2006 LLNL Nuclear Science Summer Internship Program

June 13 - August 11, 2006

Lawrence Livermore National Laboratory
Chemistry, Materials, and Life Sciences
Glenn T. Seaborg Institute
Livermore, CA 94550, USA

Web site: http://cms.llnl.gov/review/seaborginstitute/index.html

Director: Annie Kersting
Education Coordinator: Nancy Hutcheon
Administrative Assistant: Linda Jones

Sponsors:
Office of Nonproliferation Research and Engineering (NA-22)
Advanced Fuel Cycle Initiative
LLNL: Chemistry, Materials, & Life Science

Glenn T. Seaborg Institute

The Lawrence Livermore National Laboratory (LLNL) Nuclear Science Summer Internship Program (NSSIP) is designed to give both undergraduate and graduate students an opportunity to come to LLNL for 8 weeks during the summer for a hands-on research experience. Each student conducts research under the supervision of a staff scientist, attends a weekly lecture series, interacts with other students, and presents their work in poster format at the end of the program. Students also have the opportunity to participate in LLNL facility tours (e.g. National Ignition Facility, Center of Accelerator Mass-spectrometry) to gain a better understanding of the multi-disciplinary science that is on-going at LLNL. A summer highlight is a weekend trip to Yosemite national park, where the students enjoy a weekend of hiking and socializing.

Currently called NSSIP, this program began eight years ago as the Actinide Summer Program. The program is run within the Glenn T. Seaborg Institute in the Chemistry, Materials, and Life Sciences Directorate at LLNL. The goal of NSSIP is to facilitate the training of the next generation of nuclear scientists and engineers to solve critical national security problems. We select students who are majoring in physics, chemistry, nuclear engineering, chemical engineering and environmental sciences. Students engage in research projects in the disciplines of actinide and radiochemistry, isotopic analysis, radiation detection, and nuclear engineering in order to strengthen the 'pipeline" for future scientific disciplines critical to NNSA.

This is a competitive program with over 170 applicants for the 15-20 slots available. Students come highly recommended from universities all over the country. For example, this year we had students from UC Berkeley, U Michigan, Stanford, Columbia University, Georgia Tech, UC Davis, Reed College, Middlebury and MIT. We advertise with mailers and email to all the nuclear science and chemistry departments in the U.S. We also host students for a day at LLNL who are participating in the D.O.E. sponsored "*Summer School in Nuclear Chemistry*" course held at San Jose State University.

This year students conducted research on such diverse topics as: cryogenic neutron spectrometers, nuclear forensics, cladding for advanced fission reactors, sorption of radionuclides, principal component analysis of spent reactor fuel, semiconductor detectors, actinide separations chemistry, and anti-neutrino monitoring of nuclear reactors.

Graduate students are invited to return for a second year at the mentor's discretion. For the top graduate students in our program, we encourage the continuation of all collaborations between graduate student, faculty advisor and laboratory scientists. This creates a successful pipeline of top quality students from universities across the U.S. To date, 15 graduate students have continued to conduct their graduate research at LLNL. Three recent career hires were former summer students.

Seminar Schedule 2006

Wed June 21	Ken Moody, LLNL	*“Nuclear Forensics”*
Wed June 28	Mike Pivovaroff, LLNL,	*“From High Energy Astrophysics to Homeland Security – Making the Link”*
Thur June 29	Armando Alcaraz, LLNL	*“Forensics Sciences and national security at LLNL”*
Wed July 5	Rick Ryerson, LLNL	*“Raising Tibet”*
Thur July 6	John Perkins, LLNL	*“Fusion, the Ultimate Energy Source”*
Tues July 11	Mike Carter, LLNL	*“Countering Nuclear Terrorism – A Grand Challenge in Homeland Security”*
Thurs July 13	Ed Moses, LLNL	*“NIF, The National Ignition Facility”*
Wed July 19	Bob Criss, Washington University	*"Earth's Radioactivity, Heat Flow and Convection"*
Thur July 20	Bob Maxwell, LLNL	*“Nuclear Magnetic Resonance Center”*
Tues July 25	Eric Gard, LLNL	*“Bio-Aerosol Mass Spectrometry”*
Thur July 27	Alex Hamza, LLNL	*“Nanomaterials Synthesis and Characterization”*
Wed Aug 2	Glenn Knoll, University of Michigan	*“Fundamentals of Radiation Detection”*
Thur Aug 3	Jim Rathkopf, LLNL	*“Principles of Nuclear Weapons Physics”*
Thur Aug 10	Jon Maienschien, LLNL	*“High Explosives Research”*

Summer Students 2006

Student	Major	University	Year
Amy Coffer	Nuclear Engineering	U Michigan	Undergrad
Bhushar Mookerji	Physics/Electrical Engineering	MIT	Undergrad
Elaine Hart	Materials Science	Stanford	Graduate
Daniel Heineck	Electrical Engineering	Oregon State University	Graduate
Greg Brennecka	Nuclear Chemistry	U Rochester	Graduate
Jeremy Waen	Chemistry	Reed College	Graduate
John Clements	Environmental Engineering	Clemson	Graduate
Kevin Stein	Physics	UC Berkeley	Undergrad
Martin Robel	Nuclear Engineering	UC Berkeley	Graduate
Paul Koster van Groos	Environmental Engineering	UC Berkeley	Graduate
Rion Graham	Physics	UCLA	Undergrad
Tsuguo Aramaki	Physics	Columbia	Graduate
William Coleman	Physics	LSU	Graduate
Shadi Ghrayeb	Nuclear Engineering	Rensselaer Polytechnic Institute	Undergrad
Alex Johnson	Nuclear Engineering	Georgia Tech	Undergrad
Nathanael Kuo	Electrical Engineering	U of Illinois	Undergrad
Benjamin Westbrook	Physics	U San Francisco	Undergrad
Ryan Till	Mechanical Engineering	U Arizona	Undergrad
Megan Brinkmeyer	Chemistry	St. Mary’s College	Undergrad

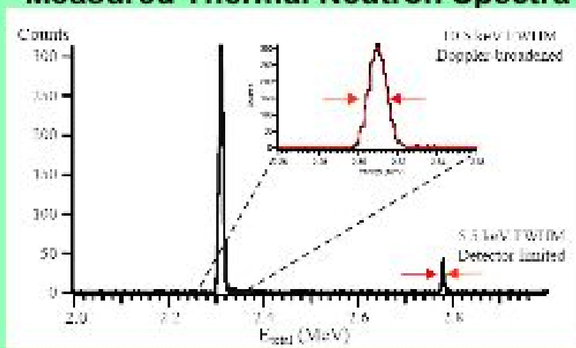
Unfolding a Cryogenic Neutron Spectrometer

Amy Coffer, Nuclear Science Summer Internship Program
Thomas Niedermayr, I-Div, Physics and Advanced Technologies

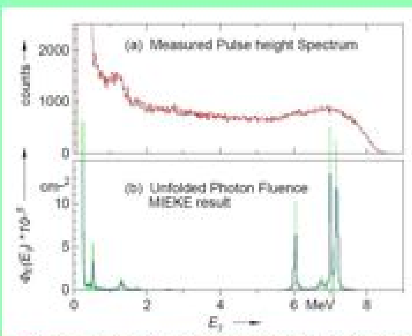
The Detector

- Nuclear Attribution
- Microcalorimetry
- High Resolution ~ 0.1%
- Compact and Commercially Available

Measured Thermal Neutron Spectra



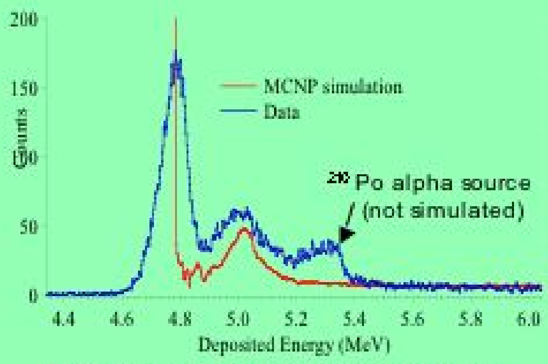
Unfolding



Example of unfolding a low resolution detector

- Increase System Performance
- Know Detector Response
- Mathematical Technique
- More Accurate and Precise Info

Simulation



MCNP simulation matches detector measurement

- Quality of Unfolding
- MCNP
- Maximum Entropy
- PTB Computer Code

The optical constants of Gallium stabilized d-phase plutonium metal between 0.7 and 4.3 eV measured by spectroscopic ellipsometry *using a double-windowed experimental chamber*

Glenn T. Seaborg Institute

Bhaskar Mookerji[1,2] and Wigbert J. Siekhaus[3]

[1]Undergraduate Summer Institute [2]Massachusetts Institute of Technology; Cambridge, MA 02139

[2]Material Science and Technology Division, Chemistry and Material Science Directorate, LLNL; Livermore, CA 94550

Abstract

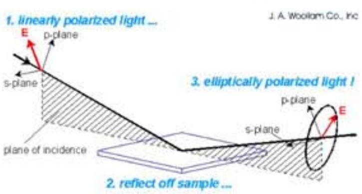
A double-windowed vacuum-tight experimental chamber was developed, and calibrated on the spectroscopic ellipsometer over the energy range from 0.7 to 4.5 eV using a silicon wafer with approximately 25 nm oxide thickness to remove the multiple-window effects from measurements. The ellipsometric measurements were done such that incident and exit beam were at 65 degree from surface normal. The plutonium sample (3 mm diameter, 0.1 mm thick) was electro-polished and mounted into the sample chamber in a glove box having a nitrogen atmosphere with less than 100ppm moisture and oxygen content. The index of refraction *n* and the extinction coefficient *k* decrease from 3.7 to 1 and 5.5 to 1.1 respectively as the photon energy increases from 0.7 to 4.3 eV.

