ANNUAL REPORT 2011 Yoshikawa Lab., IMR, Tohoku Univ.

# ANNUAL REPORT 2011

## April 2011 - March 2012

# Yoshikawa Lab.

Since 2007

IMR, Tohoku University

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# Preface

The report in your hands is intended to give a general impression about research activity of the *Yoshikawa Laboratory* in the Institute for Materials Research (IMR), Tohoku University. This is the first report of my laboratory as official Research Division in IMR (originally established in April, 2007 as *Yoshikawa Group* at IMRAM). It contains a summary of our various activities and copies of our selected publications. We hope that you will find it interesting and informative.

This year, major part of the time is devoted to earthquake disaster reconstruction, however, thanks to the kind help from all over the world, all the equipments were somehow restored by the end of 2011.

Current issue covers our activity within academic year from April 2011 to March 2012. Within this period we put our efforts to develop our key technologies and understand the science behind them. Highlights of this year are three items. One is the further progress of $^{6}$LiCAF for neutron detection as an alternative of $^{3}$He. Second is the establishment of micro-pulling down method for halide materials. Now, it is available to grow hygroscopic materials like $LaBr_3$ by the micro-pulling down method. Third one is a new topic. National project about shaped crystal growth of langasite type CNGG and CTGS for combustion pressure sensor has been started.

When we try to develop novel scintillator crystals, there are plenty of topics, which we have to study, such as design of host lattice from both solid state chemistry point of view and physical point of view. In the multicomponent garnet study, we found that the relative position between band gap and position of dopant level are key issue. Moreover, single crystal growth technology, optical characterization, understanding of luminescence processes, searching for the suitable photo detector, reflector, light guide and so on are also very important.

This year, we attempted to apply this concept to other topics such as piezoelectric materials and got positive results.

These activities are always supported by our colleagues from all over the world through the fruitful collaboration.

Details of these studies can be found in the photos and papers published within the above period and included in this report.

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

Akira YOSHIKAWA

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

March, 2012

# *Research Digest*

# Research Activities in 2011

**Development of**
**Scintillator, Laser, Piezoelectric crystals**
**Crystal growth technology and device application**

**Int'l collaboration**

Inst. Phys. (Czech), CB, Univ. Lyon1 (France), Milan-Bicocca Univ. (Italy),
Soltan Institute for Nuclear Studies (Poland), Delft Univ. of Tech. (The Netherlands)

**Fluoride Scintillators**

**Oxide Scintillators**

Neutron imager, VUV scintillator

PEM PET/MRI

**Halide Scintillators**

**Multidisciplinary Research for "Characterization" and "Device application"**

**Transparent Ceramics Scintillators**

Combustion sensor, SAW filter, …

G-PMT, APD, MPPC, MSGC, GaN, …

**Piezoelectric Crystals**

**Development of new photodetectors**

**Univ., National Inst.**

Univ. of Tokyo (Kamiokande, Takahashi Lab.), Kyoto Univ.(Tanimori Lab.),
Nagoya Univ. (Iguchi Lab., Uritani Lab.), Osaka Univ.(Sarukura Lab.),
Hiroshima Univ.(Fukazawa Lab.), Kyushu Univ.(Ishibashi Lab.), Waseda Univ. (Kataoka Lab.)

**Company**

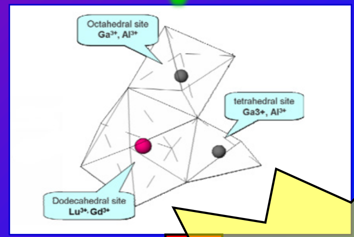
Tokuyama, Furukawa, TDK, MGC, Kobe Steel, Koike, Murata Manufacturing,
Canon, Chiyoda technol, Nihon Kessho Kogaku, Tanaka metal, Furuya metal,
Akita Seiko, Aoyama Seiko, TEP, Toei, Star Seiki, Koshuha Nestle



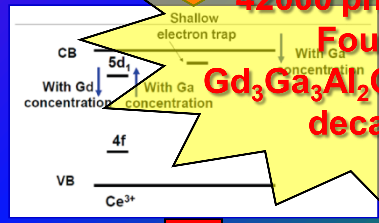
# Oxide scintillators

## Materials for gamma-ray detection

### Multicomponent Ce-doped garnet scintillators

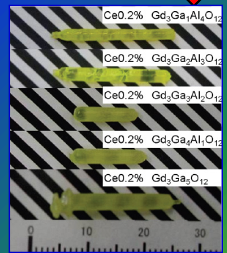


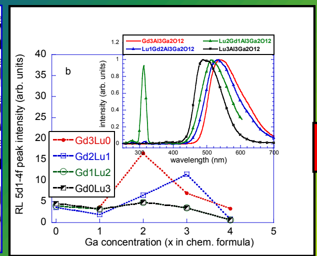

Balanced composition - band gap engineering and energy-level positioning




**High light yield 42000 photons/MeV Found for $Gd_3Ga_3Al_2O_{12}$:Ce!! 90ns decay time**


Composition screening by Micro-pulling down





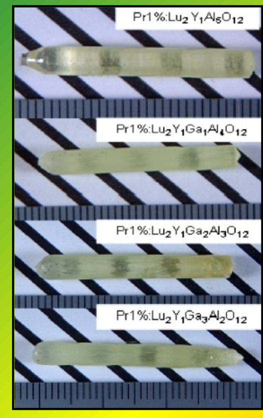
Emission peak intensity and position on Ga concentration.

K. Kamada et al.,
Crystal Growth and Design 11 (2011) 4484-4490

Czochralski growth – high quality crystals

$Gd_3Ga_3Al_2O_{12}$:Ce

### Multicomponent Pr-doped garnet scintillators



Faster response (20ns) than Ce-doped materials, good light yield

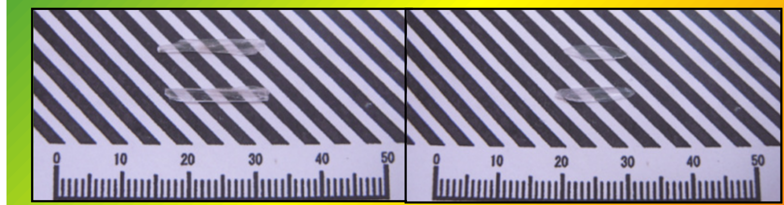
| | Theoretical density ($g/cm^3$) | L.Y. (photon/MeV) | decay time (ns) |
|---|---|---|---|
| Pr:LuAG standard | 6.73 | 20000 | 20.2ns (83%) 168ns (17%) |
| **$Lu_2Y_1Ga_0Al_5O_{12}$** | **5.98** | **24500** | **19.3ns (78%) 93.7ns (22%)** |
| $Lu_2Y_1Ga_1Al_4O_{12}$ | 6.22 | 16000 | 18.1ns (84%) 80.5ns (16%) |
| **$Lu_2Y_1Ga_2Al_3O_{12}$** | **6.45** | **20500** | **17.4ns (91%) 82.8ns (9%)** |
| $Lu_2Y_1Ga_3Al_2O_{12}$ | 6.69 | 8800 | 7.9ns (17%) 20.4ns (83%) |
| $Lu_2Y_1Ga_4Al_1O_{12}$ | 6.92 | 0 | *N.D |
| $Lu_2Y_1Ga_5Al_0O_{12}$ | 7.16 | 0 | *N.D |

K. Kamada et al.,
IEEE Trans. Nucl. Sci. 59 (2012) 2130 - 2134

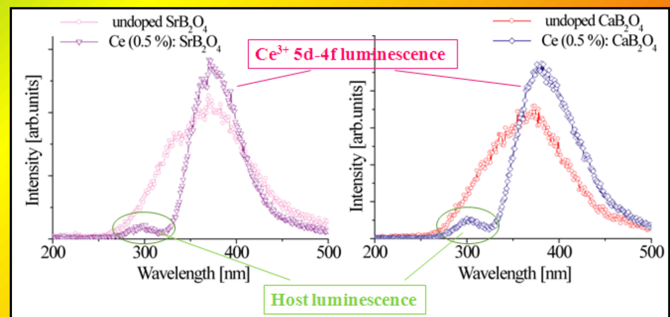
**Support Industry by "Japan Science and Technology Agency(JST)"**

## Materials for neutron detection

### B-containing crystals

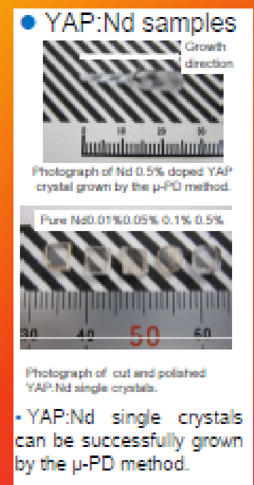


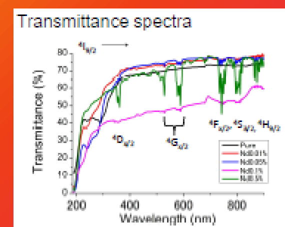
0.5 % Ce-doped $SrB_2O_4$ 0.5 % Ce-doped $CaB_2O_4$

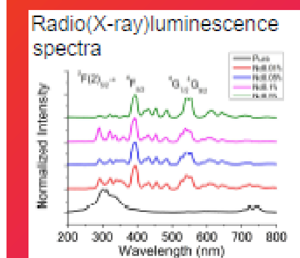



## Materials for X-ray detection

### Nd:YAP crystals



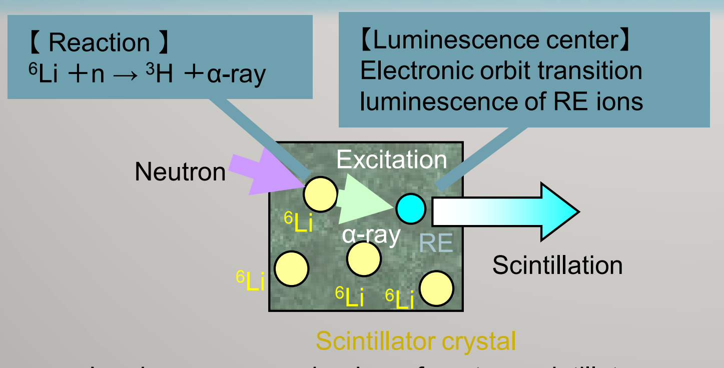



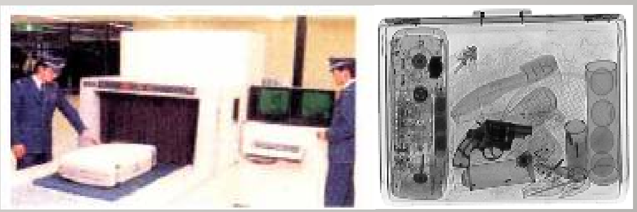

**Radio-luminescence spectra under X-ray irradiation showed several emission peaks and the strongest peak was observed at 395 nm ($^2F(2)_{5/2}$–$^4F_{5/2}$ transition).**

# Development of fluoride scintillator

## Neutron scintillators alternatives to $^3He$

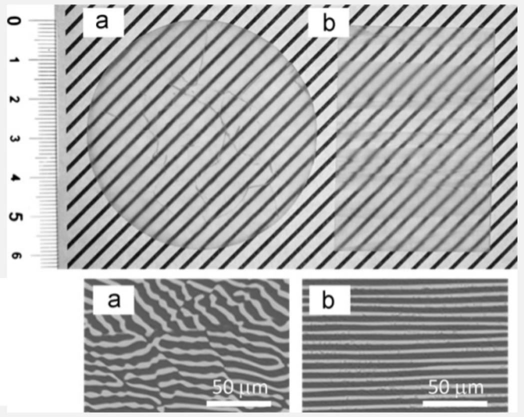



Luminescence mechanism of neutron scintillator.



**Baggage-screening at Airport by using the neutron systems**

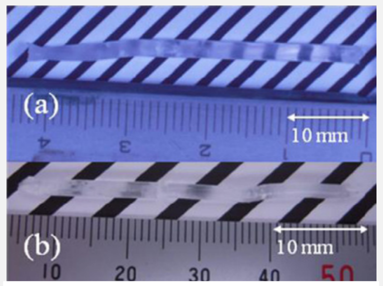
## Eutectic composite scintillators




**Photograph and SEM image of $LiF/CaF_2$:Eu composite cut across (a) and along (b) the solidification direction.**

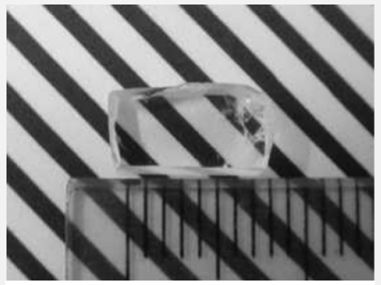
N. Kawaguchi, et al., Nucl. Instrum. Meth-A., 652, 209-211, (2011).

## Other fluoride crystals



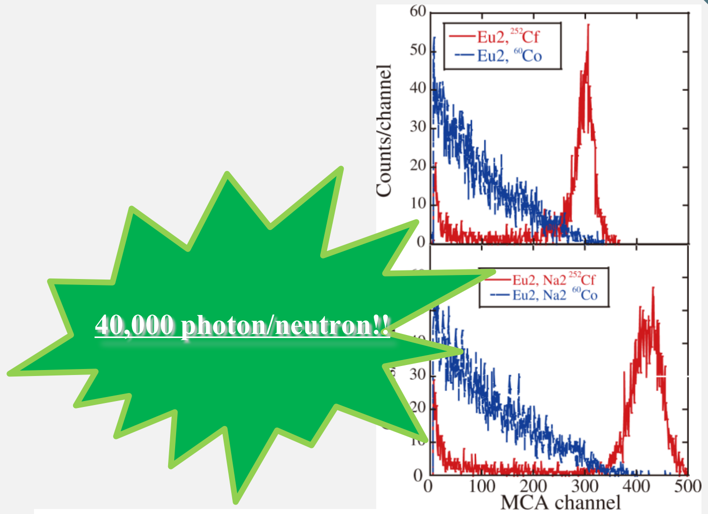

**The $K_6Li(Y_xCe_x)F_5$ ((a): x = 0.003, (b): x = 0.02) single crystals grown by the l-PD method.**

Y. Furuya, et al., Opti. Mater., 33(6), 855-881, (2011).



**The Sr7%:Ce15%:$GdF_3$ specimen cut from the single crystal grown by the CZ method (the scale is in mm).**

A. Fukabori, et al., Cryst. Griwth, 318, 1175-1178 (2011),




**The light yield of Eu 2%, Na 2%-doped LiCAF (bottom) reached 40,000 ph/n, which was about 30% higher than that of Eu:LiCAF (top).**

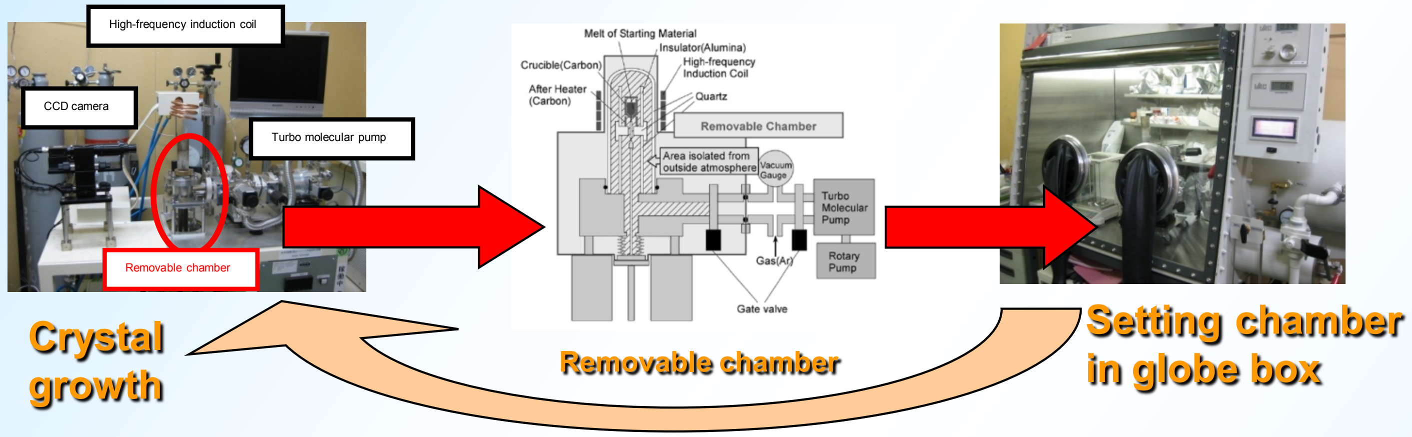
T. Yanagida, et al., Appli. Phys. express ,4(10), 106401, (2011) .


Support Industry by "Japan Science an d Technology Agency (JST)" with Tokuyama Co.


# Halide scintillators

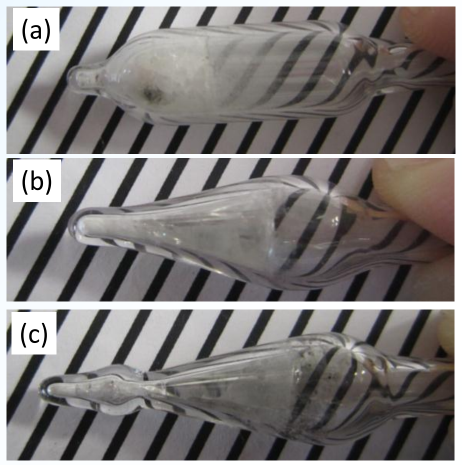
Aim of this work was to prepare new halide materials for radiation detectors (scintillators and photodetectors) and study their optical properties. Single crystals of $SrI_2$, Eu:$SrI_2$, $LaBr_3$, $RbPb_2Cl_5$, Nd:$RbPb_2Cl_5$ and Yb:$RbPb_2Cl_5$ were prepared by the micro-pulling-down (μ-PD) method and by the vertical Bridgman method. Grown crystals are shown below.

## Experimental arrangement of halide m-PD machine.



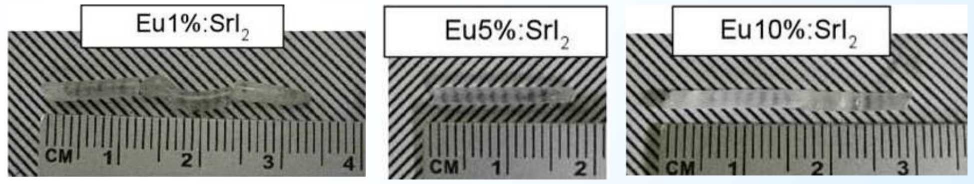

**Halides** are hygroscopic materials which easily react with air and air moisture under formation of oxy- and hydroxy-halides, though careful handling of starting materials under protective argon atmosphere in globe box is required.

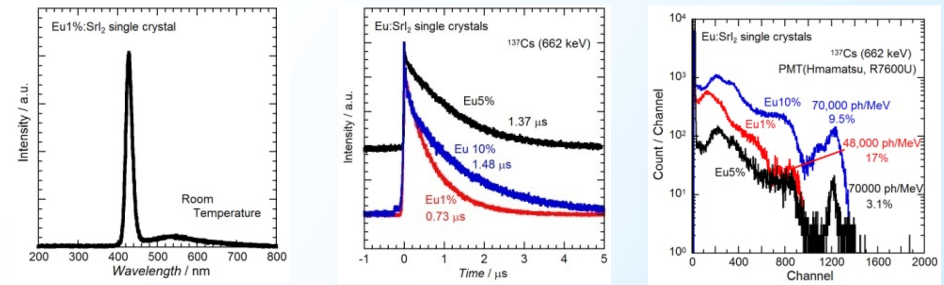
## Crystals prepared by vertical Bridgman method



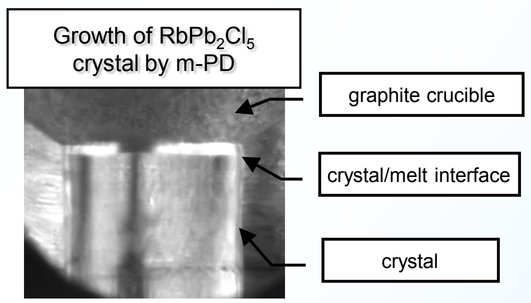

**$LaBr_3$** crystals prepared in different quartz ampoules: (a) normal,(b) conical and (c) conical with necking.

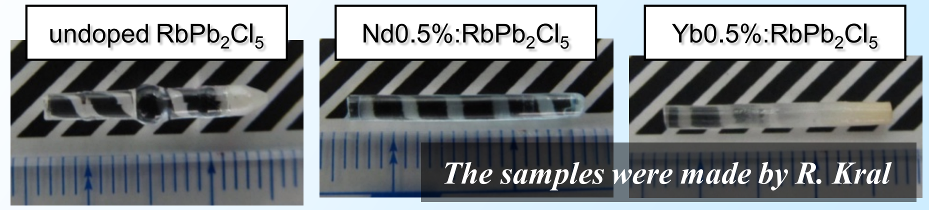
## Chloride and iodide single crystals grown by micro-pulling-down method



**$SrI_2$** crystals were studied by **X-ray diffraction** (XRD) that confirmed presence of only $SrI_2$ crystal phase, **radioluminescence** measurements showed $Eu^{2+}$ emission peak around 430 nm, under $^{137}$Cs **γ-ray irradiation** light yields were 48,000 ~ 70,000 ph/MeV, energy resolutions were 3.1 ~ 17% and decay times were 0.73 ~ 1.48 μs.



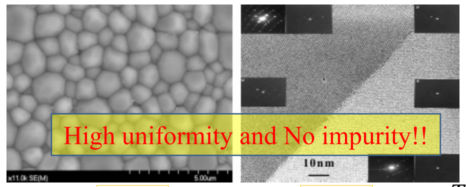

Grown **$RbPb_2Cl_5$** single crystal was colorless and transparent, Nd:$RbPb_2Cl_5$ was blue and Yb:$RbPb_2Cl_5$ was colorless and opaque in its upper part due to segregation of the Yb(III). **XRD** measurements showed presence of only $RbPb_2Cl_5$ crystal phase.

