



Scintillation Materials Research Center

Annual Report

November 2005 – October 2006



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Scintillator Materials Research Center

Contact information

Mailing address:

Scintillation Materials Research Center
The University of Tennessee
124 Perkins Hall
Knoxville, TN 37996-2000

Physical addresses:

SMRC office: 301 Science and Engineering Research Facility (SERF)
Characterization Lab: 302 SERF
Synthesis Lab: 306 SERF

Telephone numbers:

Chuck Melcher: 974-0254
Merry Spurrier: 974-0267
Fax: 974-4998

E-mail:

SMRC@utk.edu
cmelcher@utk.edu
mspurrier@utk.edu

Website:

<http://www.engr.utk.edu/smrc/>

Highlights

The Center was established on October 31, 2005. Some highlights of our first year are shown below:

Laboratory opening

The SMRC operated temporarily in laboratories at Siemens Medical Solutions - Molecular Imaging in west Knoxville while facilities on campus were under construction.

The University funded extensive infrastructure improvements to the third floor of the Science and Engineering Research Facility including plumbing, electrical, and structural modifications. De-ionized, potable, and chilled water lines were installed, as well as drain lines and lines for growth station water-cooling and nitrogen delivery.

Electrical renovation included the installation of 480V and 208V service for the crystal growth station and other furnaces, and additional 120V circuits for various instruments. Structural work included the installation of new walls, cabinets, benchtops, and flooring. Also, the computer network was upgraded and expanded. In May of 2006, the SMRC moved into its permanent location, and equipment donated by Siemens, including a state of the art Czochralski crystal growth station and numerous characterization instruments, was installed in June.



Crystal growth technology

The past year saw major developments in the crystal growth technology of lutetium oxyorthosilicate (LSO). LSO, a major scintillator used in Positron Emission Tomography, uses cerium as the activation center with typical decay times in the 42 -45 ns range. We have discovered that by co-doping with optically inactive ions, the scintillation decay time can be shortened to a previously unachievable 28-30 ns, while also significantly improving the light yield. A patent is pending due to the commercial advantages that this technology offers for efficient large-scale crystal manufacture. Additional discoveries made during the past year have resulted in increased understanding of the impact of co-doping on crystal growth processes which may also have commercial impact.

New research projects

A primary objective of the SMRC is to support the research of faculty and graduate students in the area of scintillation materials. Professor Philip Rack and his graduate student, Jonathan Peak, both supported by the SMRC, are now applying their experience with luminescence and thin film combinatorial techniques to build libraries of new scintillation materials. The intent is to develop this method into an efficient screening process for the identification and optimization of potential new scintillators.



In addition to combinatorial synthesis, we also began work on a variety of other projects including optimization of Czochralski crystal growth processes for additional scintillators, investigation of scintillation properties of neutron-sensitive glass fibers for security applications, sol-gel synthesis, and a cooperation with Lawrence Berkeley National Laboratory for the refinement of a novel technique for measurement of activator site charge states via synchrotron measurements.

Students

The SMRC currently supports three graduate students along with one undergraduate, and also provided summer internships for three students. Their research encompasses various aspects of scintillation materials synthesis and characterization, including X-ray diffraction and scanning electron microscopy, sputter deposition and sol-gel synthesis, luminescence and optical characterization, investigation of defects, and modeling of light transport in single crystals and detectors. The SMRC is committed to ongoing support of both graduate and undergraduate student research in the coming years.

Patents

The SMRC staff currently have two patents pending. Two additional invention disclosures were submitted during the past year; it is anticipated that applications will be filed in the near future.

Publications/Presentations

The SMRC staff authored or co-authored 10 papers over the last year; in addition, the SMRC Director was an invited speaker at the 1st European Conference on Molecular Imaging Technology.

About the SMRC

The Scintillation Materials Research Center (SMRC) is a unique multidisciplinary research facility formed by a collaboration of the University of Tennessee and CTI Molecular Imaging (now merged with Siemens Medical Solutions Molecular Imaging). We are located in the University's Science and Engineering Research Facility, and are part of the College of Engineering. Our mission is to discover and develop new scintillation materials which will provide the foundation for the



Figure 1 The Science and Engineering Research Facility

next generation of gamma-ray, x-ray, and neutron detectors. New radiation detectors will have a major impact on future medical imaging systems, homeland security inspection and monitoring systems, neutron and high energy particle physics experiments, and remote exploration for new energy resources.

CTI Molecular Imaging (CTI) is a company founded in 1983 by four alumni of UT's College of Engineering. After winning Time Magazine's "Medical Invention of the Year" award in 2000 and introducing their proprietary LSO scintillator technology to medical imaging, CTI embarked in 2003 on a number of joint research projects with faculty in the college to expand research in scintillation materials. CTI and Siemens Medical Solutions merged in 2005, forming SMSMI.

The SMRC combines the academic resources of the university with the scintillator research legacy of CTI to develop innovative materials for state-of-the-art radiation sensors and imaging systems. The center's research activities include the synthesis and characterization of new material compositions, fundamental investigation of scintillation mechanisms, development of novel crystal growth techniques, applications of nanotechnology to ceramic scintillator fabrication, modification of existing optical materials for scintillator applications, mathematical modeling of materials and synthesis processes, optimization of existing scintillator crystals for specific applications, and advances in scintillating fiber technology.

