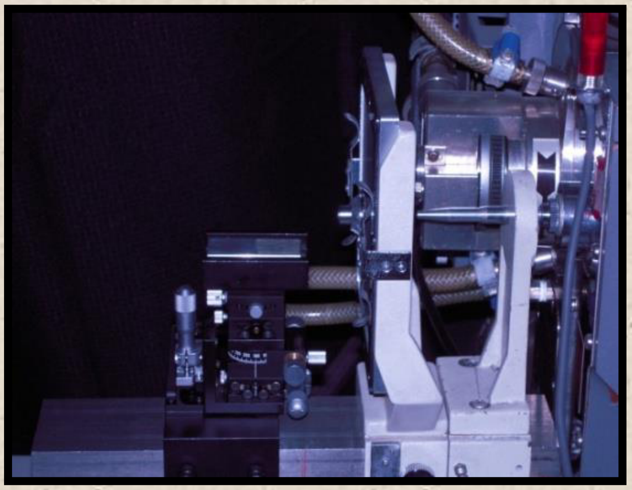
Analyzer & Mapping & Point and ID & Broad Area Automatic Analysis

SEM, Hitachi S-3400N

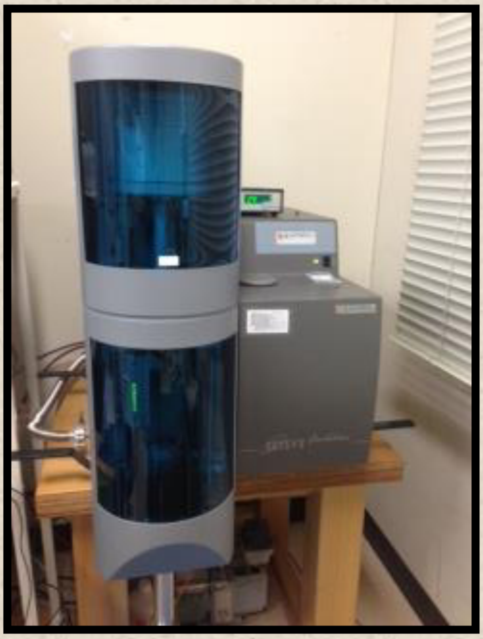
Thin Film HRXRD
RIGAKU

Powder XRD
RIGAKU

Rotary Type XRD
RIGAKU



Laue Camera
Rigaku
R-AXIS DS3



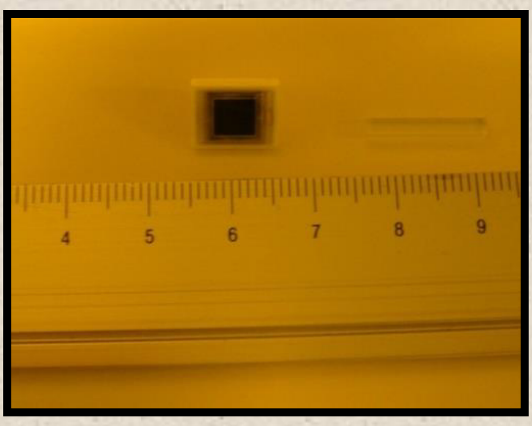
TG-DTA
SETARAM

Piezoelectric Constant
Measurement System

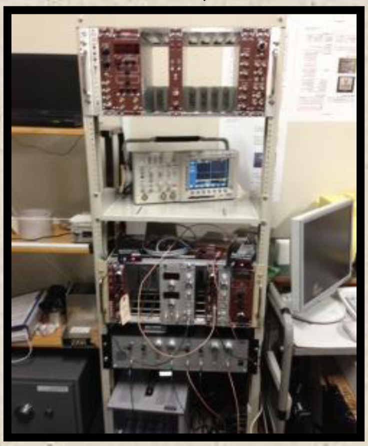
IP Reader
Rigaku R-AXIS DS3C

Absorption and fluorescence spectrometer

UV-VIS spectrophotometer
Shimadzu,UV-2550



Si Avalanche Photodiode



Radiation measurement modules
(Light yield, Scintillation decay time)

Edinburg Instruments Spectrometer: Photoluminescence & Decay time

Furnaces & Ovens

Drying oven

AS ONE, DO-450-V

Temperature: 100º C

YAMATO, FO200

DENKEN, KDF007F

Temperature: 1100℃

Anneal furnace

Nisshin-giken,

Temperature: ~1600℃

SiC furnace

Motoyama Super-C

Max temperature: ~1450ºC

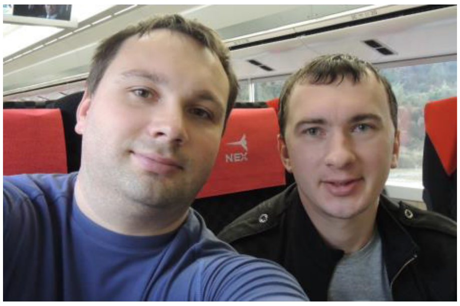
High Temperature Furnace
V&H Technology

Muffle Furnaces
ISUZU, STR-12K

Tube Furnace
ALPHA

Messages from foreign visitors

Message for 2013 annual report
from Iaroslav Gerasymov and Evgeny Galenin (ISMA NANU, Kharkov, Ukraine)



We had met Prof. Yoshikawa at the conference on Advanced Scintillation Materials in September 2013 in Kharkov (Ukraine). At that time, he invited us to visit his laboratory at Tohoku University in Sendai for joint experiments on gadolinium pyrosilicate crystal growth by the μ-PD method. Almost immediately after that, we had started preparations for the visit with great assistance of Hiroshi Uemura, who made excellent job to complete all the formalities in time.

We had departed from Kharkov on December 8, when our city was full of snow, and the next day we arrived to spring-like warm Sendai. The first colleague we met in Sendai was Dr. Valery Chani, who escorted us to the lab. We were very happy to meet with the members of the laboratory and to be introduced to their research. At first day in IMR, we were also lucky to participate at the research meeting of Yoshikawa Lab and to discuss there various aspects of the laboratory activities, including luminescent properties of mixed garnets and fabrication of novel functional materials.

During the rest of the week, we participated in about ten crystal growth experiments with assistance and under guidance of Prof. Kamada. His introduction to principles of GPS crystal growth by the μ-PD was priceless. He was also very helpful explaining us the role of the μ-PD method in selection of optimal composition of the crystals considering their crystalline quality and physical properties.

Additional pleasant surprise for us was a welcome party that was held in honor of our visit. This informal event accompanied with delicious plum wine and a bowl of soup from Prof. Kamada should be also considered as contribution to Ukrainian-Japanese cooperation.

From our point of view, the great success of the Yoshikawa Lab is based on high professionalism of its leader, a great friendly team of highly qualified enthusiasts, and excellent growth and characterization equipment. As a result, fundamental and practical achievements of this research group are well recognized by the world scientific community.

Before leaving Sendai, we discovered that the city was covered with snow. This reminded us snowy winter in Ukraine. After departure from Narita airport, we shared our impressions about visiting Japan and enjoyed view of beautiful Fuji mountain from the window of the airplane.

Travel to Japan and visit the Yoshikawa Laboratory gave us the opportunity to meet new people and new culture. That was an unforgettable experience.

Looking forward to further cooperation,
Iaroslav Gerasymov and Evgeny Galenin.

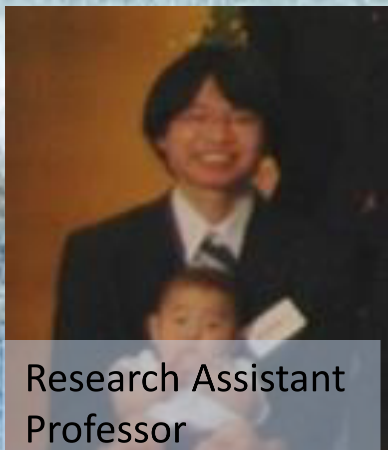
Members

Members of Yoshikawa Lab.

Professor
A. Yoshikawa

Associate Professor
Y. Yokota

Associate Professor
K. Kamada

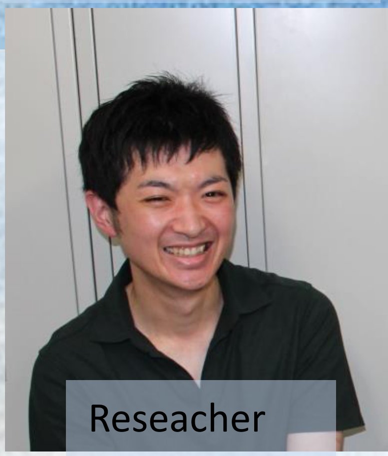


Research Assistant Professor
S. Kurosawa

Research Assistant Professor
J. Pejchal

Technical Counselor
H. Uemura

Reseacher
Y. Shoji



Reseacher
H. Nagato

Reseacher
S. Hayasaka

Secretary
M. Sasaki

Secretary
K. Toguchi

Secretary
Y. Takeda

Secretary
Y. Saijo

Second grade Doctoral course
A. Yamaji

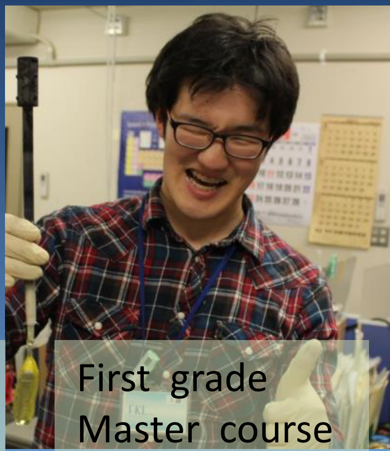
Second grade Master course
S. Suzuki

Second grade Master course
M. Seki

Second grade Master course
K. Nishimoto

Second grade Master course
A. Suzuki

First grade Master course
K. Hishinuma



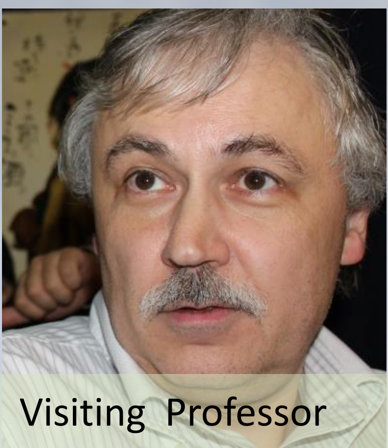
First grade Master course
T. Kudo

Reseacher
M. Satoh

Reseacher
T. Nihei

Visiting Professor
Prof. M. Nikl

Visiting Professor
Prof. G. Boulon



Visiting Professor
Prof.
V. V. Kochurikhin

Visiting Resarcher
Dr. V. Chani

Visiting Resarcher
Dr. A. Medvedev

２０１３年度　吉川研究室　Members
（２０１３ Academic year）

Professor		***教授***	
Prof.	Akira Yoshikawa	吉川　彰	NICHe 兼任
Associate Professors		***准教授***	
Prof.	Yuui Yokota	横田　有為	NICHe
Prof.	Kei Kamada	鎌田　圭	NICHe
Research Assistant Professors		***助教***	
Dr.	Shunsuke Kurosawa	黒澤　俊介	NICHe 兼任
Dr.	Jan Pejchal	ヤン　ペジャール 金研	NICHe
Researchers		***研究員***	
	Yasuhiro Shoji	庄子　育宏	
Dr.	Valery Chani	ヴァレリー　チャニ	
	Hisakazu Nagato	長門　久和	
	Shoki Hayasaka	早坂　将輝	
Advisors		***研究顧問***	
Dr.	Touetsu Shishido	宍戸　統悦	
Dr.	Masae Kikuchi	菊地　昌枝	東北福祉大学　特任教授
Technical Counselor		***技術参事***	
	Hiroshi Uemura	上村　博	
Secretaries		***秘書***	
	Megumi Sasaki	佐々木　愛美	
	Keiko Toguchi	戸口　景子	
	Yuka Takeda	武田　悠佳	2013.11.月から
	Yumiko Saijo	西条　由美子	2013.6 月まで