Introduction

The optical constants (*n* and *k*, coefficients of refraction and extinction, respectively) of plutonium metal are of scientific and practical interest, but not readily available. They are difficult to measure, because only small quantities can be handled since the material is highly radioactive. Moreover, safety regulations require that plutonium be doubly contained at all times.

To accurately measure these properties, we first design a double-windowed experimental cell that secures a plutonium sample. A variable angle spectroscopic ellipsometer (J.A. Woollam Co., In. Lincoln, Nebraska 68508) will then be calibrated to apply numerical corrections at measurement-time to account for systematic errors introduced by doubled Pyrex windows. A pre-existing bulk metal model can then be applied to model our plutonium data once it has been taken.

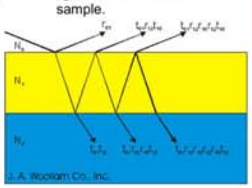
Introduction to Ellipsometry



Ellipsometry uses polarized light to characterize thin films and surfaces. Incident light interacts with the sample, changing from a linear polarization state to an elliptical polarization state. This polarization change is determined by measuring the light reflected from the sample.

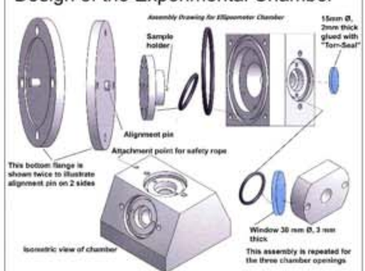
$$\rho = \tan(\psi)e^{i\Delta} = \frac{\tilde{R}_p}{\tilde{R}_s}$$

Ellipsometry measures two parameters, ψ and Δ, which are related to Fresnel reflection coefficients. An infinite, convergent series of transmitted and reflected light sums to these coefficients.



Experimental Methods and Analysis

Design of the Experimental Chamber



The experimental chamber holds the Pu sample in two separate enclosures and ensures its precise and reproducible alignment on the ellipsometer's working platform. The doubled Pyrex windows exposes 3mm wide samples to light incident at 65 degrees relative to the sample's surface normal vector.

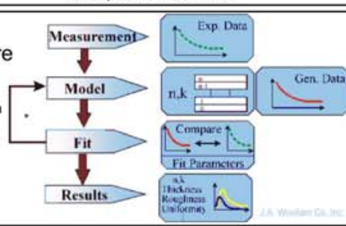
Sample Preparation and Window Calibration

The Pu sample was mechanically and electrochemically polished in a moisture-free, N_2 glove-box. The sample was approximately 101 μm thick and 3mm in diameter. Transmission electron microscopy on similar samples indicated an oxide thickness of a few nm.

Before the Pu measurement, a silicon wafer standard (3mm Ø with a 25 nm thick SiO_2 layer) was cleaned in air using acetone and was then mounted horizontally in the bottom of the ellipsometer cell. The cell and silicon wafer were then mounted vertically in the ellipsometer, during which a software calibration script determines a "window effect correction function" that is used to correct Δ and Ψ data from unknown samples in the chamber.

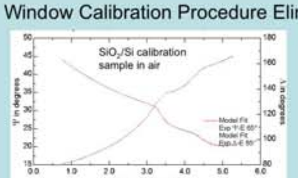
Ellipsometry analysis procedure

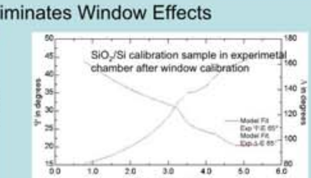
Following measurement, the Pu sample is modeled as bulk metal sample using initial data provided for well-polished, bulk gold. The model's generated constants are adjusted until they fit the experimental data.



Results and Discussion

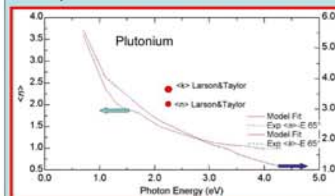
Window Calibration Procedure Eliminates Window Effects





The two figures show experimental data taken from a 1cm² oxidized Si wafer mounted first in air, and then in the experimental cell. A model consisting of bulk silicon, a 1 nm thick Si/SiO_2 interface layer and a SiO_2 layer of adjustable thickness was applied. In the cell, the measured oxide thickness (25.808 +/- 0.018 nm) did not differ significantly from the given value of 25.000 nm. The sample in open air measured 25.484 +/- 0.0456 nm.

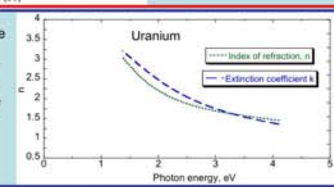
Optical Constants: Plutonium and Uranium



This is the first set of optical constants of Pu measured over a wide energy range. They disagree with earlier data[1]. Those data were, however, taken through two thick pyrex windows *without* window corrections

[1] D. T. Larson, D. L. Cash, Journal of Nuclear Materials 24 (1967), 232-233.

Uranium was analyzed before with the same instrument over a smaller energy range. Plutonium has lower *n* and *k* values than uranium over the spectrum from 0.7 to 4.5 eV.

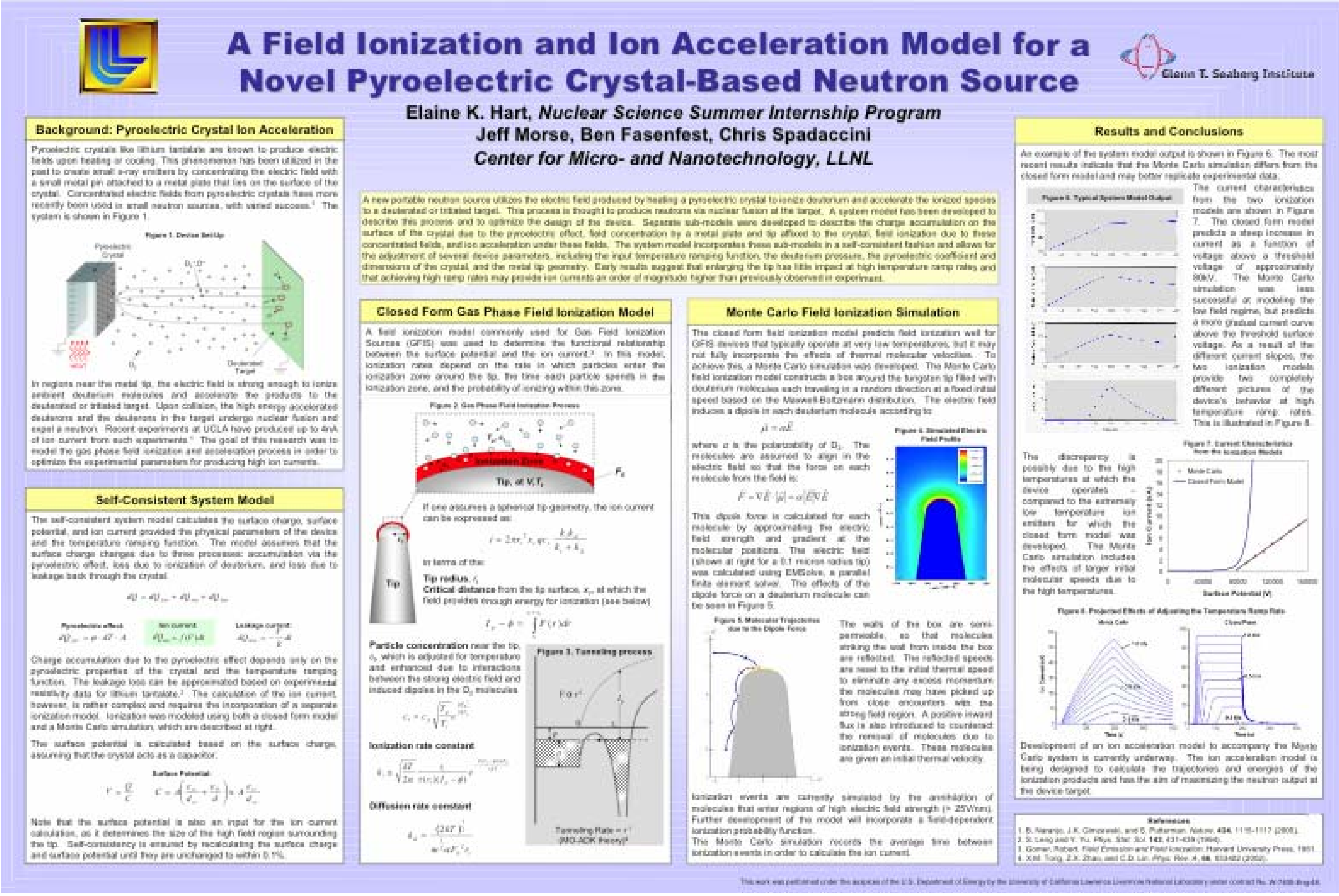


Conclusions

- We have demonstrated the reliability of a window calibration procedure for double-window experimental chambers that are needed for optical analysis of transuranium elements
- We have used it to determine the unknown optical constants of Pu metal.
- With this new data we can extend our procedure to determining the optical constants of plutonium surface oxides, part of a larger project to fully characterize the optical properties of plutonium.