The center draws upon the extensive high-technology resources of the Knoxville/Oak Ridge area and offers unique training and educational opportunities for students as well as providing a mechanism for collaborative research with industrial partners and academic research groups.

Research capabilities and expertise

The SMRC has broad capabilities and expertise in both material synthesis and material characterization. An extensive collection of laboratory equipment permits the research staff and students to both synthesize new materials and measure their properties in-house, thus facilitating research by allowing prompt feedback.

Material Synthesis

The primary piece of equipment in the SMRC Material Synthesis Laboratory is a Cyberstar Oxypuller 05-03 R & D Czochralski crystal growth system, sized for 50-60 mm crucibles. The system is fully computerized for optimal process control and features:

- A removable bellows, permitting use as either an open or closed growth chamber
- A dedicated cooling water system with high precision (within 0.1°F) temperature control
- High stability, compact 30 kW Huttinger MF (10 kHz range) power supply with closed loop control
- Optimized for 50-60 mm crucible diameter



Additional material synthesis equipment includes:

- A Dycor quadrupole mass spectrometer system used for real-time monitoring of crystal growth stations and annealing furnace atmospheres and volatile melt components. The system includes a Dymaxion RGA with enclosed ion source, capillary inlet manifold, compound turbomolecular pump, dry pump, and pump controller; 1-100 AMU range.
- A Sartorius density determination kit for verification of composition and monitoring of phase changes.
- A Leica 6SD microscope with dual light source is available for physical examination of samples as well as pre-growth inspection of iridium parts.
- High temperature (up to 1700°C) annealing furnaces with atmosphere control and monitoring capabilities.



Additional capabilities available via cooperation with the Materials Science and Engineering Department (MSE) include combinatorial synthesis of thin films via a sputtering system that was specially designed with four individual rf magnetron sources confocally arranged toward a common substrate with in-situ heat and bias capability. Using this approach, nearly an entire binary, ternary, and quaternary phase diagram can be deposited onto a single 100 mm diameter substrate. This method can then be used as part of a rapid screening process for the identification of potential new scintillators and to accelerate the composition optimization process, thereby facilitating the development of new materials.

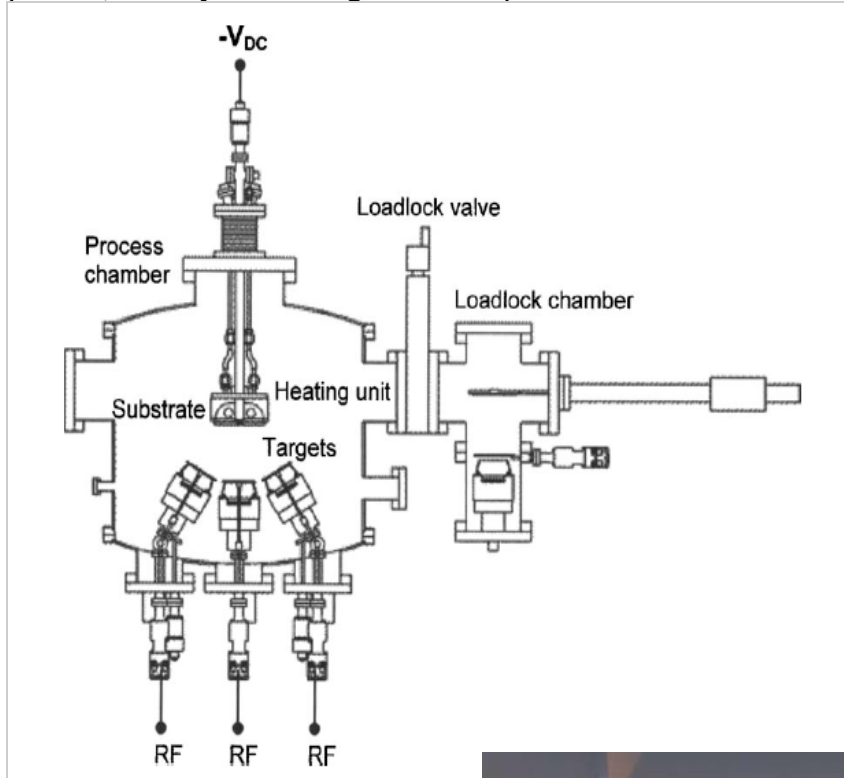


Figure 2 Schematic of sputtering

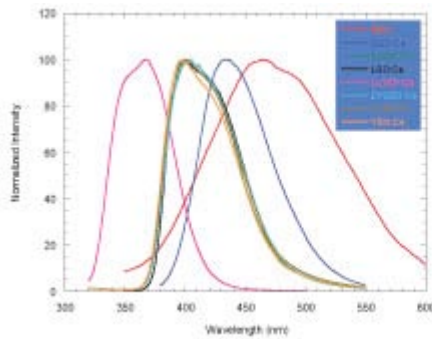


Figure 1. The sputtering system in the process of depositing a $Y_3Al_5O_{12}:\text{Gd}$ library.