# Transparent Ceramics Scintillator

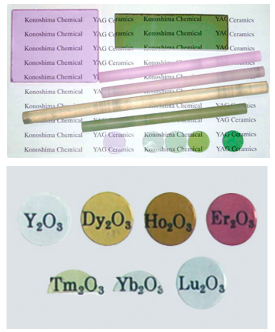
## ✓Advantages of ceramics

- Having better chemical uniformity than single crystals.
- Being able to be produced with high concentration of dopant.
- Befitted industrial production because of relatively large size and low cost.
- Economical especially for high-melting point materials.

Recently, the technique of the powder synthesis and sintering has been significantly improved!!

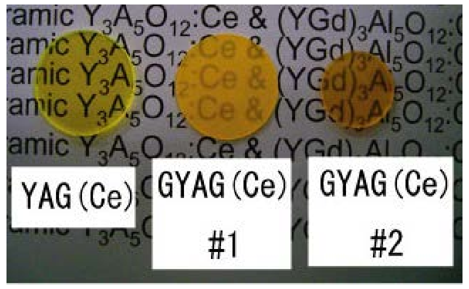


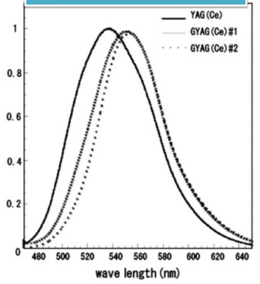

SEM TEM

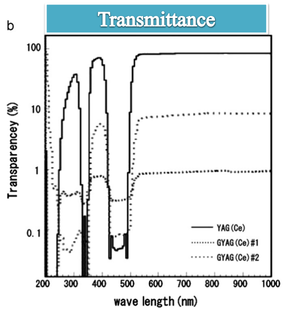


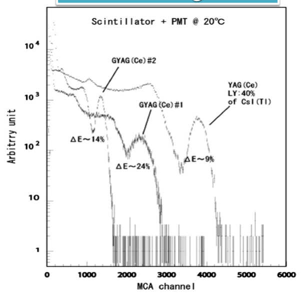

*Transparent ceramics by Konoshima chemical Co., Ltd.*

## ✓Garnet ceramics

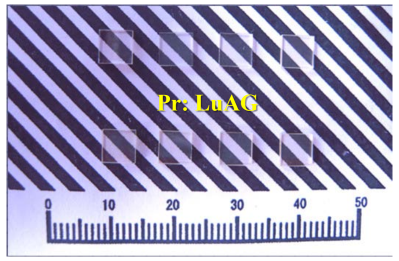


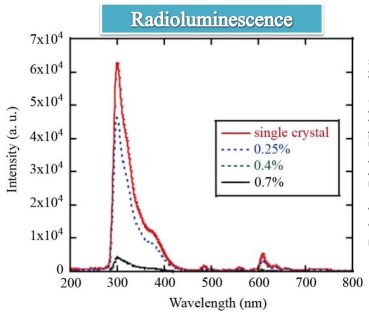



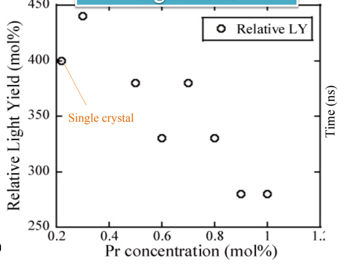



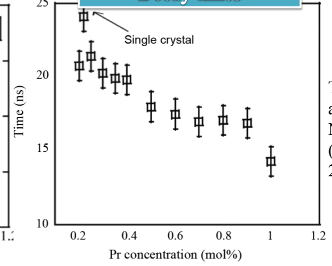




T. Yanagida et al., Nucl. Instr. and Meth. A 579 (2007) 23–26.

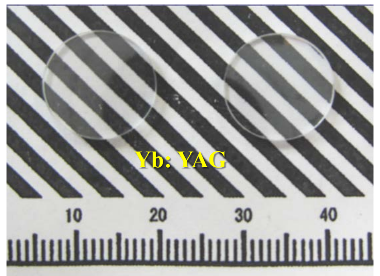


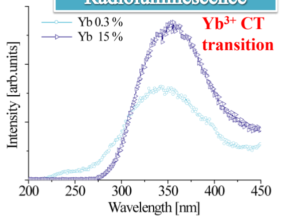



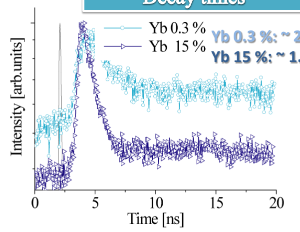



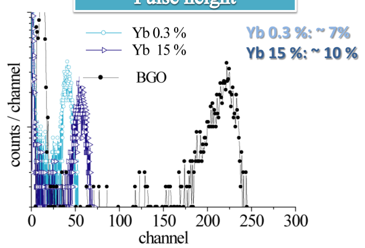




T. Yanagida et al., IEEE Trans. Nucl. Sci., 56 (2009) 2955-2959.



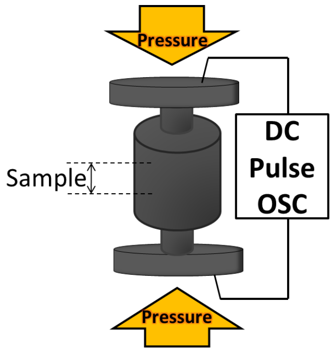



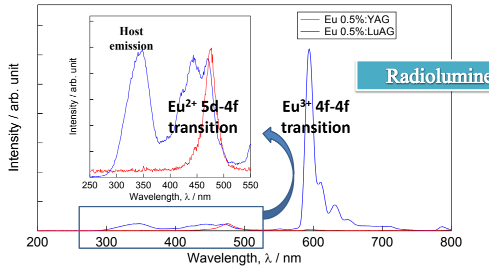

藤本 裕発表, 日本セラミックス協会2012年年会

## ✓Spark Plasma Sintering (SPS)



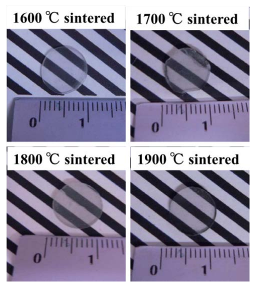

**Advantage of SPS**

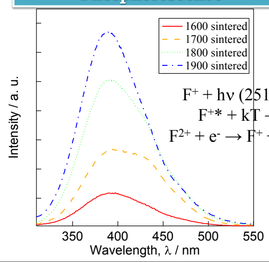
- A rapid consolidation rate and that is appropriate for densification of variety of ceramics
- High reductive condition due to carbon die and punch.
- Easily high density solidification.

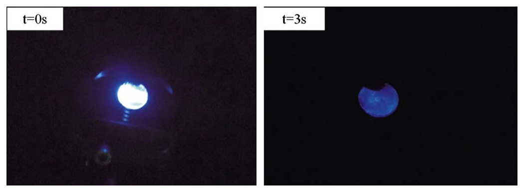

杉山 誠発表,
日本セラミックス協会
2012年年会

**MgO**
**Phosphorescent luminescence**
**(not scintillator)**




$F^+ + h\nu\ (251\ nm) \rightarrow F^{+*}$
$F^{+*} + kT \rightarrow F^{2+} + e^-$
$F^{2+} + e^- \rightarrow F^+ + h\nu\ (390\ nm)$



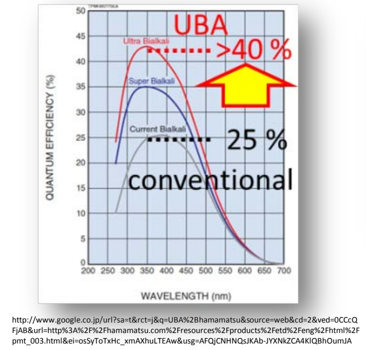

S. Wakahara et al., Opt. mater., 2012 in press.

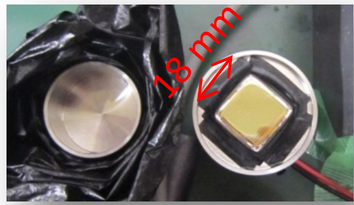


# Photo detectors

We investigate radiation reactions of our scintillators using a photomultiplier tube and semi-conductor, and we have heat .

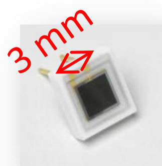
## 1, Photomultiplier Tube & Semi-conductor




Ultra Bialkali (UBA) R7600U

We use Ultra Bialkali Photomultiplier tube (PMT) R7600U as a standard PMT for pulse height measurement.




APD


MPPC and the readout circuit

Measurement set up for the APD and MPPC

### APD (avalanche photodiode)

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

### MPPC (Multi-Pixel Photon Counter)

- ✓ Good photon counting

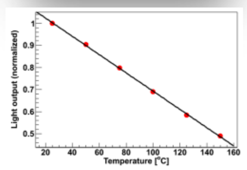
## 2, Black box

We make 2 black boxes to measure the photon-counting with a MPPC, luminescence in other lab.

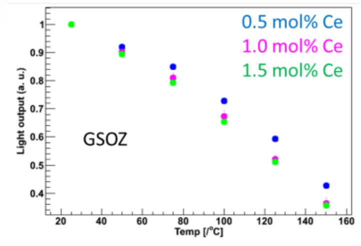
## 3, Measurement of Temperature Dependence

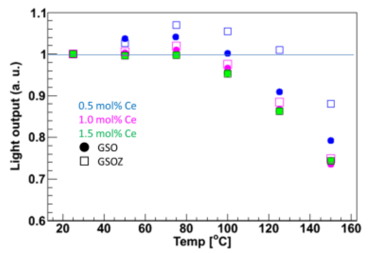
- ✓ We investigated the temperature dependence of the light output and the decay time for several scintillator

Thermostat chamber (Isuzu HPPC-40-20)




Calibration system with LED light




light output vs. temperature

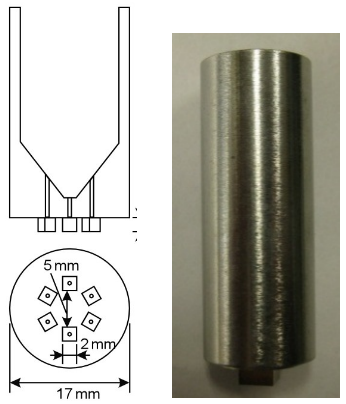
# Development of Shaped Piezoelectric crystals

**Previous μ-PD furnace for growth of shaped crystals**

- Large space for installation
- High cost of manufacturing
- High power consumption
- Difficult technique of crystal growth

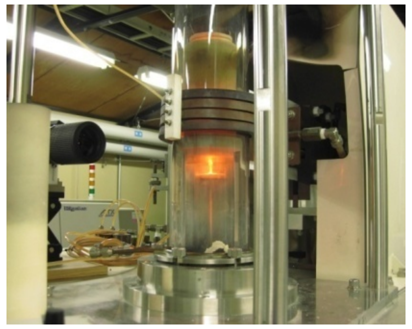
Development of new μ-PD furnace for mass-production of shaped crystals

**Crucible**




**Generator**

**Furnace**

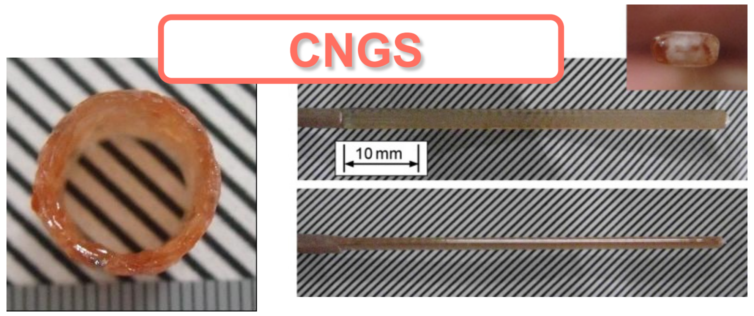


## New μ-PD furnace

### Shaped crystals grown by new μ-PD furnace

**Appearance**

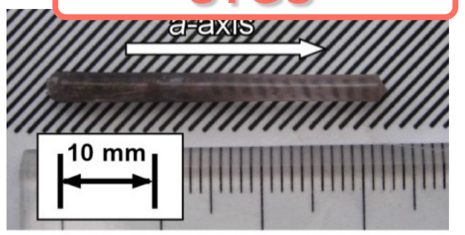
**CNGS**



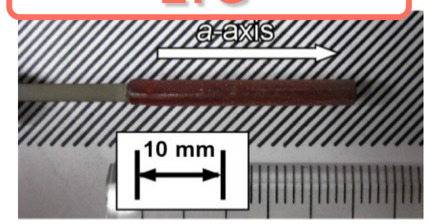

**CTGS**


**SNGS**


**STGS**




**LTG**



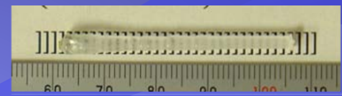

Support Industry by "Ministry of Economy, Trade and Industry"
[illegible]

# Crystal Gallery

## OXIDE

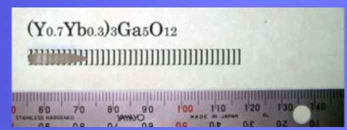
### ～ Garnet type ～

$Y_3Al_5O_{12}$



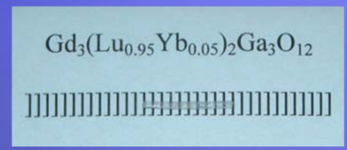
$Lu_3Al_5O_{12}$

$Ce:Lu_3Al_5O_{12}$

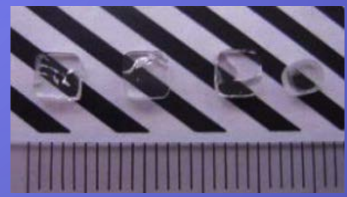



$Y_3Ga_5O_{12}$

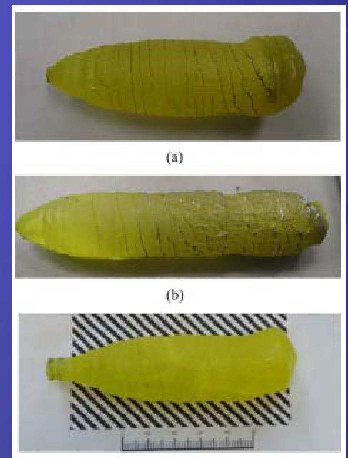
$Ho:Lu_3Al_5O_{12}$



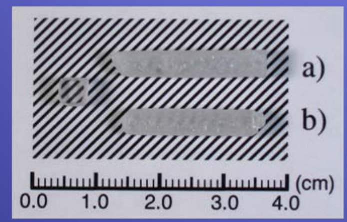

$Gd_3Lu_2Ga_3O_{12}$



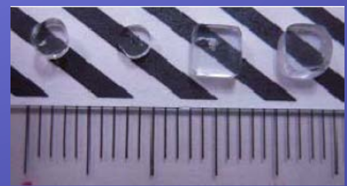
$Tm:Lu_3Al_5O_{12}$



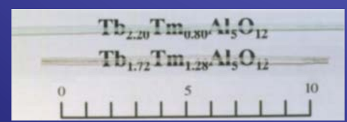

$Ce:GAGG$



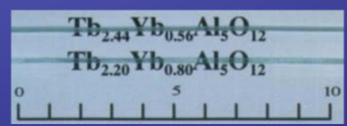

$Lu_3Ga_5O_{12}$



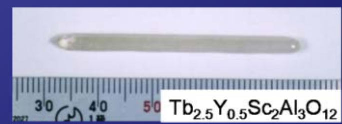
$Nd:Lu_3Al_5O_{12}$



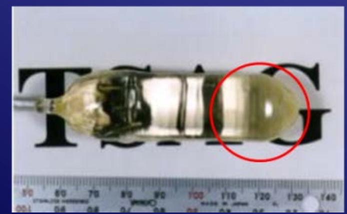
$Tb_{3-x}Tm_xAl_5O_{12}$



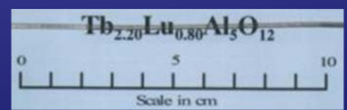
$Tb_{3-x}Yb_xAl_5O_{12}$




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



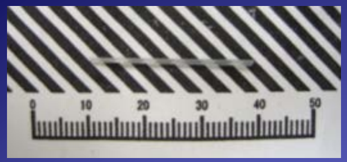
$Tb_{2.2}Sc_{2.8}Al_3O_{12}$




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

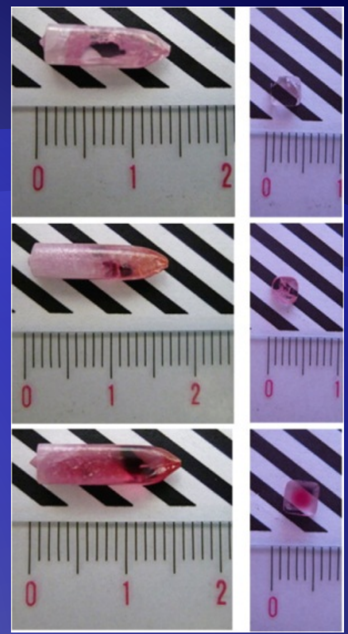
$Tb_{3-x}Sc_{2+x}Al_3O_{12}$

## ～Perovskite type～

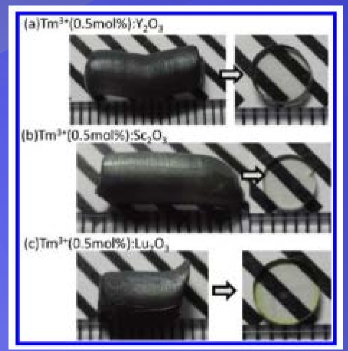


$YAlO_3$

$(Lu, Y)AlO_3$



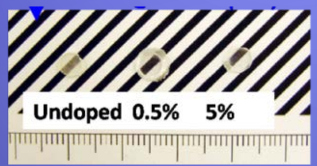

$Tm{:}YAlO_3$



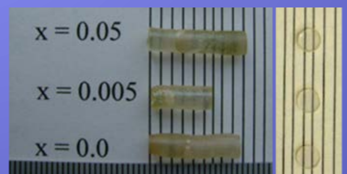
$Cr{:}YAlO_3$

$TbAlO_3$

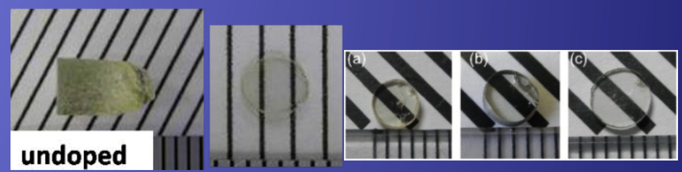
## ～Sesquioxide type～




$Sc_2O_3$

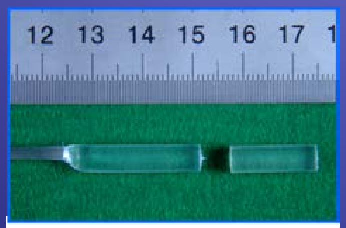



$Y_2O_3$

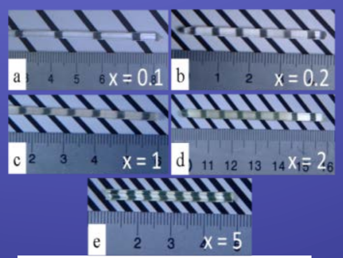



$Lu_2O_3$

## ～Spinel type～

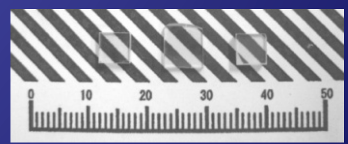


Rod-shaped Ti-doped $MgAl_2O_4$



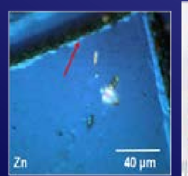

Rod-shaped Mn-doped $MgAl_2O_4$

## ～Vanadate type～

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

## ～ZnO type～



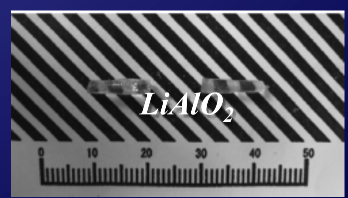
LPE ZnO single crystalline film

ZnO single crystal

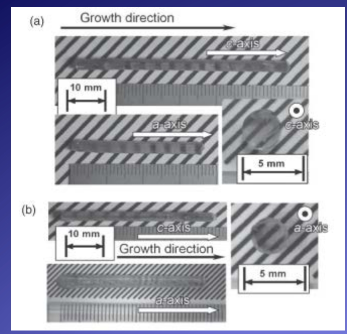
## ～Apatite type～

$Ca_8La_2(PO_4)_6O_2$

## ～Aluminate type～



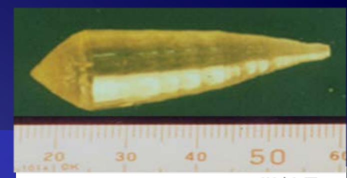

## ～Langasite type～




$Ca_3NbGa_3Si_2O_{14}$ and $Sr_3NbGa_3Si_2O_{14}$

$La_3Ga_{5.3}Al_{0.2}SiO_{14}$(LGAS)単結晶

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



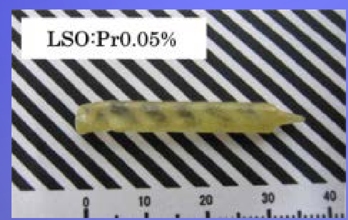
$Sr_3TaGa_3Si_2O_{14}$ (STGS)単結晶

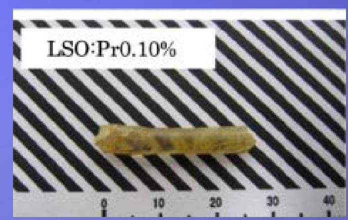
$La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA)単結晶

$La_3Nb_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LNGA)単結晶

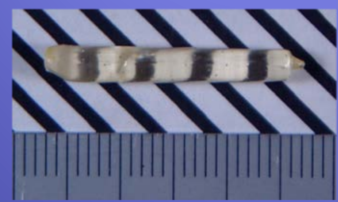
$La_{2.95}Ba_{0.05}Ta_{0.525}Ga_{5.475}O_{14}$ 単結晶

## ～Silicate type～



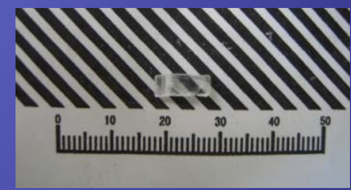







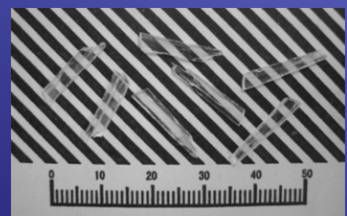
$Lu_2SiO_5$

$Y_2SiO_5$

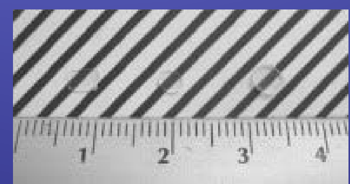


$Gd_2SiO_5$

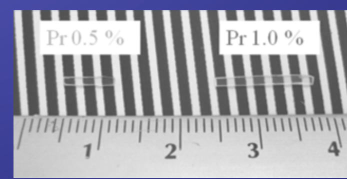
## ～Borate type～



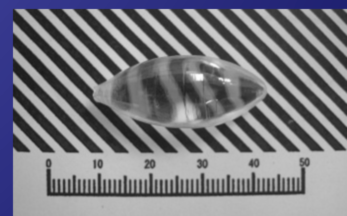
uncoped $CaB_2O_4$



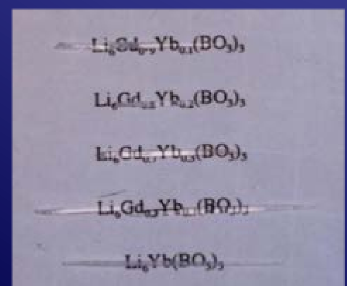
$Tm{:}Ca_3(BO_3)_2$



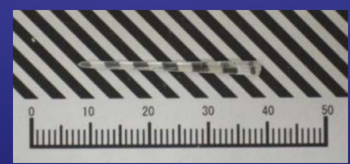

$Pr{:}Ca_3(BO_3)_2$



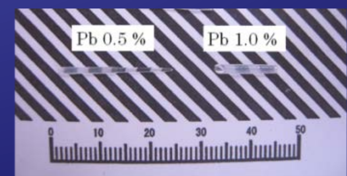
$Ce{:}Ca_3(BO_3)_2$



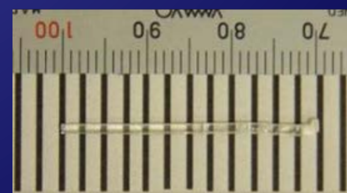
$Li_6Yb(BO_3)_3$, $Li_6Gd(BO_3)_3$



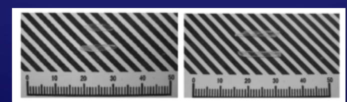
$YCa_4O(BO_3)_3$




$Pb{:}YCa_4O(BO_3)_3$

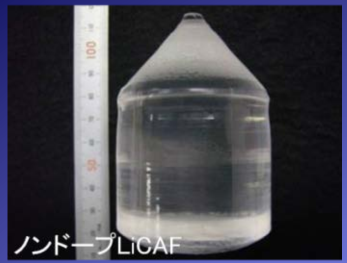


$Li_6Y(BO_3)_3$



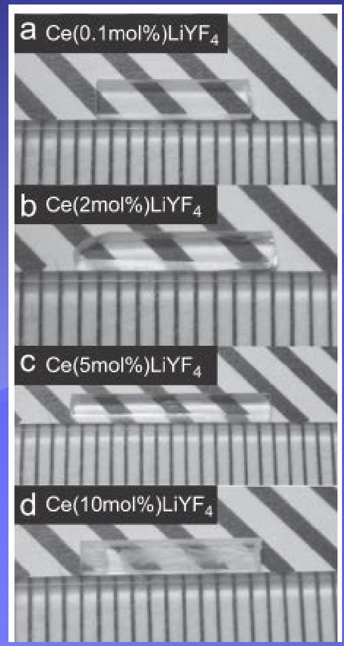
undoped and Ce-doped $:SrB_2O_4$

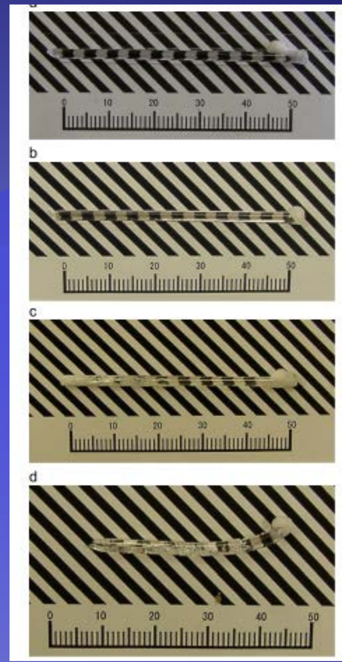
# Fluoride



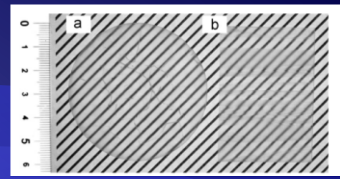


*Ce:LiCAF*

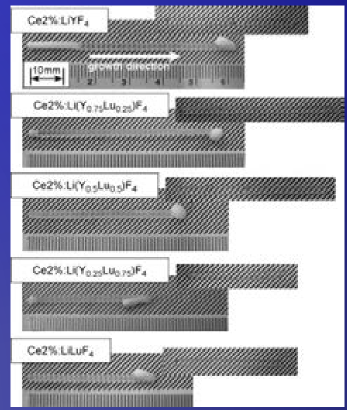


*Ca0.5%, Ce1%, 3%, 7%, 10% co-doped $Y_{0.5}Gd_{0.5}F_3$*

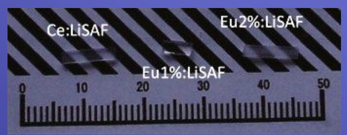


*$LiF/CaF_2$:Eu*

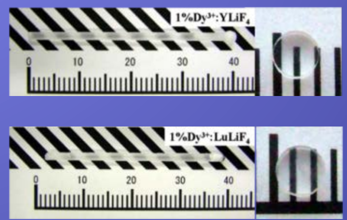




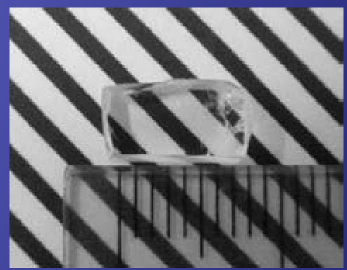
*Eu:LiCAF*



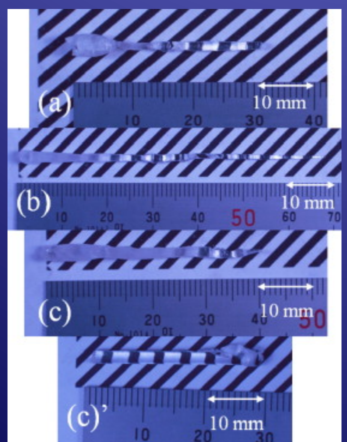

*Eu, Ce:LiSAF*



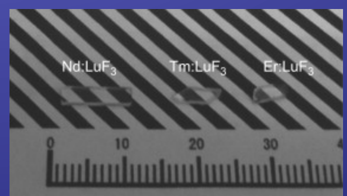
*Ce:YLF*

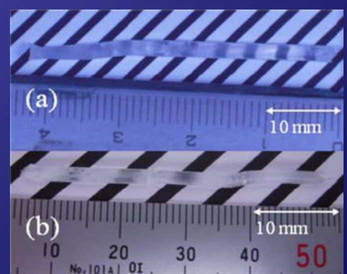


*Sr7%:Ce15%:GdF*



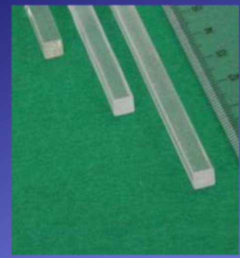

*$(Na_xCa_{1-2x}Lu_x)F_2$ (a: x=1/6, b: x=1/4, c: x=1/3 ) and c':$(Na_xCa_{1-x-y}Lu_{y-0.05}Ce_{0.05})F_{2+y-x}$ (x=0.32,y=0.43)*

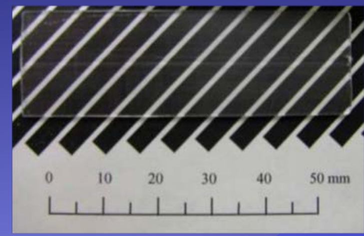


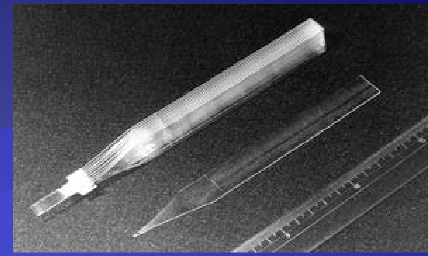



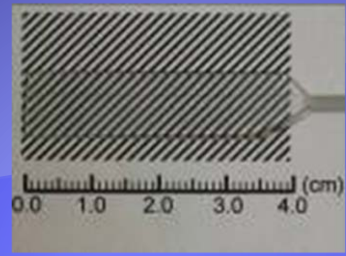

*$K^6Li(Y1_{-x}Ce_x)F_5$ ((a): x = 0.003, (b): x = 0.02)*

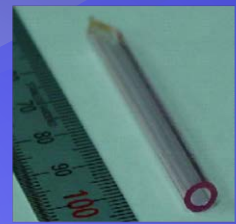
# Shaped Crystal

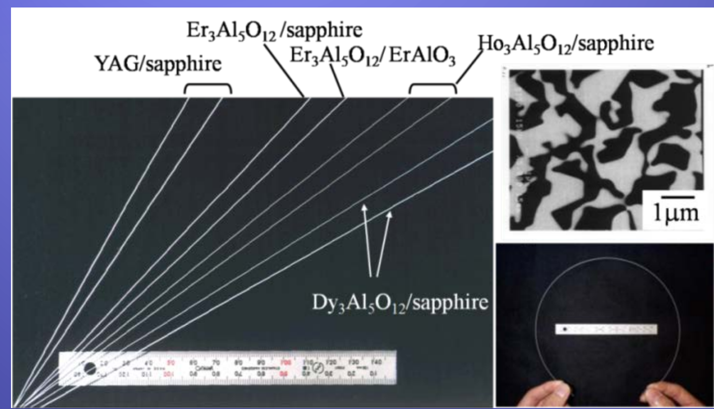








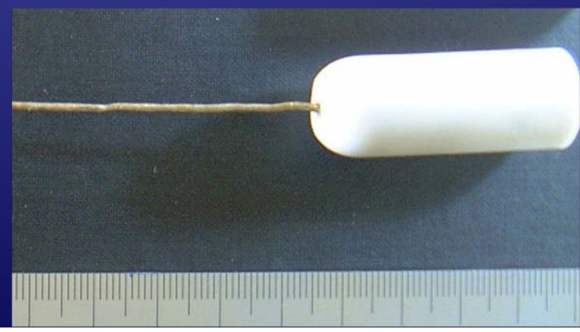








# Others

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)

# *Message from Foreign Participants*

**Message for 2011 annual report from Robert Kral**

(Institute of Physics Academy of Sciences of the Czech Republic, Prague, Czech Republic)

In autumn 2011, I visited Prof. Yoshikawa's laboratory for my fourth time for 10 weeks. Actually, this stay was planed for March 2011, but unfortunately it had to be postponed to autumn 2011 due to the devastating earthquake, which stroke Japan on March $11^{th}$. When the earthquake stroke and the tsunami hit Miyagi area and other prefectures, I was at that time in Vienna international airport waiting for my flight to Narita airport and connecting flight to Sendai. First, I could not believe that it was happening, because the day before I was emailing with my colleague Dr. Pejchal from the laboratory. But after I could not reach him or anyone else from the laboratory, I realized that the situation was very serious. And when I saw pictures of the flooded and burning Sendai airport, there was no possibility I could continue in my trip. I canceled my flight to Japan and returned back to Prague. At home I watched the news where I could see the real extensive and destructive power of the earthquake. It was a relief, when I received a message from Dr. Pejchal that he and everybody in the laboratory were all right and no one was injured.

I returned to Sendai in autumn and I was amazed how quickly Sendai city, Miyagi area and all Japan recovered from the disaster. During this stay, I proceeded in preparation of halide single crystals within continuing collaboration of Dr. Nikl (Institute of Physics Academy of Sciences of the Czech Republic) and Prof. Yoshikawa in their bilateral project "New material conceptions for fast scintillators" (2009-2012) from the Czech Science Foundation. Purpose of this project was to prepare new halide materials by micro-pulling-down method and study their optical properties for scintillation applications. We were focused on preparation of rubidium lead chloride ($RbPb_2Cl_5$) single crystals pure and doped with RE elements such as Nd(III), Yb(III), Pr(III) and Dy(III). All starting materials were synthesized and purified in our Laboratory of Crystal Growth at the Institute of Physics AS CR, Prague, Czech Republic.

This stay was also interesting that I could witness a successful moving of Prof. Yoshikawa's laboratory from its previous place to the new laboratory at the Institute of Material Research (IMR), Tohoku University. I am always looking forward to visit and to work in Prof. Yoshikawa's laboratory, there is a very pleasant atmosphere and support from all the laboratory members whenever I needed help. Each stay, I could learn something new and I could improve my skills in crystal growth.



**Message for 2011 annual report from Jan Pejchal**

After almost 3 years of staying in this research group, the last year was affected by the big earthquake on March 11, 2011. A week after the disaster, I returned to my home country for a while, because my house was affected by later aftershock and I was not sure if it was safe to stay there. In April, when I returned back to Sendai, I was really impressed how the Japanese people could cope with such a big disaster. Japanese nation is very strong and can set a good example to the others. As my stay was planned until the middle of May, I was trying to continue some experiments. Of course the research in the lab was not running in the full extent, as some of the equipment was damaged due to the earthquake. On May $14^{th}$, exactly after 3 years of stay, I returned back to Czech Republic. I would never think that I would give farewell to Japan in this way.

There I continued my research on luminescence of scintillators, still keeping close contacts with Sendai. I had an opportunity to meet the members of Yoshikawa lab in various international conferences, such as International conference on scintillation materials SCINT2011 or International workshop on crystal growth IWCGT5, both held in Germany.

On $1^{st}$ October 2011, I came to Sendai again for 2 months in the framework of planned business trip. However, I was very happy when Prof. Yoshikawa offered me a position for half year. I gladly accepted, as it was again a great opportunity for me to improve my knowledge and skills in crystal growth here in this lab and share my experience on luminescence studies.

In the end of the year a big moving of the whole laboratory from IMRAM to the IMR building was performed. At that time, almost all the equipment was recovered from the damage caused by the earthquake. I really appreciate the effort and good job of all the members of Yoshikawa laboratory.

Another good news was that I could prolong my stay one year more. So I can still continue with research here and more profound my knowledge and experience in this successful research group, while enjoying Japanese life and culture. I hope our cooperation will still continue.

Sincerely Yours

Jan Pejchal

# *Members*

# Members of Yoshikawa Lab

Professor
**A. Yoshikawa**

Technical Counsellor
**H. Uemura**

Research Assistant Professor
**Y. Yokota**

Associate Professor
**T. Yanagida**

Secretary
**A. Imai**

Post Doctral Fellow
**S. Kurosawa**

Post Doctral Fellow
**J. Peichal**

Engineer
**K. Toguchi**

Researcher
**A. Yamaji**

Post Doctral Fellow
**Y. Futami**

Second grades Ph.D student
**Y. Fujimoto**

Third grades Ph.D student
**A. Fukabori**

First grades Master course student
**S. Wakahara**

Second grades Master course student
**M. Sugiyama**

Researcher
**D. Totsuka**
NK&K NIHON KESSHO KOGAKU CO., LTD

Researcher
**N. Kawaguchi**
TOKUYAMA

Researcher
**K. Fukuda**
TOKUYAMA

Researcher
**K. Kamada**
FURUKAWA

Researcher
**M.Sato**
TDK

Research Fellow
**H. Ogino**
東京大学 THE UNIVERSITY OF TOKYO

2011 年度　吉川研究室
**Members (2011 academic year)**

*4. 1. 2011*

| | | | |
|---|---|---|---|
| **Professor** | | **教授** | |
| Dr. | Akira Yoshikawa | 吉川　彰 | NICHe 兼任 |
| **Associate Professor** | | **准教授** | |
| Dr. | Takayuki Yanagida | 柳田　健之 | |
| **Research Assistant Professor** | | **助教** | |
| Dr. | Yuui Yokota | 横田　有為 | |
| **Adviser** | | **顧問** | |
| Prof | Masae Kikuchi | 菊地　昌枝 | |
| Prof | Touetsu Shishido | 宍戸　統悦 | |
| **Post Doctral Fellow** | | **博士研究員** | |
| Dr. | Jan Pejchal | ヤン　ペジャール | Czech |
| Dr. | Shunsuke Kurosawa | 黒澤　俊介 | |
| Dr. | Yoshisuke Futami | 二見　能資 | |
| **Technical Counsellor** | | **技術参事** | |
| | Hiroshi Uemura | 上村　博 | |
| **Secretaries** | | **秘書** | |
| | Akiko Imai | 今井　亜希子 | |
| | Keiko Toguchi | 戸口　景子 | |
| **Researcher** | | **研究員** | |
| | Akihiro Yamaji | 山路　晃広 | |



| | | | |
|---|---|---|---|
| **Graduate Students** | | **大学院生** | |
| | Akihiro Fukabori | 深堀　明博 | D3 |
| | Yutaka Fujimoto | 藤本　裕 | D2 |
| | Makoto Sugiyama | 杉山　誠 | M2 |
| | Shingo Wakahara | 若原　慎吾 | M1 |
| **Researchers** | | **民間等共同研究員** | |
| Dr. | Kentaro Fukuda | 福田　健太郎 | 株式会社トクヤマ／<br>Tokuyama Co., Ltd. |
| | Noriaki Kawaguchi | 河口　範明 | 株式会社トクヤマ／<br>Tokuyama Co., Ltd. |
| Dr. | Kei Kamada | 鎌田　圭 | 古河機械金属株式会社／<br>Furukawa Co., Ltd. |
| | Daisuke Totsuka | 戸塚　大輔 | 日本結晶光学株式会社／<br>Nihon kesshokougaku Co., Ltd. |
| | Masato Sato | 佐藤　真人 | TDK 株式会社<br>TDK Co., Ltd. |
| **Visiting Professors/Researchers** | | **客員教授** | |
| Prof. | Georges Boulon | ジョージ　ブーロン | France |
| Prof. | Martin Nikl | マーチン　ニクル | Czech Republic |
| Dr. | Vladimir V.Kochurikhin | ヴラディミール カチューリッヒン | Russia |

# *Research Life*

I joined the Yoshikawa laboratory one year ago and I am studying the scintillation materials. When I joined, we were moving to IMR and because of the big earthquake before, we had to organize and repair a lot of equipment. So it was hard time for me in the first month. At present, however, I am able to enjoy my research life attending many conferences (both of domestic and international) and writing some scientific papers. I studied the crystal growth and scintillation properties of $LiSrAlF_6$ host crystal for thermal neutron detector. In order to increase the light output of Eu or Ce doped crystal, I grew the alkali metal co-doped ones successfully by Micro-Pulling Down method. Additionally, I studied the phosphorescent properties of ceramics as sub-theme. Nobody in our laboratory has researched the phosphorescent materials, so it was interesting and challenging study for me. I also enjoyed some events such as cherry-blossom, beer party in IMR, ski trip and so on.

All of the members in this laboratory are kind and help me when I need assistance, therefore I really appreciate it and I'll do my best in studies and work for this laboratory until my graduation.

**Shingo Wakahara**
**(M1)**



# *Prizes and Awards*

## 受賞等
## *Prizes and Awards*

1. 横田 有為
   田中貴金属グループ「貴金属に関わる研究助成金」MMS 賞（2011 年 5 月 31 日）

2. 杉山 誠
   ″Nd 添加 LuAG 単結晶を用いた X 線撮像検出器の試作″ 放射線夏の学校　優秀ポスター賞（2011 年 8 月 3 日）

3. 黒澤 俊介
   ″真空紫外線発光シンチレータとガス検出器による 2 次元高計数率検出器の開発″ 放射線夏の学校　最優秀ポスター賞（2011 年 8 月 3 日）

4. 藤本 裕
   第 31 回応用物理学会講演奨励賞（2011 年 11 月 15 日）

5. 杉山 誠
   東北大学工学研究科長賞（2012 年 3 月 21 日）

表彰状

MMS賞

東北大学　横田　有為　殿

あなたは、2010年度「貴金属に関わる研究助成金制度」において、貴金属が貢献できる新しい技術・商品の実用化に向けての研究・開発テーマにふさわしい内容を応募され、審査の結果、頭書の通り優秀な成績を収められましたので、ここにこれを賞します。

2011年5月31日

TANAKAホールディングス株式会社
田中貴金属販売株式会社
田中貴金属インターナショナル株式会社
代表取締役社長　岡本　英彌

東北大学

工修第20902号

# 学 位 記

杉 山　誠

1988年3月23日生

本学大学院工学研究科材料システム工学専攻の
博士課程の前期２年の課程を
修了したので修士（工学）の
学位を授与する

2012年3月27日

東北大学

# *List of Collaborative Research*

## *Visits by International Collaborators 2011*

| Affiliation | Researcher | Research Theme |
|---|---|---|
| General Physics Institute (Russia) | Dr. V. Kochurikhin | Growth of bulk single crystals and automatic diameter control of Czochralski growth |
| CB Lyon1 Univ. (France) | Pr. G. Boulon<br>Dr. T. Epicier | Laser and nonlinear materials and their spectroscopic properties |
| Institute of Physics ASCR (Czech Republic) | Dr. M. Nikl<br>Mr. R. Král | Characterization of various scintillator materials |
| TPS corporation (Korea) | Mr. Won Gun Lee | Growth of bulk single crystals by Czochralski method |
| North –Caucasus State Technical Univ. (Russia) | Pr. B. Sinelnikov | Growth of bulk single crystals |
| Sungkyunkwan Univ. (Korea) | Pr. Dae Ho Yoon | Growth of bulk single crystals by micro-pulling-down method |

## *Seminar at Yoshikawa Laboratory 2011*

| Date | Affiliation | Speaker | Title of speech |
|---|---|---|---|
| March 7 | Institute of Physics ASCR (Czech Republic) | Dr. M. Nikl | LiCaAlF6 – based Scintillators. Luminescence and Scintillation Mechanisms & Defects |



## *Visits to International Collaborators 2011*

| Affiliation | Researcher | Period of stay |
|---|---|---|
| Institute of Physics ASCR (Czech Republic) | Dr. Pejchal | May 9 - October 14<br>(except August 25 - 31) |
| Pennsylvania State University Materials Research Institute (U.S.A) | Pr. Yoshikawa,<br>Dr. Yokota | June 25 - July 6 |
| Technische Universitat Munchen (Germany) | Pr. Yoshikawa | July 25 - 29 |
| DESY in Hamburg (Germany) | Dr. Pejchal | August 25 - 31 |
| Institute of Physics ASCR (Czech Republic) | Pr. Yoshikawa,<br>Assoc. Pr. Yanagida,<br>Dr. Yokota, Dr. Kurosawa<br>Mr. Yamaji, Mr. Wakahara | September 10 - 20 |

# *Research Funds*

# 平成 23 年度 研究資金
# *Research funds (2011 fiscal year)*

【内閣府 (研究管理機関：日本学術振興会)】

The Cabinet Office (In charge: Japan Society for the Promotion of Science)

1. 最先端・次世代研究開発支援プログラム
   Funding Program for Next Generation World-Leading Researchers (NEXT Program)

「次世代癌治療用近赤外線発光シンチレータの系統的研究開発」
"Systematic study of infrared photon emitting scintillators for cancer therapy"

研究代表者：吉川 彰
Project Leader: A. Yoshikawa
メンバー：柳田 健之、横田 有為、藤本 裕、山路 晃広、杉山 誠、戸塚 大輔
Member: T. Yanagida, Y. Yokota, Y. Fujimoto, A. Yamaji, M. Sugiyama, D. Totsuka
150,080,000 yen/3years, 2011.2 ~ 2013.3
( 90,744,000 yen in 2011FY)



【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

「ランガサイト型圧電結晶の形状制御単結晶作製装置及び作製技術の開発」
“Developments of growth furnace and technique on shape-controlled langasite-type piezoelectric single crystals”
プロジェクト副代表者：吉川 彰
Project Sub Leader: A. Yoshikawa (NICHe)
メンバー：横田 有為、二見 能資、佐藤 真人
Member: Y. Yokota, Y. Futami, M. Sato
Total: 98,080,000 yen/1year, 2011.2 ~ 2011.12
( 17,400,000 yen for our team)

「難加工性機能性合金の形状制御結晶育成技術の開発」
“Developments of functional metallic products by shape-controlled growth techniques”
プロジェクト副代表者：吉川 彰
Project Sub Leader: A. Yoshikawa (NICHe)
メンバー：横田 有為、二見 能資
Member: Y. Yokota, Y. Futami
Total: 69,830,000 yen/2years, 2012.1 ~ 2013.3
( 19,580,000 yen for our team)

## 【JSTプロジェクト】

Japan Science and Technology Agency

1. JST研究成果最適展開支援事業（A-STEP）【FS】探索タイプ
   JST Adaptable and seamless technology transfer program through target-driven R&D

   「電荷移動遷移を用いたTOF-PET用シンチレータの開発」
   "Development of new scintillators for TOF-PET based on Charge transfer and Charge transition phenomena"
   プロジェクト代表者：柳田 健之　Project Leader: T. Yanagida(NICHe)
   1,300,000 yen/1year, 2011.8-2012.3

   「形状制御Pr:LuAGシンチレータ単結晶育成技術の開発」
   "Development of shape-controlled crystal growth of Pr:LuAG scintillator"
   プロジェクト代表者：横田 有為　Project Leader: Y. Yokota
   1,300,000 yen/1year, 2011.8-2012.3

   「Pr 添加 YAP シンチレータの高特性化および大口径化研究」
   "Study on Pr:YAP scintillating crystals of greater diameter and improved performance"
   プロジェクト代表者：二見 能資　Project Leader: Y. Futami
   700,000 yen/1year, 2011.12-2012.3

2. JST産学イノベーション加速事業【先端計測分析技術・機器開発】
   要素技術開発
   "Development of Systems and Technology for Advanced Measurement and Analysis Technology"
   "Development Program for Advanced Measurement and Analysis (Program-T)"

   「熱-熱外中性子用高効率シンチレータ検出器の開発」
   (名古屋大学からの再委託研究)
   "Development of highly efficient scintillator for thermal-epithermal neutrons"
   プロジェクト副代表者：柳田 健之　Project Sub Leader: T. Yanagida(NICHe)
   Total: 52,000,000 yen/3years, 2010.9-2014.3
   ( 5,820,000 yen for our team)



## 【文部科学省科学研究費補助金】

Ministry of Education, Culture, Sports, Science and Technology

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. 若手研究（A）Grants-in-Aid for young scientists (A)
   研究代表者: 吉川 彰　Project Leader: A. Yoshikawa
   5,000,000 yen/year, 2011.4-2012.3

   研究代表者: 柳田 健之　Project Leader: T. Yanagida
   5,600,000 yen/year, 2011.4-2012.3

2. 若手研究（B）Grants-in-Aid for young scientists (B)
   研究代表者: 横田 有為　Project Leader: Y. Yokota
   1,500,000 yen/year, 2011.4-2012.3

3. 挑戦的萌芽　Challenging Exploratory Research
   研究代表者: 柳田 健之　Project Leader: T. Yanagida
   1,200,000 yen/year, 2011.4-2012.3

4. 特別研究員奨励費
   研究代表者: 藤本 裕　Project Leader: Y. Fujimoto
   700,000 yen/year, 2011.4-2012.3

   研究代表者: 黒澤 俊介　Project Leader: S. Kurosawa
   800,000 yen/year, 2011.4-2011.3

## 【企業・財団・個人からの受託・共同研究，寄付金及び小型プロジェクト】

## Funds from industry, Foundations, personal donation and small project

1. 株式会社トクヤマ

   Tokuyama Corporation

2. 古河機械金属株式会社

   Furukawa Co. Ltd.

3. TDK 株式会社

   TDK Corporation

4. 日本結晶光学株式会社

   Nihon Kessho Kogaku Co.Ltd

5. 三菱ガス化学株式会社

   Mitsubishi Gas Chemical Co.Ltd

6. キヤノン株式会社

   Canon Inc.

7. 田中貴金属インターナショナル株式会社

   Tanaka Kikinzoku International Co.Ltd

8. (有)ワールドエンジニアリングシステム

   World Engineering System Ltd

9. 公益財団法人 村田学術振興財団

   The Murata Science Foundation

10. 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.

    「中性子検出器用新規シンチレータ結晶の開発」

平成 23 年度申請特許

## *List of patents*

1. シンチレーター及び放射線検出器

   吉川　彰、柳田　健之、横田　有為、福田　健太郎、河口　範明
   (Akira Yoshikawa, Takayuki Yanagida, Yuui Yokota, Kentaro Fukuda, Noriaki Kawaguchi)

2. フッ化物単結晶、発光素子及びシンチレーター

   吉川　彰、柳田　健之、横田　有為、藤本　裕、石津　澄人、河口　範明、福田　健太郎
   (Akira Yoshikawa, Takayuki Yanagida, Yuui Yokota, Yutaka Fujimoto, Sumito Ishizu, Noriaki Kawaguchi, Kentaro Fukuda)

3. Ｘ線シンチレーター用材料

   吉川　彰、柳田　健之、藤本　裕、杉山　誠、戸塚　大輔
   (Akira Yoshikawa, Takayuki Yanagida, Yutaka Fujimoto, Makoto Sugiyama, Daisuke Totsuka)

4. 放射線検出器

   吉川　彰、柳田　健之、鎌田　圭、佐藤　浩樹、堤　浩輔、遠藤　貴範、伊藤　繁記
   (Akira Yoshikawa, Takayuki Yanagida, Kei Kamada, Hiroki Sato, Kousuke Tsutsumi, Takanori Endo, Shigeki Ito)

他　１０　件　（　合計　１４　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**

| | |
|---|---|
| 日本結晶成長学会<br>Japanese Association for Crystal Growth Cooperation | 理事（新技術・新材料分科会担当）<br>Trustee (responsible for new technology and new materials branch) |
| | 編集委員<br>Member of the editorial staff |
| 日本学術振興会第 161 委員会<br>No. 161 committee, Japan Society for the Promotion of Science | 運営委員<br>Manager |
| 応用物理学会<br>The Japan Society of Applied Physics | プログラム編集委員<br>Programming Committee |
| 第 7 回発光・受光及び放射線変換素子に関する国際学会<br>The 7th International Conference on Luminescent Detectors and Transformers of Ionizing Radiation (LUMDETR 2009) | 科学諮問委員<br>Scientific Advisory Committee |
| Europhysical Conference of Defects in Insulating Materials (EURODIM 2010) | 国際諮問委員<br>International Advisory Committee |
| International Conference on Defects in Insulating Materials (ICDIM 2012) | 国際諮問委員<br>International Advisory Committee |



# *List of Presentations*

A. International conference and symposia
国際学会

1. Akira Yoshikawa
"Development of novel rare earth doped fluoride and oxide scintillators, and 2 dimensional imaging "
The Second International Conference on RARE EARTH MATERIALS (REMAT), 2011.6.13-15, Wroclaw, POLAND(Invited)(2011)

2. J. Pejchal, Y. Fujimoto, V. Chani, T. Yanagida, Y. Yokota, A. Yoshikawa, M. Nikl, A. Beitlerova
"Modications of micro-pulling-down method for the growth of selected Li-containing crystals for neutron scintillator and VUV scintillation crystals. "
5th International Workshop on Crystal Growth Technology (IWCGT-5), 2011. 6.26- 6.30, Berlin, Germany(2011)

3. A. Yoshikawa, T. Yanagida, N. Kawaguchi, Y. Fujimoto, Y. Yokota, K. Fukuda, K. Watanabe, A. Yamazaki, A. Uritani, T. Iguchi, M. Nikl
"Crystal growth and scintillation properties of 6LiCAF single crystal, as a candidate for neutron detection alternatives to 3He "
The 16th International Conference on Luminescence and Optical Spectroscopy of Condensed Matter (ICL'11), 2011.6.26- 7. 1, Michigan, USA(Invited)(2011)

4. Martin Nikl, Jiri A. Mares, Akira Yoshikawa, Hiraku Ogino, Karel Nejezchleb, Karel Blazek, Anna Vedda
"Ten Years Of The LuAG-based Scintillators Development. State Of Art And Prospects. "
The 16th International Conference on Luminescence and Optical Spectroscopy of Condensed Matter (ICL'11), 2011.6.26- 7. 1, Michigan, USA(Invited)(2011)

5. Akira YOSHIKAWA, Yuui YOKOTA, Noriaki KAWAGUCHI, Kentaro FUKUDA, Takayuki YANAGIDA, Yutaka FUJIMOTO, Ken-ichi WATANABE, Atsushi YAMAZAKI, Akira URITANI, Tetuo IGUCHI
"Czochralski Growth of RE Doped LiCAF and Their Scintillation Properties as a Candidate for 3He Alternatives "
The 5th Asian Conference on Crystal Growth and Crystal Technology (CGCT-5), 2011.6.27-7.1, Suntec, Singapore (Invited)(2011)



6. Marilou CADATAL-RADUBAN, Toshihiko SHIMIZU, Kohei YAMANOI, Kohei TAKEDA, Kohei SAKAI, Minh HONG PHAM, Tomoharu NAKAZATO, Nobuhiko SARUKURA, Kentaro FUKUDA, Toshihisa SUYAMA, Takayuki YANAGIDA, Akira YOSHIKAWA, Fumio SAITO
"Micro-pulling Down Method-grown Er3+:licaalf6 as Prospective Vacuum Ultraviolet Laser Material "
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43. 横田 有為, 二見 能資, 佐藤 真人, 遠田 一重, 柳田 健之, 小野寺 晃, 吉川 彰
"板状 Ca3NbGa3Si2O14 圧電結晶の結晶育成と特性評価"
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44. 黒澤俊介、柳田祥男、窪 秀利、谷上幸次郎、福田健太郎、河口範明、石津澄人、中川正則、須山敏尚、柳田健之、横田有為、吉川 彰
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45. 杉山誠，柳田健之，藤本裕，横田有為，吉川彰
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2012 年春季 第 59 回 応用物理学関係連合講演会, 2011. 3.15-18，東京，早稲田大学
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46. 戸塚大輔,柳田健之,杉山誠,藤本裕,横田有為,吉川彰
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47. 二見 能資，柳田 健之， 藤本 裕，杉山 誠,Jan Pejchal，黒澤 俊介，横田 有為，吉川彰,伊藤 暁彦，後藤 孝
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48. 若原慎吾，柳田健之，横田有為，藤本裕，杉山誠，二見能資，吉川彰
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# *List of Publications*

1. *JOURNAL OF CERAMIC PROCESSING RESEARCH* Volume: 12 Issue: 2 Pages: 169-172 Published: APR 2011
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JH. Mun , A. Jouini , A. Novoselov , A. Yoshikawa , T. Fukuda

2. *JOURNAL OF CRYSTAL GROWTH* Volume: 320 Issue: 1 Pages: 63-68 Published: APR 1 2011
Crystal growth and characterization of (NaxCa1-2xLux)F-2 single crystals
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3. *NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 634 Issue: 1 Pages: 59-63 Published: APR 1 2011
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4. *OPTICAL MATERIALS* Volume: 33 Issue: 6 Pages: 877-881 Published: APR 2011
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7. *NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 635 Issue: 1 Pages: 53-56 Published: APR 11 2011
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8. *DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 633 Supplement: 1 Pages: S36-S39 Published: MAY 2011
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15. *JAPANESE JOURNAL OF APPLIED PHYSICS* Volume: 50 Issue: 9 Special Issue: SI Article Number: 09ND03 Published: SEP 2011

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T Yanagida , A Yamaji , N Kawaguchi , Y Fujimoto , K Fukuda , S Kurosawa , A Yamazaki , K Watanabe , Y Futami , Y Yokota , A Uritani , T Iguchi , A Yoshikawa , M Nikl

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19. *NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 251-255 Published: OCT 1 2011

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27. *OPTICAL MATERIALS* Volume: 34 Issue: 1 Pages: 75-78 Published: NOV 2011

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28. *APPLIED PHYSICS EXPRESS* Volume: 4 Issue: 12 Article Number: 126402 Published: DEC 2011

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K Kamada , T Yanagida , J Pejchal , M Nikl , T Endo , K Tsutumi , Y Fujimoto , A Fukabori , A Yoshikawa

44. *CRYSTAL GROWTH & DESIGN* Volume: 12 Issue: 1 Pages: 142-146 Published: JAN 2012

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Y Fujimoto , T Yanagida , N Kawaguchi , S Kurosawa , K Fukuda , D Totsuka , K Watanabe , A Yamazaki , Y Yokota , A Yoshikawa

45. *NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 664 Issue: 1 Pages: 127-131 Published: FEB 1 2012

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46. *OPTICAL MATERIALS* Volume: 34 Issue: 4 Pages: 627-631 Published: FEB 2012

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D Totsuka , T Yanagida , M Sugiyama , J Pejchal , Y Fujimoto , Y Yokota , A Yoshikawa

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Crystal growth and scintillation properties of Ce-doped sodium calcium lutetium complex fluoride

S Wakahara , Y Furuya , T Yanagida , Y Yokota , J Pejchal , M Sugiyama , N Kawaguchi , D Totsuka , A Yoshikawa

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M Ieda , T Ishimaru , S Ono , K Yamanoi , M Cadatal-Raduban , T Shimizu , N Sarukura , K Fukuda , T Suyama , Y Yokota , T Yanagida , A Yoshikawa



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T Yanagida , K Fukuda , Y Fujimoto , N Kawaguchi , S Kurosawa , A Yamazaki , K Watanabe , Y Futami , Y Yokota , J Pejchal , A Yoshikawa , A Uritani , T Iguchi

50. *JOURNAL OF INSTRUMENTATION* Volume: 7 Article Number: C03013 Published: MAR 2012

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S Kurosawa , K Taniue , H Sekiya , H Kubo , C Ida , K Miuchi , T Tanimori , T Yanagida , Y Yokota , A Yoshikawa , K Fukuda , N Kawaguchi , S Ishizu , M Nakagawa , T Suyama, J Pejchal

# *Copies of Selected Papers*

*JOURNAL OF CERAMIC PROCESSING RESEARCH* Volume: 12 Issue: 2 Pages: 169-172 Published: APR 2011

## Crystal growth and thermal conductivity of an Tm3+-doped Y2O3 for IR eye-safe laser

JH. Mun [1], A. Jouini [2], A. Novoselov [3], A. Yoshikawa [3], T. Fukuda [3]

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2. Univ Lyon 1, CNRS, UMR 5620, F-69622 Villeurbanne, France
3. Tohoku Univ, Inst Multidisciplinary Res Adv Mat, Aoba Ku, Sendai, Miyagi 9808577, Japan

**Abstract:**

Refractory undoped and Tm3+-doped (0.15, 1, 3 and 5 mol. /0) Y2O3 single crystals were grown by the micro-pulling-down method. Chemical analysis showed a homogeneous distribution of Tm3+ dopant along the crystal rod. The dependence of thermal conductivity on Tm3+ concentration in Tm3+:Y2O3 was characterized. The value decreases when the Tm3+ concentration increases in the host but still stays high enough (7.46 Wm(-1)K(-1)) when doped with Tm3+ (5 mol.%), which represents a promising material for an infrared eye-safe laser application.

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*JOURNAL OF CRYSTAL GROWTH* Volume: 320 Issue: 1 Pages: 63-68 Published: APR 1 2011

## Crystal growth and characterization of (NaxCa1-2xLux)F-2 single crystals

Y Furuya [1], H Tanaka [1], K Fukuda [1,2], N Kawaguchi [1,2], Y Yokota [1], T Yanagida [1], V Chani [1], M Nikl [3], A Yoshikawa [1,4]

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**Abstract:**

(NaxCa1-2xLux)F-2 (x=1/6, 1/4, 1/3) and (NaxCa1-x-yLuy-0.05Ce0.05)F2+y-x (x=0.32, y=0.43) single crystals were grown from the melt using the precise atmosphere controlled type Micro-Pulling-Down (mu-PD) method to investigate their potential as a host material of scintillators. The grown crystals were of single-phase with fluorite-type structure (Fm-3m, Z=4) as confirmed by XRD. The crystals demonstrated 60-90% transmittance above 200 nm wavelength and their absorption edges were approximately 145-155 nm. Their densities were 4.5-5.3 g/cm(3), and their effective atomic numbers were 56.7-61.4. Ce doped sample demonstrated better scintillation properties compared to Ce doped CaF2, such as higher light yield and shorter decay time. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 634 Issue: 1 Pages: 59-63 Published: APR 1 2011

## Time- and wavelength-resolved luminescence evaluation of several types of scintillators using streak camera system equipped with pulsed X-ray source

Y Furuya [1], T Yanagida [1], Y Fujimoto [1], Y Yokota [1], K Kamada [1], N Kawaguchi [1,2], S Ishizu [2], K Uchiyama [3], K Mori [3], K Kitano [4], M Nikl [5], A Yoshikawa [1,6]

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5. Inst Phys ASCR, Prague 16253 6, Czech Republic
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**Abstract:**

To design new scintillating materials, it is very important to understand detailed information about the events, which occurred during the excitation and emission processes under the ionizing radiation excitation. We developed a streak camera system equipped with picosecond pulsed X-ray source to observe time- and wavelength-resolved scintillation events. In this report, we test the performance of this new system using several types of scintillators including bulk oxide/halide crystals, transparent ceramics, plastics and powders. For all samples, the results were consistent with those reported previously. The results demonstrated that the developed system is suitable for evaluation of the scintillation properties. (C) 2011 Elsevier B.V. All rights reserved.

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*OPTICAL MATERIALS* Volume: 33 Issue: 6 Pages: 877-881 Published: APR 2011

## Crystal growth and thermal neutron scintillation response of Ce doped (KLiYF5)-Li-6

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**Abstract:**

(KLi)-Li-6(V1-xCex)F-5(x = 0.003, 0.02) single crystals were grown from the melt using the precise atmosphere control type Micro-Pulling-Down (mu-PD) method to examine their potential as a new thermal neutron scintillators. The grown crystals were single-phase materials as confirmed by XRD. The crystals demonstrated 40-60% transmittance above 320 nm and Ce3+ 5d-4f luminescence observed around 340 nm when exited by a-ray. The radio luminescence measurements under thermal neutron excitation (Cf-252) demonstrated the light yield of 890 (Ph/neutron) and the decay time excited by alpha-ray exhibited 20 and 259 ns.(C) 2011 Elsevier B.V. All rights reserved.

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## Crystal growth and scintillation properties of Nd-doped Lu3Al5O12 single crystals with different Nd concentrations

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**Abstract:**

Nd 0.1%, 0.5%, 1% and 3% doped Lu3Al5O12 (Nd:LuAG) single crystals were grown in the nitrogen atmosphere by the micro-pulling down (mu-PD) method. The grown crystals had a single-phase confirmed by powder XRD analysis. In absorption spectra, some weak absorption lines due to Nd3+ 4f-4f transitions were observed and their intensity increased with the increase of Nd concentration. When excited by Am-241 alpha-ray, a broad emission peak due to defects in the host lattice at 320 nm and some sharp lines due to Nd3+ 4f-4f transitions at wavelength longer than 400 nm were observed. The decay time profiles of Nd:LuAG under gamma-ray excitation were well approximated by two exponential function of 340-760 ns and 3-5 mu s for each sample. By pulse height measurement using Cs-137, Nd 0.5%:LuAG showed the highest light yield of 7600 +/- 760 photons/MeV. (C) 2011 Elsevier B.V. All rights reserved.

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## Crystal growth and scintillation characteristics of the Nd3+ doped LiLuF4 single crystals

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**Abstract:**

Sixty millimeter diameter single crystal of Ne doped LiLuF4 was successfully growh by the Czochralski technique. No remarkable absorption due to unfavorable impurities was observed from optical absorption measurements in the vacuum ultra-violet spectral region. The high crystallinity and homogeneous luminescence characteristics were found from X-ray rocking curve and cathode-ray luminescence respectively. X-ray excited luminescence spectrum was measured and the significant 4f(2)5d-4f(3) luminescence at 182 nm was observed in the grown crystal. The pulse height spectrum was taken upon gamma-ray irradiation. As a result, the grown crystals demonstrated sufficient response to the gamma-ray showing the light yield of 420 +/- 30 photons/MeV. The decay curve under alpha-ray irradiation was also investigated and described by two component decay kinetics which consists of the decay constants of 34 and 450 ns. (C) 2011 Elsevier B.V. All rights reserved.

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 635 Issue: 1 Pages: 53-56 Published: APR 11 2011

## Comparative study of optical and scintillation properties of YVO4, (Lu0.5Y0.5)VO4, and LuVO4 single crystals

Y Fujimoto [1], T Yanagida [1], Y Yokota [1], V Chani [1], VV Kochurikhin [3], A Yoshikawa [1,2]

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**Abstract:**

Optical and scintillation properties of YVO4, (Lu0.5Y0.5)VO4, and LuVO4 single crystals grown by the Czochralski (CZ) method with RF heating system are compared. All vanadate crystals show high transmittance (similar to 80%) in the 400-900 nm wavelength range. In both photo- and radio-luminescence spectra, intense peak around 400-500 nm, which was ascribed to the transition from triplet state of VO43-, was clearly observed. The main decay time component was about 38 mu s (YVO4), 18 mu s ((Lu0.5Y0.5)VO4), and 17 mu s (LuVO4) under 340 nm excitation. The scintillation light yields of YVO4. (Lu0.5Y0.5)VO4, and LuVO4 crystals (obtained from the Cs-137 excited pulse height spectra) were evaluated to be about 11,200, 10,700, and 10,300 ph/MeV, respectively. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 633 Supplement: 1 Pages: S36-S39 Published: MAY 2011

## A new imaging device based on UV scintillators and a large area gas photomultiplier

H Sekiya [1,2], C Ida [3], H Kubo [3], S Kurosawa [3], K Miuchi [3], T Tanimori [3], K Taniue [3], A Yoshikawa [4], T Yanagida [4], Y Yokota [4], K Fukuda [5], S Ishizu [5], N Kawaguchi [5], T Suyama [5]

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**Abstract:**

A new type high spatial resolution radiation detector based on a UV scintillator+a UV imaging gas photomultiplier is presented. The prototype UV detector consists of a 10 cm x 10 cm mu PIC, 2 GEMs and a semitransparent Csl photocathode deposited on a MgF2 window. The effective photo-sensitive area is phi 34 mm and the readouts are 400 mu m pitch strips. A newly developed 20 mm size LaF3(Nd) crystal which emits 172 nm photons is coupled to the detector for the first step. The detector was tested in pulse mode operation with 5.5 MeV alpha particles from Am-241. The single photoelectrons were successfully detected and the images of the crystal shape were clearly obtained. 

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*CRYSTAL GROWTH & DESIGN* Volume: 11 Issue: 6 Pages: 2404-2411 Published: JUN 2011

## Growth of Tm3+-Doped Y2O3, Sc2O3, and Lu2O3 Crystals by the Micropulling down Technique and Their Optical and Scintillation Characteristics

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**Abstract:**

Single -crystal growth of Tm3+-doped Y2O3, Sc2O3, and Lu2O3 sesquioxides, with a melting point around 2400 degrees C, by the micropulling down technique is reported. Crystal quality and composition of the crystals were evaluated by rocking curve and energy-dispersive X-ray (EDX) analyses, respectively. Optical properties including optical transmittance and photoluminescence decay profiles were also examined. Furthermore, scintillation performance of the crystals including pulse height spectra and radioluminescence spectra under alpha-ray and gamma-ray excitation was evaluated.

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## Crystal growth and optical properties of the Nd3+ doped LuF3 single crystals

K Fukuda [1,2], S Ishizu [2], N Kawaguchi [1,2], T Suyama [2], T Yanagida [1], Y Yokota [1], M Nikl [3], A Yoshikawa [1,4]

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**Abstract:**

Thirty millimeter diameter single crystal of Nd3+ doped LuF3 was grown using LiF as solvent. The single phase crystallization was confirmed by the powder X-ray diffraction, and high structural perfection was demonstrated by X-ray rocking curve (XRC) measurements. FWHM of XRC for 220 reflection was 32 arcsec. No remarkable absorption due to unfavorable impurities was observed from optical absorption measurements in the VUV spectral region. The crystal showed the VUV luminescence peaking around 178 nm that is consistent with the 4f(2)5d-4f(3) transition of Nd3+ ion. The luminescence intensity of Nd:LuF3 under X-ray irradiation was significantly higher than that of reported VUV scintillators such as Nd:LaF3 or Nd:LiLuF4. 

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*OPTICAL MATERIALS* Volume: 33 Issue: 8 Pages: 1243-1247 Published: JUN 2011

## Basic study of Europium doped LiCaAlF6 scintillator and its capability for thermal neutron imaging application

T Yanagida [1], N Kawaguchi [2,4], Y Fujimoto [4], K Fukuda [2], Y Yokota [4], A Yamazaki [3], K Watanabe [3], J Pejchal [4,5], A Uritani [3], T Iguchi [3], A Yoshikawa [1,4]

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**Abstract:**

Eu2+ 0.1, 0.5, 1, and 2 mol% doped LiCaAlF6 single crystalline scintillators were grown by the micro-pulling down (mu-PD) method. Eu2+ 2 mol% doped LiCaAlF6 was also prepared using the Czochralski method. In the transmittance spectra, 4f-5d absorption lines appeared around 200-220 and 290-350 nm. An intense emission at 375 nm due to Eu2+ 5d-4f transition was observed under Am-241 alpha-ray excitation. When Cf-252 excited pulse height spectra were measured, Eu 2% doped one showed the highest light yield of 29,000 ph/n with 1.15 mu s decay time. Using the 2 inch phi Czochralski grown one coupled with the position sensitive photomultiplier tube covered by Cd mask with various size (1, 2, 3, and 5 mm) pin holes, thermal neutron imaging was examined. As a result, the spatial resolution turned out to be better than 1 mm. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 643 Issue: 1 Pages: 57-63 Published: JUL 1 2011

## The development and performance of UV-enhanced APD-arrays for high resolution PET imaging coupled with pixelized Pr:LuAG crystal

M Yoshino [1], J Kataoka [1], T Nakamori [1], H Matsuda [1], T Miura [1], T Katou [1], Y Ishikawa [2], N Kawabata [2], Y Matsunaga [2], K Kamada [3], Y Usuki [3], A Yoshikawa [4], T Yanagida [4]

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**Abstract:**

The development of high-resolution, UV-enhanced avalanche photodiode (APD) arrays usable in high-resolution PET imaging is underway. These APD arrays were specifically designed as photosensors capable of direct coupling with pixelized Pr-doped Lu3Al5O12 (Pr:LuAG) scintillators. An excellent quantum efficiency (QE) of 55% was achieved at the peak emission of Pr:LuAG (310 nm), namely, a substantial improvement from the QE <= 5% as measured with the conventional Hamamatsu reverse-type APDs (S8664 series). Each APD device has 8 x 8 (TYP1) and 12 x 12 (TYP2) pixel structures with active areas of 3 x 3 mm(2) and 2 x 2 mm(2) in each pixel, respectively. A gain uniformity of +/- 8% and low dark noise of <= 2 nA/pixel have been achieved, measured at +25 degrees C. We also report on the large size single crystal growth of improved Pr:LuAG scintillators and the preliminary performance test of the same. An energy resolution of 4.2% (FWHM) was obtained for 662 key gamma-rays for 10 x 10 x 10 mm(3) crystal, measured with a PMT employing a super-bialkali photocathode. We made a test module consisting of a UV-enhanced APD-array (either TYP1 or TYP2) optically coupled with an 8 x 8 (or 12 x 12) pixel Pr:LuAG matrix. The linearity between the output signals and incident gamma-ray energy of TYP1 and TYP2 gamma-ray detectors were only 0.27 and 0.33%, as measured at +25 degrees C for various gamma-ray sources, respectively. Energy resolutions of 7.0 +/- 0.2% (FWHM) and 9.0 +/- 0.6% (FWHM) were, respectively, obtained for TYP1 and TYP2 detector arrays for 662 keV gamma-rays. The uniformity of the pulse height distributions was also measured at less than 8% for both detectors. Finally, we measured the coincidence timing resolution of these gamma-ray detectors and obtained 4.0 +/- 0.1 ns (FWHM) for the 511 key annihilation quanta from a Na-22 source. These results suggest that UV-enhanced APD-arrays coupled with Pr:LuAG scintillators could be a promising device for future application in nuclear medicine. 

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## Crystal growth and scintillation properties of Pr-doped oxyorthosilicate for different concentration

D Totsuka [1,2], T Yanagida [4], Y Fujimoto [1], J Pejchal [1,3], Y Yokota [1], A Yoshikawa [1,4]

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**Abstract:**

0.05, 0.1 and 0.25 mol% Pr (with respect to Lu) doped Lu(2)SiO(5) (LSO) single crystals were grown by the micro-pulling down (mu-PD) method. The grown crystals were transparent, and a slight segregation of Pr(3+) was observed both in the crystal cross-section and growth direction. Transparency in the visible wavelength range was about 80% in all the crystals. Intense absorptions related with the Pr(3+) 4f-5d transitions were observed around 230 and 255 nm, and weak absorptions due to the 4f-4f transitions were detected around 450 nm. In radioluminescence spectra, the Pr(3+) 5d-4f transitions were observed around 275and 310 nm, and emissions due to the 4f-4f transition were observed around 500 nm. In the pulse height analysis using (137)Cs gamma-ray excitation, Pr 0.1% doped sample showed the highest light yield of 2,800 ph/MeV. In the decay time measurements using different excitation sources (photoluminescence, X- and gamma-ray), two different processes related to the 5d-4f emission peaks were found. Fast decay component corresponds to direct excitation of Pr(3+) (4-6 ns) and slower component (25 ns) reflects the energy migration process from the host lattice to the emission center. (C) 2011 Elsevier B.V. All rights reserved.

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## Absence of Host Cation Segregation in the (Gd,Y)(3)Al5O12 Mixed Garnet Optical Ceramics

G Boulon [1,2], T Epicier [3], W Zhao [1,4], VI Chani [2], T Yanagida [5], A Yoshikawa [2,5]

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**Abstract:**

Recently non-uniform distribution of dopants in transparent optical ceramics was observed. It was demonstrated that dopants with low segregation coefficients (K < 0.2) with respect to the melt crystal growth are segregated to the grain boundaries when the grains of the ceramics are developed. This communication discusses segregation of host cations that form crystal matrix. Experimental examination of spatial distribution of Gd3+ and Y3+ host cations in Ce3+-doped (Gd, Y)(3)Al5O12 ceramic sample demonstrated that no detectable redistribution of Gd3+ and Y3+ cations in the vicinity of the grain boundaries was detected. These results are in good agreement with comparable segregation behavior of Gd3+ and Y3+ [K(Y3+ approximate to K(Gd3+] when the garnet solidification is performed from liquid phase. This observation also confirms qualitative match of segregation in liquid and solid states. (C) 2011 The Japan Society of Applied Physics

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*JAPANESE JOURNAL OF APPLIED PHYSICS* Volume: 50 Issue: 9 Special Issue: SI Article Number: 09ND03 Published: SEP 2011

## Growth of Shape-Controlled Ca3NbGa3Si2O14 and Sr3NbGa3Si2O14 Single Crystals by Micro-Pulling-Down Method and Their Physical Properties

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**Abstract:**

Shape-controlled langasite-type piezoelectric single crystals, Ca3NbGa3Si2O14 (CNGS) and Sr3NbGa3Si2O14 (SNGS), were grown by the micro-pulling-down (mu-PD) method and their physical properties were investigated. Columnar-shaped CNGS and SNGS crystals with a-and c-axes in the growth direction were grown and the diameters of the grown columnar-shaped crystals were controlled to be approximately 3mm from the initial to the later part. The grown crystals without cracks indicated relatively high crystallinities and a single phase of the langasite-type structure in the powder X-ray diffraction patterns. Meanwhile, in the outside area of the crystals, parts of second phases were detected and energy dispersive X-ray spectroscopy measurements indicated that the second phases of the CNGS and SNGS crystals were the Ca-Nb-O and Sr-Nb-O systems, respectively. The d(11) of the CNGS crystal indicated 3.98 pC/N, which was coincident with those of previous reports. (C) 2011 The Japan Society of Applied Physics

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## Monophase domain, fibers single crystals grown by the micro-pulling down technique and optical characterisation of LiGd1-xYbx(WO4)(2)

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**Abstract:**

We have determined the single phase domain of LiGd1-xYbx(WO4)(2). The lattices parameters decrease as a function of Yb3+ substitution in Gd' sites. Transparent LiGd(1-x)Ab(x)(WO4)(2) fibers single crystals were successfully grown by the micro-pulling down technique (mu-PD). The Yb3+-doped LiGd(WO4)(2) fibers single crystals have been pulled under stationary stable growth conditions corresponding to flat crystallization interface with meniscus length equal to 120 mu m. The fibers diameters varied from 0.5 to 1 mm depending on the capillary die diameter, pulling rate and the molten zone temperature. Fibers single crystals free of defects are observed for Ytterbium concentration in the melt up to 5 at%. Above this limit, inclusions and cracks appear and the optical quality of the fibers were deteriorated. The emission spectra of Yb3+-doped LiGd(WO4)(2) were investigated. (C) 2011 Elsevier B.V. All rights reserved.

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## Europium and Sodium Codoped LiCaAlF6 Scintillator for Neutron Detection

T Yanagida [1], A Yamaji [4], N Kawaguchi [2,4], Y Fujimoto [4], K Fukuda [2], S Kurosawa [4], A Yamazaki [3], K Watanabe [3], Y Futami [4], Y Yokota [4], A Uritani [3], T Iguchi [3], A Yoshikawa [1,4], M Nikl [5]

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**Abstract:**
A recent study has revealed that a Eu-doped LiCaAlF6 (Eu:LiCAF) crystalline scintillator showed good scintillation response under thermal neutron exposure. We discover here that Eu and Na codoped LiCAF exhibits superior scintillation properties to Eu: LiCAF. We grew Eu 2% and Na 0.5, 1, and 2% codoped LiCAF by the micropulling down method. In radioluminescence spectra, they showed intense emission peaking at 370nm due to Eu2+ 5d-4f transition. In particular, the light yield of Eu 2% Na 2%-doped LiCAF reached 40000 ph/n, which was about 30% higher than that of Eu: LiCAF. (C) 2011 The Japan Society of Applied Physics

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*CRYSTAL GROWTH & DESIGN* Volume: 11 Issue: 10 Pages: 4484-4490 Published: OCT 2011

## Composition Engineering in Cerium-Doped (Lu,Gd)(3)(Ga,Al)(5)O-12 Single-Crystal Scintillators

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**Abstract:**
The Ce-doped (LuyGd1-x)(3)(Ga-y,Al1-y)(s)O-12 single crystals were grown by the micropulling down method. Their structure and chemical composition were checked by X-ray diffraction (XRD) and electron probe microanalysis (EPMA) techniques. Optical, luminescent, and scintillation characteristics were measured by the methods of time-resolved luminescence spectroscopy, including the light yield and scintillation decay. Balanced Gd and Ga admixture into the Lu3Al5O12 structure provided an excellent scintillator where the effect of shallow traps was suppressed, the spectrally corrected light yield value exceeded 40 000 photons/MeV, and scintillation decay was dominated by a 53 ns decay time value which is close to that of Ce3+ photoluminescence decay. This study provides an excellent example of a combinatorial approach where targeted single-crystal compositions are obtained by a flexible, time saving, and cost-effective crystal growth technique.

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 251-255 Published: OCT 1 2011

## Scintillation properties of Ce-doped LuLiF4 and LuScBO3

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**Abstract:**

The crystals of 1 mol% Ce-doped LuLiF4 (Ce:LLF) grown by the micro-pulling down (mu-PD) method and 1 mol% Ce-doped LuScBO3 (Ce:LSBO) grown by the conventional Czochralski (Cz) method were examined for their scintillation properties. Ce:LLF and Ce:LSBO demonstrated similar to 80% transparency at wavelengths longer than 300 and 400 nm, respectively. When excited by Am-241 alpha-ray to obtain radioactive luminescence spectra, Ce3+ 5d-4f emission peaks were detected at around 320 nm for Ce:LLF and at around 380 nm for Ce:LSBO. In Ce:LSBO, the host luminescence was also observed at 260 nm. By recording pulse height spectra under gamma-ray irradiation, the absolute light yield of Ce:LLF and Ce:LSBO was measured to be 3600 +/- 400 and 4200 +/- 400 ph/MeV, respectively. Decay time kinetics was also investigated using a pulse X-ray equipped streak camera system. The main component of Ce:LLF was similar to 320 ns and that of Ce:LSBO was similar to 31 ns. In addition, the light yield non-proportionality and energy resolution against the gamma-ray energy were evaluated. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 256-259 Published: OCT 1 2011

## Basic study of single crystal fibers of Pr:Lu3Al5O12 scintillator for gamma-ray imaging applications

T Yanagida [1], K Kamada [2], N Kawaguchi [1,3], Y Fujimoto [1], K Fukuda [1,3], Y Yokota [1], V Chani [1], A Yoshikawa [1]

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**Abstract:**

Single-crystalline fibers were grown from 0.25, 0.70, and 1.50 mol% Pr-doped Lu3Al5O12 (LuAG) melts by the micro-pulling down (mu-PD) method with a diameter of 0.3-0.5 mm and a length of about 200 mm. They were cut to 10 mm long specimens, and their scintillation properties, including light yield and decay time profile, were examined. These results were compared with corresponding properties of the specimens (0.8 x 0.8 x 10 mm(3)) cut from the bulk crystals produced by conventional Czochralski (CZ) growth. The mu-PD-grown fibers demonstrated relatively low light yield and had the same decay time constant when compared with those of the samples cut from the CZ-grown crystals. The fiber crystals were used to assemble scintillating arrays with dimensions of empty set 0.5 mm(2) x 20 pixels and empty set 0.3 x 10 mm(2) x 30 pixels coated by a BaSO4 reflector. After optical coupling with a position sensitive photomultiplier tube, the fiber-based arrays demonstrated acceptable imaging capability with a spatial resolution of about 0.5 mm. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 319-322 Published: OCT 1 2011

## Thermal neutron detection with Ce3+ doped LiCaAlF6 single crystals

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**Abstract:**

Cerium-doped LiCaAlF6 (Ce:LiCAF) crystals have been studied as scintillators in application to thermal neutron detection. Three crystals: high-doping Ce:LiCAF, low-doping Ce:LiCAF with 50% enrichment of Li-6 (both 10 mm x 10 mm x 2 mm, rectangular) and high-doping Ce:LiCAF with 95% enrichment of Li-6 (050.8 mm x 2 mm, discus) coupled to Photonis XP5300B PMT, were tested. The response of these crystals to neutrons emitted from a paraffin moderated (PuBe)-Pu-238 source has been investigated. Thermal neutron peaks have been found at a Gamma Equivalent Energy (GEE) of similar to 2.5 MeV for high-doping Ce:LiCAF (50% 6Li), similar to 2 MeV for low-doping Ce:LiCAF (50% Li-6) and similar to 1.9 MeV for high-doping Ce:LiCAF (95% Li-6). The light output of Ce:LiCAF was also measured (175-250 phe/MeV from sample to sample). Lithium-6 glass GS20 from Saint Gobain was used as a reference scintillator (050 mm x 2 mm, circle). Relative neutron efficiency, normalized to that of GS20 lithium glass, as well as gamma-neutron intrinsic efficiency for all tested samples was calculated. Intrinsic efficiency on thermal neutron detection for small Ce:LiCAF samples was estimated at about 32-35% of that of GS20 and for large Ce:LiCAF sample as about 82% of that of GS20. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 351-354 Published: OCT 1 2011

## Thermal neutron imaging with rare-earth-ion-doped LiCaAlF6 scintillators and a sealed Cf-252 source

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**Abstract:**

Thermal neutron imaging with Ce-doped LiCaAlF6 crystals has been performed. The prototype of the neutron imager using a Ce-doped LiCaAlF6 scintillating crystal and a position sensitive photomultiplier tube (PSPMT) which had 64 multi-channel anode was developed. The Ce-doped LiCaAlF6 single crystal was grown by the Czochralski method. A plate with dimensions of a diameter of 50 x 2 mm(2) was cut from the grown crystal, polished, and optically coupled to PSPMT by silicone grease. The Cf-252 source ( <1 MBq) was sealed with 43 mm of polyethylene for neutron thermalization. Alphabet-shaped Cd pieces with a thickness of 2 mm were used as a mask for the thermal neutrons. After corrections for the pedestals and gain of each pixel, we successfully obtained two-dimensional neutron images using Ce-doped LiCaAlF6. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 435-438 Published: OCT 1 2011

## Neutron-gamma discrimination based on pulse shape discrimination in a Ce:LiCaAlF6 scintillator

A Yamazaki [1], K Watanabe [1], A Uritani [1], T Iguchi [2], N Kawaguchi [3], T Yanagida [4], Y Fujimoto [4], Y Yokota [4], K Kamada [4], K Fukuda [3], T Suyama [3], A Yoshikawa [4,5]

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**Abstract:**

We demonstrate neutron-gamma discrimination based on a pulse shape discrimination method in a Ce:LiCAF scintillator. We have tried neutron-gamma discrimination using a difference in the pulse shape or the decay time of the scintillation light pulse. The decay time is converted into the rise time through an integrating circuit. A Cf-252 enclosed in a polyethylene container is used as the source of thermal neutrons and prompt gamma-rays. Obvious separation of neutron and gamma-ray events is achieved using the information of the rise time of the scintillation light pulse. In the separated neutron spectrum, the gamma-ray events are effectively suppressed with little loss of neutron events. The pulse shape discrimination is confirmed to be useful to detect neutrons with the Ce:LiCAF scintillator under an intense high-energy gamma-ray condition. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 209-211 Published: OCT 1 2011

## Fabrication and characterization of large size (LiF)-Li-6/CaF2:Eu eutectic composites with the ordered lamellar structure

N Kawaguchi [1], K Fukuda [1], T Yanagida [2], Y Fujimoto [2], Y Yokota [2], T Suyama [1], K Watanabe [3], A Yamazaki [3], A Yoshikawa [2,4]

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**Abstract:**

As alternative candidates for the He-3 neutron detectors, (LiF)-Li-6/CaF2:Eu eutectic composites were fabricated and their scintillation properties were evaluated. Large size LiF/CaF2:Eu eutectic composites of 58 mm diameter and 50 mm thickness were produced by Bridgman method. The composites had a finely ordered lamellar structure along the solidification direction. The lamellar structure was controlled by the direction and the rate of solidification, and it was optimized to improve the scintillation properties. Better results were achieved when thinner lamellar layers were aligned along the scintillation light path. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 652 Issue: 1 Pages: 238-241 Published: OCT 1 2011

## Scintillation and optical properties of Pb-doped YCa4O(BO3)(3) crystals

Y Fujimoto [1,2], T Yanagida [1], Y Yokota [1], N Kawaguchi [4], K Fukuda [1,4], D Totsuka [5], K Watanabe [6], A Yamazaki [6], V Chani [1], A Yoshikawa [1,3]

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**Abstract:**

This communication reports optical properties and radiation responses of Pb2+ 0.5 and 1.0 mol%-doped YCa4O(BO3)(3) (YCOB) single crystals grown by the micro-pulling-down (mu.-PD) method for neutron scintillator applications. The crystals had no impurity phases according to the results of X-ray powder diffraction. These Pb2+-doped crystals demonstrated blue-light luminescence at 330 nm because of Pb2+ S-1(0)-P-3(0.1) transition in the photoluminescence spectra. The main emission decay component was determined to be about 250-260 ns under 260 nm excitation wavelength. When irradiated by a Cf-252 source, the relative light yield of 0.5% Pb2+-doped crystal was about 300 ph/n that was determined using the light yield of a reference Li-glass scintillator. 

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*CRYSTAL GROWTH & DESIGN* Volume: 11 Issue: 11 Pages: 4775-4779 Published: NOV 2011

## Crystal Growth of Na-Co-Doped Ce:LiCaAlF6 Single Crystals and Their Optical, Scintillation, and Physical Properties

Y Yokota [1], Y Fujimoto [1], T Yanagida [2], H Takahashi [3], M Yonetani [4], K Hayashi [4], I Park [4], N Kawaguchi [1,5], K Fukuda [5], A Yamaji [1], Y Fukazawa [4], M Nikl [6], A Yoshikawa [1,2]

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**Abstract:**

We have grown Ce- and Na-co-doped LiCaAlF6 (Ce,Na:LiCAF) single crystals with various Ce and Na concentrations by the micro-pulling-down (mu-PD) method. Physical properties such as crystallinity, transmittance, photoluminescence, scintillation, and radiation resistance were investigated to evaluate the effects of charge compensation by Na co-doping in the Ce-doped LiCaAlF6 (Ce:LiCAF) crystal. Ce2%:LiCAF, Ce1%Na1%:LiCAF, Ce1.5%Na1.5%:LiCAF, and Ce2%Na2%:LiCAF crystals with no visible cracks and inclusions were prepared. The Ce1%Na1%:LiCAF crystal showed high crystallinity comparable to the crystal grown by the Czochralski method. In the transmittance and photoluminescence spectra of all crystals, an absorption peak around 270 nm and emission peaks around 285 and 310 nm originating from Ce3+ ions were observed, respectively. With increasing Ce and Na concentrations, the light yield systematically increased, and scintillation decay times decreased. Improvement of radiation resistance accomplished by charge compensation due to Na co-doping is clearly demonstrated.

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## Crystal growth and scintillation properties of (NaxCa1-2xLux-yNdy)F-2 single crystals

Y Furuya [1], S Wakahara [1], M Sugiyama [1], H Tanaka [1], K Fukuda [1,2], N Kawaguchi [1,2], Y Yokota [1], T Yanagida [3], A Yoshikawa [1,3]

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**Abstract:**

NaxCa1-2xLux-yNdyF2 single crystals were grown from the melt using the precise atmosphere control type Micro-Pulling-Down (mu-PD) method to investigate their potential as a vacuum-ultraviolet (VUV) scintillators. The grown crystals were single-phase materials with fluorite-type structure (Fm-3m, Z = 4) as confirmed by XRD. The crystals demonstrated 80-90% transmittance above 200 nm wavelength and Nd3+ 5d-4f luminescence (when exited by X-ray) observed around 185 nm. The radioluminescence measurements under 5.5 MeV alpha-ray excitation (Am-241) demonstrated the light yield of 48 [Ph/5.5 MeV-alpha] and the decay time of 6.4-7.7 ns. 

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## Ultrafast Transparent Ceramic Scintillators Using the Yb3+ Charge Transfer Luminescence in RE2O3 Host

T Yanagida [1], Y Fujimoto [2], S Kurosawa [2], K Watanabe [4], H Yagi [3], T Yanagitani, [3], V Jary [5], Y Futami [2], Y Yokota [2], A Yoshikawa [1,2], A Uritani [4], T Iguchi [4], M Nikl [5]

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**Abstract:**

We report a new discovery of Yb3+-doped ultrafast scintillators based on the Yb3+ charge transfer luminescence. Transparent ceramic Yb3+-doped Y2O3, Sc2O3, Lu2O3, and Yb2O3 were prepared by sintering. When irradiated by gamma- and X-rays, they showed a well detectable photoabsorption peak in the pulse height spectra and ultrafast scintillation decay dominated by the decay time of about 1 ns. For the first time, the Yb3+-doped oxide materials show detectable scintillation in the pulse height measurement at room temperature with ultrafast scintillation decay. 

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## Evaluations of pure and ytterbium doped transparent ceramic complex perovskite scintillators

T Yanagida [1], Y Fujimoto [2], Y Yokota [2], A Yoshikawa [1,2], S Kuretake [3], Y Kintaka [3], N Tanaka [3], K Kageyama [3], V Chani [2]

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**Abstract:**
Optical and scintillation properties of complex perovskite transparent ceramic scintillators of pure and Yb3+ doped Ba(ZrMgTa)O-3 and (LaSr)(AlTa)O-3 ceramics are reported. The materials were produced by Murata Manufacturing. Their optical properties including transmittance and photoluminescence spectra were evaluated. The ceramics demonstrated high transparency (50-70%) in visible wavelength region. Additionally, strong emission peaks were observed at 470 nm in Ba(ZrMgTa)O-3 under 284 nm excitation and at 500 nm in (LaSr)(AlTa)O-3 under 324 nm excitation. The photoluminescence decay times of Ba(ZrMgTa)O-3 and (LaSr)(AlTa)O-3 samples were 14 and 16 mu s, respectively. Judging from these optical properties, Yb3+ emission was not observed in these materials. In radio luminescence spectra, all specimens exhibited the same emission peaks with relatively higher emission intensity. Am-241 5.5 MeV alpha-ray induced pulse height spectra of the samples were also measured, and (LaSr)(AlTa)O-3 demonstrated similar to 500 ph/5.5 MeV alpha with 10 mu s shaping time constant. 

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## Scintillation properties of Tm-doped Lu3Al5O12 single crystals

M Sugiyama [1], Y Fujimoto [1], T Yanagida [2], D Totsuka [1,3], Y Yokota [1], A Yoshikawa [1,2]

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**Abstract:**
Using the micro-pulling-down (mu-PD) method, Tm-doped Lu3Al5O12 (Tm:LuAG) single crystals were grown to examine their scintillation properties. In transmittance spectra, they exhibited about 80% transparency in the wavelengths longer than 320 nm and five absorption lines due to Tm3+ 4f-4f transitions were observed. Am-241 alpha-ray excited radioluminescence spectra were measured and intense 4f-4f emission peaks were observed with the host emission. When excited by Cs-137 gamma-Ray to obtain pulse height spectra, Tm 1% doped LuAG showed the highest light yield coupled with a photomultiplier (PMT) or a silicon avalanche photodiode (Si-APD). The light yield was estimated to be 5800 and 7300 photons/MeV for PMT and Si-APD, respectively. Decay time profiles consist of two exponential components and the fast and slow components are considered to be attributed to the host and the combination of the host and Tm3+ 4f-4f emission, respectively. 

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## Growth and characterization of strontium metaborate scintillators

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**Abstract:**
The undoped and 0.5% Ce(3+)-doped strontium metaborate SrB(2)O(4) single crystals has been grown successfully by micro-pulling down method with radio frequency (RF) heating system, and scintillation characteristics including optical properties and radiation response were studied for these crystals. The Ce(3+)-doped SrB(2)O(4) crystal showed absorption band around 240-320 nm, which is corresponding to the 4f-5d transition of Ce(3+). Intense emission band at 375 nm due to the Ce(3+) 5d-4f transition was observed under (241)Am 5.5 MeV alpha-ray excitation. The scintillation decay time showed fast (50 ns) and slow (1430 ns) components ascribed to the Ce(3+) 5d-4f transition and lattice defect in the crystal, respectively. The scintillation light yield of Ce(3+)-doped SrB(2)O(4) was calculated to be about 1000 ph/n under (252)Cf irradiation. 

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*OPTICAL MATERIALS* Volume: 34 Issue: 2 Pages: 448-451 Published: DEC 2011

## Evaluation of characterization of rare-earth doped sesquioxide ceramic scintillators

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**Abstract:**
We investigated basic optical and scintillation properties of pure Y(2)O(3), Tm(3+)-doped Y(2)O(3), pure Lu(2)O(3) and Nd(3+)-doped Lu(2)O(3) transparent ceramics made by a sintering method. All ceramic samples showed 60-80% transparency, and some absorption bands due to Nd(3+) 4f-4f transition were observed in Nd(3+):Lu(2)O(3) ceramic. Both Tm(3+):Y(2)O(3) and Nd(3+):Lu(2)O(3) ceramics showed sharp luminescence lines corresponding to the 4f-4f transition under 285 nm (Tm(3+):Y(2)O(3)) and 340 nm (Nd(3+):Lu(2)O(3)) excitation. The photoluminescence decay times were calculated to be about 24 mu s for Tm(3+):Y(2)O(3) and 1 mu s for Nd(3+):Lu(2)O(3), respectively. In radioluminescence measurements. Tm(3+) and Nd(3+) 4f-4f luminescence were observed for Tm(3+)-doped Y(2)O(3) and Nd(3+)-doped Lu(2)O(3) ceramics under (241)Am 5.5 MeV alpha-ray excitation. Finally scintillation light yield was investigated with pulse height analysis. 

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## Fundamental optical constants of Nd-doped Y2O3 ceramic and its scintillation characteristics

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**Abstract:**

Transparent Nd:Y2O3 ceramics were produced by vacuum sintering. Their fundamental optical constants including optical transmittance, reflectivity, absorption coefficient, refractive index, extinction coefficient, and dielectric constant were evaluated. Scintillation characteristics including radio-luminescence and light yield under alpha (Am-241 5.5 MeV) excitation are reported as well.


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## Development of novel rare earth doped fluoride and oxide scintillators for two-dimensional imaging

A Yoshikawa [1,2], T Yanagida [2], Y Yokota [1], K Kamada [1], N Kawaguchi [1,3], K Fukuda [1,3], A Yamazaki [4], K Watanabe [4], A Uritani [4], T Iguchi [4], G Boulon [1,5], M Nikl [6]

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**Abstract:**

Two topics were focused. The first one was about the gamma-ray scintillator, Pr(3+): Lu(3)Al(5)O(12) (LuAG). The second one was about neutron scintillator, Ce(3+):(6)LiCaAlF(6) and Eu(2+):(6)LiCaAlF(6)((6)LiCAF). Those scintillators have been developed very recently for modern imaging applications in the medical and homeland security fields. In both cases, the rare earth ions are playing the crucial role as emission centers. Pr(3+) in LuAG provided fast 5d -> 4f transition providing noticeably shorter decay time than that of Ce(3+). Among several candidate hosts, LuAG showed the best performance. Bulk crystal growth, basic scintillation properties, two-dimensional gamma-ray imaging and positron emission mammography (PEM) application were demonstrated. Due to the international situation, the homeland security was compromized by illicit traffic of explosives, drugs, nuclear materials, etc. and the ways to its improvement became an important R&D topic. For this purpose the Ce and Eu doped LiCAF appeared competitive candidates. Especially, when substitution of (3)He neutron detectors was considered, the discrimination ability of gamma-ray from alpha-ray was important. Bulk crystal growth, basic scintillation properties and two-dimensional neutron imaging were demonstrated.

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*RADIATION MEASUREMENTS* Volume: 46 Issue: 12 Special Issue: SI Pages: 1480-1483 Published: DEC 2011

## Radiophotoluminescence from silver-doped phosphate glass

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**Abstract:**

Glass dosimeter utilizing radiophotoluminescence (RPL) is one of accumulation type solid state dosimeters, which is based on luminescence phenomenon of silver (Ag+ ions)-doped phosphate glass exposed to ionizing radiation. In this study, to clarify the emission mechanism of yellow and blue RPL peaks, optical properties of Ag+-doped glass, such as optical absorption spectrum. RPL excitation spectrum before and after X-ray irradiation as well as the lifetime of both RPL peaks are measured. From the results, we discuss the emission mechanism of yellow (peaked at 2.21 eV) and blue (peaked at 2.70 eV) RPL using a proposed energy band diagram for RPL emission and excitation in Ag+-doped phosphate glass. It is found that the radiative lifetime of blue RPL is three orders of magnitude faster than that of yellow RPL (C) 2011 Elsevier Ltd. All rights reserved.

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## Comparative study of transparent ceramic and single crystal Ce doped LuAG scintillators

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**Abstract:**

Transparent ceramic Ce 0.5% doped Lu3Al5O12 (LuAG) scintillator grown by the sintering method and single crystalline Ce doped LuAG grown by the Czochralski method are prepared. They are cut to the physical dimensions 4 x 4 x 2 mm(3). Their transmittance and radio luminescence spectra are evaluated. They are both transmissive in wavelength longer than 500 nm and intense Ce3+ 5d-4f emission appears around 520 nm. When Cs-137 gamma-ray is irradiated, 662 key photo-absorption peaks are clearly observed in each sample. The transparent ceramic one shows higher light yield than that of the single crystalline one. The absolute light yield of the ceramic sample is turned out to be 14800 +/- 1500 ph/MeV. The decay time constants are evaluated under pulse X-ray excitation. The main component of the decay time of ceramic and single crystalline one are determined as 37 and 46 ns, respectively. (C) 2011 Elsevier Ltd. All rights reserved.

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## Scintillation characteristics of Tm3+ in Ca-3(BO3)(2) crystals

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**Abstract:**
Basic optical properties and radiation responses of undoped, Tm3+ 1.0% and 2.0% activated Ca-3(BO3)(2) (CBO) crystalline scintillator prepared by the micro-pulling down (mu-PD) method are reported. Tm3+: CBO crystals showed three weak absorption bands around 190, 260 and 350 nm, owing to the Tm3+ 4f-4f transition. Strong blue luminescence peaks at 360 and 460 urn which are ascribed to the D-1(2)-H-3(6) and D-1(2)-F-3(4) transitions of Tm3+ respectively were observed under Am-241 5.5 MeV alpha-ray excitation. The scintillation light yield of 2.0% Tm3+-doped CBO crystal was evaluated to be about 250 ph/n from the Cf-252 excited pulse height spectrum. 

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*RADIATION MEASUREMENTS* Volume: 46 Issue: 12 Special Issue: SI Pages: 1708-1711 Published: DEC 2011

## Ce and Eu-doped LiSrAlF6 scintillators for neutron detectors

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**Abstract:**
Ce 1%, Eu 1%, and Eu 2%-doped LiSrAlF6 (LiSAF) single crystals were grown by the micro-pulling-down method for thermal neutron applications. The crystals were transparent, 2.0 mm in diameter and 20-40 mm in length. Neither visible inclusions nor cracks were observed. Their transmittance spectra were measured. The strong absorption lines were observed at 200, 240, and 300 nm for Ce:LiSAF due to Ce3+ 4f-5d transition. In Eu:LiSAF, 200 (4f-5d) and 300 (4f-4f) nm absorption lines were detected. The samples demonstrated strong emission peaks at 300 nm (Ce:LiSAF) and 370 nm (Eu:LiSAFs) when they were irradiated with Am-241 alpha-rays simulating the alpha-particles from the Li-6(n, alpha) reaction. Thermal neutron responses were examined under Cf-252 irradiation. The absolute light yield of Ce, Eu 1%, and Eu 2% crystals were 3400, 18000, and 30000 ph/n, respectively. Main components of the scintillation decay time of Ce, Eu 1%, and Eu 2%-doped LiSAFs were 63, 1293, and 1205 ns. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 659 Issue: 1 Pages: 258-261 Published: DEC 11 2011

## Scintillation properties of Nd3+, Tm3+, and Er3+ doped LuF3 scintillators in the vacuum ultra violet region

T Yanagida [1], N Kawaguchi [2,3], K Fukuda [2], S Kurosawa [3], Y Fujimoto [3], Y Futami [3], Y Yokota [3], K Taniue [4], H Sekiya [5], H Kubo [4], A Yoshikawa [1,3], T Tanimori [4]

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**Abstract:**

In order to develop novel vacuum ultra violet (VUV) emitting scintillators, we grew Nd 0.5%, Tm 0.5%, and Er 0.5% doped LuF3 scintillators by the mu-pulling down method, because LuF3 has a very wide band gap and Nd3+, Tm3+, and Er3+ luminescence centers show fast and intense 5d-4f emission in VUV region. Transmittance and X-ray induced radioluminescence were studied in these three samples using our original spectrometer made by Bunkou-Keiki company. In the VUV region, transmittance of 20-60% was achieved for all the samples. The emission peaks appeared at approximately 180, 165, and 164 nm for Nd3+, Tm3+, and Er3+ doped LuF3, respectively. Using PMT R8778 (Hamamatsu), we measured their light yields under Am-241 alpha-ray excitation. Compared with Nd:LaF3 scintillator, which has 33 photoelectrons/5.5 MeV alpha, Nd:LuF3 and Tm:LuF3 showed 900 +/- 90 and 170 +/- 20 ph/5.5 MeV-alpha, respectively. Only for the Nd doped one, we can detect Cs-137 662 key gamma-ray photoabsorption peak and the light yield of 1200 +/- 120 ph/MeV was measured. We also investigated their decay time profiles by picosecond pulse X-ray equipped streak camera, and the main decay component of Nd:LuF3 turned out to be 7.63 ns. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 659 ssue: 1 Pages: 355-360 Published: DEC 11 2011

## Luminescence properties and gamma-ray response of the Ce and Ca co-doped (Gd,Y)F-3 single crystals

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**Abstract:**

The Ca0.5% and Ce1%, 3%, 7%, 10% co-doped Gd0.5Y0.5F3 single crystals were grown by the mu-PD method. In the Ca0.5% and Ce3% co-doped sample, Ce3+-perturbed luminescence at 380 nm was observed with 32.4 ns photoluminescence decay time. The energy transfer in the sequence of the regular Ce3+ -> (Gd3+)(n)-> the perturbed Ce3+ sites was evidenced through observation of decay time shortening of the regular Ce3+ and Gd3+ centers and the change between the Gd3+ and Ce3+-perturbed emission intensity. The gamma-ray excited scintillation response of the Ca0.5%, Ce7% co-doped Gd0.5Y0.5F3 sample was investigated with the help of the pulse height spectra and the light yield, energy resolution and non-proportionality was evaluated in the interval of energies of 59.4-1274 keV. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 659 Issue: 1 Pages: 368-372 Published: DEC 11 2011

## Crystal growth and scintillation properties of Ce and Eu doped LiSrAlF6

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**Abstract:**

Ce and Eu doped LiSrAlF6 (LiSAF) single crystals for the neutron detection with different dopant concentrations were grown by the micro-pulling-down method (mu-PD). In Ce:LiSAF, intense emission peaks due to Ce3+ 5d-4f transitions were observed at approximately 315 and 335 nm in photo- and a-ray induced radio-luminescence spectra. In case of Eu:LiSAFs, an intense emission peak at 375 nm due to Eu2+ 5d-4f transition was observed in the radio-luminescence spectra. The pulse height spectra and decay time profiles were measured under Cf-252 neutron irradiation to examine the neutron response. The Ce 3% and Eu 2% doped LiSAF showed the highest light yield of 2860 ph/n with 19 ns main decay time component and 24,000 ph/n with 1610 ns. 

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 659 Issue: 1 Pages: 399-402 Published: DEC 11 2011

## Performance test of Si PIN photodiode line scanner for thermal neutron detection

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**Abstract:**

Thermal neutron imaging using Si PIN photodiode line scanner and Eu-doped LiCaAlF(6) crystal scintillator has been developed. The pixel dimensions of photodiode are 1.18 mm (width) x 3.8 mm (length) with 0.4 mm gap and the module has 192 channels in linear array. The emission peaks of Eu-doped LiCaAlF6 after thermal neutron excitation are placed at 370 and 590 nm, and the corresponding photon sensitivities of photodiode are 0.04 and 0.34 A/W, respectively. Polished scintillator blocks with a size of 1.18 mm (width) x 3.8 mm (length) x 5.0 mm (thickness) were wrapped by several layers of Teflon tapes as a reflector and optically coupled to the photodiodes by silicone grease. JRR-3 MUSASI beam line emitting 13.5 meV thermal neutrons with the flux of 8 x 10(5) n/cm(2) s was used for the imaging test. As a subject for imaging, a Cd plate was moved at the speed of 50 mm/s perpendicular to the thermal neutron beam. Analog integration time was set to be 416.6 mu s, then signals were converted by a delta-sigma A/D converter. After the image processing, we successfully obtained moving Cd plate image under thermal neutron irradiation using PIN photodiode line scanner coupled with Eu-doped LiCaAlF(6) SCintillator. 

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## Scintillator-oriented combinatorial search in Ce-doped (Y,Gd)(3)(Ga,Al)(5)O-12 multicomponent garnet compounds

K Kamada [1], T Yanagida [2], J Pejchal [1], M Nikl [3], T Endo [1], K Tsutumi [1], Y Fujimoto [1], A Fukabori [1], A Yoshikawa [1,3]

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**Abstract:**

Ce-doped (YyGd1-y)(3)(GaxAl1-x)(5)O-12 (x = 0, 1, 2, 3, 4 and y = 1, 2, 3) single crystals are grown by the micro-pulling down method. X-ray diffraction and electron probe microanalysis techniques are employed to check their structure and chemical composition, respectively. Optical and photoluminescence characteristics are measured and radioluminescence spectra, light yield and scintillation decay measurements are further made to evaluate the scintillation performance. We show that balanced Gd and Ga admixture in the Y3Al5O12 structure can considerably increase the scintillation efficiency, and the spectrally corrected light yield value exceeds 44 000 photon MeV-1. Scintillation decay times approach that of Ce3+ photoluminescence decay and an additional less intense slower component is also observed. Physical aspects of energy transfer process and 5d(1) excited state depopulation are discussed. The micro-pulling down technique is shown as an ideal tool for a directed combinatorial search for targeted single crystal compositions to reveal those with the highest figure-of-merit for a given application field.

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*CRYSTAL GROWTH & DESIGN* Volume: 12 Issue: 1 Pages: 142-146 Published: JAN 2012

## Characterizations of Ce3+-Doped CaB2O4 Crystalline Scintillator

Y Fujimoto [1], T Yanagida [2], N Kawaguchi [3], S Kurosawa [1], K Fukuda [3], D Totsuka [4], K Watanabe [5], A Yamazaki [5], Y Yokota [1], A Yoshikawa [1,2]

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**Abstract:**

Single crystals of undoped and 0.5% Ce3+-doped calcium metaborate CaB2O4 have been grown successfully by micropulling down (mu-PD) technique. The material can find its application in the neutron detection. After the single-phase of CaB2O4 was confirmed using X-ray diffraction analysis, optical and scintillation characteristics were investigated. In the transmittance spectra, Ce3+-doped crystals showed absorption bands around 270 and 320 nm which are ascribed to the transition from 4f ground state to Sd excited state of Ce3+. Under the Am-241 5.5 MeV alpha-ray excitation, strong emission peak at 370 nm because of the Ce3+ Sd-4f transition was observed for Ce3+-doped crystal, while the undoped crystal showed broad intrinsic emission band around 300- 400 nm which is caused by the lattice defects in the host crystal. The absolute light yield was calculated to be about 2200 photons per neutron under Cf-252 neutron irradiation.

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*NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION A-ACCELERATORS SPECTROMETERS DETECTORS AND ASSOCIATED EQUIPMENT* Volume: 664 Issue: 1 Pages: 127-131 Published: FEB 1 2012

## Crystal growth and scintillation properties of Er-doped Lu3Al5O12 single crystals

M Sugiyama [1], Y Fujimoto [1], T Yanagida [2], D Totsuka [1,3], S Kurosawa [1], Y Futami [1], Y Yokota [1], V Chani [1], A Yoshikawa [1,2]

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**Abstract:**

Er-doped Lu3Al5O12 (Er:LuAG) single crystalline scintillators with different Er concentrations of 0.1, 0.5, 1, and 3% were grown by the micro-pulling-down (mu-PD) method. The grown crystals were composed of single-phase material, as demonstrated by powder X-ray diffraction (XRD). The radioluminescence spectra measured under Am-241 alpha-ray excitation indicated host emission at approximately 350 nm and Er3+ 4f-4f emissions. According to the pulse height spectra recorded under gamma-ray irradiation, the 0.5% Er:LuAG exhibited the highest peak channel among the samples. The gamma-ray excited decay time profiles were well fitted by the two-component exponential approximation (0.8 mu s and 6-10 mu s). (C) 2011 Elsevier B.V. All rights reserved.

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## Investigations of optical and scintillation properties of Tm3+-doped YAlO3

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**Abstract:**

Pure, 0.1, 0.5 and 1 mol% Tm-doped YAP single crystalline scintillators were grown by the mu-PD method. The XRD analysis confirmed the lattice constants decrease with the Tm concentration. In the transmittance measurement, the absorption bands due to the Tm3+ 4f-4f transitions were observed at 265, 360, 485, 690 and 800 nm and they were ascribed to the transition from the H-3(6) ground state to its I-1(6), D-1(2), (1)G(4), F-3(3) and H-3(4) excited states, respectively. Strong emission peak due to the I-1(6)-F-3(4) transition of Tm3+ appeared at 350 nm under X-ray irradiation. The photoluminescence decay time constants related to this transition were evaluated to be from 15.3 to 17.3 mu s and the scintillation decay time constants under gamma-ray excitation were estimated to be from 17.5 to 18.8 mu s. The Tm 1% doped crystal exhibited the highest light yield of 15, 100 +/- 1500 photons/MeV when excited by Cs-137 gamma-ray radiation. (C) 2011 Elsevier B.V. All rights reserved.

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*OPTICAL MATERIALS* Volume: 34 Issue: 4 Pages: 729-732 Published: FEB 2012

## Crystal growth and scintillation properties of Ce-doped sodium calcium lutetium complex fluoride

S Wakahara [1], Y Furuya [1], T Yanagida [2], Y Yokota [1], J Pejchal [3], M Sugiyama [1], N Kawaguchi [1], D Totsuka [1,4], A Yoshikawa [1,2]

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**Abstract:**

0.1%, 0.5%, 1% and 3% Ce doped scintillation crystals based on NaF-CaF2-LuF3 solid solutions were grown from the melt using Micro-Pulling-Down (mu-PD) method. The grown crystals were transparent and their transmittance was approximately 80% for the wavelengths longer than 320 nm. Concerning the scintillation properties, radio-luminescence peaks of the crystals were detected at approximately 330 nm. The light yield of the crystals was also measured using gamma-ray (Cs-137 and Na-22) as the excitation source. 

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*JAPANESE JOURNAL OF APPLIED PHYSICS* Volume: 51 Issue: 2 Article Number: 022603 Published: FEB 2012

## Optical Characteristic Improvement of Neodymium-Doped Lanthanum Fluoride Thin Films Grown by Pulsed Laser Deposition for Vacuum Ultraviolet Application

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**Abstract:**

Neodymium-doped lanthanum fluoride (Nd3+:LaF3) thin films were successfully grown on MgF2(001) substrates by pulsed laser deposition. Photoluminescence spectra revealed a dominant peak at 173 nm with a decay time of 7.8 ns, which is similar to the results obtained from a bulk Nd3+:LaF3 crystal. Improvements in crystalline quality and vacuum ultraviolet (VUV) luminescence quantum efficiency were achieved by substrate heating, with optimum results being obtained at 400 degrees C. These results would open up possibilities in the development of a light-emitting device operating in the VUV region. 

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*OPTICAL MATERIALS* Volume: 34 Issue: 5 Pages: 868-871 Published: MAR 2012

## Eu-doped (LiF)-Li-6-SrF2 eutectic scintillators for neutron detection

T Yanagida [1], K Fukuda [2], Y Fujimoto [4], N Kawaguchi [2,4], S Kurosawa [4], A Yamazaki [3], K Watanabe [3], Y Futami [4], Y Yokota [4], J Pejchal [4], A Yoshikawa [1,4], A Uritani [3], T Iguchi [3]

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**Abstract:**

Eu2+ 0.05%, 0.1%, and 0.2% activated LiF-SrF2 eutectic scintillators were prepared by the Bridgman method using Li-6 enriched (95%) raw material. The alpha-ray-induced radio luminescence spectra showed intense emission peak at 430 nm due to an emission from Eu2+ 5d-4f transition in the Eu:SrF2 layers. When excited by Cf-252 neutrons, all the samples exhibited almost the same light yields of 5000-7000 ph/n with a typical decay times of several hundreds ns. (C) 2011 Elsevier B.V. All rights reserved.

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## Development of a new imaging device using a VUV scintillator and a gas photomultiplier with a mu-PIC and GEM

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**Abstract:**

We have developed a new imaging device consisting of a VUV scintillator coupled to a gas photomultiplier (gas PMT) with a CsI photocathode, micro pixel chamber (mu-PIC) and gas electron multipliers (GEMs). Generally, the VUV scintillator has a short decay time of less than similar to 10 nsec. Thus the new detector could be used under the high rate counting (similar to 10 MHz) in the hard X-ray to soft-gamma ray region. Our goal in this paper is to obtain a soft gamma-ray image at 0.1 MeV with our gas PMT combined with the VUV scintillator as a high rate counter, and we have optimized electric fields in the gas PMT and developed a new VUV scintillator with a higher light output. In order to obtain a higher collection efficiency of photoelectrons and suppress the ion feedback in the gas PMT, we first optimized the electric field. Then we decided the electric field in the drift, transfer, and induction region to be 0.25, 1.0 and 3.0 kV/cm, respectively. The total gas gain of the gas PMT was approximately 2 x 10(5), and the gas PMT was estimated to have a quantum efficiency (QE) of 0.7% at 178 nm. Additionally, as a consequence of new VUV scintillators search, Nd:LuLiF4 and Nd:LuF3 with a volume of 10 mm x 10 mm x 5 mm were found to have higher light outputs than Nd:LaF3, which is a conventional VUV scintillator, by a factor of 2.1, and 2.6, respectively, and the Nd:LuF3 irradiated with 5.5-MeV alpha-rays had a light output of approximately 400 photons. Finally, we succeeded in obtaining the crystal images upon 5.5 MeV alpha and 0.122 MeV gamma rays excitation from Am-241- and Co-57 sources, respectively, using the gas PMT with the Nd:LuF3.

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http://iopscience.iop.org/1748-0221/7/03/C03013/

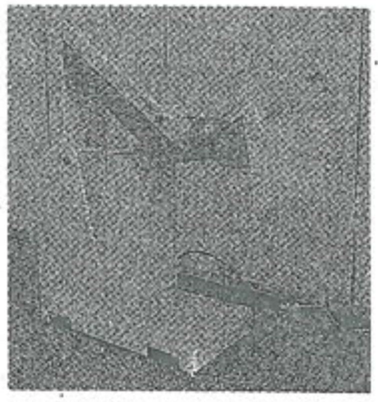
# *TV, Journals and Newspaper Items*

## 乳がん診断装置

# 13年初頭めど投入

## LuAG結晶利用 高分解能で低価格に

### 古河機械金属



分解能、データ処理性能を大幅に向上し筐体のさらなる小型化も実現した

古河機械金属は2013年初頭をめどに、LuAG（ルテチウム・アルミニウム・ガーネット）結晶を利用した乳がん診断装置「PEM」（陽電子撮像乳がん検診装置）を発売する。PEMは、乳がん検診で利用されるマンモグラフィーとは異なり、X線使用による被曝や検診時の苦痛がないことが特徴だが、現時点では高価格であることに加え、微小ながんを発見するための分解能が低いことが課題となっている。同社は結晶と機器をマッチングさせるための技術的ノウハウを有することを武器に、高分解能でリーズナブルなPEMを市場投入することで、乳がんの早期発見に寄与する。

古河機械金属はPEMの開発について、09年にスタートした新エネルギー・産業技術総合開発機構（NEDO）の3カ年プロジェクト「極初期乳がん・リンパ節がん診断を拓く次世代高解像度PEM装置の開発」に参画している。同社はLuAG結晶のブロック作製とPEMの組み立てを担当。東北大学と神戸高専、仙台画像検診クリニックと連携し、高分解能化と低価格化を実現するPEMの開発を推進している。

計画では、この8月末にPEMの筐体を完成させ9月に画像出力を確認、10月初旬に臨床試験を実施し、12月に薬事承認を申請、承認が下り次第PEMを発売する予定。12年末頃に承認が下りると想定されることから、13年初頭の発売を見込んでいる。

従来のPEM試作機では、2×2×15ミリメートルのLuAG結晶を1万2240本使用することで分解能2ミリメートルを確認。画像化のためのデータ処理時間も、それまでの2時間を5分前後へと短縮していた。

現在製作している試作機は、同本数のLuAG結晶を使用しながら、分解能を1ミリメートルに、データ処理時間を1分以内とするなど、大幅な性能向上を達成している。さらに検出器に搭載している真空管を半導体に置き換えることで、検出器の厚みを150ミリメートルから75ミリメートルへと半減、筐体のさらなる小型化を実現した。

PEMの販売は同社が行う予定だが、販売チャネル拡大のためOEM供給などさまざまな施策を検討していく。価格に関しては、5000万円以下という低価格での提供を目指す。

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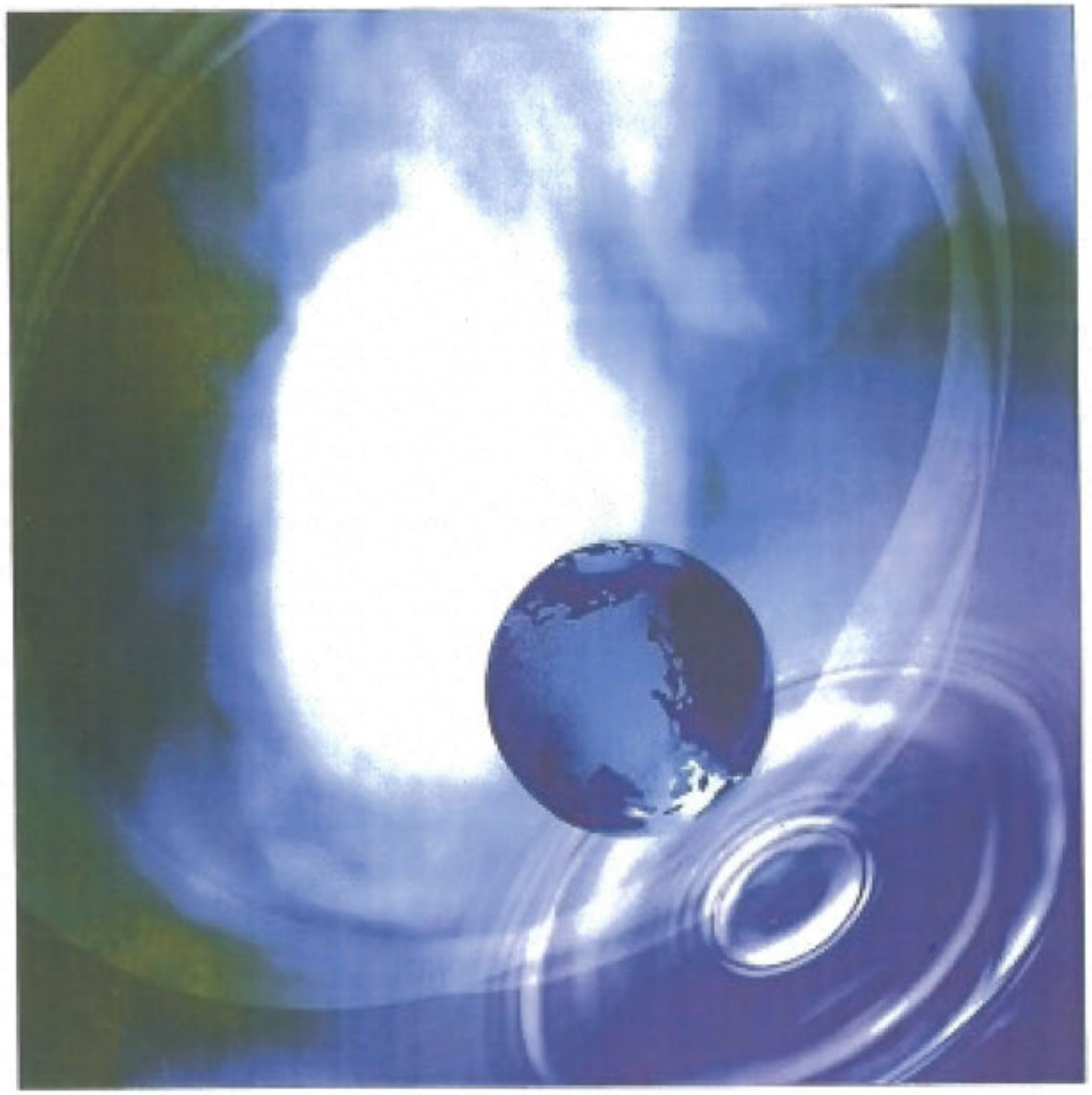
# FBNews

## 放射線安全管理総合情報誌

2011 3 No.411



株式会社千代田テクノル



## 第6回 個人モニタリングに係る国際ワークショップが開催されました

(The 6th International Workshop on Individual Monitoring of Ionizing Radiation)

昨年11月29日から30日の２日間に亘り、大洗パークホテルにて「第６回 個人モニタリングに係る国際ワークショップ」が開催されました。この国際ワークショップ（以下、「WS」という）は、放射線に関わる研究分野の第一線でご活躍されている先生方により組織され、組織委員長を当社大洗研究所の山本幸佳所長が務め、また、海外からの組織委員を Maria RANOGAJEC-KOMOR 先生（クロアチア、Rudjer Boskovic Institute）にお引き受けいただいて運営されています。さらに、当社の細田社長および竹内常務が組織委員に加わり、当社がスポンサーとなって全面的にバックアップすると共に事務局もお引き受けしています。今回の WS には、アメリカ合衆国、インドネシア、オーストラリア、オーストリア、韓国、クロアチア、スイス、中国、ドイツ、ハンガリー、フィリピン、フィンランド、ブラジル、ポーランド、マレーシア、そして日本の16ヵ国より、27機関、18大学から、総勢105名（海外参加者29名、国内の社外参加者40名）の皆様にご参加いただきました。

WS では、24件（前回は24件）の講演と22件（前回は13件）のポスター発表がありました。また、機器展示会場では、マンモグラフィ用ガラス線量計（線量計測事業本部）、Dose Ace 測定システム（AGC殿）、サーベイメータや可搬型モニタリングポスト（アロカ殿）、RPL 可視化装置「みえるくん」（大洗研究所）を展示しました。WS 終了後には、当社のガラスバッジ測定施設および校正施設をご見学いただきました。今回の WS では、各国の放射線管理や被ばくの状況、新しい放射線検出器の開発・基礎研究など、産業分野～医療分野～環境分野とかなり幅広い分野の講演・発表が行われました。

会期中は活発な質疑応答や意見交換が行われました。１日目は、Anatoly Rosenfeld 教授（オーストラリア、Wollongong 大学）、Stephen W. S. McKeever 教授（アメリカ、オクラホマ州立大学）による特別講演を中心に10件の講演がありました。２日目は、Marco Silari 博士（スイス、欧州合同素粒子原子核研究機構（CERN））、Francesco d'Errico 教授（アメリカ、エール大学）による特別講演を中心に14件の講演がありました。ここでは、紙面の都合上、特別講演の内容の概略のみご紹介いたします。

Anatoly Rosenfeld 教授は、「Advanced Semiconductor Dosimetry for Radiotherapy and Radiation Protection」というタイトルで、放射線治療における Si 半導体検出器を用いた線量計測手法の開発について講演されました。従来、放射線治療の現場では線量計測には電離箱が使われていますが、Si 半導体検出器は、それに比べて18,000倍（同じ放射線有感体積を持つ電離箱に比べて）も放射線に対する感度が高く、小型化が容易であるという長所を持っています。さらに、放射線検出器と放射線信号処理回路を一体化することが可能であり、１つの IC チップの上にこれらを多数組み込むことが可能です。先生は、異なる用途のために様々な Si 半導体検出器を開発されました。例えば、皮膚線量の計測用途として、MOSFET（Metal-Oxide-Semiconductor Field-Effect Transistor、トランジスタの一種）型線量計の一種である MOskin™ 線量計を開発されました。MOskin™ 線量計の応答は電離箱の応答に非常によく一致したこと、放射線治療時の線量をリアルタイムで測定でき、計画線量との差異は５%以内であったなどの説明をされました。この線量計は、高線量率小線源療法（HDR）、強度変調放射線治療（IMRT）の際に実際に使われているそうです。その他、DMG 線量計（IMRT用途）、Brachypix 線量計（目の小線源療法用途）、Neutronpix 線量計（中性子検出用途）の開発事例紹介がありました。検出器の開発だけでなく、半導体検出器の基礎理論や将来への展望についても説明があり、今後の展開が非常に楽しみな講演内容でした。

施設見学の様子

Stephen W. S. McKeever 教授は、「Principles and Recent Developments in the use of Stimulated Luminescence in Radiation Dosimetry」というタイトルで、OSL 線量計の原理や応用について講演されました。講演の中では、はじめに $Al_2O_3$：C の性質に関する説明がありました。一般的なガンマ線やベータ線の他に、重粒子線に対する OSL 応答が詳しく調べられ、この結果が宇宙空間や重粒子線を用いた放射線治療時の Dosimetry に役立っているとの説明がありました。さらに、石英やアルカリハライド（KBr, KCl）も OSL 物質として知られていると説明されました。特に、Eu を添加したアルカリハライドは $Al_2O_3$：C に比べて OSL の蛍光寿命が１/1000程度と短いために、リアルタイムの線量測定に有効であるとの紹介がなされました。

元 IAEA 事務次長 町末男先生からは、「Nuclear Power Policy of Japan and World Trends」というタイトルで、我が国の原子力政策や世界の原子力政策の潮流についてのご講演を頂きました。我が国は、2030年までに MOX 燃料の活用、再処理工場の稼動、高速増殖炉の商用化を通じてエネルギー自給率を高めつつ、エネルギーの70%を非化石燃料系エネルギーに転換していくことを目標にしている、との説明がありました。私たちの暮らしに直接関係がある話としては、新車の70%をハイブリッドあるいは電気自動車にする、照明は100% LED 化を進めるという計画があるとのことでした。我が国同様、世界各国でも、エネルギーの非化石燃料化のために原子力発電所の新設や改造が計画されています。2030年には、現在稼働している原子力発電所の数倍の数の発電所が地球上で稼働していると予測もあり、特にアジア地域における進展が大きいそうです。そこで我が国は、これらアジア諸国に対して IAEA、OECD/NEA などの国際機関を通しての方法、二国間・多国間協定による方法で人材育成や技術協力を行っているとの説明がありました。

Marco Silari 博士は「Bonner Sphere Spectrometry for the Characterization of Neutron Fields」というタイトルでボナー球を用いた中性子場の評価方法について講演されました。はじめに、ボナー球の測定原理と測定可能な中性子エネルギーについて説明され、先生が開発された高エネルギー中性子測定用ボナー球の紹介を行われました。高エネルギー中性子測定用ボナー球は、ポリエチレン層と鉛層を組み合わせた構造になっており、高エネルギー中性子と鉛の相互作用で生じる２次中性子を効率良く測定できるように工夫されているそうです。計算機シミュレーションによる方法や準単色中性子場を利用した実験により、ボナー球の基礎特性を綿密に調べた後、実際の高エネルギー中性子場（CERN-EU High-Energy Reference Field（CERF））を使って実証試験をされた結果を説明されました。理論的な裏打ちをされたものづくり技術の高さに感心する内容でした。







Francesco d'Errico 教授は「Advances in Special Nuclear Material Detection」というタイトルで、中性子バブル線量計を使ったテロ対策用核物質探知システムの開発に関して講演されました。ソ連邦崩壊を契機に、核兵器技術の流出が世界的な問題となっています。特にアメリカを標的としたテロの増加、北朝鮮による核開発など危機迫るものがあります。現在、荷物に紛れ込ませた核物質を検出するためにプローブビーム（中性子線や高エネルギーX 線）を荷物に照射し、核物質から出て来る即発あるいは遅発中性子をシンチレーション検出器などで検出しています。先生は、操作が単純、誤作動がない、２次元計測が容易、といった特長を持つ中性子バブル線量計をシンチレーション検出器に代わる核物質探知システムの検出器とするべく、開発を進められているそうです。

今回の WS では、講演のみならずポスターセッションも充実しました。今回の WS では、前回の反省を踏まえてポスターセッションの時間を確保しました。発表件数は前回を大きく上回り、22件となりました。シンチレーターの開発、新しいガラス線量計の開発や基礎原理の研究、放射線管理に関する研究などがありました。当社大洗研究所からは、次の４件のポスター発表が行われました。佐藤裕一研究員による「Dose Response of a Three-Dimensional Polymer Gel Dosimeter Composed of Three Types of Monomers with Gellan Gum」、宮本由香研究員による「RPL characteristics of $Ag^{+-}$doped phosphate glass dosimeter」、篠崎和佳子研究員による「Improved our Individual Neutron Dosimeter "WNP" with Newly Adopted CR-39 Material」そして牧大介研究員による「Microscopic dose measurement with thin radiophoto-luminescence glass plate」です。これらの発表は、当社の線量計開発の様子を広く知っていただくよい機会となりました。


ワークショップに参加された社外の研究者の皆様

ポスター発表の様子

私どもは、この WS を通して、国産技術であるガラス線量計による個人線量測定システムが世界中に広まり、各国の放射線防護のお役に立てること、加えて、この WS が各国参加者の情報交換や交流の場となり、そこから放射線安全利用の輪が益々広がることを切に願っております。

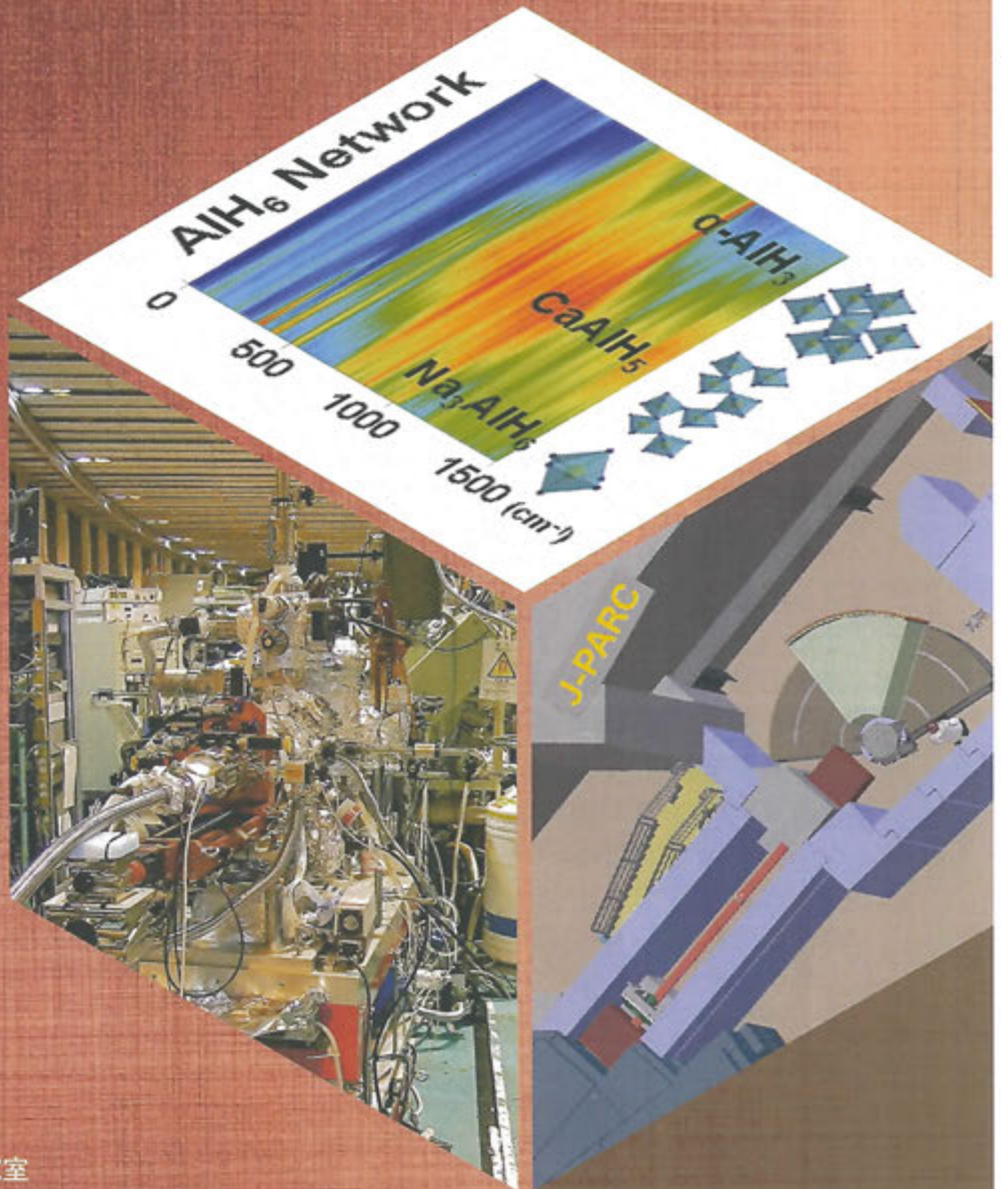
（大洗研究所：牧　大介）

# KinkeN

IMR NEWS

2011 AUTUMN

vol. 66

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～材料科学の灯火を東北にかかげて～

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写真提供：上・右／山田研究室　左／野尻研究室

TOHOKU UNIVERSITY

# Front Line
## 最先端研究紹介

第96回総合科学技術会議（平成23年2月10日開催）において、「最先端・次世代研究開発支援プログラム（ライフ・イノベーション）」に採択される研究者が決定し、金属材料研究所からは吉川彰教授が選ばれました。このプログラムは将来、世界をリードすることが期待される潜在的可能性を持った研究者に対する支援制度です。

## 次世代癌治療用近赤外線発光シンチレータの系統的研究開発


先端結晶工学研究部　吉川　彰

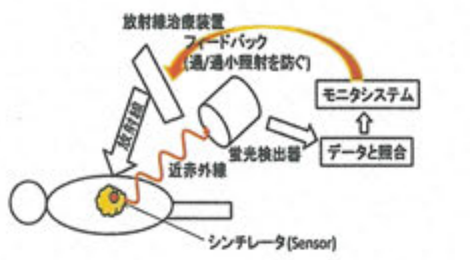

世界一高齢化が進む我が国では患者の肉体的負担が少ない放射線による癌治療は重要です。現在の放射線治療では照射した放射線量を正確に計測できず、最適な放射線量による治療を行えないという問題があります。そこで、本研究では人体に無害な近赤外線を用いて放射線量のリアルタイム計測を可能にし、適切な放射線量を用いたより安心・安全で効果的な癌治療に役立てることを考えました。

研究開発目標としては、放射線が当たると人体に無害な近赤外線で発光し、20cmの肉厚（腹囲125cm相当）をも透過し、体外から検出可能なほど明るく光る赤外発光材料を開発することとしています。

赤外発光材料を用いたリアルタイム放射線量計の研究が世界初の試みである上に、赤外に発光するシンチレータ材料の研究自体も前例は殆ど無く、学術的にも新規性の極めて高い研究です。先行研究では拳大の線量計を体内に埋込むものや、光ファイバーを体内から引き出すシステム等が想定されておりますが、これらに比して体への負担が少なく、臨床現場のイノベーションを念頭に置いていることも特色です。

長波長発光のシンチレータなので、既存のものよりも$\Delta E_g$の小さな物質を母材とすることで、放射線からのエネルギーデポジットの際に、より多くのe-hペアを生成することを考えております。また、赤外域での発光はレーザー等で多くの研究があるため、賦活剤までのエネルギー輸送が済んだ際に発光効率が良いものがどれかは豊富なデータベースがあります。材料開発は、これらを考慮に入れつつ発光中心の候補・母材の候補を絞り、その後は、材料開発の王道である絨毯爆撃方式で最適材料・組成の探索を行います。その際、研究室で独自開発したマイクロ引下法という迅速単結晶作製法を活用します。当該法は従来法のCZ法やブリッジマン法に比して、数十倍程度高速に単結晶を作製することができる画期的な融液成長法です。

本研究の枠外には本学大学病院との共同研究体制も構築されておりますので、実用化レベルの材料が開発され、リアルタイム線量計が完成した場合、大学病院にて数万人規模の臨床データを蓄積することで、将来的には日本人の典型的な体型と腫瘍の種類を分類し、シミュレーションのみで治療計画を立てて癌治療を行うことも可能になると期待されます。




赤外線発光シンチレータを利用したリアルタイム体内線量計測システムの概念図。

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The Flux Growth Society of Japan



研究室紹介

## 東北大学 金属材料研究所 先端結晶工学研究部

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**先端結晶工学研究部**

金属材料研究所[創立 1916 年，現職員数約 400 名(非常勤含む)]は大学の本部がある片平キャンパス内にあります。仙台駅から徒歩で 20 分弱であり，外部から御来所頂く際にも便利な立地です。

先端結晶工学研究部は吉川が着任するに当たり新設されたもので，市民社会の安全・安心を支える高機能機器の具現化を可能にする結晶材料の創成を目指しながら新規結晶の開発研究を行い，外部からの刺激に対する応答を系統的に理解し，特性の優れた結晶材料はデバイス化，実機搭載などを積極的に進めて行くことを使命としています。組織名称に“工学”が入っているのは「自然科学の知見を利用して，人類の幸福に資する材料・デバイスを開発することを常に意識する」という研究室のスタンスを表しています。実際に現物を創って見せること，特性の評価結果を実際に取得して体系的にまとめて議論することを重要視しており，研究室にいる間も居室ではなく，実験室に長時間いることが奨励されています。研究室の座右の銘として「つくれなければ，価値はない」を掲げています。

国内外の大学・国研との共同研究はもちろんですが，国内外の企業との共同研究も積極的に努めております。

構成人数は，創設初年度の 2011 年 4 月 1 日現在で常駐メ

写真 震災後も変わらぬ姿を見せてくれた桜の前に整列する先端結晶工学研究部のメンバー：前列左から二見能資，柳田健之，上村博(知財)，吉川彰，横田有為，黒澤俊介，2 列目左から，戸口景子(事務)，藤本裕，Jan Pejchal，山路晃広，Vladimir V. Kochurikhin，若原慎吾，戸塚大輔，杉山誠





ンバー22 名(教授 1 名，准教授 1 名，助教 1 名，研究員 5 名，技術参事 1 名，秘書 2 名，研究顧問 1 名，博士課程 4 名，修士課程 2 名，民間等共同研究員 4 名)です。

教育は大学院工学研究科・材料システム工学専攻・材料機能制御プロセス学講座(協力講座)を担当しております。

**主な研究活動**

研究は人類の幸福のために役に立つ結晶を創成することが大目的です。その意味で，実用化を意識した産学連携の研究を多く進めております。ただし，産学連携研究体制の中で，我々が担当するのは比較的基礎研究の部分となっております。具体的には材料設計，ものつくり，特性評価，デバイス化といった要素技術の上流から下流までを研究室内で全て行う一気通貫研究体制を特徴としております(図 1)。この体制の構築にはあらゆる智を総合することが必要で，化学系，物理系両方のスタッフが必須であり，研究室内で異分野融合を行っております。

この一気通貫研究体制により，研究室で開発したガンマ線用シンチレータ Pr:LuAG[図 2(a)]は古河機械金属(株)が実用化を担当してくれ，東大，東工大，放医研，仙台画像検診クリニック等との共同研究を実施しております。2009 年 3 月には乳癌用 PET への実機搭載が実現しました[図 2(b)]。また，研究室で開発した中性子線用シンチレータ LiCAF[図 2(c)]は(株)トクヤマが実用化を担当し，名大の協力も得て $^{3}He$ 代替の新規中性子検出器の鍵となる材料として注目を集めております。広大の御協力を得て年内には PoGO-Lite という国際協力グループによる宇宙探査用の気球実験に搭載されることも決定し，基礎学問の世界にも堅実な貢献を果たしております。

現在，研究室で担当している公的プロジェクトは最先端・

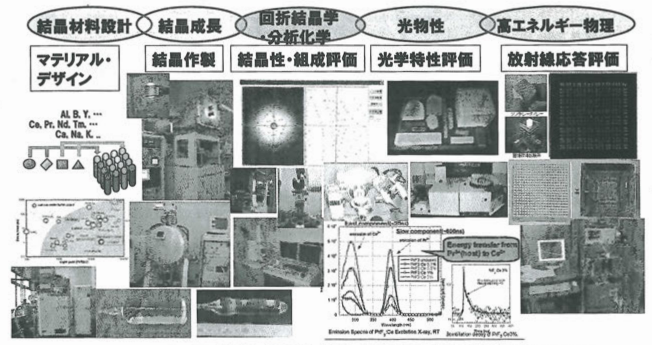


図 1 研究室の体制：物質設計，結晶成長，結晶性・組成評価，光物性，放射線応答と，開発に必要な要素技術の川上から川下まで網羅し，異分野融合体制で新規材料を創り，機能性を追求する。

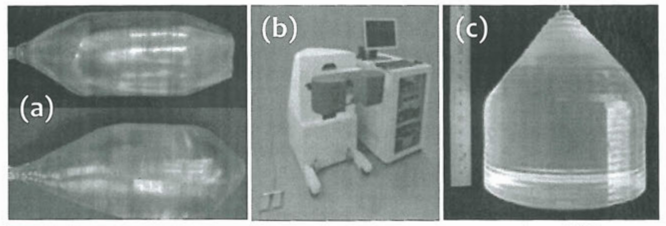


図 2 (a) γ 線用シンチレータ(Pr:LuAG)，(b) Pr:LuAG を搭載した乳癌用 PET 装置，(c) 中性子用シンチレータ LiCAF 単結晶





次世代研究開発支援プログラム「次世代癌治療用近赤外線発光シンチレータの系統的研究開発」と戦略的基盤技術高度化支援事業「燃焼圧センサー用ランガサイト型圧電結晶の形状制御単結晶作製装置及び作製技術の開発」であり，結晶作製装置のみならず，放射線や圧力と言った外からの刺激に対する物質の応答を正しく理解し，高効率で応答する結晶材料の開発とそれを用いたデバイス化を目指しております。必要な組成分析，結晶性評価，光物性評価，放射線応答評価，電気抵抗評価，圧電評価等の装置を揃え，デバイス化や装置化が得意な企業からの民間等共同研究員にも御助力を頂きながら研究に取り組んでいます。

最後に，東日本大震災の影響に関しまして申し述べます。御多分に漏れず，我々の研究室も合成装置のほぼ全てが影響を受けました。所の方針もあり，建物の健全性が確認される 3 月下旬までは避難のステージにおりましたが，4 月以降は，徐々に復興のステージへと歩を進めております。国内外の多くの皆様から励ましの御言葉や支援物資を頂きましたお陰で，4 月現在で 3 割程度の装置に対して応急処置が済みまして実験を一部再開しております。通常の状態とはかけ離れた状況ですが，不便な部分は根性と気合いでカバーして，仙台から復興の狼煙を上げることが出来る様，日々精進して参る決意でおります。また，今回の震災では風評被害などが多く伝えられており，改めてパブリックアウトリーチ活動の重要性を認識致しました。こちらも重要視しながら研究活動を進めて参る所存です。

なお，先端結晶工学研究部のホームページは http://yoshikawa-lab.imr.tohoku.ac.jp/です。

# 2011 年度（平成 23 年度）吉川研究室行事

| 月 | 吉川研究室 | 学会 | 研究会・講演会 |
|---|---|---|---|
| 4 | | | 4/7-9 阪大レーザー研研究会<br>4/22 学振１６１委員会(東京) |
| 5 | 引っ越し | | 4/9-11 阪大レーザー研シンポジウム 2011 |
| 6 | | 6/13-18 REMAT(Wroclaw, POLAND)<br>6/25-30 IWCGT-5(Berlin, GERMANY)<br>6/30-7/6 ICL11(Ann Arbor, USA) | 6/4-5 放射線物理研究会(名古屋) |
| 7 | | 7/31-8/3 ACCGE(Monterey, USA) | 7/24-30 国際サマースクール<br>(ミュンヘン工科大学) |
| 8 | | 8/28-9/1 応用物理学会秋季学術講演会<br>(山形) | 8/3 放射線物理研究会(秋保温泉) |
| 9 | | 9/10-17 SCINT2011(Giessen, GERMANY) | 9/22 学振１６１委員会(東京) |
| 10 | | 10/22-31 IEEE NSS/MIC2011<br>(Valencia, SPAIN) | 10/18-19 発光材料国際シンポジウム(作並温泉)<br>10/21 THCH Biz EXPO(名古屋) |
| 11 | | 11/14-17 The 7th Laser Ceramics<br>Symposium(SINGAPORE) | 11/2-5 NCCG-41(つくば)<br>11/19-20 第 1 回戦略的放射線物理学研究会(浜松)<br>11/25 学振１６１委員会(東京) |
| 12 | 12/27 忘年会 | | 12/1-2 GCOE 国際会議(金研)<br>12/2-5 大洗国際ワークショップ(大洗)<br>12/9-11 光物性研究会(熊本) |
| 1 | | | 1/11-12 学振１６１委員会(浜松)<br>1/20-21 TDK 進捗＆技術検討会議(秋保温泉)<br>1/23-26 研究会「放射線検出器とその応用」(つくば)<br>1/27 放射線医学総合研究所･PET 研究会講演(稲毛) |
| 2 | | | |
| 3 | 3/7-8 研究室シンポジウム＆スキー旅行(蔵王) | 3/14-18 応用物理学関連講演会(東京)<br>3/19-21 日本セラミクス協会 2012 年会(京都)<br>3/27-30 PRE'12 Workshop(京都) | 3/2-3 学振１６１委員会(東京) |



# *Events and Memorieas*

# Events of Yoshikawa laboratory

## Visiting Laser Laboratory
Osaka univ., April 7, 2011

## Moving to IMR
Tohoku univ., May 13, 2011

## Hanami (Cherry-Blossom viewing)
Tohoku univ., April 15, 2011



## International symposium on luminescent materials
Sakunami, Oct 18-19, 2011

## Summer school of radiation
Akiu, August 1-3, 2011

## SCINT
## Germany, 11-16, Sep, 2011


## IEEE / NSS 2011
## Spain, 23~31, Oct.,2011




## Ski trip
## Zao, March 7, 2012

## Farewell party
## March 2, 2012

吉川研究室 研究報告書

# *2011 Research Reports of the Yoshikawa Laboratory*

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New Industry Creation Hatchery Center (NICHe)
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