Acknowledgments

This work was performed under the auspices of the U. S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48. The Seaborg Institute at LLNL supported B. Mookerji.
We gratefully acknowledge the support of Gerry Cooney and James Hilfiker of J.A. Woollam Co. Inc. for software support.
Special thanks to Martin Stratman for design/machining of the experimental cell and Mark Wall for handling the the plutonium samples.


A Field Ionization and Ion Acceleration Model for a Novel Pyroelectric Crystal-Based Neutron Source
Glenn T. Seaborg Institute
Elaine K. Hart, Nuclear Science Summer Internship Program
Jeff Morse, Ben Fasenfest, Chris Spadaccini
Center for Micro- and Nanotechnology, LLNL
Background: Pyroelectric Crystal Ion Acceleration
Self-Consistent System Model
Closed Form Gas Phase Field Ionization Model
Monte Carlo Field Ionization Simulation
Results and Conclusions
Figure 3. Tunneling process
Ionization Zone
Tip, at V, T
Tip

Pillar-Based Neutron Detector

Daniel Heineck, Nuclear Science Summer Internship Program

Rebecca Nikolic, Catherine Reinhardt, EETD

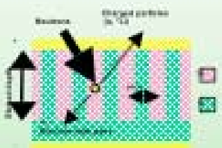
Lawrence Livermore National Laboratory

Glenn T. Seaborg Institute

This part of the pillar-based neutron detector project aims to find a reliable ohmic contact process with the goal of a specific contact resistance of 10⁻ _-cm² for both sides of the detector. For the P+ side, Aluminum, layered Ni/Ti/Au, Pd/Ti/Au, and Pt/Ti/Au schemes are tested at multiple annealing temperatures. For the N+ side, Aluminum and a layered Ti/Pt/Au are examined. The Pt/Ti/Au metal scheme proves to be the most effective contact method for the P+ side, whereas the N+ contact design is not yet determined. The Pt/Ti/Au contact with a 3.5 minute anneal at 300°C has a specific contact resistance of 6.6x10⁻ _-cm².

Introduction:

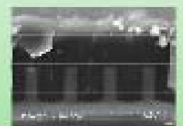
- Boron 10 absorbs incident neutrons, releasing a ^{7}Li and an _ particle in opposite directions
- The energetic ions then collide with silicon atoms in the pillars, exciting the outer shell electrons out of their lattice sites.
- High electric field accelerate electrons to the positive terminal of the device
- Pillar design is chosen in order to maximize the number of ion collisions in the silicon
- Low resistance (10⁻ _-cm²), low leakage electrical contacts are critical to ensure maximum signal to noise ratio for the detector



Work Function (_) is the energy difference between E_F and E_0

N-Type Silicon _s	4.12 eV
P-Type Silicon _s	5.08 eV
Metal Work Functions	_m
Aluminum	4.28 eV
Titanium	4.33 eV
Palladium	5.12 eV
Nickel	5.15 eV
Platinum	5.65 eV
N+ Metal Scheme	_s > _m
P+ Metal Scheme	_s < _m

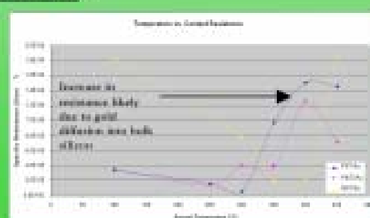
As shown on n-type material, electrons tunneling through the potential barrier are field emissions. Ohmic contacts are designed to fit this category. Electrons that must attain sufficient kinetic energy to pass over the barrier are thermionic emissions.



Pillar structures after boron 10 has been deposited in the gaps. The CVD process development is done by Barry Cheung, University of Nebraska. Samples are then lapped and polished back to the pillar height.

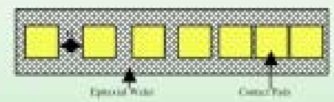
Results:

The Platinum/Titanium/Gold metal scheme has the lowest R_C of all the front side metal schemes chosen since it has a low eutectic temperature with silicon and has the highest work function, which reduces the potential barrier between the metal and the semiconductor. This value is only 6.6x10⁻ _-cm² when the sample is annealed at 300°C for 3.5 minutes. Palladium and nickel base layers also show promise and their processes will be further examined.

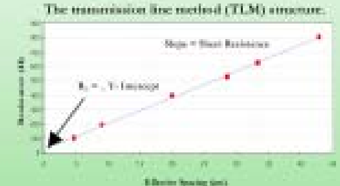


Method:

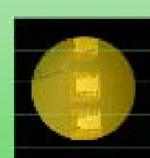
1. Mask small epitaxial samples with a lift-off photoresist pattern
2. Deposit metal schemes onto epitaxial samples using E-beam evaporation
3. Characterize specific contact resistance (R_C) by measuring the transmission line method (TLM) structures on samples before sintering process
4. Measure R_C for sintering temperatures ranging from 250°C to 450°C for 30 seconds
5. Vary anneal time for each scheme at respective optimal temperature.



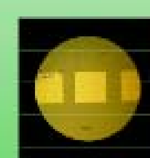
The transmission line method (TLM) structure.



Distance vs. resistance graph used to calculate R_C



Right and left are a Pt/Ti/Au contact after a 300°C 3.5 minute anneal and a Ni/Ti/Au contact after a 375°C 30 second anneal. The edges of the nickel sample show gold diffusion into the titanium, whereas the lower temperature platinum sample does not.



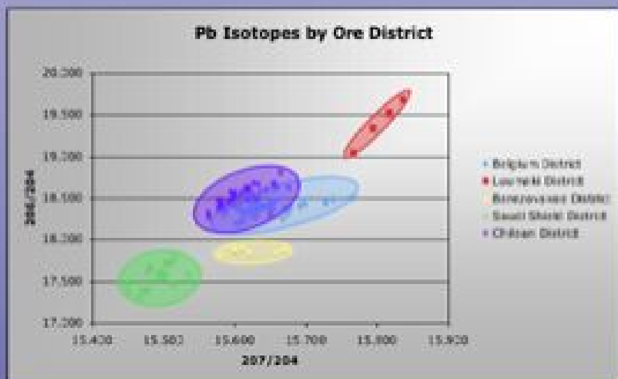
Discussion:

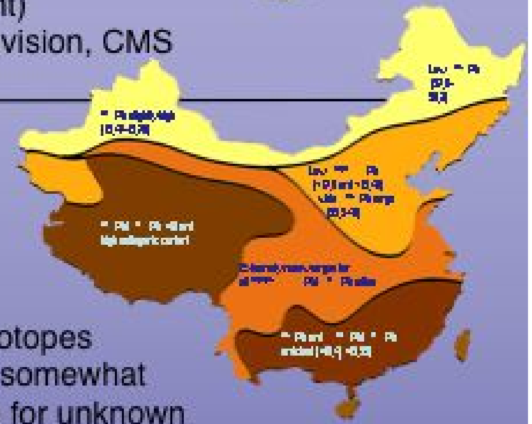
In order to reduce the specific contact resistance by an order of magnitude, variations in the thickness of the current Pt/Ti/Au layers will be examined. To complete the crystallization of the $PtSi_2$ silicide, longer anneal times will be examined. Once the goal contact resistance is met, an X-ray diffraction analysis of the surface chemistry will occur. Typically, N+ silicon and aluminum make excellent contacts at very low (200-350°C) sintering temperatures. However, the N+ back side contacts are still a work in progress. Once the ohmic contact process is completed, a basic electrical model of the sensor will be developed.

Applications of Mass Spectrometry in Nuclear Forensics

Greg Brennecka - University of Rochester (NSSIP student)
Erick Ramon - Chemical Biology and Nuclear Science Division, CMS
Glenn T. Seaborg Institute

Geolocation using a Pb isotope database

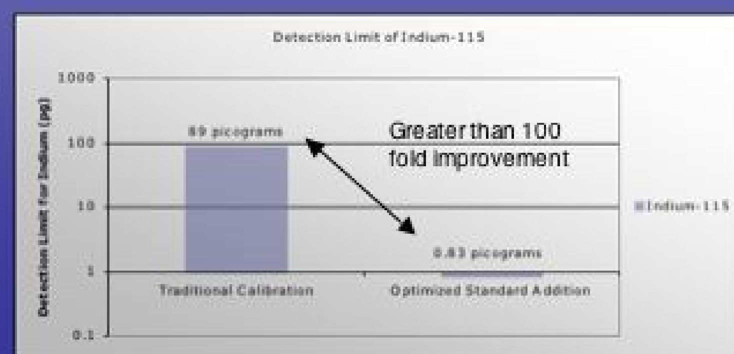




Natural variance of Pb isotopes worldwide can provide a somewhat unique isotopic signature for unknown samples containing Pb (less than 5 ppb)

Improved analytical capability for characterizing elemental impurities in uranium oxide

- Uranium oxides contain elemental impurities that can be used to determine provenance and handling procedures.
- Signal suppression from a high uranium matrix reduces our ability to accurately measure trace element impurities.
- A new technique was developed which reduces signal suppression and improves data quality.