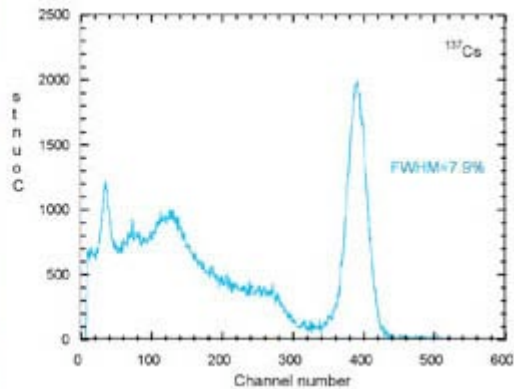
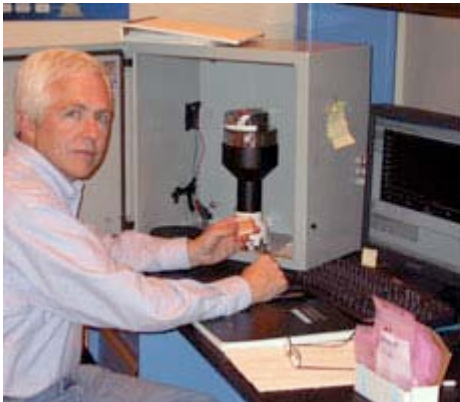
Material Characterization

The Material Characterization Laboratory is equipped with instruments designed to measure scintillation, optical, and physical properties. Many measurements can be conducted at a wide range of sample temperatures. The measurement equipment includes:

- A Varian Cary 5000 UV-VIS-IR spectrophotometer for studies of electronic energy levels and optical properties. This instrument operates at wavelengths from 175 nm to 900 nm, with nitrogen purging capability in both the monochromator and sample compartments.
- A Hitachi F4500 scanning fluorescence spectrophotometer for emission and excitation measurements. This instrument measures fluorescence, phosphorescence, and luminescence in solid or liquid samples over a 200-900 nm wavelength range.

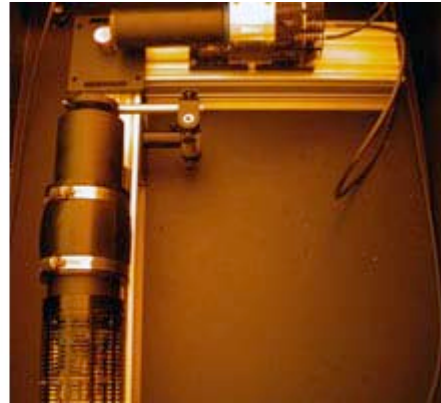
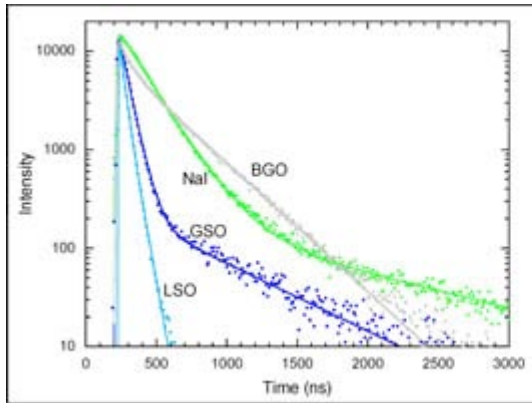


- Scintillation light yield and energy resolution measurements can be done in the UV-VIS-IR wavelengths. Non-proportionality measurements can also be done with a wide range of excitation energies.



- Absolute light output measurements.

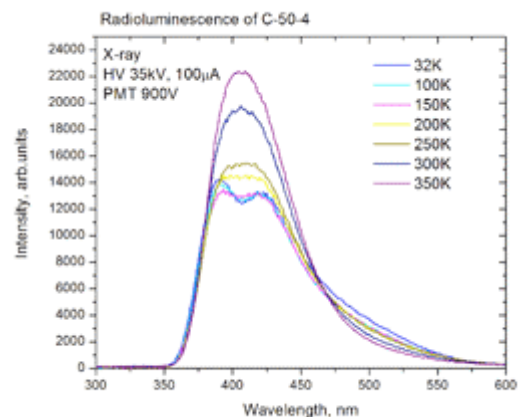
- A dedicated system is available for time-correlated single photon counting for investigation of scintillation kinetics; measurements can also be done at temperatures ranging from 9K to 800K.



- A system for thermoluminescence measurements is available for investigation of electron/hole trap parameters and the role of charge carriers in the scintillation process. This system includes an ARS compressor and cryostat designed for measurements in a 9K-800K temperature range, a LakeShore controller capable of precision temperature control, and an X-ray generator capable of continuous operation.