Students		*大学院生*	*Grade*
	Akihiro Yamaji	山路　晃広	D2
	Kei Nishimoto	西本　けい	M2
	Mafuyu Seki	関　真冬	M2
	Akira Suzuki	鈴木　彬	M2
	Shotaro Suzuki	鈴木　祥太朗	M2
	Kosuke Hishinuma	菱沼　康介	M1
	Tetsuo Kudo	工藤　哲男	M1

Researchers (Company)		*民間共同研究員*	
Dr.	Andrey Medvedev	アンドレイ　メドベージェフ	Fomos-Materials
Dr.	Kentaro Fukuda	福田　健太郎	株式会社トクヤマ
	Kou Onodera	小野寺　晃	TDK 株式会社
	Masato Satoh	佐藤　真人	TDK 株式会社
	Takayuki Nihei	二瓶　貴之	TDK 株式会社
	Tatsuya Iwasaki	岩崎　達哉	株式会社キヤノン
	Ryota Ohashi	大橋　良太	株式会社キヤノン

Visiting Professors		*短期滞在：客員教授・客員研究員*	
Prof.	Gorges Boulon	ジョージ　ブーロン	France
Prof.	Martin Nikl	マーチン　ニクル	Czech Republic
Prof.	Vladimir V. Kochurikhin	ヴラディミール　カチューリッヒン	Russian Fed.
Prof.	Christophe Dujardin	クリストフ　ドゥジャルダン	France

Research life

Research Life

Tetsuo Kudo

During my first year in Yoshikawa laboratory, I have been studying crystal growth and evaluation of piezoelectric materials. I found the research life in this laboratory very exciting.

Crystal growth has been a big challenge for me. In the beginning my research, I could not make good crystal to evaluate. However, thanks a useful advice from many researchers in Yoshikawa laboratory, I was successful in growing a crystal. First time after success in growing large bulk crystal by Czochralski method, I was so excited and I wanted to research so much about crystal growth.

For this year, I had many opportunities to study about crystals. For example, I went Poland and Russia to participate in international conference “ICCGE-17”. In the conference, I had discussed about my research and listened to interesting presentations.

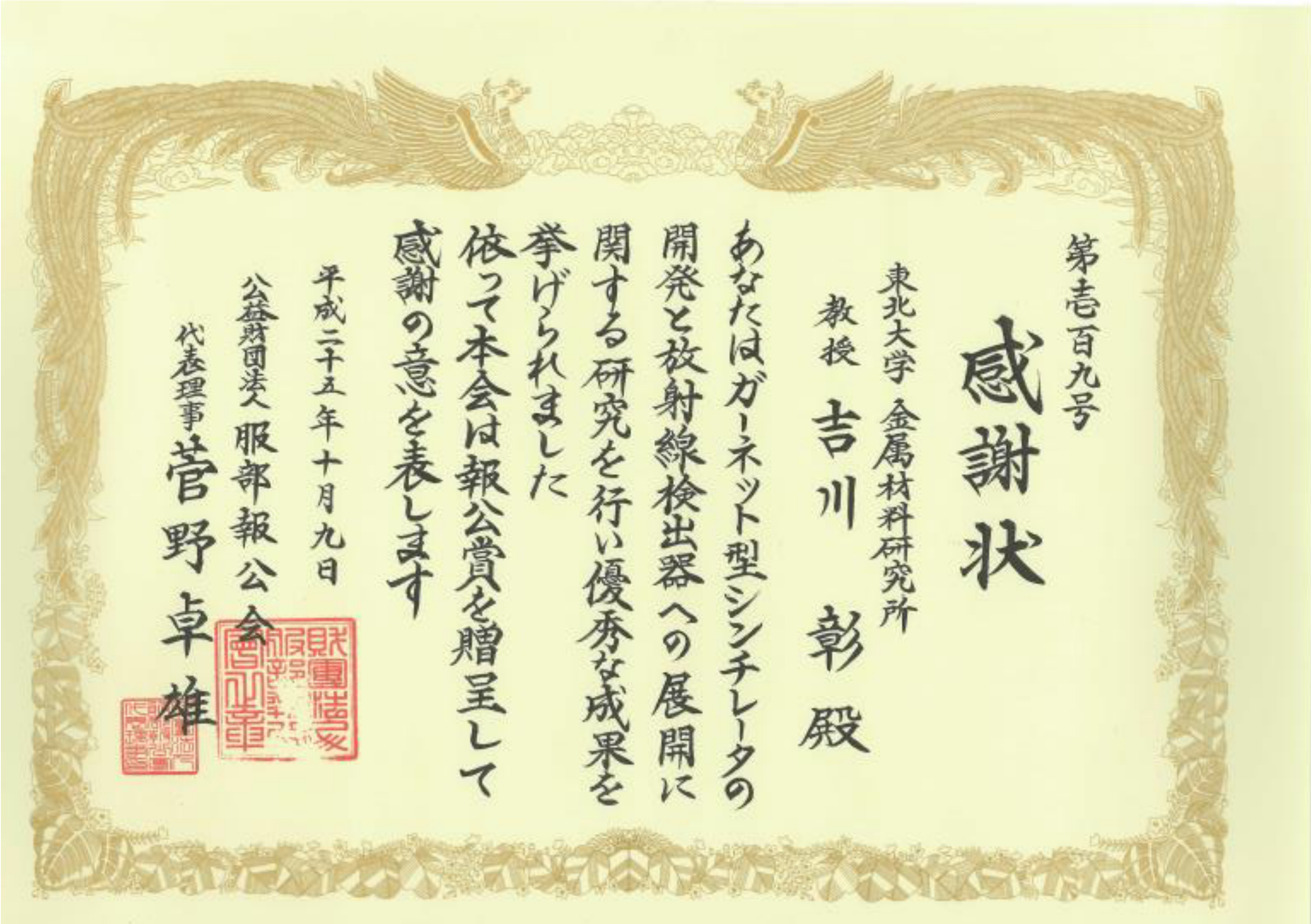
Next year, I will be 2nd grade master course student. For accomplishment to write master thesis, I am going to research much more than the last year and learn many things in Yoshikawa laboratory.

Kousuke Hishinuma

One year has passed since I came to Yoshikawa Laboratory. There are many respectable people in this lab and they are living a busy life. At first, I had never heard the word “scintillator” until I come this lab. That was a new material for me and strongly attracted my attention.

I am studying directionally solidified eutectic scintillators. The motivation of the study is to develop materials for high resolution radiation imaging taking the advantage of the light-guiding ability caused by eutectic micro-structure. I feel that is very difficult but challenging. I will explore the promising eutectic materials. Thank you for your reading.

Prizes and awards



第壱百九号

感謝状

東北大学 金属材料研究所
教授 吉川 彰 殿

あなたはガーネット型シンチレータの
開発と放射線検出器への展開に
関する研究を行い優秀な成果を
挙げられました
依って本会は報公賞を贈呈して
感謝の意を表します

平成二十五年十月九日

公益財団法人 服部報公会
代表理事 菅野卓雄

List of research collaborations

Visits by International Collaborator 2013

Affiliation	Researcher	Research Theme
General Physics Institute (Russia)	Dr. V. Kochurikhin	Growth of bulk single crystals and automatic diameter control of Czochralski growth
General Physics Institute (Russia)	Dr. M. Ivanov Mrs. L. Gushchina	Growth of bulk single crystals
Fomos Materials (Russia)	Dr. A. Medvedev	Langasite-type piezoelectric single crystals and their piezoelectric properties
LPCML, CB Lyon 1 Univ. (France)	Pr. G. Boulon	Ceramic laser materials with a nonpress vacuum sintering method
LPCML, CB Lyon 1 Univ. (France)	Pr. G. Boulon	Ceramic scintillator materials with a nonpress vacuum sintering method
Svetcha (Canada)	Dr. V. Chani	Growth of bulk oxide single crystals
Institute of Physics ASCR (Czech Republic)	Dr. M. Nikl	Characterization of various scintillator materials
Institute for Scintillation Materials (Ukraine)	Mr. I. Gerasymov Mr. E. Galenin	Growth of bulk halide scintillator crystals

Seminars at Yoshikawa Laboratory 2013

Date	Affiliation	Speaker	Title of speech
June 5 –July 24	Institute of Physics ASCR (Czech Republic)	Dr. M. Nikl	Prof. Nikl's Seminar (total 5 times) Luminescent materials and Optical spectroscopy of inorganic solids
Mar. 6-7 2014	Institute of Physics ASCR (Czech Republic)	Dr. M. Nikl	Inorganic scintillation materials

Visits to International Collaborator 2013

Affiliation	Researcher	Period of stay
Scintillation Materials RC The University of Tennessee, (USA)	Assoc. Prof. Kamada Mr. Shoji Mr. Yamaji	July 17-19
Siemens Corporation, Tennessee (USA)	Assoc. Prof. Kamada Mr. Shoji Mr. Yamaji	July 17-19
Institute of Physics ASCR (Czech Republic)	Prof. Yoshikawa Assoc. Prof. Yokota	July 21-26
Institute of Physics ASCR (Czech Republic)	Assist. Prof. Pejchal	August 8-26
General Physics Institute (Russia)	Prof. Yoshikawa Mr. Shoji Mr. Nishimoto Mr. S. Suzuki	August 8-10
Fomos Materials (Russia)	Prof. Yoshikawa Mr. Shoji Mr. Nishimoto Mr. S. Suzuki	August 8-10
TPS corporation (Korea)	Prof. Yoshikawa	August 18 - 20
Institute for Scintillation Materials (Ukraine)	Prof. Yoshikawa	September 26 - 28

Research funds

平成 25 年度　研究資金
Research funds (2013 fiscal year)

【経済産業省－東北経済産業局】

Tohoku Bureau of Economy, Trade and Industry
The Ministry of Economy, Trade and Industry

1. 戦略的基盤技術高度化支援事業（サポイン）
   Funding Program for Strategic Support Industry

   「難加工性機能性合金の形状制御結晶育成技術の開発」
   "Development of functional metallic products by shape-controlled growth techniques"

   期間　Term:　2012.1 - 2014.3
   本年度 Total:　26,929,000 yen, 2013.4 - 2014.3

2. 先端技術実証･評価設備整備費等補助金
   （企業等の実証･評価等設備の開発）
   Funding Program for the small and medium enterprise for promotion of innovation

   「無坩堝で高融点酸化物シンチレータ結晶の製造を実現するスカルメルト法溶解技術の開発」
   "Development of Skull Melt method to produce high-melting point oxide scintillator crystals without crucible"

   株式会社ｼﾞｰ･ｲｰ・ｴｽからの再委託研究
   Truster: GES Co.Ltd

   期間　Term:　2014.3 - 2014.12
   本年度 Total:　15,000,000 yen for our team, 2014.3 - 2014.12

【JST プロジェクト】

Japan Science and Technology Agency

1. JST研究成果展開事業 【先端計測分析技術・機器開発】
Development of Systems and Technology for Advanced Measurement and Analysis Technology

「無人ヘリ搭載用散乱エネルギー認識型高位置分解能ガンマカメラの実用化開発」
"Research and development of high position resolution gamma camera borne by an unmanned helicopter employing photon scattering for radiation survey"

古河機械金属株式会社からの再委託研究
Truster: Furukawa Co. Ltd.
期間　Term:　2012.4 - 2015.3
本年度 Total:　21,250,000 yen for our team, 2013.4 - 2014.3

2. JST研究成果最適展開支援プログラム （A-STEP）シーズ育成タイプ
Adaptable and seamless technology transfer program through target-driven R&D

「核物質セキュリティ用^{3}He 代替中性子計測装置の開発」
"Development of neutron scanning apparatus for homeland security using scintillator instead of ^{3}He gas detector "
期間　Term:　2012.10 – 2015.3
本年度 Total:　14,500,000 yen for our team, 2013.4 - 2014.3

3. JST研究成果展開事業 【先端計測分析技術・機器開発】
Development of Systems and Technology for Advanced Measurement and Analysis Technology

「高エネルギー分解能・高スループットの国産放射測定検査装置」
"Development of radiometry tester with high energy resolution and high efficiency"

株式会社千代田テクノルからの再委託研究
Truster: Chiyoda Technol Corporation

期間　Term:　2013.10 - 2016.3
本年度 Total:　29,900,000 yen for our team, 2013.10 - 2014.3

4. JST研究成果展開事業
（研究成果最適展開支援プログラム（A-STEP）
実用化挑戦ステージ実用化挑戦タイプ　（中小・ベンチャー開発）

Foundation for Small and Medium Enterprise Promotion
(Supporting small and medium-sized and the venture company of the research and development.)