Sorption Studies of Eu and Cs with Urban Surfaces i.e. How to Clean Up the Worst of Messes

Jeremy G. Waen, Nuclear Science Summer Internship Program

Mark Sutton, CBND, CMS, Bob Fischer, RHWM, SEP, Brian Viani, Geochemistry, E&E

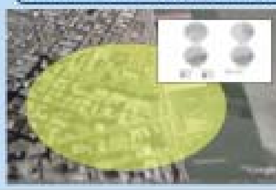
Glenn T. Seaborg Institute

Reed College

August 10th, 2006

Abstract

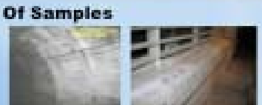
The Issue: Terrorist-Dispersed Radionuclides

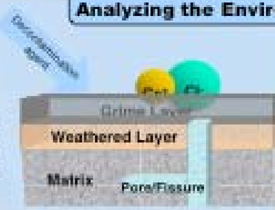


The Chemistry Behind The Threat

The Approach: Analyzing the Environment

A Plethora Of Samples

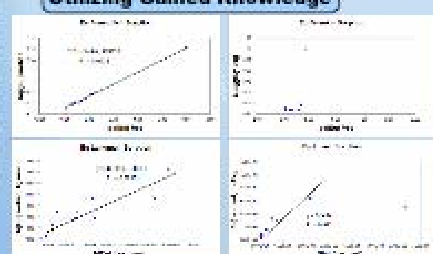




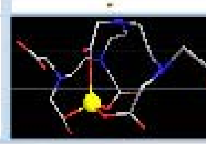
Looking Below The Surface

The Solution: Utilizing Gained Knowledge

Sorption Studies Of Radionuclide Analogs



Discussion, Conclusions, & Future Plans



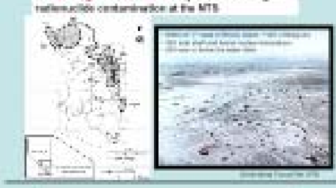
Optimization of Ion-Exchange Separations of Neptunium, Plutonium, and Uranium in Groundwater Samples from the Nevada Test Site

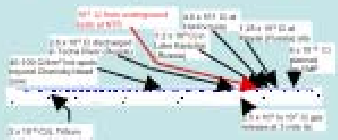
John P. Clements, Nuclear Science Summer Internship Program
Mavrik Zavarin, Pihong Zhao, Chemistry & Material Science
Glenn T. Seaborg Institute

Recent reactive transport model simulations of the Cambric underground nuclear test have predicted Np concentrations at three wells (RNM-1, RNM-2S, and UE-5n) that are inconsistent with reported Np concentrations at those sites. To resolve this discrepancy, we have improved our detection limits and optimized our procedures for Np, Pu, and U separation from Nevada Test Site (NTS) groundwater. Np preconcentration and separation was performed on groundwater samples from the three wells of interest, RNM-1, RNM-2S, and UE-5n. Np preconcentration from 10 liter groundwater samples was performed using $KMnO_4$ and $FeCl_3$ as [illegible] scavengers. The Mn/FeOx precipitates were dissolved in concentrated HNO_3/HF, dried, redissolved into a 3M HNO_3/0.22M $NH_2OH \cdot HCl$/0.02 M Fe(II) solution, and loaded onto Eichrom TEVA resin columns. The Hydroxylamine HCl is able to reduce Pu to Pu(III), while leaving U as U(VI), and reducing Np(V) to Np(IV). Np(IV) is favorable to TEVA column sorption. 3M HNO_3 effectively removed Pu(III) and U(VI) from the column. Np elution was accomplished with 0.02M HCl + 0.2M HF. Samples are awaiting ICP-MS analysis; based on ^{239}Np gamma counting, >50% recovery has been achieved.

Introduction

The Underground Test Area Project is evaluating radionuclide contamination at the NTS.





Recent data indicate that Np may be present at RNM-1, RNM-2s, and UE-5n. Reactive transport models suggest little or no Np transport to UE-5n. We are attempting to resolve this discrepancy.

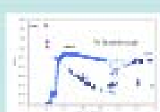
Methods

Preconcentration:

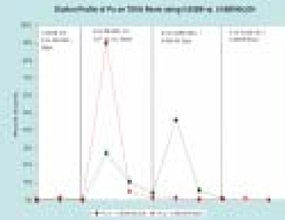
- We considered preconcentration based on current LANL and LLNL procedures:
  - Manganese oxide
  - Iron oxide
- MnO_2 was chosen for preconcentration and the following were investigated:
  1. How much MnO_2 is required to quantitatively scavenge Np from 10 L groundwater samples collected from NTS wells?
  2. Is the addition of Fe(II) into the solution beneficial to Np scavenging?

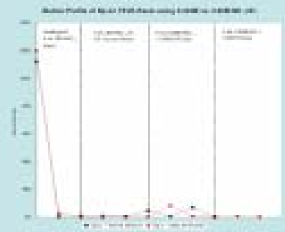
Column Separation:

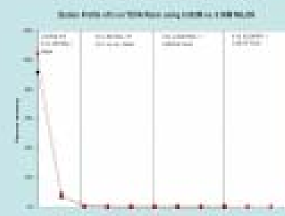
- Np separation effectiveness was investigated:
  1. What oxidation or reduction chemistry is required to effectively separate Np in NTS type groundwater matrices? (Hydroxylamine concentrations needed for reducing Np(V) to Np(IV) and what effect it has on Pu)
  2. What role does Fe(II) play in Np(V) reduction to Np(IV) and to the final separation on the TEVA column?
  3. Can ^{239}Np ($t_{1/2}$=2 days) be effectively used to calculate ^{237}Np recovery from groundwater?
- Optimized column separation:
  1. Loading in a 3M HNO_3/0.22M NH_2OH/0.02M Fe(II) solution.
  2. Wash with 3M HNO_3 to remove U and Pu.
  3. Elute Np with 0.02M HCl + 0.2M HF



Results







Discussion/Conclusions

- The addition of Fe into solution appears to be necessary for Np reduction/separation.
  - 0.002M NH_2OH, 9.0%
  - 0.06M NH_2OH, 5.1%
  - 0.22M NH_2OH/0.02M FeII, >60%
- A relatively high NH_2OH concentration is required for effective Pu and Np reduction (> 0.1M and preferably 0.22-0.33M)
- Uranium (which is a major interference to ICP analysis of Np) can be effectively removed in 8-10 bed volumes of 3M HNO_3.
- Excess $NaNO_2$ is required to effectively oxidize Pu(III) to Pu(IV), if it is desirable to sorb Pu to the TEVA resin. The nitrite must be sufficient to destroy all hydroxylamine in solution as well as oxidize Pu.
- At least 0.2M HF is needed to effectively remove Np from the column
- Final methods for NTS groundwater samples achieved ~55% Np recovery using ^{239}Np tracer
- ^{237}Np analysis is underway

LSST - The Next Level of Ground-Based Detection

Kevin Stein, Nuclear Science Summer Internship Program
Stephen Asztalos, I-Division, Physics and Advanced Technology
Lawrence Livermore National Laboratories

August 10, 2006

The Project

The Large Synoptic Survey Telescope (LSST) is a proposed telescope that will help astronomers solve certain cosmological problems. LSST, with its 8.4 meter diameter, will have a resolution of 0.146 arc seconds and a field of view of three square degrees. It will be able to detect a 24th magnitude star with only a ten second exposure. These specifications will allow astronomers to indirectly search for dark matter and observe distant supernovae that give clues to the shape of the universe. LSST is projected to be completed in 2012. Simulations were needed to examine how an industry standard detection program, Source Extractor, would respond to LSST images and what corrections would have to be made to obtain correct data.

Figure 1: Planned site for LSST

Figure 2: LSST Design

Discussion

The results of all of these simulations show astronomers how to correct the data that they will get from LSST. The analysis shows that astronomers won't have a problem making the ground-breaking observations that LSST is being built to make.

Figure 8: Drawing of LSST in use

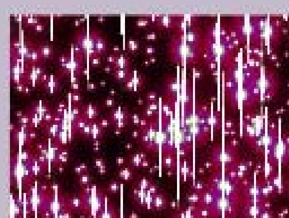


Figure 7: Pretty simulated image

Methods

* Using a randomly generated list of stars, a simulation of an LSST image was created using a program named Sky Maker.
* This image was then convolved with an atmospheric point-spread function (PSF). At first this PSF was manually created but later the program Arroyo was used to create a sophisticated PSF.
* The sources in the convolved image were extracted using Source Extractor.
* The brightnesses (magnitudes) and the ellipticities of the extracted stars were compared to the input data.

