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# Materials Modeling for High-Performance Radiation Detectors

V. Lordi

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**MATERIALS MODELING FOR HIGH-PERFORMANCE RADIATION DETECTORS**

LL13-MatModelRadDetect-PD2Jf

**Vincenzo Lordi (PI)****Nicole Adelstein, Joel Varley, Keith Ray, Daniel Åberg, Michael Skarlinski, Eunae Cho****1. INTRODUCTION**

Limitations in the performance of room-temperature gamma radiation detectors are intimately related to the properties of the materials used to fabricate them. In particular, atomic-scale defects in the materials that can depend on crystal growth or device processing conditions often ultimately determine the performance of a given detector. Understanding the origins and consequences of these various defects is essential to optimizing the materials and achieving maximum device performance (*e.g.*, energy resolution and sensitivity).

This project aims to enable rational materials design for select high-payoff challenges in radiation detection materials by using state-of-the-art modeling techniques bridging multiple resolutions. Three specific high-impact challenges are addressed: (i) design and optimization of electrical contact stacks for thallium bromide (TlBr) detectors to stabilize temporal response at room-temperature; (ii) design of host glass for large-volume, low-cost, high-performance glass scintillators; and (iii) determination of the electrical impacts of dislocation networks in cadmium zinc telluride (CZT) that limit its performance and usable single-crystal volume.

**2. THALLIUM BROMIDE**

TlBr detectors show great promise for high-resolution room-temperature gamma spectroscopy, but rapid decay of their performance over hours to days has stymied practical implementation. Recent results have indicated that various surface treatments and the choice of metal used for electrical contacts can have a dominant impact on the longevity of detectors. We seek a detailed mechanistic understanding of the effects of electrical contact design and fabrication, to enable rational optimization of the contacts for maximum detector lifetime.

**3. GLASS SCINTILLATORS**

Glass scintillators may hold great promise for low-cost, large-volume, shapeable detectors. However, the light yield and energy resolution of glass scintillators to-date have been more than an order of magnitude worse than for crystalline scintillators. Moreover, their performance has been far worse than the theoretical limits. Glass scintillators are fabricated by dissolving rare-earth dopants into a glass matrix. Their operation relies on gamma ray absorption in the glass that excites electrons that then must migrate to a dopant atom and emit light. The limiting process is the transport of electrons in the glass, so that they reach an activator before decaying without emitting light. We aim to discover the nature of electron transport in specific glass materials and how it may be enhanced by tuning composition, structure, or forming process. Ultimately, such knowledge can enable the fabrication of high-performance glass scintillators.

**4. CADMIUM ZINC TELLURIDE**

CZT is the most mature room-temperature gamma radiation detector material, but high-yield growth of large uniform crystals that exhibit the best spectroscopic quality remains problematic.

One of the remaining unknowns affecting performance is the electrical impact of dislocation networks in the material. Dislocations are one-dimensional crystal defects that sometimes also include impurities. How dislocations affect electrical transport in the crystal (*e.g.*, carrier lifetimes) is not understood, but is of critical importance to detector performance. We aim to use atomic-scale simulations of dislocations in CZT to determine their electrical properties and assess the performance limitations associated with their presence.

## 5. RESULTS, DISCUSSION AND CONCLUSIONS

### 5.1 THALLIUM BROMIDE ELECTRICAL CONTACTS

During the past year, two aspects of the electrical contacts were studied: (i) the impact of metal choice from the point of view of being a source of impurities in TlBr; and (ii) the materials properties of chlorine-treated interfaces.

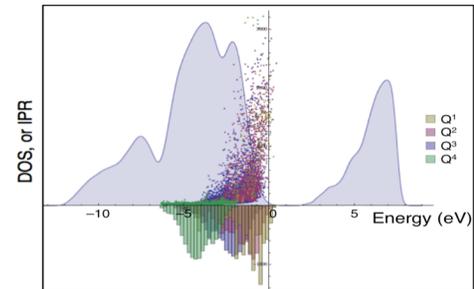
Experimental results have shown that detector performance and longevity vary with choice of contact metal, and also that migration of metal into TlBr is possible. We used simulations to assess the electronic properties of more than a dozen different metal impurities in TlBr. We determined each metal's migration rate, whether it contributes to loss of detector signal current (carrier trapping), and whether it incorporates into TlBr as a charged species. Our analysis allowed us to predict the highest performance metals for both the anode and cathode electrodes, using the assumptions above. For the anode, we ranked indium (In), nickel (Ni), and platinum (Pt) highest; for the cathode, chromium (Cr), titanium (Ti), and molybdenum (Mo) were best. These predictions were validated by experiments that showed improved longevity of devices using Pt/Pt or Ni/Cr contacts.