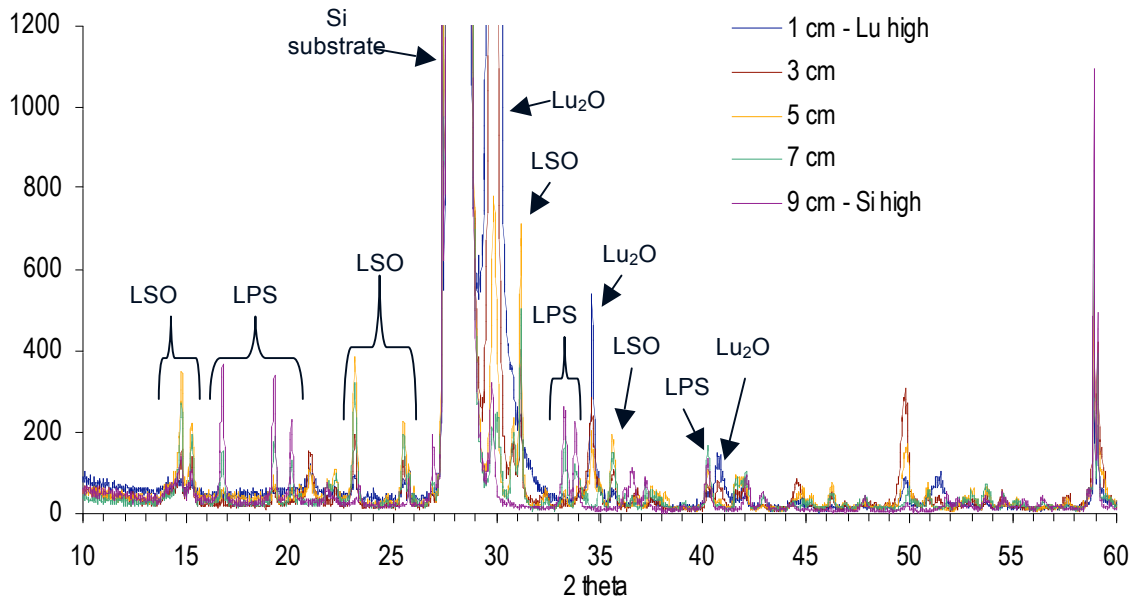


- Radioluminescence measurements regime.

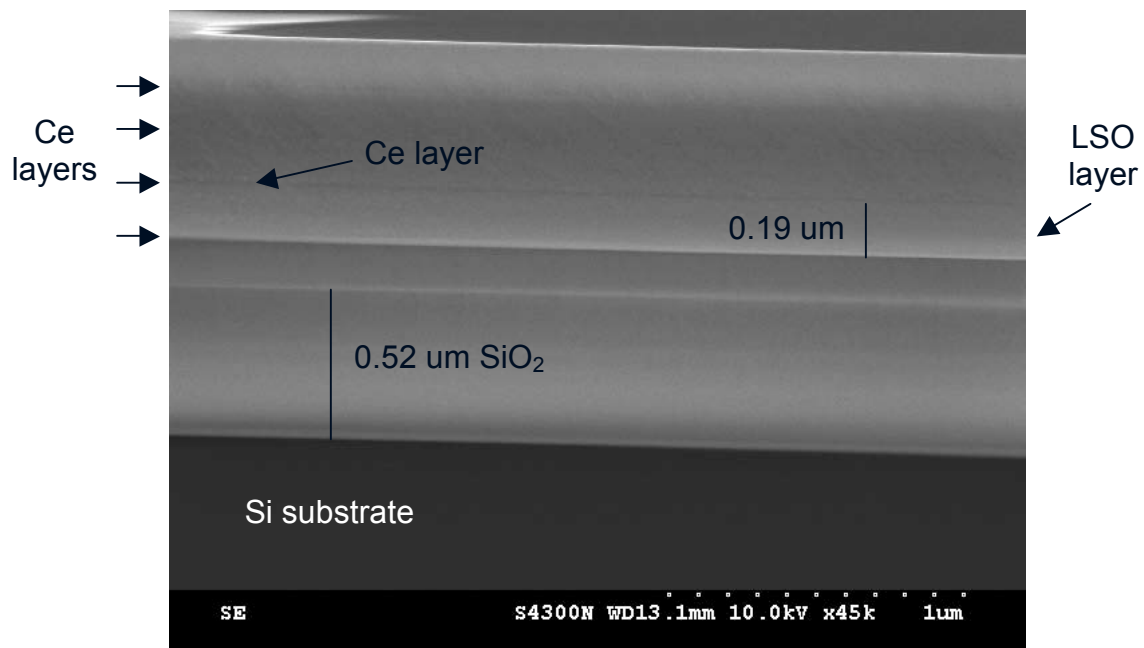


Additional capabilities via cooperation with MSE include X-ray diffraction (XRD) and scanning electron microscopy (SEM).