Figure 3: Initial image

Figure 4: Arroyo PSF

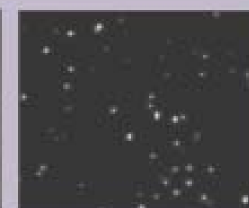


Figure 5: Convolved image

Results

* As magnitude increases, systematic error in detection increases.
* Worse seeing (higher FWHM) adds to the systematic error
* Added ellipticity doesn't have as much of an effect
* Source ellipticity is expected to be dependent on the length of the exposure

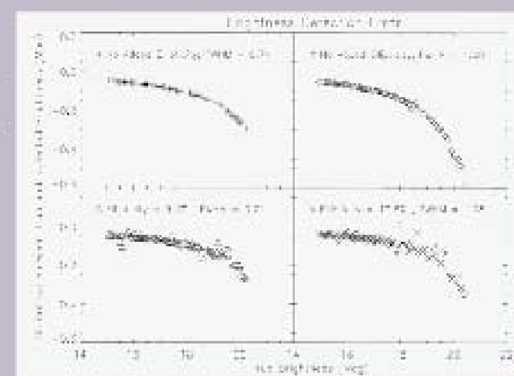


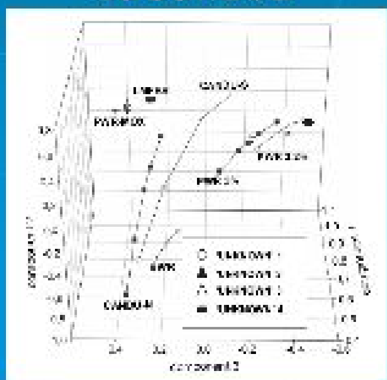
Figure 6: Results of initial simulations

This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

Characterization of Spent Reactor Fuel Using Principle Components Analysis

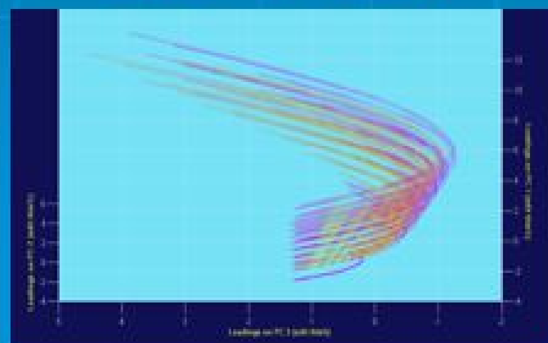
Martin Robel
University of California, Berkeley, Department of Nuclear Engineering
Lawrence Livermore National Laboratory Nuclear Science Summer Internship Program
Mentor: Mike Kristo, C&MS Division

Idealized



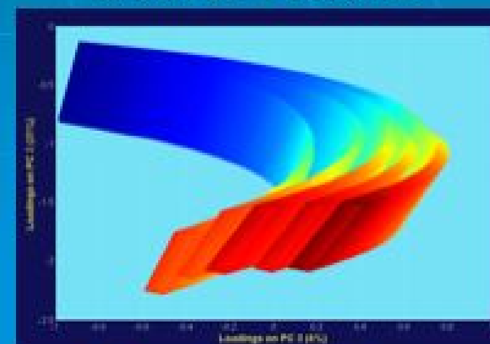
Note clear separation

Closer to Reality

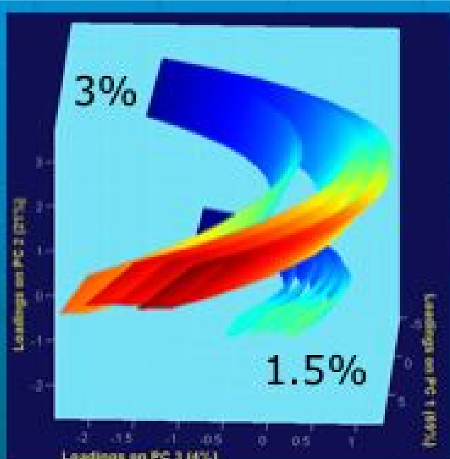


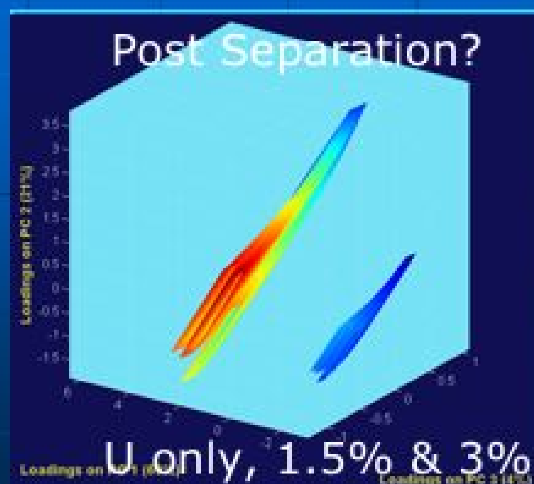
How to discern?

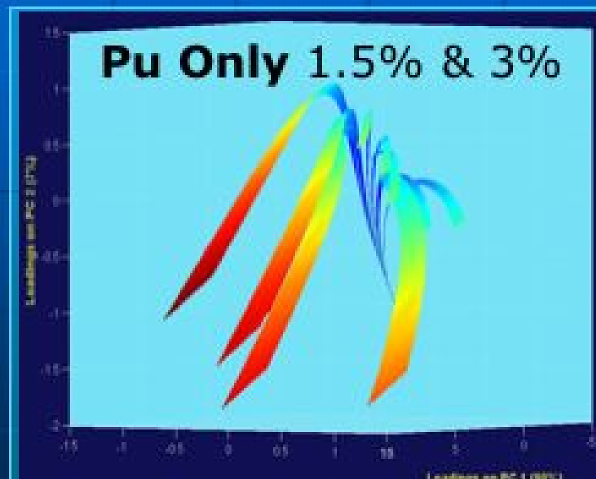
Clearer Picture



5 Reactors, 3%







Moving Toward Accurate Mercury Isotope Analysis: Evaluating Transport and Transformation Processes

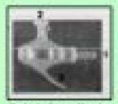
Paul G. Koster van Groos, Nuclear Summer Science Internship Program (LLNL), University of California, Berkeley
James R. Hunt, Department of Civil and Environmental Engineering, University of California, Berkeley
Ross Williams, Chemical Biology and Nuclear Science Division, Chemistry and Materials Science Directorate, Lawrence Livermore National Laboratory
Brad K. Esser, Chemical Biology and Nuclear Science Division, Chemistry and Materials Science Directorate, Lawrence Livermore National Laboratory

Glenn T. Seaborg Institute

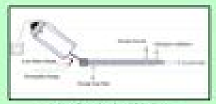
Two different sample introduction systems, an Aridus desolvating nebulizer and a PFA nebulizer with cyclonic spray chamber, in conjunction with a GV Instruments IsoProbe multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS), were evaluated for use in mercury isotope analysis. Results indicate that a significant fraction of mercury is lost within the desolvating nebulizer, and that both systems, albeit to differing degrees, suffer from memory effects that could influence isotopic measurements. Our measurements indicate that two or more fractionating processes could be occurring within the sample introduction systems. We suggest that these include the disproportionation of mercurous (Hg_2^{2+}) ions, as well as diffusion-limited sorption of elemental mercury (Hg^0). Future experiments will seek to eliminate these fractionating processes within the analytical method by complexing mercuric (Hg^{2+}) ions with strong chelators. Other experiments should verify the degree to which the processes identified above fractionate mercury.

Introduction

Methods

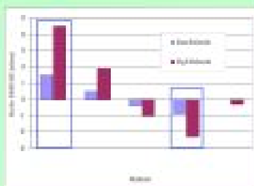


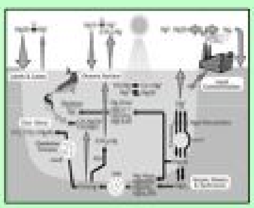
Cyclonic Spray Chamber

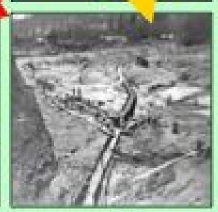


Aridus Desolvating Nebulizer

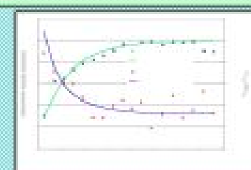
Environmental Context

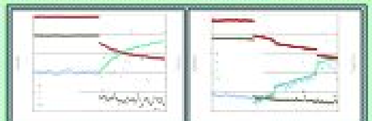


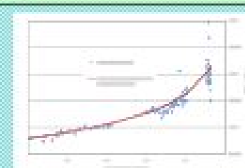




Results

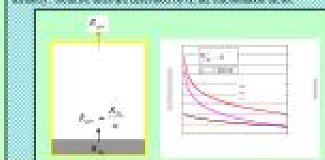






Discussion

Rayleigh Fractionation of Isotopes



Disproportionation

Diffusion-limited sorption

Conclusions

Compton-Electron Tracking Imaging Gamma-Ray Detector

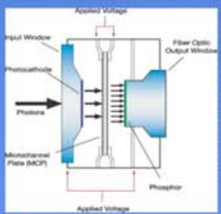
Rion E. Graham, Nuclear Science Summer Internship Program

Glenn T. Seaborg Institute

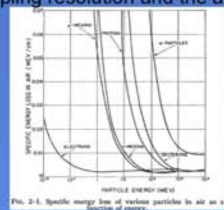
Ron Wurtz, I-Division, PAT

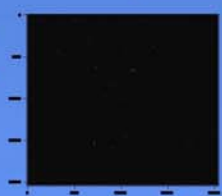
ABSTRACT

We have developed a novel Compton imaging gamma-ray detector targeted at energies in the 2.6 MeV range. The current configuration consists of a 1024-by-1024 pixel intensified CCD camera coupled to a Terbium-doped-glass cubic fiber array. Measured photon-count characteristics of MIP tracks allow for predictions of spatial and energy resolutions of Compton electron events within the cube through a simple conversion of specific-energy loss. A detailed quantitative comparison between the system's native sampling resolution and the angular resolution of electron tracks is in progress.