In addition to in-migration of metal impurities from the contacts, chemistry at the contact interface can form compounds that modify the electrical characteristics of the device or modify the propensity for metal impurity migration. Such interface chemistry may be intentionally controlled via chemical treatment of the surface prior to depositing metal. Hydrochloric acid (HCl) treatment has been shown to convert the TlBr surface to a chlorinated TlBrCl alloy, with enhanced device longevity. Using atomistic simulations, we determined a role played by Cl treatment is to block injection of holes from metal into TlBr. The TlBrCl material also favorably alters the properties of metal impurities in the interface region. These mechanistic understandings help enable design of future contact stacks with optimal engineered properties for detector longevity.

### 5.2 GLASS SCINTILLATOR HOST

Since little is known fundamentally about the correlation of composition, atomic structure, and electronic transport properties of glass materials, our first task was to establish a methodology to generate, assess, and validate atomistic models of glass materials, including computational knobs as proxies for experimental variability. We tested computational melt-quench-anneal procedures using various descriptions of the interatomic interactions to obtain structural models of silica and sodium silicate glasses, as validation. We obtained structures in good agreement with experiments. We also assessed the sensitivity of various parameters for generating models of glass structures that vary from nominal experimental structures.

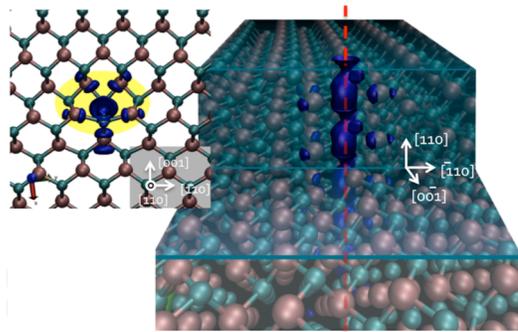
To study the electronic properties of the glass materials related to electron transport that limits scintillator performance, we developed a new methodology to correlate atomic structural features with electronic states associated with carrier trapping. More precisely, we analyzed the localization of electronic states (carrier trapping propensity) throughout the so-called band tails (ranges of excitation energies associated with poor electron transport) and could identify atomic motifs responsible for the most carrier trapping. We exercised this methodology on a sodium silicate glass, which is not particularly a good scintillator glass, but for which good experimental data exists for comparison. Future work will focus on performing similar analyses on known-better and known-worse glass scintillator hosts to discover correlations that can indicate desired structures for optimal carrier transport. Then, strategies for developing a glass composition and/or forming process to favor such structures can be developed, using both simulations and experiments to investigate control over variability of atomic structures of glasses.



**Figure 1. Electronic states near the band edges in sodium silicate glass show varying degrees of electron trapping associated with structural features denoted by  $Q^n$  numbers.**

### 5.3 DISLOCATIONS IN CADMIUM ZINC TELLURIDE

During the past year, we generated atomistic models of seven different likely dislocations in CZT using cadmium telluride as a model system, calculated their relative stabilities, and evaluated their electronic properties. To perform this work, we developed and tested methods to accurately treat simulations containing tens of thousands of atoms by combining methodologies. We found that the most stable dislocation structure, a *straight screw dislocation*, is fairly innocuous electrically and does not contribute to carrier capture. However, several other possible structures (*straight edge dislocations*) can capture electrons or holes. Edge dislocations contain either Cd or Te dimers in their cores and consequently may trap electrons or holes to varying degrees. An interesting result is that certain straight edge dislocations can form a conducting channel for trapped carriers along the dislocation line, actually enhancing transport in that direction, particularly if the dislocations can be aligned in a crystal.



**Figure 2. Holes captured by certain edge dislocations in CZT are conducted down the dislocation line.**

## 6. PUBLICATIONS

J.B. Varley, A.M. Conway, L.F. Voss, E. Swanberg, R.T. Graff, R.J. Nikolic, S.A. Payne, V. Lordi, and A.J. Nelson, "Effect of chlorination on the TlBr band gap for improved room temperature radiation detectors," *submitted* (2014).

V. Lordi, D. Åberg, M. Skarlinksi, and E. Cho, "Atomic and Electronic Structure of Dislocations in Cd(Zn)Te," extended abstract for Nuclear Science Symposium & Room-Temperature Semiconductor Detector Workshop, Seattle, Nov. 10-14, 2014.