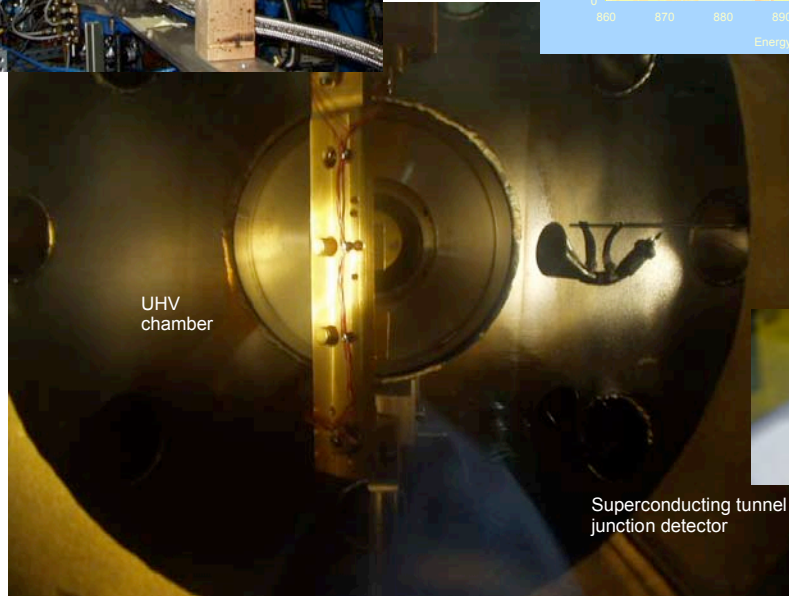
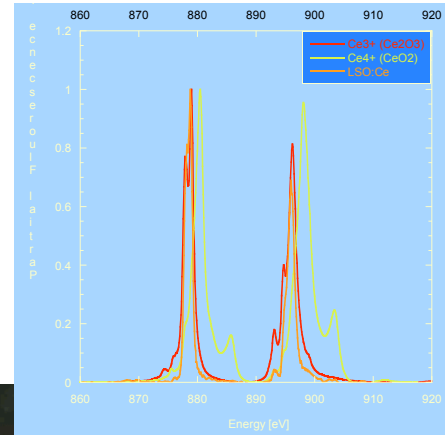
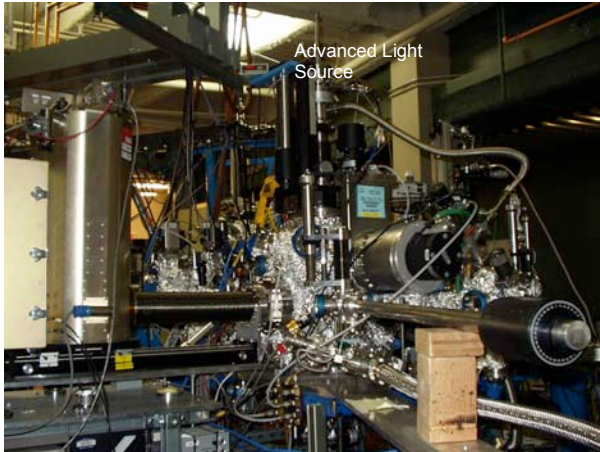
XRD of Lu₂O₃ to SiO₂ gradient w/ .5 um SiO₂ diffusion layer on Si Substrate.



SEM of deposited structure



Additional recent collaborative work has been done at the Advanced Light Source synchrotron at Lawrence Berkeley National Laboratory; a novel measurement technique to determine luminescence center oxidation state in inorganic scintillators has been developed.



Personnel

The Scintillation Materials Research Center fully supports two staff members, and offers partial support to two others. The SMRC currently supports one undergraduate and three graduate students; three summer students were also employed by the center in 2006.

In addition, SMRC staff have continued to develop relationships with scientists in related fields, some of whom were regular collaborators over the past year. Potential collaborations with Prof. David Townsend of the Cancer Imaging and Tracer Development program at the UT Medical Center are currently being explored.

Staff

The full-time center staff includes:

- Chuck Melcher – Director of the SMRC and Research Professor in Materials Science and Engineering
- Merry Spurrier – Research Associate III, SMRC

Partially supported staff includes:

- Philip Rack – Associate Professor in Materials Science and Engineering
- Frank Holiway - Materials Science and Engineering Accounting Specialist

Collaborating scientists

- Lars Eriksson - Professor, Karolinska Institute in Stockholm, Sweden and Scientific Advisor, Siemens Medical Solutions Molecular Imaging
- Piotr Szupryczynski – Staff Scientist, Siemens Medical Solutions Molecular Imaging and Assistant Research Professor in the Materials Science and Engineering.
- Stephan Friedrich - Lawrence Berkeley National Laboratory

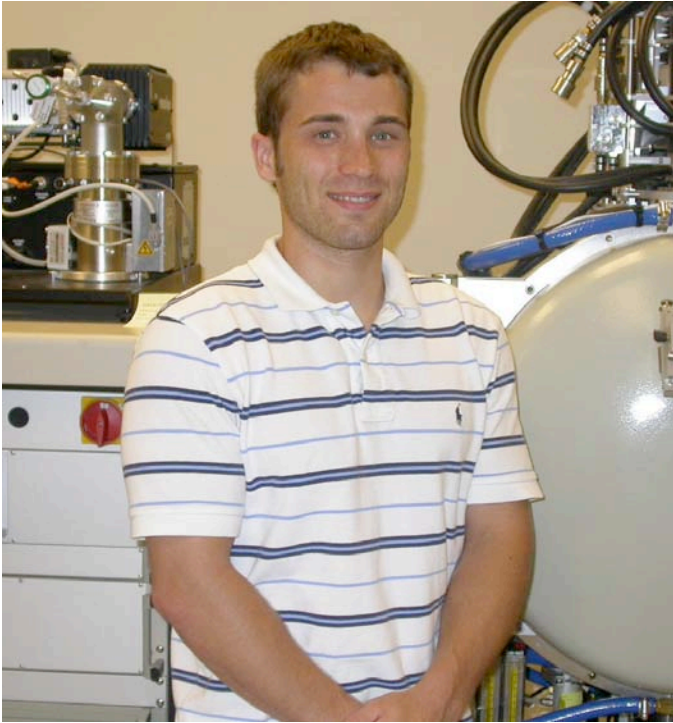
Students

The graduate students supported by the SMRC are Jonathan Peak, Harold Rothfuss, and Kan Yang. They work on projects ranging from thin films to timing measurements to afterglow; see the following pages for more detailed information.

The undergraduate student employed by the SMRC is Paul Cutler, who is responsible for sample preparation including cutting and polishing, as well as performing scintillation measurements.

The summer students employed by the SMRC during the past year were Emily Sheats, currently a graduate student in MSE, Matt Pharr, currently an undergraduate student at Rice University and Paul Cutler. These students were invaluable during our laboratory move, assisting with everything from disassembling equipment and packing boxes to unpacking and organizing the new labs.