The use of an intensified CCD allows for the detection of a single photon. In addition, the camera's 50% Q.E. value means that half of the photons coming from the intensifier will be measured. In contrast, the former cameras were not intensified and had a Q.E. of only about 10%, producing images like those to the right. The difference in track resolution and intensity in stunning.





$$\left(\frac{MeV}{cm}\right)_{MIPS:class}\left(\frac{photons}{cm}\right)_{MIPS}^{-1}\cos(\theta) = \ldots$$

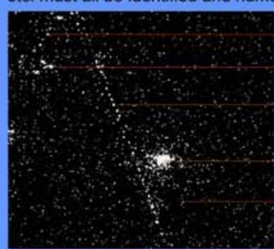
$$\left(\frac{MeV(E)}{cm}\right)_{electron:class}\left(\frac{1}{\ldots}\right) = \frac{photons}{cm} = \text{spatial resolution}$$

<u>Measured Spatial Resolution</u>: **382*cos(θ) photon/cm**, electron @ 1MeV or **26/(cos(θ)) μm/photon**, electron @ 1Mev

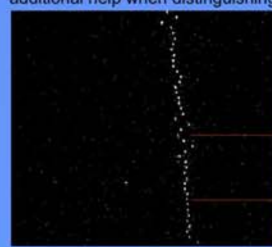
<u>Future Work</u>: Our first course of action is to purchase a second camera to be used in a coincidence-triggered simultaneous imaging configuration (similar to images seen on the left.) Once functional, we will be able to reconstruct continuous tracks for even greater energy and angular resolution. The correlation of the pictures will also help to further reduce noise and spurious events. Additionally, we hope to obtain a several-Mev gamma-ray source to produce electron tracks of sufficient length to test our predicted resolution.

The cubic fiber array (fibercube) is made of Tb-doped glass fibers, each 15μm in diameter, which are grouped into bundles 10 fibers high. These bundles are then joined to form '2-D' layers which are in turn stacked orthogonally to form the cube.

Image reduction is a vital step in the quantitative analysis of desired events. The bias, dark-current events, leaked visible photons, photocathode events, etc. must all be identified and numerically eliminated.



The MIP track shown below is from a 10ms exposure. There is a balancing between probability of occurrence and non-MIP noise that determines the optimal exposure length. The back-end mirror helps to intensify only those events that are within the cube, providing additional help when distinguishing particle tracks.



This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

Low Power, Low Noise Preamplifier for Semiconductor Detectors

~ for the future radiation detection; gamma-ray, X-ray, dark matter search ~

Glenn T. Seaborg Institute

Tsuguo Aramaki, Columbia University

Lorenzo Fabris, I-Division

Lawrence Livermore National Laboratory

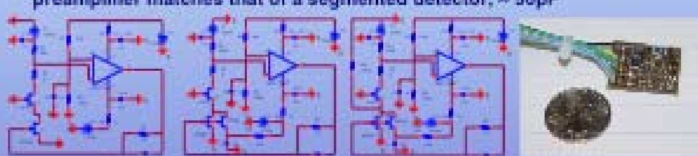
Nuclear Science Summer Internship Program

Segmented semiconductor detectors are very powerful instruments to detect both radiation and charged particles, due to their high energy-resolution. To enhance this characteristic, we are developing a low power, low noise preamplifier by using two state-of-the-art, very low noise JFETs as the input stage of the preamplifier. This way, the total input capacitance of the preamplifier is a good match to that of the detector's segments, while preserving the same low noise performance of the single device. We were able to achieve an ideal result with a low FWHM noise for Ge and Si detectors. This will be widely applicable for several radiation detection project, such as the Compton Imaging project for the Department of Homeland Security and the GAPS project, an indirect dark matter search and a collaboration between LLNL and Columbia University.

Objective:

How much can we reduce the noise of preamplifier by using two JFETs as the input stage?

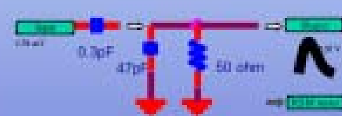
- Use two JFETs as the input stage so that the input capacitance of the preamplifier matches that of a segmented detector, ~ 50pF



scheme of the 1-JFET preamplifier, 2-JFET with additional power and board

Methods:

- Inject 178 mV into 0.3pF capacitance, 47pF detector capacitance
  This simulates 1 MeV signal in a Ge detector
- Set the gain of the spec-amp such that V_{peak} = 2.35 V
- Remove the source and measure the FWHM noise at rms meter
  1mV rms = 1keV FWHM
- Measure the noise in the different conditions; Peaking Time 0.5 us -15 us



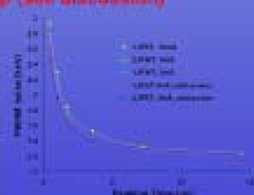
- 1-JFET with 10 mA, 2-JFET with 5 mA each
- 1-JFET with 5 mA (R3 = 1.6k)
- 2-JFET with 5 mA each with additional power (reduce the noise contribution of the op-amp)

Results:

- As shown below, in the same condition, the 2-JFET preamplifier has 4-11% less noise at fast peaking time.
- As peaking time is longer, the noise becomes equal or a little bit worse than 1-JFET, but this is due to 1/f noise from the connection with the board.

⇨ make the components of the input stage stand-up (see discussion)

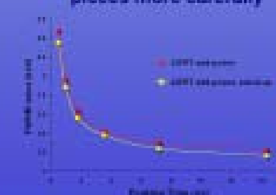
FWHM noise (keV)



FWHM noise vs. Peaking Time

Discussion:

- As shown below, if we make the pieces stand-up, the noise is reduced and the 2-FET preamplifier shows better results even at slow peaking time.
- As a further improvement, we need to consider the arrangement of the pieces more carefully



FWHM noise of 2-JFET and 2-JFET stand-up

Conclusion:

2-JFET as input stage is helpful to reduce the noise.

Also, the FWHM, 1.1 - 2.3 keV FWHM of 1 MeV signal, is very practical for the readout of the segmented semiconductor radiation detection with many channels

Contextually-Aware Expert-System for Automated Nuclear Threat Assessment

Shadi Z. Ghrayeb, Rensselaer Polytechnic Institute,

S. Labov , M. Pivovaroff I-Division, Physics & Advanced Technologies Directorate
Lawrence Livermore National Laboratory,

Rensselaer

Glenn T. Seaborg Institute

The detonation of a nuclear or radiological device in the United States would have a catastrophic effect. These national laboratories, universities, industries and research centers have been collaborating in finding solutions to the issues of dealing with countering clandestinely delivered nuclear and radiological threats. Current detection monitors deployed at border-crossings and other strategic locations are still susceptible to high false-alarms rates, due to the detection of radioactive nuisance sources from industrial, medical or naturally occurring radioactive material (NORM). High false alarms limit the monitors' effectiveness at the screening of cargo, trucks and personal vehicles. Our goal is to develop a detailed categorization system called, "Contextually-Aware Expert-System for Automated Threat Assessment" for guiding the accuracy of an instrument's identification. The system involves a program that captures the deep knowledge and understanding possessed by a human expert, and based on particular observables, returns a specific answer or response that fits the data. This system will be provided as effective support of the field agents so that they can process an order of magnitude more events per unit time while halving the number of calls to the primary reach-backs.

Introduction:

This project combines ambiguous output collected from border-crossing detectors with contextual information in a novel expert system to help resolve radiation alarms.

In the instance of such an alarm, a human expert is often required to intervene to resolve the situation. The expert considers the spectroscopic signal while incorporating contextual information and past experience in similar situations to make a determination to pass the shipment or initiate a detailed search. This project will develop a computerized system that captures the deep knowledge and understanding possessed by a human expert where the project returns a specific answer or response to fit that data.

Radiation Portal Monitor (RPM)

Traffic through RPM

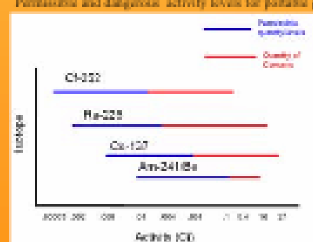
Results:

We have begun assembling databases on commonly used nuclear material. In addition to the many analysis of nuisance sources we have collected, we are collecting lists of manufactures, users and shippers of nuclear materials. This includes information about possible applications for each isotope in commercial and medical uses. These data will provide inputs to the user and the expert system regarding the normal use of these materials, and will help in the anomaly detection analysis.

Cs-137 Spectrum

Spectrum analysis identifies Isotope

Permissible and dangerous activity levels for portable gauges



Expert system determines the threat level

Methods:

As part of my role in this project I will perform a mapping of the natural radiation. background. A BNC SAM-935 detector system was mobilized into a backpack unit. Sensor data is sent to a hand-held palm device and transmitted to a remote database for storage. Included in the transmission are time of fix, GPS coordinates, count rate, and spectrum information. This technique of measuring radiation levels and analyzing the collected spectrum will be familiar to the secondary inspections of cargo performed by border patrol custom agents.

In analyzing the collected spectrum, artificial intelligence will be used to provide information about the manifest to implement into our system.

Discussion:

In the instance of such an alarm, a human expert is often required to intervene to resolve the situation. Our plan is to refine our system towards our eventual goal of implementing a fully-automated system to support the Advanced Spectroscopic Portal monitors in the field. This requires a combination of a deep understanding of a specific problem and a hierarchical series of rules.