「高温域で劣化しない資源探査用シンチレーター」
"Scintillator for Resources Exploration Equipment with high performance at high temperature"

株式会社Ｃ＆Ａからの再委託研究
Truster: C&A Corporarion.
期間　Term:　2013.12 - 2017.3
本年度 Total:　7,000,000 yen for our team, 2013.12 - 2014.3

5. JST研究成果展開事業
FS 探索タイプ
FS stage

「低コストPET用アレイカメラを実現する高エネルギー分解能を有するパイロシリケート型ピクセルシンチレータの開発」
"Development of pyrosilicate type pixel scintillator with a high energy resolution allowing fabrication of low cost array camera for PET application"

期間　Term:　2013.4 - 2014.3
本年度 Total:　1,300,000 yen for our team, 2013.4 - 2014.3

「次世代周波数資源の活用に向けた光ファイバー一体型高効率 THz 光機能素子の開発」

“Development of next-generation optical fiber-integrated THz devices”

期間　 Term:　2013.4 - 2014.3
本年度 Total:　650,000 yen for our team, 2013.4 - 2014.3

6. JST復興促進プログラム（A-STEP）

Funding Program for Revitalization Promotion

「燃焼圧センサ用ランガサイト型圧電結晶の形状制御結晶育成用坩堝の開発」
横田 有為 (Yuui Yokota)

「環境モニタ・医療用カメラへの応用をめざした高阻止能・高エネルギー分解能を持つシンチレータの開発」
黒澤　俊介（Shunsuke Kurosawa）

「ナノ秒以下の時間分解能をもつシンチレータの開発と医療への応用」
（Jan PEJCHAL）

期間　Term:　2012.4 - 2014.3
本年度 Total:　600,000 yen for our team, 2013.12 - 2014.3

【NEDO プロジェクト】

New Energy and Industrial Technology Development Organization

1．平成２５年度課題設定型産業技術開発費助成金
（希少金属代替・低減技術実用化開発助成事業）

Rare Metal Substituted Materials Development Project, 2013

ＴＤＫ株式会社からの再委託研究
Truster: TDK Corporation
期間　Term: 2013.10 - 2015.3
本年度　Total: 20,000,000 yen for our team, 2013.10 - 2014.3

【復興庁】　Reconstruction Agency

1．平成２４年度地域イノベーション戦略支援プログラム
Funding Program for "Invest Japan" promotion for reconstruction

宮城県インテリジェントコスモス研究機構からの再委託プログラム
Truster: Intelligent Cosmos Research Institute, Miyagi pref.

「次世代自動車のための人材育成プログラム」
"Manpower training program for innovation in automotive industry"

期間　Term:　2012.4 - 2017.3
本年度　Total:　10,600,000 yen for our team, 2013.4 - 2014.3

【厚生労働省科学研究費補助金】

Ministry of Health, Labour and Welfare
科学研究費助成　Health Science Research Grants

「非侵襲血中 RI 濃度測定を可能にするウエアラブル・サブミリ解像度 PET 装置の開発」
“Development of wearable and sub-millimeter resolution type PET apparatus that enables non-invasive measurement of RI concentration in blood”

東京大学からの再委託研究
Truster: Tokyo University
期間　Term:　2013.4 - 2016.3
本年度　Total:　20,000,000 yen, 2013.4 - 2014.3

【文部科学省科学研究費補助金】

Ministry of Education, Culture, Sports, Science and Technology

1. 日本学術振興会　Japan Society for the Promotion of Science
   科学研究費助成　Grants-in-Aid for Scientific Research

1-1.　若手研究（B）Grants-in-Aid for young scientists (B)
横田　有為　(Yuui Yokota)
600,000 yen, 2013.4-2014.3

1-2　萌芽　Grants-in-Aid for young scientists (Sprout)
吉川　彰　(Akira Yoshikawa)
1,600,000 yen, 2013.4-2014.3

1-3　特別研究員　Fellowship
山路　晃広　(Akihiro Yamaji)
1,000,000 yen, 2013.4-2014.3

【国立大学法人　事業化推進事業型研究】

事業化推進型研究補助金
Foundation for Business Incubation Program

期間　Term:　2014.2 - 2015.1
本年度 Total:　50,000,000 yen,　2014.2 - 2015.1

【企業・財団・個人からの受託・共同研究，寄付金および小型プロジェクト】

Funds from industry, foundations, personal donation and small projects

1. 株式会社トクヤマ　Tokuyama Corporation

2. TDK 株式会社　TDK Corporation

3. 株式会社Ｃ＆Ａ　C&A Corporation

4. キヤノン株式会社　Canon Inc.

5. 浜松ホトニクス株式会社　Hamamatsu Photonics K.K

6. TANAKA ホールディングス株式会社　TANAKA HOLDINGS Co. Ltd.

7. 大阪大学レーザーエネルギー学研究センター　共同研究
   Collaboration program with Institute of Laser Engineering, Osaka Univ.
   「真空紫外域に発光する新規発光結晶の開発」

平成 25 年度申請特許

List of patents

1. 結晶の製造方法

   吉川　彰、横田　有為、黒澤　俊介、鈴木　祥太郎、福田　健太郎、石津　澄人
   (Akira Yoshikawa, Yuui Yokota, Shunsuke Kurosawa, Shotaro Suzuki,
   Kentaro Fukuda, Sumito Ishizu)

2. シンチレーター結晶、放射線検出器および非破壊検査装置

   吉川　彰、黒澤　俊介、横田　有為、庄子　育宏、鎌田　圭
   (Akira Yoshikawa, Shunsuke Kurosawa, Yuui Yokota, Yasuhiro Shoji, Kei Kamada)

3. シンチレーターおよび放射線検出器

   鎌田　圭、吉川　彰、横田　有為、黒澤　俊介、菱沼　康介
   (Kei Kamada, Akira Yoshikawa, Yuui Yokota, Shunsuke Kurosawa, Kosuke Hishinuma)

他　外国出願　２　件

学会、学内における役員・委員等
Committees of academic societies and conferences

吉川　彰
Dr. Akira YOSHIKAWA, Professor

Optical Materials（Elsevier）	Associate Editor
Radiation Measurement（Elsevier）	Guest Editor
International Association on Inorganic Scintillators and their Applications (SCINT)	International Advisory Committee
International Conference on Luminescent Detectors and Transformers of Ionizing Radiation (LUMDETR)	Scientific Advisory Committee
日本結晶成長学会 Japanese Association for Crystal Growth Cooperation	理事（新技術・新材料分科会担当） Trustee (responsible for new technology and new materials branch)
	編集委員 Member of the editorial staff
日本フラックス研究会 Japanese Association for Flux Growth	常任理事 Trustee
日本学術振興会第 186 委員会 No. 186 committee, Japan Society for the Promotion of Science	代表幹事 Chief secretary
日本学術振興会第 161 委員会 No. 161 committee, Japan Society for the Promotion of Science	運営委員 Manager

2013 年度（平成 25 年度）吉川研究室行事

月	吉川研究室	学会	研究会・講演会
4		4/15-19　SCINT2013(China) 4/26-28　REMAT 2013(Poland)	
5		5/20-24 第 30 回強誘電体応用会議（京都）	5/20 学振１８６委員会(金沢) 5/10 学振１６１委員会(仙台)
6		6/30-7/4　CLEO-PR2013(京都)	
7		7/19-20　STAC-7（横浜） 7/21-26　ACCGE-19 (USA) 7/21-25　ISAF(Czech)	7/5-6 学振１８６委員会(札幌) 7/19 学振１６１委員会(東京)
8		8/11-16　ICCGE-17(Poland)	
9		9/16-20 第 74 回応用物理学会秋季学術講演会（京都） 9/20-23 日本物理学会 2013 年秋季大会(高知) 9/24-27 SSDM2013(福岡)	9/12　学振１８６委員会(福島)
10		10/20-23　ISLNOM-6(China) 10/27-11/2　2013 IEEE(Seoul)	10/11 学振１６１委員会(東京)
11		11/30-12/1　IWIRM9 (大洗)	11/6　学振１８６委員会(千葉) 11/6-8 第 43 回結晶成長国内会議（長野）
12	12/17 忘年会		12/6 第 8 回日本フラックス成長研究発表会（東京） 12/13-14 第 24 回光物性研究会（大阪） 12/13 学振１６１委員会(伊豆)
1			1/28-30 研究会「放射線検出器とその応用」(つくば) 1/30-31　学振１８６委員会(東京)
2			
3	3/6-7 研究室シンポジウム＆スキー旅行(秋田)	3/17-20 応用物理学関連講演会(神奈川)	3/7-8 学振１６１委員会(東京)

List of abstracts

APPLIED PHYSICS LETTERS 102, 161907, Published: APR2013

Scintillation characteristics of $LiCaAlF_6$-based single crystals under X-ray excitation

M. Nikl[1], P. Bruza[2], D. Panek[2], M. Vrbova[2], E. Mihokova[1], J. A. Mares[1], A. Beitlerova[1], N. Kawaguchi[3,4], K. Fukuda[3,4], A. Yoshikawa[3,5]

1. Institute of Physics AS CR, Cukrovarnicka 10, 16200 Prague 6, Czech Republic
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3. Tohoku Univ, IMR, 2-1-1 Katahira, Aoba Ku, Sendai, Miyagi 9808577, Japan
4. Kasumigaseki Common Gate West Tower 2-1, Kasumigaseki 3-chome, Chiyoda-ku, Tokyo 100-8983, Japan
5. Tohoku Univ, NICHe, Aramaki, Aoba Ku, Sendai, Miyagi 9808579, Japan

Abstract:
$LiCaAlF_6$-based scintillators are studied under X- and soft gamma-ray excitations. Under nanosecond pulsed soft X-ray laser excitation the scintillation decay is measured with extremely high dynamical resolution and broad time scale. The undoped $LiCaAlF_6$ shows complex temperature dependence of exciton luminescence and tunneling-driven energy transfer process in scintillation decay. In both the Ce and Eu-doped $LiCaAlF_6$ the dominant part of measured scintillation decay is due to prompt recombination of electrons and holes at the doped emission centers. Nevertheless, the measured light yield value is considerably lower with respect to the derived upper limits. Possible origin of its deterioration is discussed.

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http://scitation.aip.org/content/aip/journal/apl/102/16/10.1063/1.4803047

FERROELECTRICS 449, 147, Published: SEP2013

Effects of growth atmosphere on crystal growth and optical properties for $Ca_3NbGa_3Si_2O_{14}$ single crystals

Y. Yokota[1], M. Sato[2], K. Tota[2], S. Kurosawa[1], K. Onodera[2], A. Yoshikawa[1,3]

1. Tohoku Univ, IMR, 2-1-1 Katahira, Aoba Ku, Sendai, Miyagi 9808577, Japan
2. TDK corporation, 1-1 Okinota, Kisakata, Nikaho 0180193, Japan
3. Tohoku Univ, NICHe, Aramaki, Aoba Ku, Sendai, Miyagi 9808579, Japan

Abstract:
$Ca_3NbGa_3Si_2O_{14}$ (CNGS) single crystals were grown by the micro-pulling-down (μ-PD) method under various atmosphere, Air, Ar + 3%O_2 and Ar 100%, and the effects of growth atmosphere on crystal growth and optical properties were investigated. While the CNGS crystal grown in Air indicated orange color, the crystal grown in Ar was colorless. CNGS crystal grown in Air indicated several large absorption peaks around 360 and 500 nm in the transmittance spectrum. In contrast, the crystal grown in Ar didn't indicate these absorption peaks.

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http://www.tandfonline.com/doi/abs/10.1080/00150193.2013.822780

RADIATION MEASUREMENTS 55, 132, Published: AUG2013

Eu and Rb co-doped $LiCaAlF_6$ scintillators for neutron detection

A. Yamaji[1], T. Yanagida[2], N. Kawaguchi[3], Y. Yokota[1], Y. Fujimoto[1], S. Kurosawa[1], J. Pejchal[1], K. Watanabe[4], A. Yamazaki[4], A. Yoshikawa[1,2]

1. Institute of Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Japan
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3. Tokuyama Corporation, Shibuya 3-chome, Shibuya, Japan
4. Department of Material, Physics and Energy Engineering, Nagoya University, Japan

Abstract:
Eu and Rb co-doped $LiCaAlF_6$ (LiCAF) single crystals with different dopant concentrations were grown by the micro-pulling-down method for neutron detection. Their transmittance spectra showed strong absorption bands at 200-220 and 290-350 nm, and under ^{241}Am alpha-ray excitation, their radioluminescence spectra exhibited an intense emission peak at 373 nm that was attributed to the Eu^{2+} 5d-4f transition. These results were consistent with those for the Rb-free Eu:LiCAF. The highest light yield among the grown crystals was 36,000 ph/n, which was 20% greater than that of the Rb-free crystal. In addition, the neutron-excited scintillation decay times were 650-750 ns slower than that of the Rb-free Eu:LiCAF.

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http://www.sciencedirect.com/science/article/pii/S1350448713002151

IEEE TRANSACTIONS ON NUCLEAR SCIENCE 61, 419, Published: NOV2013

Growth and Scintillation Properties of Ce:Li(Ca,Ba)AlF_6 Scintillator Crystals

Y. Yokota[1], S. Kurosawa[1,2], K. Fukuda[3], K. Kamada[2,4], A. Yoshikawa[1,2,4]

1. New Industry Creation Hatchery Center (NICHe), Tohoku University, 6-6-10 Aramaki, Aoba-ku, Sendai, Japan
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3. Tokuyama Corporation, Shibuya 3-chome, Shibuya, Japan
4. C&A Corporation, Sendai 980-8576, Japan

Abstract:
Ba co-doped Ce:$LiCaAlF_6$ [Ce:Li(Ca,Ba)AlF_6] and $LiCaAlF_6$ [Li(Ca,Ba)AlF_6] crystals with various Ba concentrations were grown and their structures, optical and scintillation properties were investigated. As-grown Ce2%Ba1% and Ce2%Ba2%:Li(Ca,Ba)AlF_6 crystals were high transparency in all parts while an end part of as-grown Ce2%Ba5%:Li(Ca,Ba)AlF_6 crystal and all parts of as-grown Ba5%:Li(Ca,Ba)AlF_6 crystal included milky parts which decreased transmittance. Ce:Li(Ca,Ba)AlF_6 crystals indicated the emission peaks at 288 and 308 nm from which was attributable to the 5d-4f transition of Ce ion in the radioluminescence spectra under -ray irradiation. In the case of Ce1%Ba2%:Li(Ca,Ba)AlF_6 crystal, light yield under thermal neutron irradiation were improved. Decay times of Ce:Ce:Li(Ca,Ba)AlF_6 under thermal neutron irradiation systematically increased with an increase of Ba concentration.

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http://ieeexplore.ieee.org/xpl/articleDetails.jsp?arnumber=6658909

NUCLEAR INSTRUMENTS AND METHODS IN PHYSICS RESEARCH A 732, 599, Published: DEC2013

Development of a single crystal with a high index of refraction

S. Kurosawa[1,2], V. V. Kochurikhin[3], A.Yamaji[1], Y. Yokota[2], H. Kubo[4], T. Tanimori[4], A. Yoshikawa[1,2]

1. Institute of Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Japan
2. New Industry Creation Hatchery Center (NICHe), Tohoku University, 6-6-10 Aramaki, Aoba-ku, Sendai, Japan
3. General Physics Institute, Moscow, Russia
4. Department of Physics, Kyoto University, Kyoto, Japan

Abstract:
Time-of-flight Positron emission tomography (TOF-PET) is one of the next-generation medical imaging methods, which requires scintillators with a very short decay time. However, the shortest scintillation decay times are typically 20–30 ns, and these values are not sufficient for TOF-PET. Cherenkov counters are used in high energy physics and they are expected to be applied in medical imaging due to their short decay time. Here, high-refractive index materials are necessary for Cherenkov radiators to reach a high light output. We measured refractive indices of $Gd_3Ga_5O_{12}$ (GGG), $Y_3Ga_5O_{12}$ (YGG) and $Lu_3Ga_5O_{12}$ (LuGG) crystals grown by a micro-pulling-down μPD method. The GGG, YGG and LuGG crystals were found to have refractive indices of 2.5, 2.3 and 2.3 at 400nm, respectively. Then we grew a 40mm diameter GGG crystal by the Czochralski method, and the emission decay times of the GGG crystals irradiated with muons and gamma rays were 1071 ns and 1072 ns, respectively, using a photo- multiplier tube (HamamatsuR6231-100). Cherenkov light of the GGG crystal could be observed for the gamma-ray irradiation.

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http://www.sciencedirect.com/science/article/pii/S0168900213011443

IEEE TRANSACTIONS ON NUCLEAR SCIENCE 61, 293, Published: FEB2013

Czochralski growth and scintillation properties of Ce:$(Gd,Y,Lu)_3(Al,Ga)_5O_{12}$ single crystals

K. Kamada[1,2], P. Prusa[3], M. Nikl[3], K. Blazek[3], T. Endo[4], K. Tsutsumi[4], S. Kurosawa[5], Y. Yokota[1], A. Yoshikawa[1,2,5]

1. New Industry Creation Hatchery Center (NICHe), Tohoku University, 6-6-10 Aramaki, Aoba-ku, Sendai, Japan
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3. Institute of Physics AS CR, Cukrovarnicka 10, 16200 Prague 6, Czech Republic
4. Materials Research Laboratory, Furukawa Co. Ltd., Tsukuba, 305-0856, Japan
5. Institute of Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Japan

Abstract:
1-inch size Ce1%:$Gd_2Lu_1Al_2Ga_3O_{12}$, $Gd_1Lu_2Al_2Ga_3O_{12}$, $Gd_1Y_2Al_{1.5}Ga_{3.5}O_{12}$ and $Lu_2Y_1Al_2Ga_3O_{12}$ were grown by the Czhocralski (Cz) method. The EPMA techniques is employed to check their chemical composition. Luminescence and scintillation properties were also evaluated. The Ce1%: $Gd_1Y_2Al_2Ga_3O_{12}$ sample showed the highest light yield of around 40 000 photon/MeV. The scintillation decay time was 46.6 ns (63%) and 157 ns (37%).

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IEEE TRANSACTIONS ON NUCLEAR SCIENCE 61, 411, Published: FEB2014

Luminescence and Scintillation Properties of Scintillators Based on Orthorhombic and Monoclinic $BaLu_2F_8$ Single Crystals

J. Pejchal[1,2], K. Fukuda[3], S. Kurosawa[1,4], Y. Yokota[1], R. Kral[2], M. Nikl[2], A. Yoshikawa[1,4]

1. Institute of Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Japan
2. Institute of Physics AS CR, Cukrovarnicka 10, 16200 Prague 6, Czech Republic
3. Tokuyama Corporation, Shibuya 3-chome, Shibuya, Japan
4. New Industry Creation Hatchery Center (NICHe), Tohoku University, 6-6-10 Aramaki, Aoba-ku, Sendai, Japan

Abstract:
The rare-earth-doped orthorhombic $BaLu_2F_8$ vacuum-ultra-violet scintillator crystals have been studied. The fast emission around 185 nm with a decay time of several nanoseconds was due to the allowed 5d-4f transition of the Nd^{3+} ion. The high temperature phase $BaLu_2F_8$ orthorhombic crystals have been prepared by micro-pulling-down method. Unfortunately, no 5d-4f emission (neither from Nd^{3+} nor from Tm^{3+}) was observed in the crystals under excitation by ionizing radiation at all. This was explained by preferential energy transfer from the host to the lattice defect states. Further improvement of the scintillation efficiency by facilitating the energy transfer from the host matrix to the Nd^{3+} luminescence center by Tm^{3+}-codoping was attempted. The energy transfer from the Tm^{3+} ions to the Nd^{3+} ones has been proved, however, no improvement of the overall scintillation efficiency was observed.

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Neutron detection with $LiCaAlF_6$ scintillator doped with 3d-transition metal ions

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Abstract:
Capability of thermal neutron detection was examined for $LiCaAlF_6$ (LiCAF) scintillators doped with 3d-transition metal ions. Their radioluminescence spectra were measured with an ^{241}Am source to simulate ^{6}Li(n, α)^{3}H reaction. The sufficiently intense radioluminescence was observed for the Mn, Co and Cu dopants, while only a weak one was observed for Ti, V, Fe and Ni. A Mn doped LiCAF crystal, which showed the highest radioluminescence intensity, was coupled with a Si avalanche photodiode for the examination of its neutron response. It was confirmed that the average current of the photodiode clearly increased under excitation with 13.5 meV neutron flux.

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Structural and optical properties of neodymium-doped lutetium fluoride thin films grown by pulsed laser deposition

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Abstract:
Neodymium-doped lutetium fluoride (Nd^{3+}:LuF_3) thin films were successfully grown on MgF_2 (0 0 1) substrates by pulsed laser deposition (PLD). It is void of cracks that are otherwise prevalent due to structural phase transitions in Nd^{3+}:LuF_3 during thin film deposition and bulk crystal growth. Cathodoluminescence (CL) spectra revealed multiple emission peaks, with a dominant peak in the vacuum ultraviolet (VUV) region at 179 nm. This peak has a decay time of 6.7 ns. The ability to grow high quality Nd^{3+}-doped fluoride thin films would enable fabrication of VUV light-emitting devices that will enhance applications requiring efficient VUV light sources.

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Photo- and radio-excited luminescence properties of Eu-doped La_2O_3–Al_2O_3 based eutectics

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Abstract:
Eutectic crystal of 0.5% Eu-doped $30LaAlO_3$–$70Al_2O_3$ (vol %) was prepared by micro-pulling down (μ-PD) technique under nitrogen atmosphere. Being excited at a wavelength of 320 nm, the crystal exhibited intense emission band with a maximum at 450 nm which is corresponding to $4f^65d$-$4f^7(^8S_{7/2})$ transitions of Eu^{2+}. The decay time and fluorescence quantum efficiency (QE) were determined to be about 475 ns and 60%, respectively. When alpha-ray excited the crystal, both Eu^{2+} $4f^65d$-$4f^7(^8S_{7/2})$ and Eu^{3+} $4f^6$-$4f^6$ $(^5D_0$-$^7F_{1,2})$ emission peaks were observed at 435 nm and 600 nm. By the pulse height spectra, the relative scintillation light yield of the crystal was about 4% compared with that of BGO commercial scintillator.

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NUCLEAR INSTRUMENTS AND METHODS IN PHYSICS RESEARCH A 726, 155, Published:OCT2013

Spatial resolution of a μPIC-based neutron imaging detector

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Abstract:
We present a detailed study of the spatial resolution of our time-resolved neutron imaging detector utilizing a new neutron position reconstruction method that improves both spatial resolution and event reconstruction efficiency. Our prototype detector system, employing a micro-pattern gaseous detector known as the micro-pixel chamber (μPIC) coupled with a field-programmable-gate-array-based data acquisition system, combines 100μm-level spatial and sub-μs time resolutions with excellent gamma rejection and high data rates, making it well suited for applications in neutron radiography at high-intensity, pulsed neutron sources. From data taken at the Materials and Life Science Experimental Facility within the Japan Proton Accelerator Research Complex (J-PARC), the spatial resolution was found to be approximately Gaussian with a sigma of 103.48±0.77μm (after correcting for beam divergence). This is a significant improvement over that achievable with our previous reconstruction method (334±13μm), and compares well with conventional neutron imaging detectors and with other high-rate detectors currently under development. Further, a detector simulation indicates that a spatial resolution of less than 60μm may be possible with optimization of the gas characteristics and μPIC structure. We also present an example of imaging combined with neutron resonance absorption spectroscopy.

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NUCLEAR INSTRUMENTS AND METHODS IN PHYSICS RESEARCH A 726, 37, Published: OCT2013

Scintillation properties of Yb^{3+}-doped $YAlO_3$ in the temperature range from 4.2 to 175 K

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Abstract:
We measured the temperature dependence of the emission wavelength spectrum of YAP:Yb by irradiating with β-rays from a $^{90}Sr/^{90}Y$ source in the temperature range from 4.2 to 175 K. The light yield of YAP:Yb was characterized using an avalanche photodiode in the detection of 662-keV γ-rays from a ^{137}Cs source in the temperature range from 50 to 175 K. The light yield was found to increase with decreasing temperature and reached 3840 photons/MeV at a temperature of 50 K. By extrapolating the temperature dependence of the light yield using that of the integrated emission spectra, the experimental light yield was evaluated to be 4300 photons/MeV at a temperature of 4.2 K.

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Read Out Test of Pr:LuAG Scintillator Coupled to Organic Wavelength Shifter Using Si Based Photodetectors

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Abstract:
Functional possibilities of Pr:LuAG single crystal covered with plastic scintillators are demonstrated. Shift of luminescence wavelength of the hybrid scintillators towards the region of higher spectral sensitivity of photodetectors and radiation responses of the hybrid scintillators were investigated. The Pr:LuAG sample coated with bis-MSBPVD showed the better light output and energy resolution than the Pr:LuAG itself. Light output was increased up to 55% and energy resolution was also improved to 6.5%@662 keV using APD (Hamamatsu S8664-8220). In the case of MPPC (Hamamatsu S10362_33_050 3600 pixel-type), light output was increased up to 35% and energy resolution was also improved to 9.8%@662 keV. Decay curve of the Pr:LuAG sample coupled with the WLS was also measured and successfully modeled.

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Development of a Prototype Detector Using APD-Arrays Coupled With Pixelized Ce:GAGG Scintillator for High Resolution Radiation Imaging

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Abstract:
A novel digital PET scanner based on Time over Threshold method is developed. The positron emission tomography (PET) is composed of 144 channel Ce:$Gd_3Al_2Ga_3O_{12}$ (GAGG)-Avaranche photodiode (APD) detector arrays individually coupled with custom designed Time over Threshold (ToT) application-specific integrated circuit (ASIC) to realize the high count rate and good spatial resolution. Such an imaging system provides a simple front-end circuit and flexible digital signal processing like multiplexing such as a pulse train method. The measured energy resolution of the detector system was 6.7% for the 511 keV peak, and 4.25 ns time resolution was measured with a single detector module. The measured spatial resolution for a point source was 1.37 mm FWHM for our initial data with a columnar Na source.

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ACTA PHYSICA POLONICA A 124, 250, Published: DEC2013

Czochralski Growth and Properties of Scintillating Crystals

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Abstract:
The Czochralski method is one of the very few melt growth techniques that are industry friendly when considering the combination of quality, dimensions, and cost of the produced crystals suitable for their commercialization in scintillation detectors. This method is one of the oldest and most developed crystal growth processes regarding an adequate understanding the physical phenomena observed during solidification process and its practical expansion especially in the industrial scale production. It allows controllable formation of single-crystalline cylindrical ingots of various inorganic scintillation materials. The review summarizes recent progress on the Czochralski growth of a number of scintillation materials. The oxide crystals are mainly considered including the Ce and Pr-doped $RE_3Al_5O_{12}$, RE = Y, Lu, aluminum garnets and newly discovered ultraefficient Ce-doped $Gd_3(Ga,Al)_5O_{12}$ multicomponent garnet, high density $PbWO_4$ and $CdWO_4$ tungstates, Ce-doped RE_2SiO_5, RE = Y, Gd, Lu, oxyorthosilicates and $(Y,Lu)AlO_3$ aluminum perovskites and finally the classical $Bi_4Ge_3O_{12}$ scintillator. Additionally, the details of the growth of other practically important non-oxide crystals, namely the Ce and Eu-doped $LiCaAlF_6$ neutron and ultraefficient Ce-doped $LaBr_3$ scintillators, are discussed. The potential of novel micro-pulling down growth method is briefly described in the combinatorial search for new scintillator materials. Selected luminescence and scintillation characteristics including the spectra and decay kinetics, light yield and radiation resistance are also illustrated and overviewed.

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Scintillation Properties of a Non-Doped $Ca_3TaGa_3Si_2O_{14}$ Crystal

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Abstract:
In order to search for a new scintillation material consisting of tantalum (Ta) with a high atomic number of 73, we have investigated scintillation properties of langasite type crystal containing Ta. Non-doped $Ca_3TaGa_3Si_2O_{14}$ (CTGS), which is well known as a piezoelectric material, was grown by the Czochralski method. Radio-luminescence spectrum of this crystal peaked around 340 nm. Its light output was approximately1,200 photons/MeV. This crystal is expected to be a scintillation host material.

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JOURNAL OF CRYSTAL GROWTH 375, 51, Published: JUL2013

Preparation and characterization of pure and Pr(III)-doped lead chloride single crystals grown by modified micro-pulling- down method

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Abstract:
Single crystals of lead chloride pure and doped with Pr(III) were grown for the first time using the modified micro-pulling-down method. Due to hygroscopicity of both lead chloride and doping Pr(III) chloride the standard micro-pulling-down apparatus had to be equipped with a removable protective chamber. Prepared single crystals 25 mm long and 3 mm in diameter were characterized by powder X-ray diffraction and DSC thermal analysis. Optical and luminescence characteristics of lead chloride single crystals, such as absorption, radioluminescence, photoluminescence, and decay curves, were measured as well.

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Optical properties in Ag^+-doped phosphate glass irradiated with X-rays and α-particles

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Abstract:
The objective of this study is to investigate the emission mechanism of radiophotoluminescence (RPL) in the Ag^+-doped phosphate glass (glass dosimeter), which is now used as individual radiation dosimeter, because the emission mechanism of RPL in glass dosimeter has been not fully understood. We have investigated the assignments and characteristics of the X-ray induced colour centres in the Ag^+-doped phosphate glass up to now (Miyamoto et al., 2010). Optical properties such as optical absorption spectra related with X-ray and α-particles irradiation were measured for commercially available glass dosimeter.
In this study optical properties such as optical absorption spectrum as a function of X-rays and α-particles irradiation were measured for commercially available glass dosimeter. Comparison of the RPL in Ag^+-doped phosphate glass irradiated with X-rays and α-particles is discussed.

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Deep trapping states in cerium doped $(Lu,Y,Gd)_3(Ga,Al)_5O_{12}$ single crystal scintillators

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Abstract:
We study deep trapping states in Ce^{3+}-doped garnet crystals with the composition $(Lu,Y,Gd)_3(Ga,Al)_5O_{12}$, recently shown as having remarkably high light yield. We use thermally stimulated luminescence (TSL) technique above room temperature and determine the composition $Gd_3Ga_3Al_2O_{12}$ as the host showing the lowest concentration of traps. This host consistently manifests very low afterglow comparable to that of the standard BGO crystal. We also perform TSL glow peak analysis based on the initial rise technique to evaluate trap depth and other characteristics associated with TSL peaks.

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Light yield of (Lu, Y, Gd)$_3$Al$_2$Ga$_3$O$_{12}$:Ce garnets

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Abstract:
Two sets of Ce-doped multicomponent garnet scintillator samples were prepared using Czochralski method. Best performing sample from the first group was $Gd_3Al_2Ga_3O_{12}$ doped by 1% of Ce. Therefore, samples of $Gd_3Al_2Ga_3O_{12}$ with different thickness have been prepared for the second set. Light yield, its dependence on amplifier shaping time, energy resolution, and non-proportionality have been measured using hybrid photomultiplier. Best performing sample exhibits following parameters: light yield of 50 600 photons/MeV, energy resolution of 5.5%@662 keV, fast scintillation component intensity 91%, and good proportionality. Performance of other samples was negatively affected presumably either by ionization of $5d_1$ excited state of Ce^{3+} center or electron traps.

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OPTICAL MATERIALS 36, 214, Published: DEC2013

Growth and optical properties of RE-doped ternary rubidium lead chloride single crystals

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Abstract:
This paper reports on the growth of pure and Nd^{3+}, Pr^{3+}, Yb^{3+}, and Dy^{3+} doped rubidium lead chloride ($RbPb_2Cl_5$) crystals by the atmosphere-controlled micro-pulling-down method. Structural and composition measurements are reported, further completed by the absorption, radio- and photoluminescence spectra and decay measurements on the prepared single crystals. Potential of these materials and preparation method for the application in the infrared solid state laser field are discussed.

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Luminescence Properties of $Gd_3Ga_5O_{12}$:Cr Single Crystals

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Abstract:
We evaluated optical and scintillation properties of a 0.03% Cr-doped $Gd_3Ga_5O_{12}$ (GGG:Cr) single crystal grown by Czochralski method for medical use. Since human body has low absorption region from approximately 700-1100 nm, near infrared scintillators would be applied to the real time dose monitor system. In the X-ray excited radioluminescence, the emissions related to $^2E \rightarrow {}^4A_2$ transitions of Cr^{3+} were observed at around 725 nm. The scintillation light yield of GGG: Cr 0.03% single crystal under 5.5 MeV alpha-ray excitation was determined to be 30% of BGO.

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JAPANESE JOURNAL OF APPLIED PHYSICS 52, 062402, Published: MAY2013

Electronic States of Trivalent Praseodymium Ion Doped in $20Al(PO_3)_3$–80LiF Glass

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Abstract:
We investigate the photoluminescence (PL) and photoluminescence excitation (PLE) spectra of $20Al(PO_3)_3$–80LiF+Pr glass (APLF+Pr) and Pr^{3+}-doped $LiCaAlF_6$ crystal (Pr:LiCAF) in order to determine the electronic states of Pr^{3+} in APLF glass host and to improve APLF+Pr scintillation properties. Ultraviolet (UV) emission bands at around 250 and 340 nm were observed from both materials and these can be ascribed to $4f5d \rightarrow 4f^2$ transitions in Pr^{3+}. Emission at around 400 nm was also obtained and is principally attributed to $^1S_0 \rightarrow 4f^2$ transition. Difference in the emission profiles of these two materials was found to be due to the extent of the 5d band and its position relative to the $^1S_0$ state. Increasing the concentration of Pr^{3+} up to 2 mol % was found to improve UV emission ratio due to the faster cross-relaxation of 4f states. This could improve the quantum efficiency of APLF+Pr as a neutron scintillator for scattered-neutron diagnostics in laser fusion research.

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JOURNAL OF PHYSICS D: APPLIED PHYSICS 46, 365303, Published: AUG2013

Effect of the $Pr^{3+} \rightarrow Gd^{3+}$ energy transfer in multicomponent garnet single crystal scintillators

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Abstract:
Luminescence processes in the undoped and Pr^{3+}-doped $(Gd,RE)_3(Ga,Al)_5O_{12}$, RE = Lu,Y, multicomponent garnets are studied by time-resolved photoluminescence spectroscopy. Energy transfer processes between Pr^{3+} and Gd^{3+} causing significant deterioration of the scintillation performance are considered in detail. As is shown in current work, an overlap of the $5d_1$–$^3H_4$ emission transition of Pr^{3+} and ^8S–^6P_x absorption transition of Gd^{3+} results in unwanted depletion of Pr^{3+} $5d_1$ excited state and is further intensified by the concentration quenching in the Gd^{3+}-sublattice. This process explains a drastic decrease of light yield in Pr^{3+}-doped Gd^{3+}-containing multicomponent garnets observed in a previous work.

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Simulation of gas avalanche in a micro pixel chamber using Garfield++

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Abstract:
A micro pixel chamber (μ-PIC), the development of which started in 2000 as a type of a micro pattern gas detector, has a high gas gain greater than 6000 in stable operation, a large detection area of 900 cm2, and a fine position resolution of about 120 μm. However, for its development, simulation verification has not been very useful, because conventional simulations explain only part of the experimental data. On the other hand, some μ-PIC applications require precise understanding of the fluctuation of the gas avalanche and signal waveform for their improvement; therefore, there is a need to update the μ-PIC simulation. Hence, we adopted Garfield++, which is developed for simulating a microscopic avalanche in an effort to explain experimental data. The simulated avalanche size was well consistent with the experimental gas gain. Moreover, we calculated a signal waveform and successfully explained the pulse height and time-over-threshold. These results clearly indicate that the simulation of μ-PIC applications will improve and that Garfield++ simulation will easily facilitate the μ-PIC development.

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IEEE TRANSACTIONS ON NUCLEAR SCIENCE 61, 316, Published: FEB2014

Scintillation Properties of Nd^{3+}-Doped Lu_2O_3 Ceramics in the Visible and Infrared Regions

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Abstract:

Nd^{3+}-doped Lu_2O_3 (Nd: Lu_2O_3) is a candidate for an infrared scintillator because (i) Lu_2O_3 has a high density of 9.5 g/cm^3 and a high atomic number of 67 and (ii) Nd^{3+}-doped materials emit in the infrared range and the emission lines from Nd^{3+} can be used in medical applications since human body has a transparency window between 600 and 1,100 nm. However, it is extremely difficult to fabricate Lu_2O_3 single crystals using conventional crystal growth methods because of the high melting point (2,510 ° C). Using solid-state reactions, it is much easier to fabricate Lu_2O_3 into a ceramic structure. Therefore, Nd: Lu_2O_3 transparent ceramics were fabricated using a spark plasma sintering method. This technique is comparatively simple and consumes less time than other methods such as vacuum hot pressing. The scintillation properties and transmittance spectra of the as-produced ceramics were studied in both the visible and infrared regions. Radioluminescence spectra were measured in the range 800-1,200 nm. Nd^{3+} emission lines were observed in the transparency window of human body. Thus, these ceramic materials could be a candidate for medical imaging applications.

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OPTICAL MATERIALS 36, 568, Published: DEC2014

Luminescence and scintillation characteristics of $Gd_3Al_2Ga_3O_{12}:Ce^{3+}$ scintillators

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Abstract:
Cerium-doped $Gd_3Al_2Ga_3O_{12}$ single crystals were grown by the Czochralski method with 1 at.% cerium. Absorption, luminescence and scintillation characteristics were investigated. The light yield and energy resolution were measured under 662 keV γ-ray excitation. The characteristic emission band of Ce^{3+} 5d–4f transition peaking around 525 nm was observed in the photoluminescence of $Gd_3Al_2Ga_3O_{12}$:Ce. The light yield of 56,100 ph/MeV and energy resolution of 6.8% were obtained for a 5 × 5 × 1 mm3 $Gd_3Al_2Ga_3O_{12}$:Ce sample. The light yield dependence on the sample height and on the shaping time was also studied and compared with $Lu_3Al_5O_{12}$:Ce crystal.

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Perovskite fluoride crystals as light emitting materials in vacuum ultraviolet region

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Abstract:
Vacuum-ultraviolet (VUV) fluorescence from $KMgF_3$ and $BaLiF_3$ crystals excited by an extreme ultraviolet free electron laser (EUV-FEL) with 61-nm emission wavelength is studied. Cross-luminescence (CL) peaks at 8.5 eV and 7.5 eV, due to an electron from the valence band recombining with a hole in the F2p core band edge are observed in $KMgF_3$. On the other hand, $BaLiF_3$ exhibited a fluorescence peak at 7.75 eV. The band gap energy of $BaLiF_3$ is estimated from its absorption spectrum to be around 8.41 eV. Results suggest the possibility of developing VUV solid-state devices including light emitting diodes.

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RADIATION MEASUREMENTS 62, 6, Published: MAR2014

Optical and scintillating properties of $Ce:Li(Y,Lu)F_4$ single crystals

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Abstract:
We have investigated the optical and scintillating properties of Lu co-doped $Ce:LiYF_4$ single crystals with various Lu content. In the transmittance and absorption spectra, the absorption peaks at 243 nm get systematically red shifted in contrast to the peaks at 197 and 200 nm which get blue shifted with the increase in Lu content. At the same time, emission peaks at 306 nm and 200 nm under 295 nm excitation also get red shifted. The decay time of $Ce:Li(Y,Lu)F_4$ crystals under 295 nm excitation is found to be faster than that of $Ce:LiYF_4$ and $Ce:LiLuF_4$ crystals. The alpha-peak positions in the pulse-height spectra and decay times of crystals under alpha-ray irradiation are found to vary with the Lu content.

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Temperature Dependence of Neutron-Gamma Discrimination Based on Pulse Shape Discrimination Technique in a Ce:LiCaAlF$_6$ Scintillator

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Abstract:
LiCaAlF$_6$ scintillators are one of the attractive scintillators for neutron detection. To reduce the effect on gamma-rays, the Ce doped LiCaAlF$_6$ scintillators can discriminate the neutron and gamma-ray events based on the pulse shape discrimination technique. To apply the scintillators for the oil logging, the high temperature characteristics must be investigated. In this paper, the temperature dependence of the neutron-gamma discrimination based on pulse shape discrimination technique in the Ce:LiCaAlF$_6$ scintillator is investigated as one of the high temperature characteristics. The property of pulse shape discrimination in the Ce:LiCaAlF$_6$ has small temperature dependence ranging from 25°C to 150°C. We concluded that the Ce:LiCaAlF$_6$ scintillators can discriminate neutron and gamma-ray events under high temperature condition up to 150°C .

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EPJ WEB OF CONFERENCES 59, 13011, Published: NOV2013

Multichannel down-scattered neutron detector for areal density measurement

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Abstract:
A down-scattered neutron detector operating in the multichannel counting mode was developed for areal density (ρR) measurement. Equipped with a newly developed ^{6}Li glass scintillator (APLF80), the detector was tested in a fusion experiment at the GEKKO XII facility, Osaka University, Osaka, Japan. For a low-ρR fusion shot, the detector clearly discriminated the γ-rays, primary neutrons, γ-rays produced via (n, γ) reactions from the target chamber, and neutrons scattered by the target chamber. Furthermore, the observed signal was in good agreement with predictions made by Monte Carlo simulation.

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IEEE TRANSACTIONS ON NUCLEAR SCIENCES 60, 988, Published: APR2013

First Performance Results of Ce:GAGG Scintillation Crystals With Silicon Photomultipliers

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Abstract:
A new single-crystal Cerium doped $Gd_3Al_2Ga_3O_{12}$ (GAGG) scintillation crystal with high luminosity, high density and relatively fast decay time has successfully been grown. We report on the first performance results of the new GAGG scintillation crystal read out with silicon photomultipliers (SiPM) from Hamamatsu (MPPC) and FBK. The best energy resolution (511 keV peak of Ge-68) of 7.9% was attained with GAGG coupled to MPPC and 9.0% with the FBK SiPM after correcting for non-linearity. On the other hand, the best coincidence resolving time (FWHM) of polished 3 × 3 × 5 mm3 and 3 × 3 × 20mm3crystals were 464 ±12 ps and 577 ±22 ps for GAGG crystals compared to 179 ±8 ps and 214 ±6 ps for LYSO crystals respectively with MPPCs. The rise time of GAGG was measured to be 200 ps (75%) and 6 ns (25%) while the decay time was 140 ns (92%), 500 ns (7.7%) 6000 ns (0.3%).

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IEEE TRANSACTIONS ON NUCLEAR SCIENCES 60, 4582, Published: APR2013

Development of an Ultrahigh Resolution Block Detector Based on 0.4 mm Pixel Ce:GAGG Scintillators and a Silicon Photomultiplier Array

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Abstract:
Ce doped $Gd_3Al_2Ga_3O_{12}$ (Ce:GAGG) is a newly developed single-crystal scintillator which has a large light output and longer emission light wavelength. The longer wavelength of the scintillation photons will produce a larger signal when coupled to typical silicon photomultiplier (Si-PM) as the quantum efficiency of semiconductor based photodetector is generally higher for light with longer wavelength. A block detector with higher spatial resolution may thus be realized by combining Ce:GAGG with Si-PM arrays. To achieve the highest possible spatial resolution for PET and SPECT detectors, we developed an ultrahigh resolution block detector using 0.4 mm × 0.4 mm × 5 mm Ce:GAGG pixels assembled to form a 24 × 24 matrix that is coupled to an Si-PM array and evaluated the performance. All Ce:GAGG pixels were separated in the 2-dimensional position histograms for Cs-137 (662 keV) gamma photons with an average peak-to-valley (P/V) ratio of 2.4. The energy resolution was 21.6% FWHM for Cs-137 (662 keV) and 23.8% for Co-57 (122 keV) gamma photons. Since Ce:GAGG does not contain naturally occurring radioisotope (Lu), beta-gamma true coincidences can be avoided and randoms are reduced when used for PET detectors. Furthermore, this property, together with its high light output and good intrinsic energy resolution, make the scintillator suited for SPECT detectors. An ultrahigh resolution PET/SPECT hybrid system might be an interesting application using Ce:GAGG/Si-PM block detectors.

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Development of an Ultrahigh Resolution Block Detector Based on 0.4 mm Pixel Ce:GAGG Scintillators and a Silicon Photomultiplier Array

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Abstract:
Ce doped $Gd_3Al_2Ga_3O_{12}$ (Ce:GAGG) is a newly developed single-crystal scintillator which has a large light output and longer emission light wavelength. The longer wavelength of the scintillation photons will produce a larger signal when coupled to typical silicon photomultiplier (Si-PM) as the quantum efficiency of semiconductor based photodetector is generally higher for light with longer wavelength. A block detector with higher spatial resolution may thus be realized by combining Ce:GAGG with Si-PM arrays. To achieve the highest possible spatial resolution for PET and SPECT detectors, we developed an ultrahigh resolution block detector using 0.4 mm × 0.4 mm × 5 mm Ce:GAGG pixels assembled to form a 24 × 24 matrix that is coupled to an Si-PM array and evaluated the performance. All Ce:GAGG pixels were separated in the 2-dimensional position histograms for Cs-137 (662 keV) gamma photons with an average peak-to-valley (P/V) ratio of 2.4. The energy resolution was 21.6% FWHM for Cs-137 (662 keV) and 23.8% for Co-57 (122 keV) gamma photons. Since Ce:GAGG does not contain naturally occurring radioisotope (Lu), beta-gamma true coincidences can be avoided and randoms are reduced when used for PET detectors. Furthermore, this property, together with its high light output and good intrinsic energy resolution, make the scintillator suited for SPECT detectors. An ultrahigh resolution PET/SPECT hybrid system might be an interesting application using Ce:GAGG/Si-PM block detectors.

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吉川彰 東北大学教授に報公賞

吉川東北大教授に報公賞

服部報公会は２０１３年（第83回）の報公賞に東北大学金属材料研究所の吉川彰教授の「ガーネット型シンチレーターの開発と放射線検出器への展開」を採択した。贈呈式は10月9日に日本工業倶楽部で行われ、賞状並びに賞金５００万円が贈呈される。また、工学研究奨励援助として10件の研究に対し総額１０００万円が贈られる。

吉川教授は、放射線などの検出・測定に用いられるシンチレーター結晶の高速育成法「マイクロ引き下げ法」を開発、従来と比べて10倍以上のスピードで結晶成長させることに成功した。この手法を利用することで高発光で短蛍光寿命のＬＵＡＧ（ルテチウムアルミニウムガーネット）結晶を見いだすとともに、ＧＡＧＧ（ガドリニウムアルミニウムガリウムガーネット）結晶では古河機械金属と共同で高感度サーベイメーターの実用化にも成功している。

同賞は工学分野の進歩に貢献する研究成果をあげた研究者に対して贈られる。

電波新聞
２０１３年 10 月 11 日（金）

服部報公会が設立記念会
報公賞に東北大の吉川教授

服部報公会（菅原卓雄理事長）は9日、東京都内の会議場で第83回設立記念会を開催した。東北大学金属材料研究所先端結晶工学研究部未来科学技術共同研究センター兼任の吉川彰教授に報公賞（副賞として500万円）を、名古屋大学大学院工学研究科の安井隆雄助教ほか9人に工学研究奨励援助金として各100万円を贈呈した。

吉川教授（前列右から５人目）と菅原理事長（同６人目）

服部報公会は、1930年（昭和5年）、服部時計店（現セイコーホールディングス）の創業者で初代社長の服部金太郎氏が私財を投じ設立した公益事業団体。

報公賞の吉川教授は、研究テーマ「ガーネット型シンチレータの開発と放射線検出器への展開」が評価された。

各種放射線を紫外光あるいは可視光に変換する蛍光体結晶の一種であるシンチレータは、受光素子と組み合わせ放射線の検出・計測に用いられる。高エネルギー分野に加え、最近では医療や工学分野でも重要性が高まっている。特に、東日本大震災による原子力発電の事故以来、安価・小型で分解能の高い検出器の開発が望まれていた。

吉川教授は、従来法に比べ10倍以上の高速で結晶が得られる独自の「マイクロ引き下げ法」を開発。優れた特性を持つガーネット（ざくろ石）型構造の酸化物シンチレータを見いだした。さらに、実用化に成功し、放射線検出器の展開へ多大な貢献をした。

工学研究奨励援助金は、1年間で成果が見込まれ、工学の発展に寄与する研究に携わる40歳以下の研究者に贈られるもので、公募される。

受領者は、名古屋大学・安井助教のほか、大阪大学・清水一憲助教、山口大学・鈴木祐麻助教、東北大学・関剛斎助教、東北大学・高橋和貴准教授、横浜国立大学・西島喜明准教授、東京大学・花岡健二郎准教授、東京大学・福井類特任講師、三重大学・溝田功助教、東京大学・吉川健准教授。

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TOHOKU UNIVERSITY

IMR KINKEN Research Highlights 2013

Institute for Materials Research, Tohoku University

Research

Growth of Shaped Crystals and their Application in Device Fabrication

Device size–shaped crystal growth technology has been established using the micro-pulling-down method. Devices using these shaped crystals have started to become commercially available. Shaped sapphire crystals, langasite-type piezoelectric crystals for combustion sensors, scintillator crystals for pocket real-time dosimeters, and Ir-based alloy crystals for spark plugs have been grown in specially shaped crucibles.

Introductions

Various large bulk crystals have been grown from melts with the Czochralski and Bridgeman methods; these grown crystals are used in many industrial and research fields after forming processes such as cutting from the bulk crystals and polishing of the cut crystals. However, the forming processes after crystal growth greatly affect the manufacturing cost and increase the prices of the final products using the crystal elements.

The micro-pulling-down (μ-PD) method is a crystal growth method that uses a crucible with a die at the bottom. Compared to conventional methods, the μ-PD method has a great advantage in that a shaped single crystal can be grown with a specially shaped crucible [1]. Because of its potential for the near–net shape growth of a single crystal, the μ-PD method is expected to be applied in mass production.

Development of Shaped Crystals

Shaped sapphire crystals with various configurations and multiple shaped sapphire crystals were grown by the μ-PD method using molybdenum crucibles with various shaped dies [2]. All shaped crystals had high transparency and visible inclusions, and no cracks were observed in the crystals. The crystallinity of each shaped crystal was evaluated by X-ray rocking curves, and all crystals indicated a high crystallinity of less than 100 arcsec.

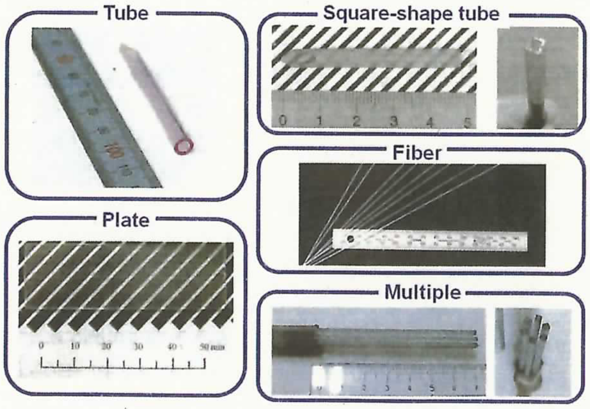

Fig. 1 Shaped sapphire crystals grown by μ-PD method.

We developed a novel Pt alloy crucible with a suitable wetting angle for a langasite-type melt. Columnar, plate, and tube-shaped $La_3Ta_{0.5}Ga_{5.5}O_{14}$ (LTG), $Ca_3NbGa_3Si_2O_{14}$ (CNGS), $Ca_3TaGa_3Si_2O_{14}$ (CTGS), $Sr_3NbGa_3Si_2O_{14}$ (SNGS), and $Sr_3TaGa_3Si_2O_{14}$ (STGS) piezoelectric crystals were grown using Pt alloy crucibles with special-shaped dies [3]-[8]. The shaped langasite-type crystals indicated comparable piezoelectric properties to previously reported results. In addition, a combustion sensor test device using the shaped langasite-type crystal was developed.

After the nuclear accident in Fukushima, the demand for dosimeters has greatly increased. However, previous dosimeters using a semiconductor had low sensitivity to radiation, and those using scintillators with high sensitivity were expensive. Therefore, we developed a growth method for shaped Ce-doped $Y_3Al_5O_{12}$ (Ce:YAG) scintillator crystals to decrease the price of dosimeters. Facets of as-grown crystals can be controlled by the crystal orientation of the seed crystal; the facet surfaces had high transparency without polishing. A dosimeter using the shaped Ce:YAG crystals without polishing was developed, and it indicated good sensitivity to radiation.

Recently, a novel Ir alloy with high oxidation resistance was developed and used for a spark plug in an engine. However, the Ir alloy is unworkable, and the thin wire had to be made from the Ir alloy cylinder by scraping. In order to develop a shaping technology for Ir alloy wire with one operation, we developed a ceramic crucible that can be used at 2400 °C without evaporation or breaking, and a shaped Ir alloy wire was grown in it.

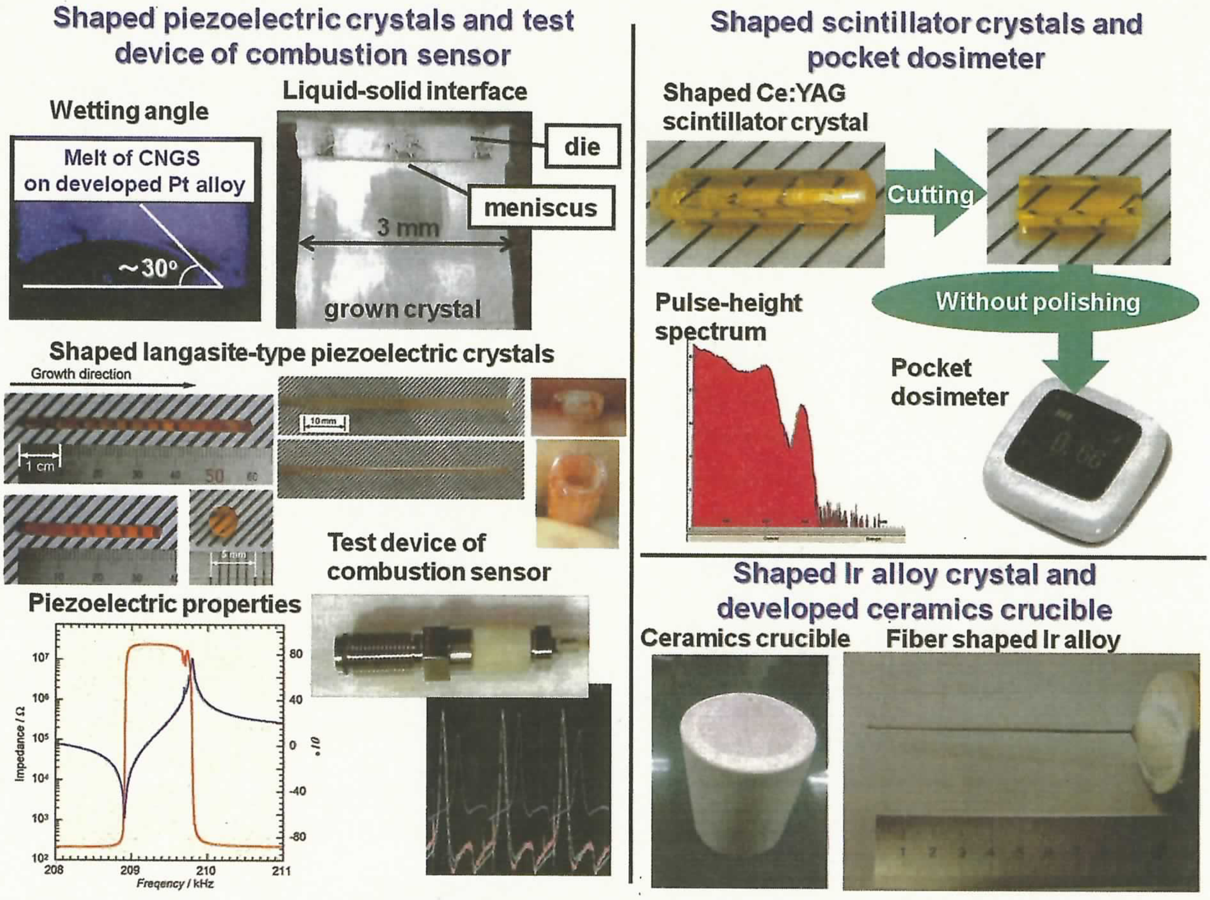

Fig. 2 Shaped piezoelectric, scintillator and Ir alloy crystals grown by μ-PD method and developed combustion sensor test device and dosimeter using shaped crystals.

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Keywords: crystal growth, piezoelectric, radiation effects
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List of events

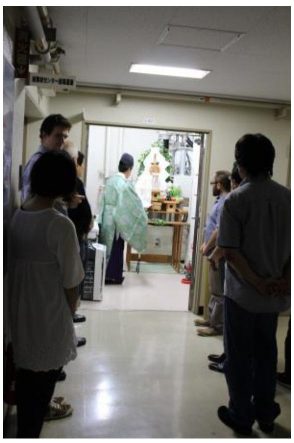
2013年度　吉川研究室　行事

月	吉川研究室内行事	学会	研究会・講演会
4	4/7-16 Kochurikhin先生　来日 4/8-19 新人ティーチ・イン 4/11 研究室内花見 4/20 金研花見大会	4/15-19 SCINT 2013 (上海) 4/26-28 REMAT 2013(ヴロツワフ)	4/15 学振124委員会
5	5/1工学マテリアル・開発系プレゼミ金研見学会	5/22-25 強誘電体応用会議(京都市)	5/20 学振186委員会 5/22金研・2013春季講演会 5/23 学内次世代自動車プロジェクト招待講演 5/24 新化学技術推進協会
6	6/3 A. Medvedev博士, M. Nikl客員教授来日 6/4 G. Boulon客員教授 来日 6/4 Welcome party 6/14 October Fest 2103 参加	6/9-18 3CG 6/24-28 iWoRID2013	
7	7/3 戸口氏　産休復帰 7/12 新Cz装置の火入れ式・祝賀会 7/12 横田先生 准教授昇進祝賀会 7/18 金研ビアパーティ参加 7/29 武田氏 加入	7/21-25 IEEE-UFFC 2013(プラハ) 7/21-26 ACCGE-19 (コロラド・キーストン)	7/5-6 学振186委員会(札幌) 7/19 学振161委員会
8	8/1 長門氏、早坂氏 加入 8/1 Welcom party 8/28-29 歓迎会	8/11-16 ICCGE-17 (ワルシャワ)	
9		9/4-6 日本セラミックス協会秋季シンポジウム 9/16-20 応用物理学会秋季学術講演会 9/21-29 SSD17 9/23-27 ASM-2013 (ハルキウ) 9/24-27 SSDM2013	9/12 学振186委員会
10	10/1 Chani先生来日 10/3 NHK 取材 10/12-13 片平まつり、きんけん一般公開参加 10/21-31 共融会フットサル大会	10/5 日本結晶成長学会・バルク成長分科会 10/20-24 ISLNOM-6 (上海) 10/27-11/2 IEEE NSS/MIC/RTSD (ソウル)	10/11 学振161委員会
11	11/5 V. Kochurikhin先生 A. Medvedev博士 来日 11/7 G. Boulon先生 来日 11/8 Pre welcome party 11/11 芋煮会 (Welcome party) 11/19 IEEEお疲れ様会、歓迎会および修論激励会	11/6-8 日本結晶成長学会	11/6 学振186委員会 11/22-21 第10回材料科学若手学校(KINKEN－WAKATE20) 11/26-28 次世代自動車国際シンポジウム 11/30-12/1 第9回放射線モニタリングワークショップ
12	12/9 E. Galenin博士およびI. Gerasymov博士来日(1W) 12/11 両博士歓迎会 12/17 忘年会 12/27 納会	12/6 日本フラックス成長学会 12/13-14 光物性研究会	
1		1/28-30 放射線検出器とその応用 (KEK)	1/29 圧電材料・デバイスシンポジウム 1/30 次世代自動車のための産学官連携イノベーション成果報告発表会 1/30-31 学術141委員会・学術186委員会合同委員会
2	2/7 修論発表会・修論打ち上げ		
3	ISFM 2013 in Tazawa-ko	3/17-20 応用物理学会春季学術講演会(相模原)	

4/11 _ Cherry blossom party

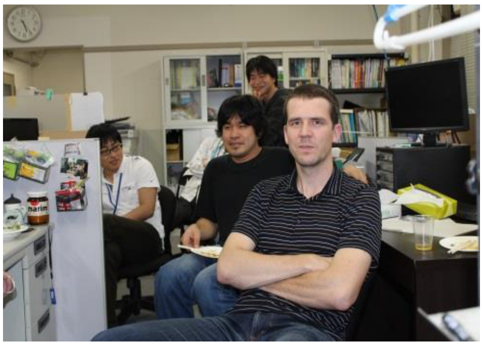
5/23_Jan san promotion / Birth day party)

7/12 _Fire ceremony (30Cz)



7/12 _Yokota Sensei Promotion party

8/1 _Welcome party



8/11-15 _ICCGE-17

9/15_In Kyoto

10/11_IMR opening to the public

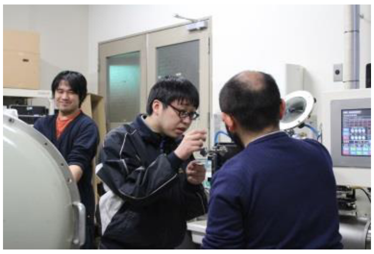
10/28-29_IEEE (in Korea)

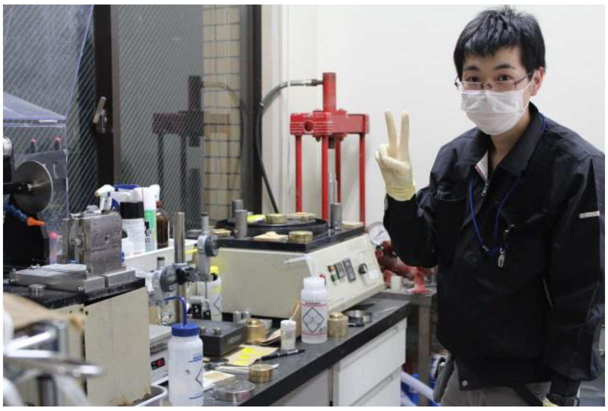
11/8 welcome party

Yoshikawa Sensei Birth day

2/8 Oral presentation of Master thesis

One day





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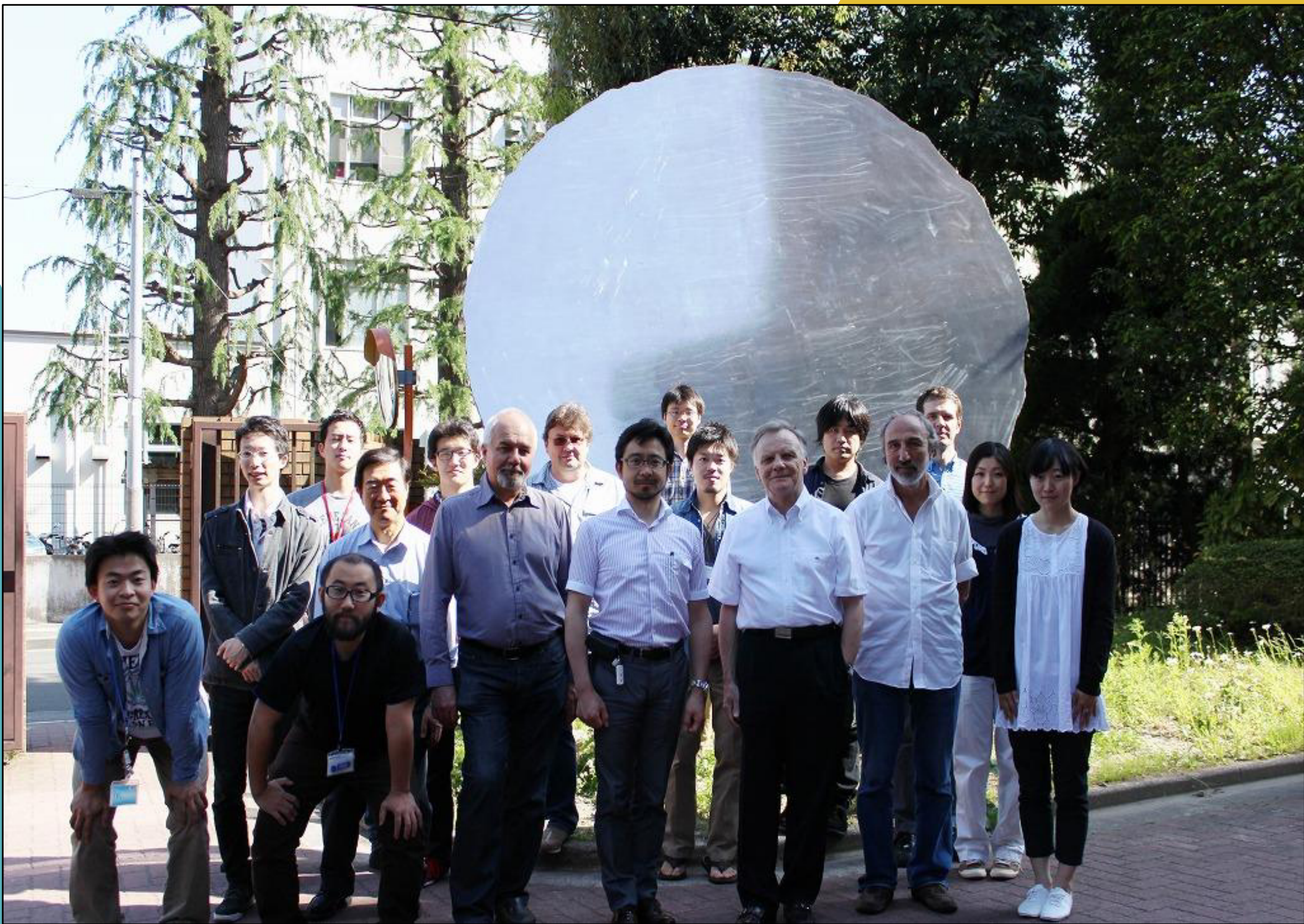

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