Jonathan Peak



Graduate Student in Materials Science and Engineering

Research interests: Combinatorial Synthesis of Scintillator Crystals

Recent work has focused on thin-film LSO:Ce as a well-characterized sample material to use as proof-of-concept for the production of sample libraries for rapid new material screening.

Undergraduate work:

Georgia Institute of Technology

BS Materials Science and Engineering (May 2005)

Co-op: Alcoa, DBS Manufacturing

Undergraduate Research Interests:

The effects of aging on mechanical and electrical properties of carbon black doped polymers; nano-silicon epoxy composites

Senior Design Project:

Simulation-based design of Multifunctional Energetic Structural Materials (MESM)

Awards:

John Covert Boyd Community Service Award

Kappa Sigma Distinguished Scholar

Mark E. Floresch Scholarship

Harold Rothfuss



Graduate Student in Materials Science and Engineering

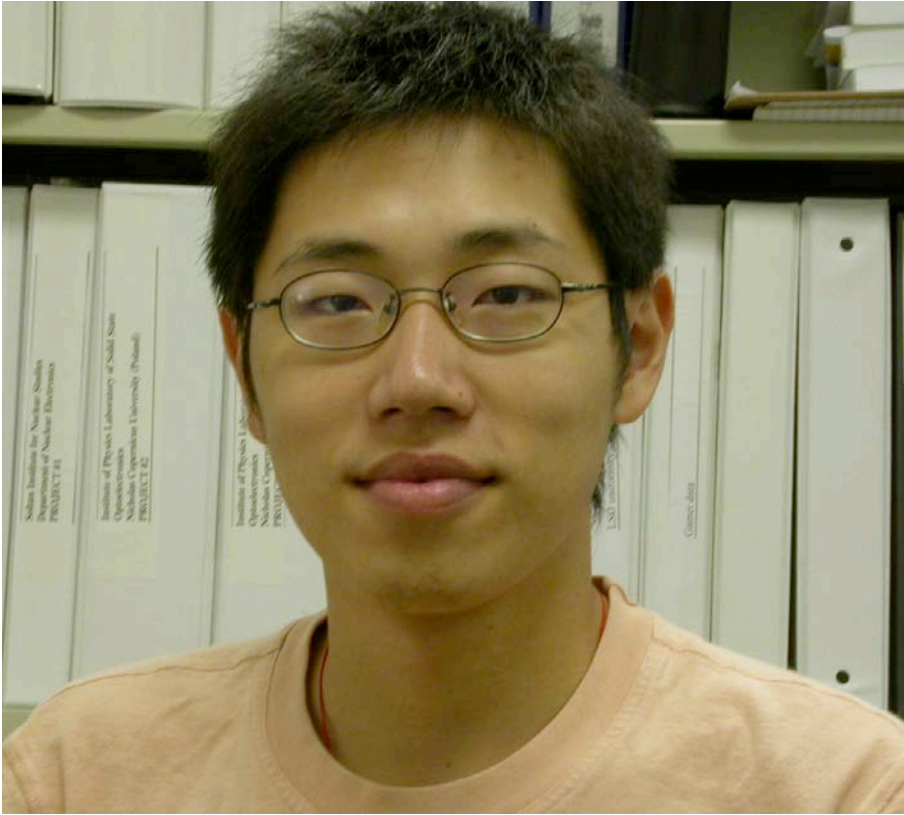
Research interests: Scintillation materials/Simulations of radiation transport in scintillators

Recent work has focused on comparisons between experimental data for energy resolution and absolute detection efficiency in LSO crystals and Monte Carlo simulation using Geant4 simulation code. Special emphasis has been placed on the introduction of non-proportionality and transport of optical photons into the simulation of light yield, in addition to the conventional electromagnetic interaction.

Undergraduate work:

University of Tennessee, Knoxville
BS in Nuclear Engineering, May 2003

Kan Yang



Graduate Student in Materials Science and Engineering

Research interests: Scintillation and crystal growth process control measurements

Recent work has focused on the development of Labview software for crystal growth process control measurements including power and water temperature measurements as well as such scintillation measurements as afterglow and thermoluminescence.

Undergraduate work:

Shanghai Jiao Tong University (SJTU), China
BS in Materials Science in Engineering

Awards:

2005 Excellent Academic Scholarship of SJTU
2004 Jiang Nan Scholarship of SJTU
2004 Excellent Academic Scholarship of SJTU

Paul Cutler



Undergraduate Student - Senior in Materials Science and Engineering, University of Tennessee; graduating in May 2007

Research interests: Crystal growth and characterization, thin film epitaxy, and nano-materials characterization

Awards and accomplishments:

Recipient of the 2006-2007 undergraduate scholarship from the Oak Ridge Chapter of ASM

Received the 2005-2006 Outstanding Junior Award from the UT Department of Materials Science and Engineering

UT-ORNL Summer Research Internship, with a project that resulted in the successful synthesis of the $\text{Lu}_4\text{Al}_2\text{O}_9$ phase via the citrate-nitrate combustion method

Summer interns 2006



Emily Sheats

Now a graduate student in MSE,
University of Tennessee

Matt Pharr

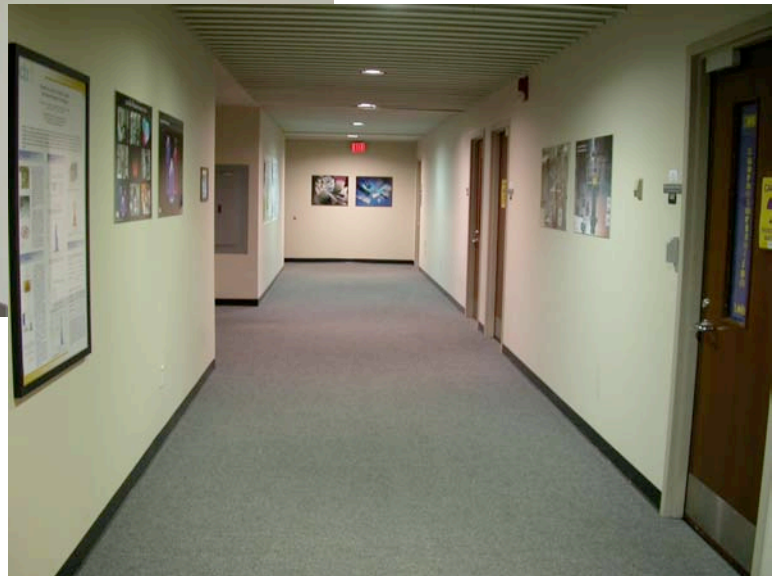
Currently an undergraduate
at Rice University, majoring
in Materials Science and
Mechanical Engineering



Laboratory relocation

The laboratory move from Siemens to UT was a major effort this year. We were given an excellent location in the Science and Engineering Research Facility, where three former classrooms were converted to office and laboratory space for the SMRC. This required extensive work from the UT Facilities group, since the SMRC laboratory equipment has significant infrastructure requirements. Installation of the electrical service was a major task, given the growth station requirement for 480V service. The building chilled, potable, and de-ionized water lines were extended to our labs and drain lines were installed, as were copper pipes for growth station water-cooling and nitrogen.

The laboratory furnishings and equipment were relocated from Siemens to UT in May; the characterization laboratory was functional shortly thereafter. The synthesis laboratory was completed a few weeks later; the first crystal was grown in the new laboratory in July.



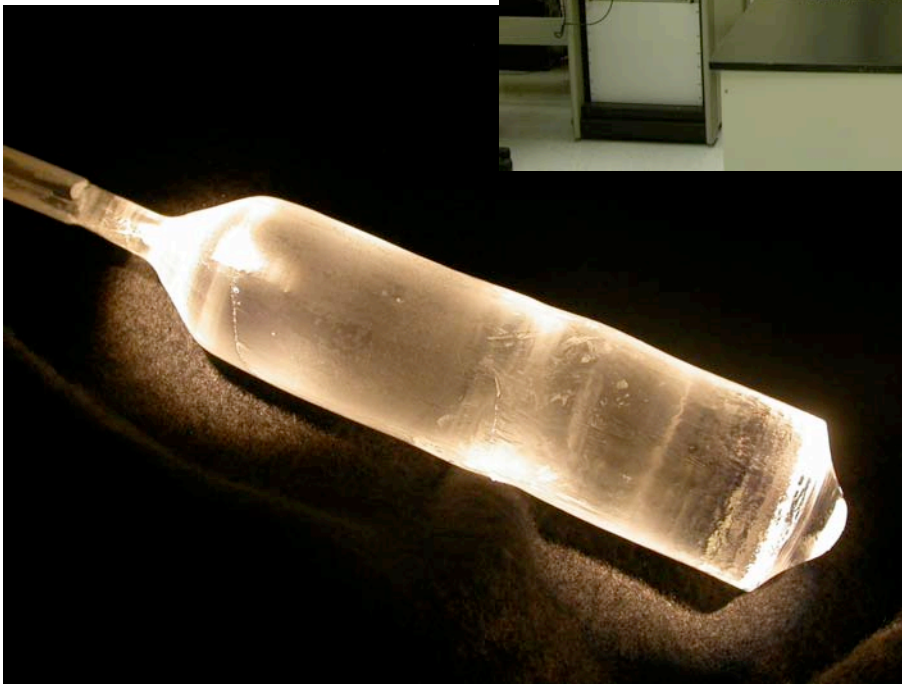
The entrance to the SMRC is located on the third floor of the Science and Engineering Research Facility.

The SMRC office space:





The Synthesis Lab



**The first LSO
boule grown at UT**

The Characterization Lab

The yellow lighting is due to the need to exclude wavelengths of light that excite the inorganic scintillators that are studied in this lab.



Patents and Invention disclosures

The SMRC staff currently have two patents pending:

- *Method for producing crystal elements having strategically oriented faces for enhancing performance.*
- *Method for improving the scintillation and optical properties of lutetium oxyorthosilicate.*

Two additional invention disclosures have been submitted through the UT Research Foundation:

- *Method for control of scintillation decay time in inorganic scintillators*
- *Suppression of Growth Instability in Inorganic Scintillators*

Publications 11/2005 – 10/2006

P. Szupryczynski, C. L. Melcher, M. A. Spurrier, A. A. Carey, M. P. Maskarinec, B. Chakoumakos, C. Rawn, R. Nutt, Ce-doped lutetium pyrosilicate scintillators LPS and LYPS, *IEEE Nuclear Science Symposium Conference Record 3*, 1310-1313 (2005).

P. Szupryczynski, C. L. Melcher, M. A. Spurrier, and A. A. Carey, Scintillation and optical properties of LuAP and LuYAP crystals, *IEEE Nuclear Science Symposium Conference Record 4*, 1305-1309 (2005).

M. Moszyński, M. Kapusta, A. Nassalski, T. Szcześniak, D. Wolski, L. Eriksson, and C. L. Melcher, New prospects for time-of-flight PET with LSO scintillators, *IEEE Nuclear Science Symposium Conference Record 5*, 2854-2858 (2005).

L. A. Eriksson, D. Townsend, M. Eriksson, M. E. Casey, M. Schmand, M. Conti, C. Michel, C. L. Melcher, B. Bendriem, T. Jones, and R. Nutt, Potentials for a fifth generation PET scanner for oncology, *IEEE Nuclear Science Symposium Conference Record 4*, 1996-2000 (2005).

S. A. Speakman, W. D. Porter, M. A. Spurrier, C. L. Melcher, Thermal expansion and thermal stability of cerium-doped Lu₂SiO₅, *Mat. Res. Bull.* 41, 423-435 (2006).

C. L. Melcher, S. Friedrich, C. Piamonteze, M. Spurrier, P. Szupryczynski, Cerium charge state analysis by X-ray absorption, in *Scint 2005: Proceedings of the Eighth International Conference on Inorganic Scintillators and Their Use in Scientific and Industrial Applications*, A. Getkin and B. Grinyov, eds., pp. 15-19 (2006).

C. L. Melcher, L. A. Eriksson, M. Aykac, F. Bauer, C. Williams, M. Loope, M. Schmand, Current and Future Use of LSO:Ce Scintillators in PET, in *Radiation Detectors for Medical Applications*, S. Tavernier et al. (eds.), Springer, 243-257 (2006).

J. S. Neal, L. A. Boatner, M. Spurrier, P. Szupryczynski, C. L. Melcher, Cerium-doped mixed-alkali rare-earth double-phosphate scintillators for thermal neutron detection, submitted to *Nucl. Instr. Meth.* (2006).

J. S. Neal, L. A. Boatner, M. Spurrier, P. Szupryczynski, C. L. Melcher, Cerium-doped mixed-alkali rare-earth double-phosphate scintillators for X- and gamma-ray detection, in *Hard X-Ray and Gamma-ray Detector Physics and Penetrating Radiation Systems VIII*, L. A. Franks et al. (eds.), Proc. Of SPIE Vol. 6319, 631907 (2006).

S. Friedrich, O. B. Drury, S. Yuan, P. Szupryczynski, M. A. Spurrier, C. L. Melcher, A 36-pixel tunnel junction soft X-ray spectrometer for scintillator material science, submitted to *IEEE Trans. Appl. Supercond.* (2006).

Presentations and reports

Invited Conference Presentation

Prof. Chuck Melcher was invited to give a plenary talk in the opening session of the 1st European Conference on Molecular Imaging Technology in Marseilles, France, on May 9, 2006. His presentation was A Brief History of LSO: From Conception to Commercial Implementation.

Review meetings and reports

In the interest of maintaining a close and effective working relationship after our relocation, we began holding informal meetings with Siemens personnel. Regularly scheduled review meetings are also held, at which the previous month's activities, results, and accomplishments are presented; PowerPoint presentations are available beginning August 2006.

Additional Reports

In addition to monthly presentations, Siemens has been provided with more detailed reports of SMRC discoveries and research as experimental data warranted. Four of these reports were completed during the past year; they were, in chronological order:

- An LSO research update, which presented information about the development of the next generation of LSO. This represents a continuation of work on the SMRC breakthrough development in LSO technology, in which co-doping and optimized growth conditions are used to very significantly improve scintillation and optical properties. The importance of this technology to large-scale manufacturing cannot be over-emphasized; if fully adopted, it results in consistently high light yield crystals with exceptionally good decay time, virtually eliminating off-color or poor scintillation quality crystals
- A detailed report on the characterization of LFS samples, as part of an assessment of the likelihood of infringement on the LSO patent
- A report on the extension of co-doping technology to an additional scintillator
- A tutorial providing basic background information regarding the improvements to LSO technology, including information needed to successfully transfer this technology to large-scale manufacturing of high quality crystals for PET

Miscellaneous

- The SMRC staff has continued to monitor new patent applications and publications relating to medical imaging and radiation detection, and has both notified Siemens of potential patent infringements and provided information to assist Siemens in evaluating any threat to their interests as these issues arose.

Financial Support

Financial support for the SMRC over the past year was provided by the following sources:

Operating expenses:

Siemens Medical Solutions Molecular Imaging	\$500,000
University of Tennessee	
Chancellor's office	\$60,000
College of Engineering	\$20,000
Materials Science and Engineering	\$40,000
<i>Total</i>	<i>\$620,000</i>

For laboratory renovation:

University of Tennessee	
College of Engineering	\$37,500
Chancellor's Office	\$37,500
<i>Total</i>	<i>\$75,000</i>

In addition to the ongoing agreement with Siemens, a subcontract with NuSAFE, Inc. under their prime contract from the Defense Threat Reduction Agency has been finalized. While no monies were received during the past year, it is anticipated that the SMRC will receive some funding from this source during the 2006-2007 center year.