We anticipate the impact of our system will be most significant when the number of measurement systems increases as more points of entry, state and local detection systems are implemented. The number of measurement systems could easily increase by factors of 10 or more, which would overwhelm the current primary reachback capabilities.

This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

UCRL-xx-xxxxxx

Monitoring Nuclear Reactors with Antineutrinos

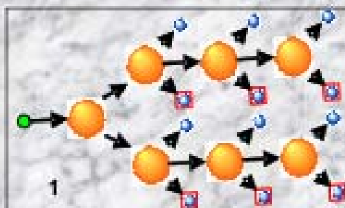
Alex Johnson, Nuclear Science Summer Internship Program

Adam Bernstein, I Division, PAT Directorate

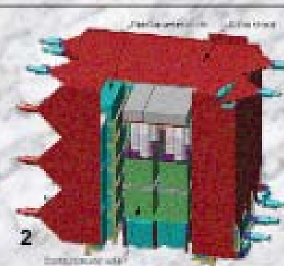
Glenn T. Seaborg Institute

Georgia Institute of Technology

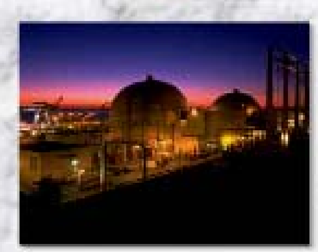
- **Theory**
  - Antineutrino Production[1]
  - Detection[2]
    - High flux of antineutrinos
    - Scintillator volume
    - Advanced shielding
  - Preventing Diversion
    - Detecting an abnormal antineutrino rate
    - Modeling probable diversion schemes
- **Experiment**
  - SONGS[3]
    - Operated for 1.5 years with little or no maintenance
    - Located 25 meters from reactor core[4]
- **Simulation**
  - Diversion Scenarios
    - Baseline – Standard reactor cycle
    - A – 3.5% replaced with 1.5%
    - C – Spent fuel replaced with fresh fuel
    - D – Reactor run at 105% power
  - ORIGEN
    - Provides burnup dependent isotopic data for each scenario
    - Predicts fission rates for each isotope per burnup step
  - Conversion[5]
- **Conclusions**
  - Due to uncertainty in simulation A and C are undetectable[6]
  - Diversion D is different from the baseline by 5%
  - Diversion D verifies methods and supports use in power monitoring
  - Can monitor power weekly to within 3%[7]



1



2

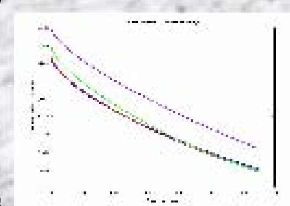


3

4

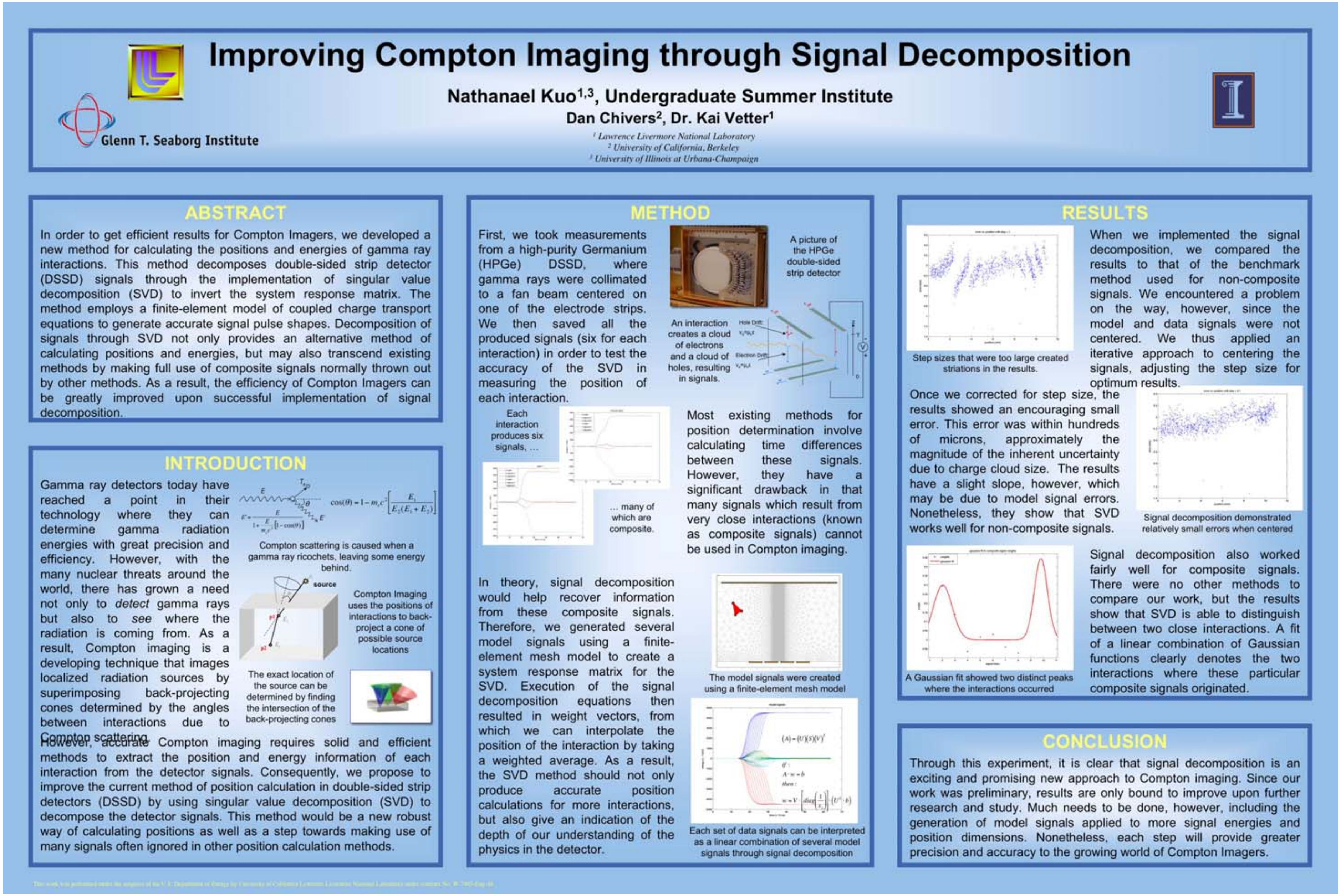
$$\frac{dN_{\nu}(E)}{\Delta E \cdot dT} = \left(\frac{N_{targets}}{4 \cdot \pi \cdot distance^2}\right) \cdot \varepsilon(E) \cdot \left(\frac{Nfissions_i}{second} \cdot \frac{dN_{\nu_i}}{dE \cdot fission} \cdot \sigma(E)\right)$$

5



6

7


Improving Compton Imaging through Signal Decomposition
Glenn T. Seaborg Institute
Nathanael Kuo[1,3], Undergraduate Summer Institute
Dan Chivers[2], Dr. Kai Vetter[1]
[1] Lawrence Livermore National Laboratory
[2] University of California, Berkeley
[3] University of Illinois at Urbana-Champaign
ABSTRACT
In order to get efficient results for Compton Imagers, we developed a new method for calculating the positions and energies of gamma ray interactions. This method decomposes double-sided strip detector (DSSD) signals through the implementation of singular value decomposition (SVD) to invert the system response matrix. The method employs a finite-element model of coupled charge transport equations to generate accurate signal pulse shapes. Decomposition of signals through SVD not only provides an alternative method of calculating positions and energies, but may also transcend existing methods by making full use of composite signals normally thrown out by other methods. As a result, the efficiency of Compton Imagers can be greatly improved upon successful implementation of signal decomposition.
INTRODUCTION
Gamma ray detectors today have reached a point in their technology where they can determine gamma radiation energies with great precision and efficiency. However, with the many nuclear threats around the world, there has grown a need not only to detect gamma rays but also to see where the radiation is coming from. As a result, Compton imaging is a developing technique that images localized radiation sources by superimposing back-projecting cones determined by the angles between interactions due to Compton scattering.
Compton scattering is caused when a gamma ray ricochets, leaving some energy behind.
Compton Imaging uses the positions of interactions to back-project a cone of possible source locations
source
The exact location of the source can be determined by finding the intersection of the back-projecting cones
However, accurate Compton imaging requires solid and efficient methods to extract the position and energy information of each interaction from the detector signals. Consequently, we propose to improve the current method of position calculation in double-sided strip detectors (DSSD) by using singular value decomposition (SVD) to decompose the detector signals. This method would be a new robust way of calculating positions as well as a step towards making use of many signals often ignored in other position calculation methods.
METHOD
First, we took measurements from a high-purity Germanium (HPGe) DSSD, where gamma rays were collimated to a fan beam centered on one of the electrode strips. We then saved all the produced signals (six for each interaction) in order to test the accuracy of the SVD in measuring the position of each interaction.
A picture of the HPGe double-sided strip detector
An interaction creates a cloud of electrons and a cloud of holes, resulting in signals.
Hole Drift
Electron Drift
Each interaction produces six signals, …
… many of which are composite.
Most existing methods for position determination involve calculating time differences between these signals. However, they have a significant drawback in that many signals which result from very close interactions (known as composite signals) cannot be used in Compton imaging.
In theory, signal decomposition would help recover information from these composite signals. Therefore, we generated several model signals using a finite-element mesh model to create a system response matrix for the SVD. Execution of the signal decomposition equations then resulted in weight vectors, from which we can interpolate the position of the interaction by taking a weighted average. As a result, the SVD method should not only produce accurate position calculations for more interactions, but also give an indication of the depth of our understanding of the physics in the detector.
The model signals were created using a finite-element mesh model
Each set of data signals can be interpreted as a linear combination of several model signals through signal decomposition
RESULTS
When we implemented the signal decomposition, we compared the results to that of the benchmark method used for non-composite signals. We encountered a problem on the way, however, since the model and data signals were not centered. We thus applied an iterative approach to centering the signals, adjusting the step size for optimum results.
Step sizes that were too large created striations in the results.
Once we corrected for step size, the results showed an encouraging small error. This error was within hundreds of microns, approximately the magnitude of the inherent uncertainty due to charge cloud size. The results have a slight slope, however, which may be due to model signal errors. Nonetheless, they show that SVD works well for non-composite signals.
Signal decomposition demonstrated relatively small errors when centered
Signal decomposition also worked fairly well for composite signals. There were no other methods to compare our work, but the results show that SVD is able to distinguish between two close interactions. A fit of a linear combination of Gaussian functions clearly denotes the two interactions where these particular composite signals originated.
A Gaussian fit showed two distinct peaks where the interactions occurred
CONCLUSION
Through this experiment, it is clear that signal decomposition is an exciting and promising new approach to Compton imaging. Since our work was preliminary, results are only bound to improve upon further research and study. Much needs to be done, however, including the generation of model signals applied to more signal energies and position dimensions. Nonetheless, each step will provide greater precision and accuracy to the growing world of Compton Imagers.

Materials in Extreme Environments: The FeCr Challenge

Ryan Till, Undergraduate Summer Institute, Arizona State University

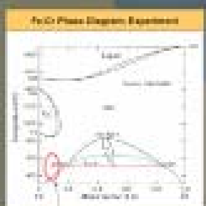
Glenn T. Seaborg Institute

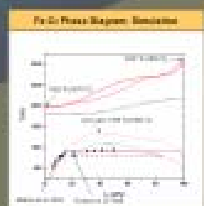
M. Caro (CBNS) and A. Caro (MSTD)
Lawrence Livermore National Laboratory

FeCr is the candidate cladding material for advanced fission reactors. Iron chromium (FeCr) alloys must withstand extreme conditions (high temperatures, high neutron flux, corrosive environments). Radiation damage significantly alters the alloys crystalline structure. Defects (interstitials and vacancies) are created by high-energy collisions of neutrons with the Fe-matrix. In this work, we utilize models and computational tools to study their behavior at the atomic level. We determine formation energies for interstitials in FeCr alloys for various Cr concentrations and configurations. Energies of Cr cluster formation are established.

Introduction:

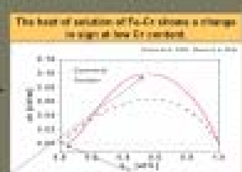
- Extreme environments (High temperature, high dose, corrosion)
- Existing structural materials are affected by radiation damage (swelling, creep, embrittlement)
- Defects are created when high energy neutrons collide with the lattice of the cladding material
- Microstructural changes must be investigated.

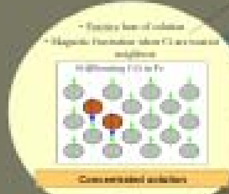




The FeCr Challenge:

-magnetic effects determine the formation energy!

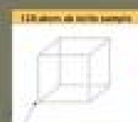




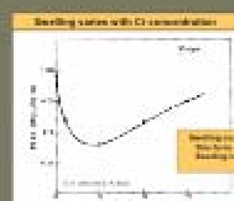
As Cr concentration increases, Cr clusters form and heat of formation becomes positive.

Methods:

- Defects are extremely small. They are below the resolution of microscopes
- Computer simulations are used as an aid.
- There are a variety of different methods to model atomic level interactions.
- Ab initio uses small samples and models each atom using Schrodinger's equation.
- Molecular Dynamics is able to tackle larger samples.



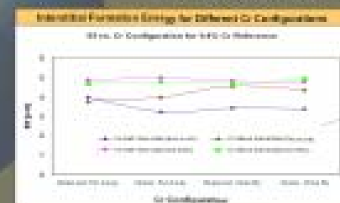
- Comparisons are done to ensure semi-empirical approach (Molecular Dynamics) captures quantum interactions (ab initio).
- Size effects occur in the small ab initio sample.
- The sample is repeated eight times to capture these effects.
- Thus, we compare similar results.

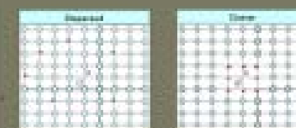


- Swelling is the result of large amounts of point defects.
- A swollen material has decreased strength.

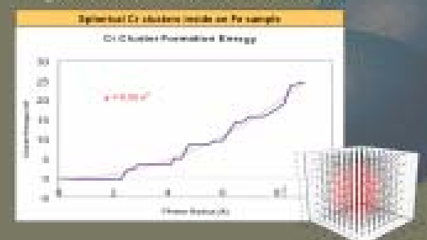
Results:

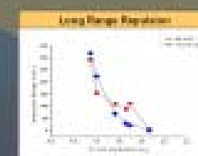
- Interstitial formation energies found using Molecular Dynamics & compared to ab initio results for various Cr configurations.





- Energy of spherical Cr clusters inside a pure Fe sample as a function of Cr cluster radius.



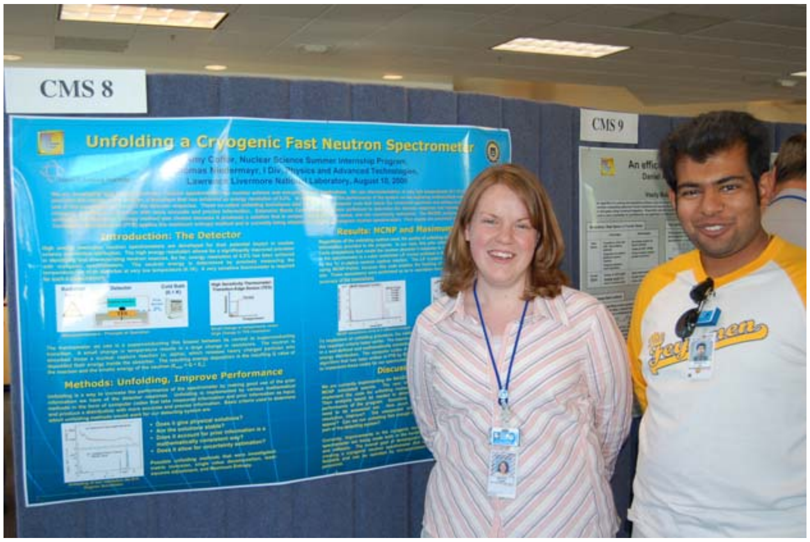


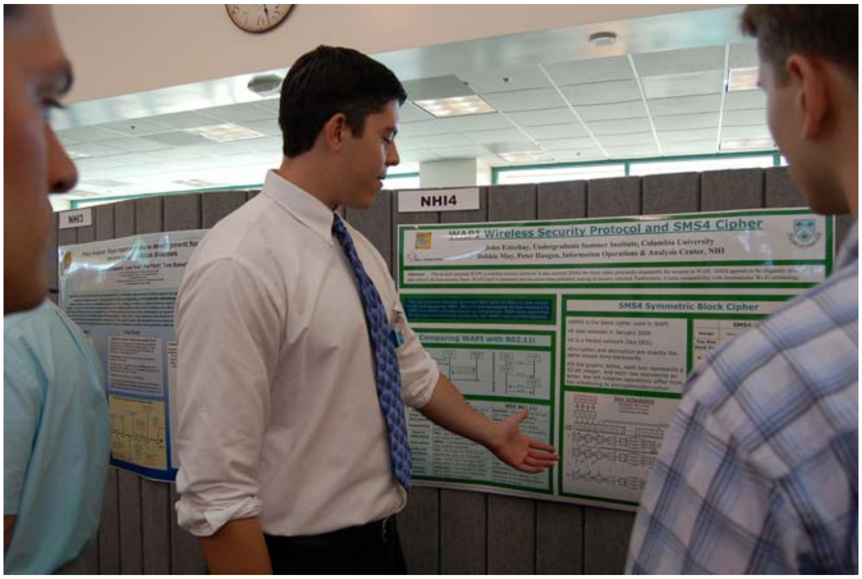
Discussion:

Comparison of results obtained from ab initio and empirical models is a powerful way to test the validity and accuracy of the empirical approach. In this work we have been able to test our Fe-Cr classic potential against a series of ab initio results on particular solute configurations. This comparison showed us some points of discrepancy that were corrected in a second, improved, version of the potential.

Additionally, we calculated the energy of increasingly bigger spherical Cr clusters embedded in Fe in order to determine the interface energy between alpha and alpha' precipitates, a parameter that enters nucleation rate theory to determine critical size of precipitates.

Summer students at LLNL 2006






Liquid Water Through Classical and Quantum Computer Simulations
University of California, Davis
CMS 32
CMS 33
Results:
Discussion:


CMS 6
Abstract
Introduction: