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Office of Nonproliferation and Verification Research & Development

SNM Movement Detection / Radiation Sensors and Advanced Materials Portfolio Review

RadSensing2011

Improved Growth of CZT Crystals

Document Number 5295420

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Sandia National Laboratories

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Participants / Budget

- **Sandia National Laboratories ($620K)**
  - **Sandia CA**
    - F. P. Doty (PI)
    - X. W. Zhou, D. K. Ward, B. M. Wong (Modeling / Simulation)
  - **Sandia NM**
    - Qiming Li (Microcopy Analysis)

Project Overview

- **Background**
  - Understanding and control of small scale defects (in particular dislocations) leads to improved CZT crystals.
- **Goals**
  - Study fine scale defects with focus on dislocations (configurations, effects on properties, formation mechanisms).
- **Technical Approaches**
  - Combine experiments with state-of-the-art simulation methods.
  - Use quantum-accuracy-enabling bond order potential (BOP) to construct atomistic simulation models.
- **Deliverables**
  - Experimental and theoretical results on effects of fine scale defects.
  - Journal publications and reports.
  - BOPs for CdTe, CdZn, TeZn and CdTeZn.
  - BOP-based molecular simulation package LAMMPS.
- **Capability Improvement to be Addressed**
  - Capability to understand and reduce detrimental defects with scales smaller than those explored in the past. This helps increase the yield of detector-grade CZT crystals.
- **Progresses**
  - Demonstrated sample-preparation-free experiments for dislocation analysis in CZT crystals.
  - A review paper for CZT modeling was accepted in Journal of Chemical Physics. Presented results in numerous conferences.
  - Developed and validated CdTe BOP.
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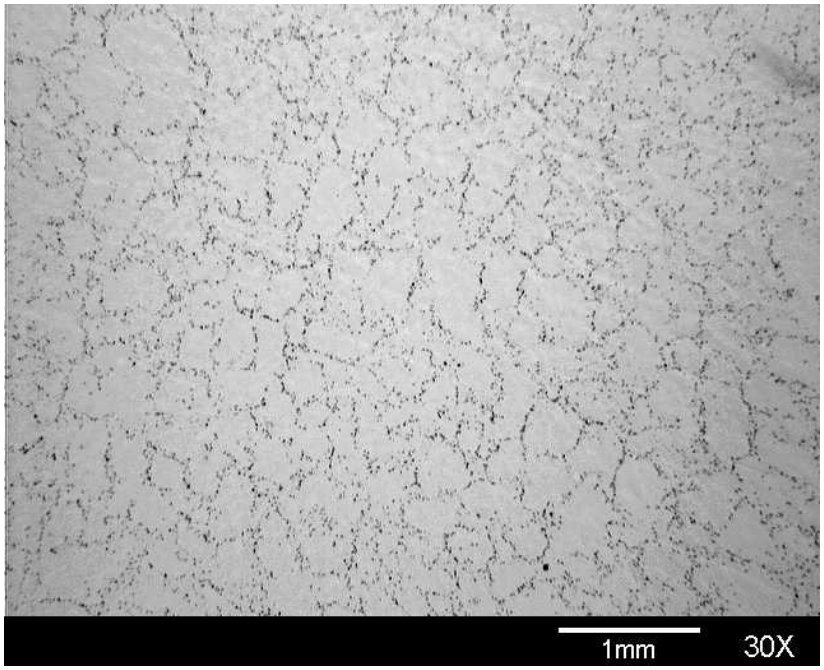
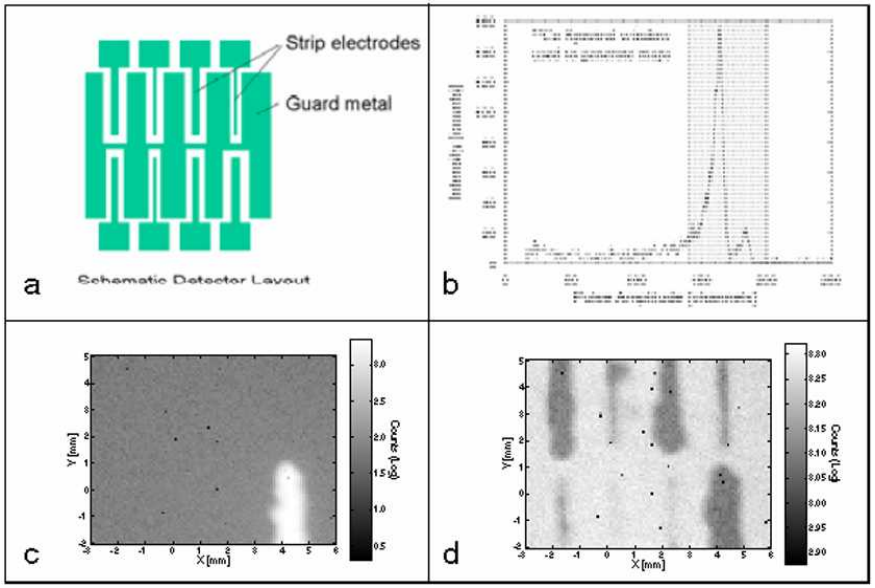
- **Technical Challenges**
  - Multi-physics, multiscale problem; Computational expenses; Gap between theoretical and practical growth conditions.

Background: Lateral Shift ~ 0.1 – 0.5 mm





Left: dislocation subgrains; Right: (a) schematic of metal on anode surface; (b) pulse height window used to form images; (c) single strip electrode response map; and (d) guard response map (collimated 122 keV photons using 250 mm aperture).

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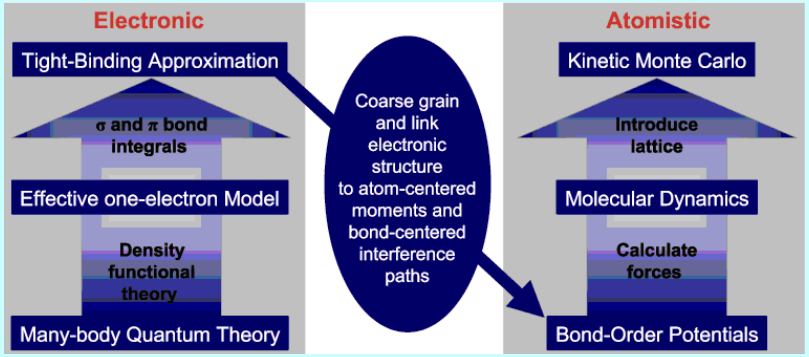
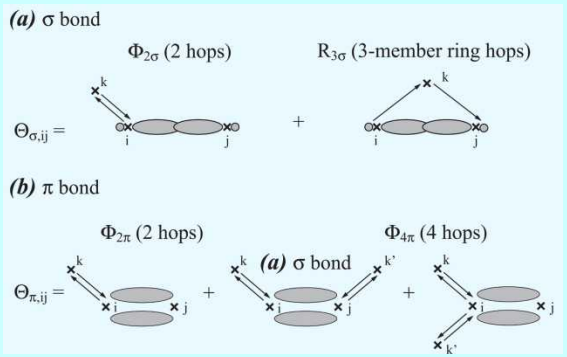
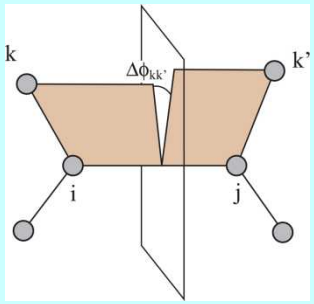
BOP Origin







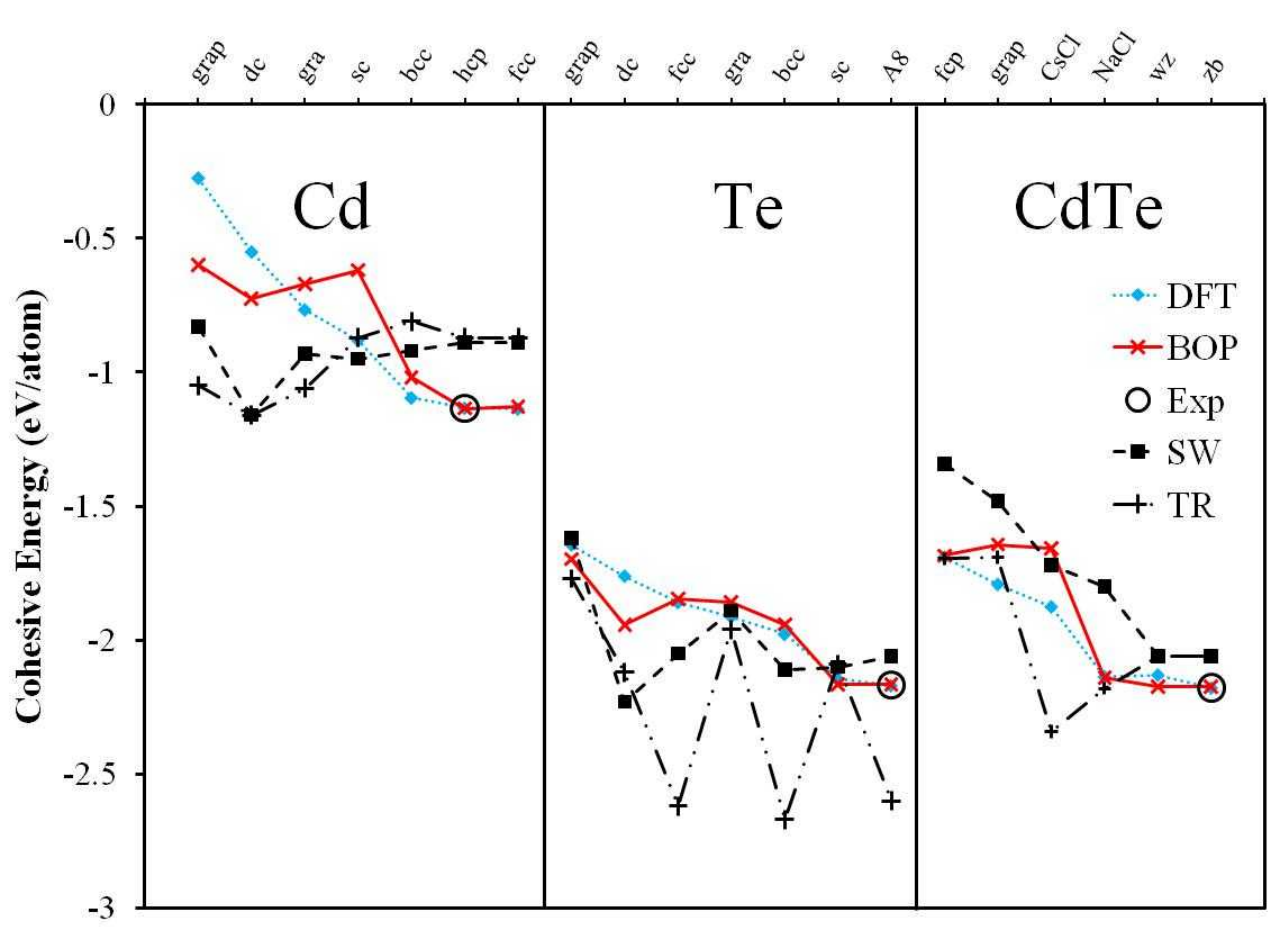
1. **Derived from quantum mechanics theory through systematic coarse-graining;**
2. **Separate treatment of σ and π bonding energies** (products of bond order* and bond integral#);
3. **The first two levels of the expanded Green function retained for the σ and π bond orders;**
4. **Up to four electron hops are considered, naturally incorporating the 3-member ring term in the σ bonding ($R_{3\sigma}$) and the dihedral angle ($\Delta\phi_{kk'}$) effect in the p bonding;**
5. **Valence effect is addressed.**
6. **Accuracy comparable to quantum mechanics and scale comparable to conventional molecular dynamics;**

* bond order: half the difference of electrons in the bond and anti-boding states.
\# bond integral: hopping probability of electrons from one orbital to another.

Ideal Atomistic Model for Defects

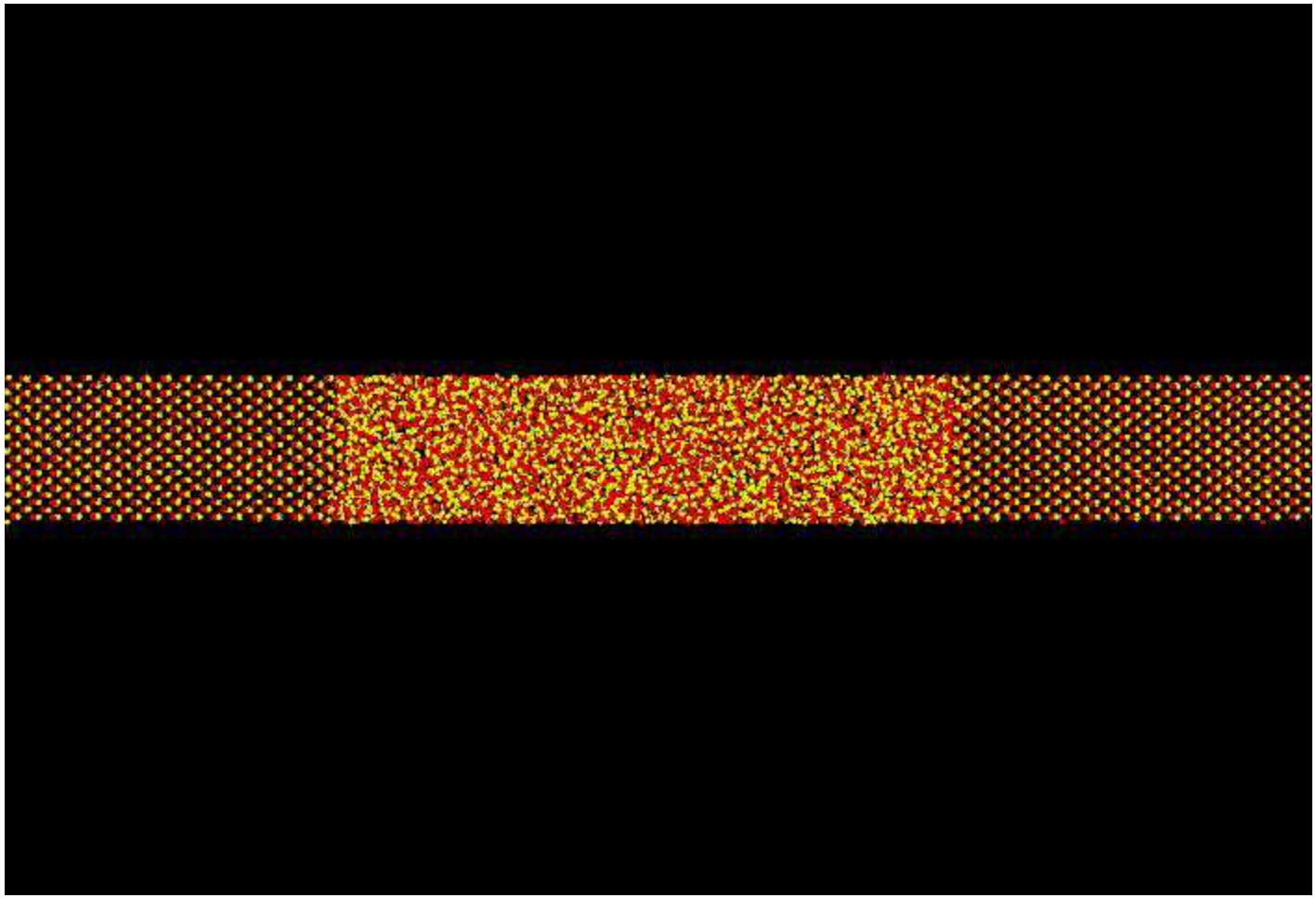
1. **We will develop a CdTe BOP and validate it against clusters** (dimer, trimer, square, tetrahedron, and chain for elements and compounds; Cd_2Te and $CdTe_2$ trimers for compounds)**, lattices** (diamond-cubic, simple-cubic, body-centered-cubic, face-centered-cubic, hexagonal-close-packed, graphite, planer-graphite, and A8 for elements; zinc-blende, wurtzite, NaCl, CsCl, binary-graphite, AuCu, CuPt, NiAs, CrB, AlSb, planar-binary-graphite, and face-centered-square for the stoichiometric compound CdTe; Ag_2O, CaF_2 for the non-stoichiometric compounds $CdTe_2$ or Cd_2Te)**, and defects** (vacancies, Cd@Te and Te@Cd antisites, Cd and Te interstitials at different locations)**;**
2. **We will also validate the BOP using melting and vapor deposition simulations.**

Cohesive energies vs. Phases

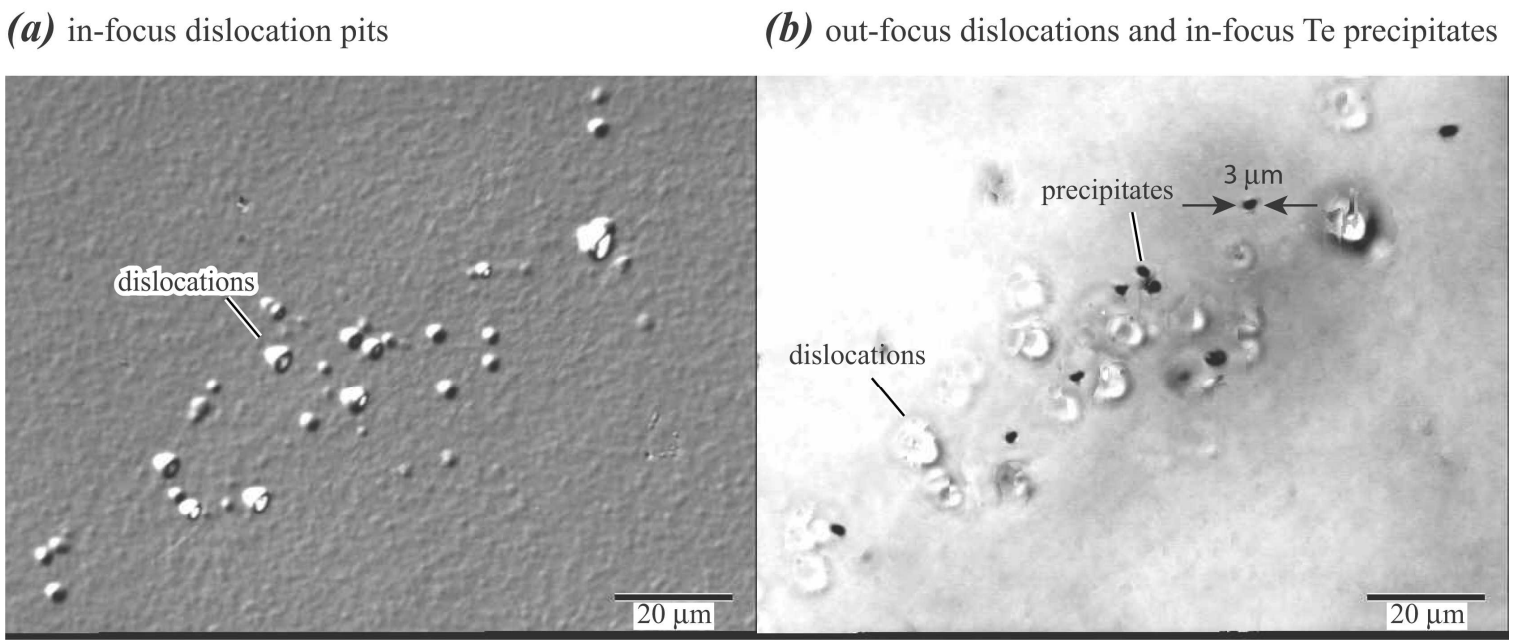


BOP greatly improved trend of energies of different Cd, Te, and CdTe phases!

CdTe Liquid Growth Simulation



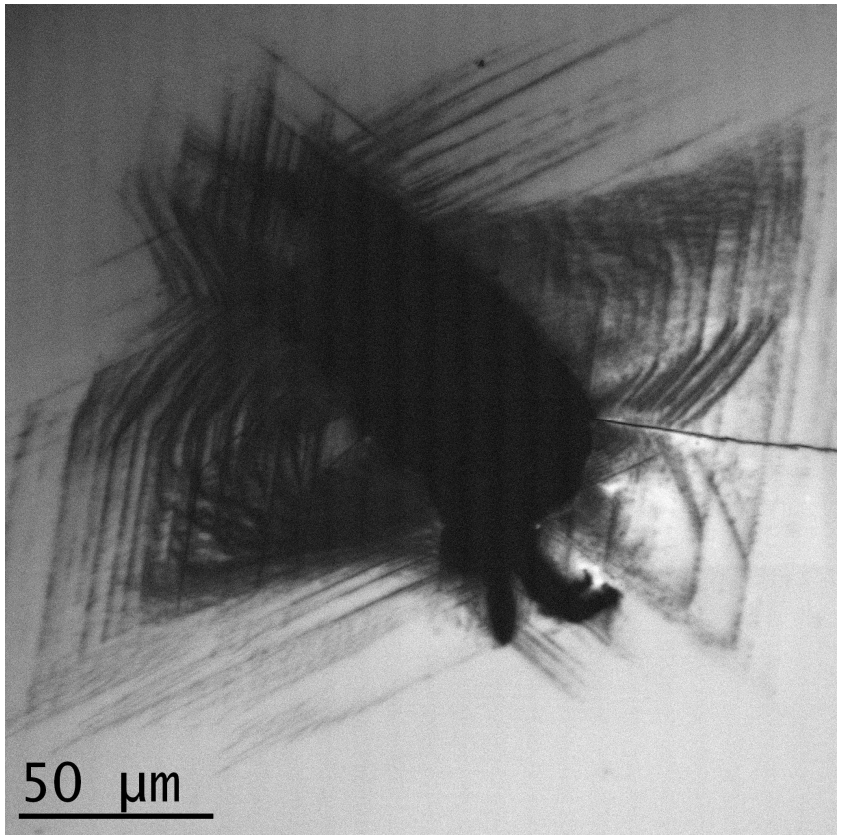
Nucleation Sites for Te Precipitates

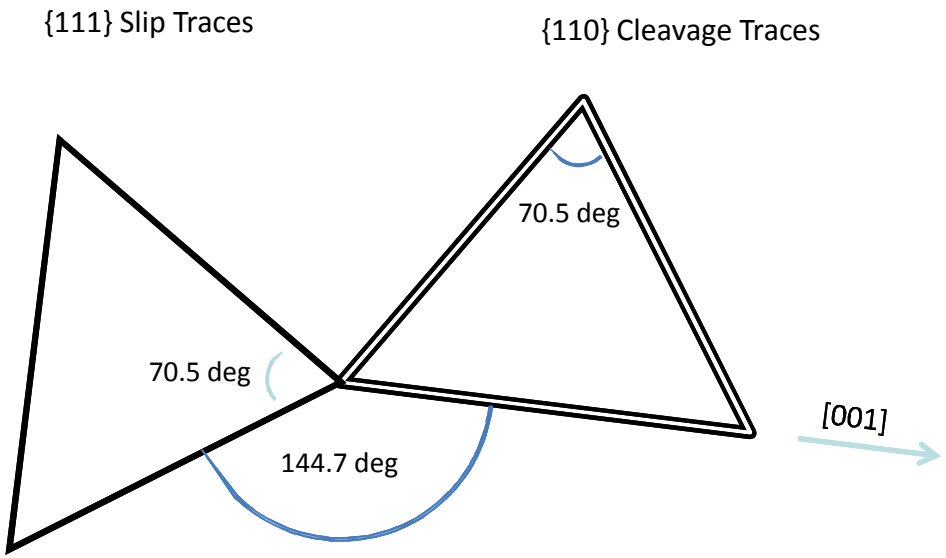


a) differential interference contrast showing dislocation etch pits on (111) surface. b) IR transmission of the same region showing 1 to 3 μm Te precipitates and out-of-focus pits

Dislocation Analysis

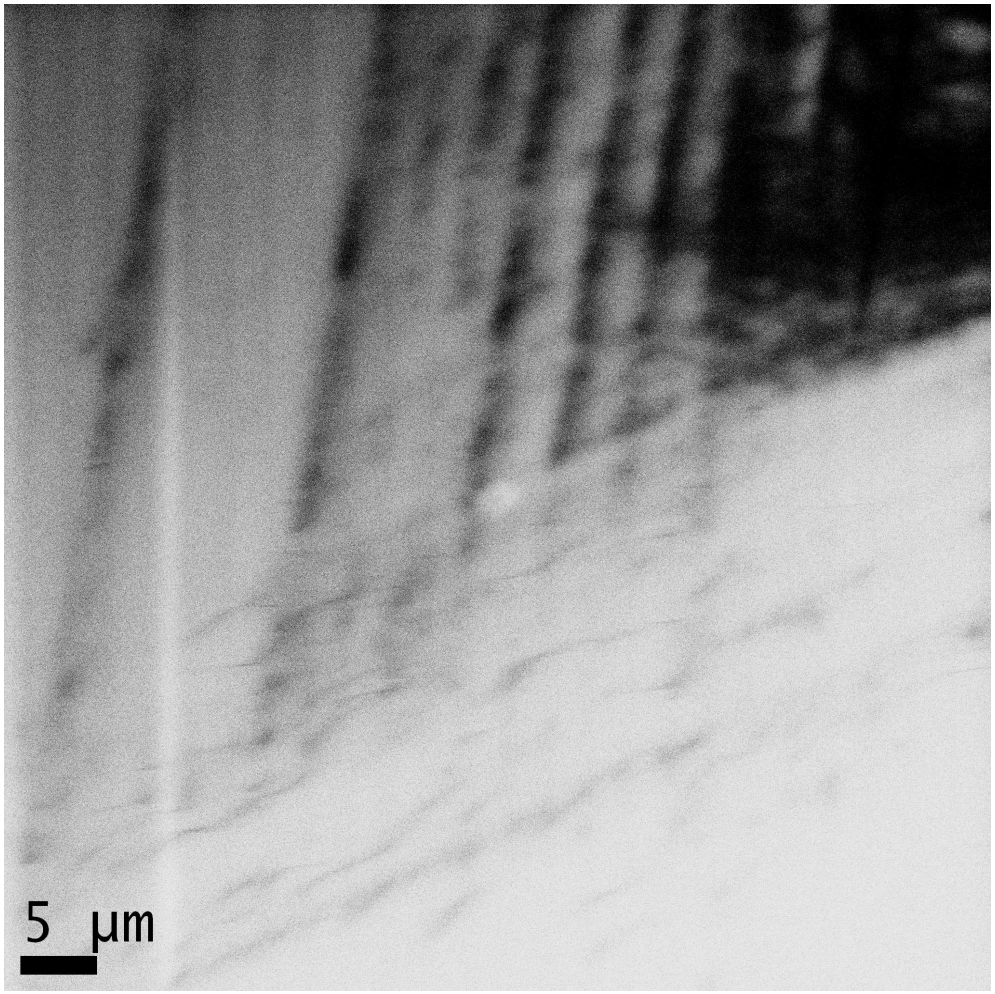
Assuming the image plane is a (110) cleavage plane





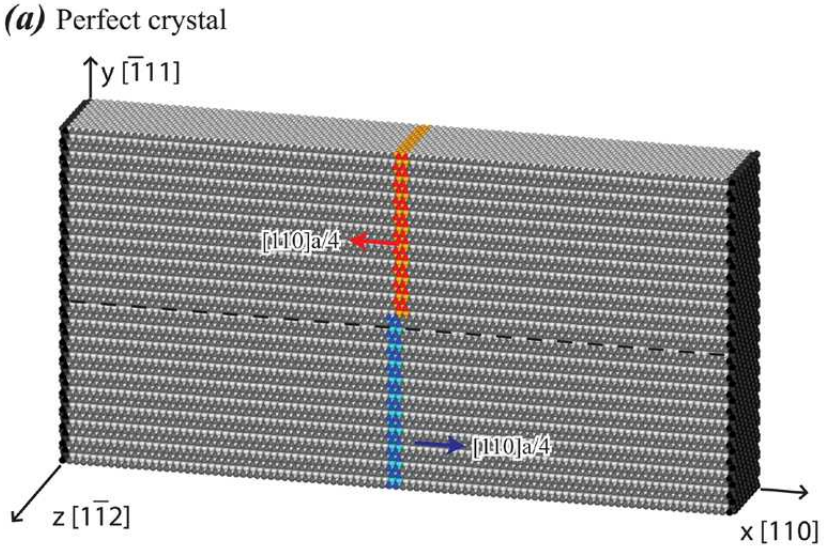
Dislocation slip planes can be determined by angles between plane traces on the (110) cleavage plane

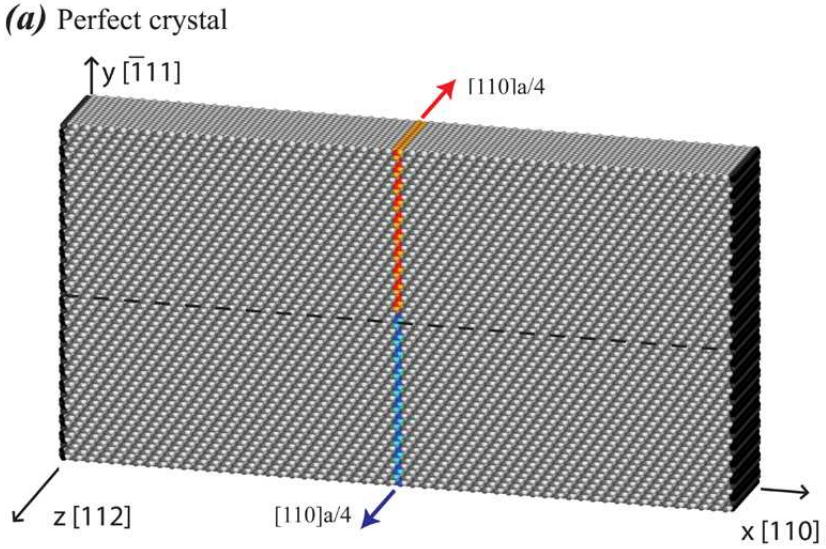
Zoom-In Scan

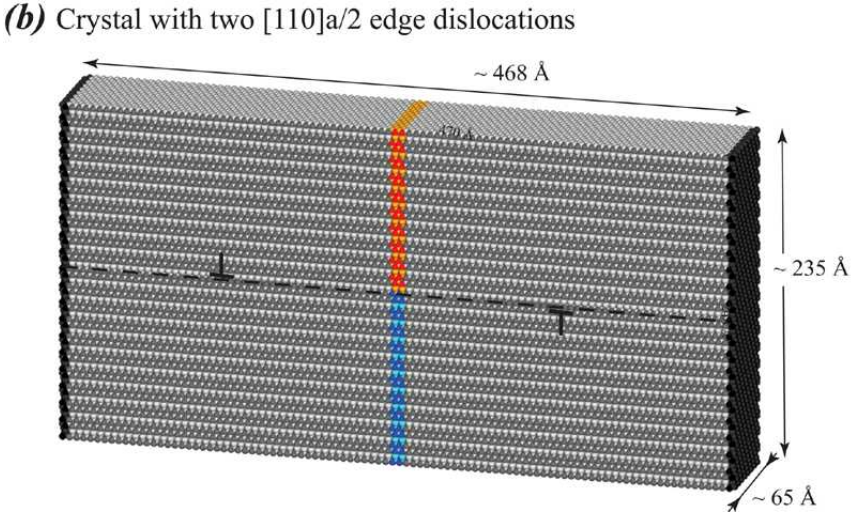


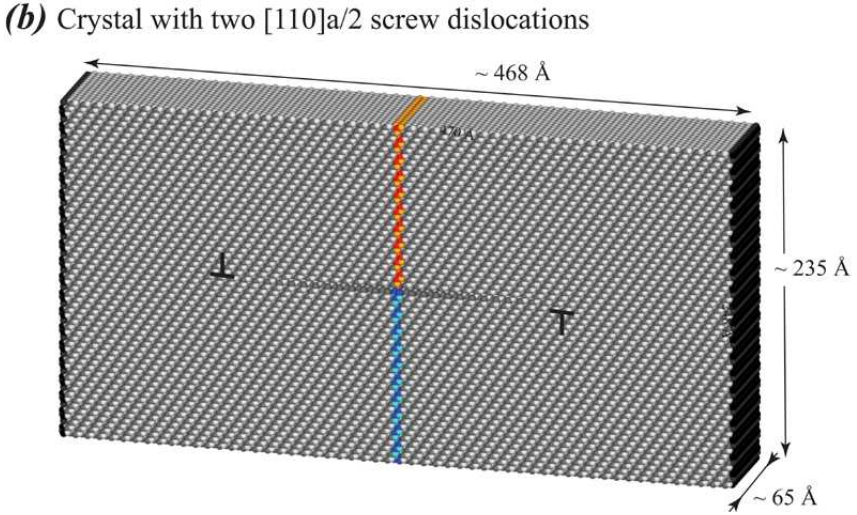
Dislocations become curvy after the zoom in scan.

Dislocation Models

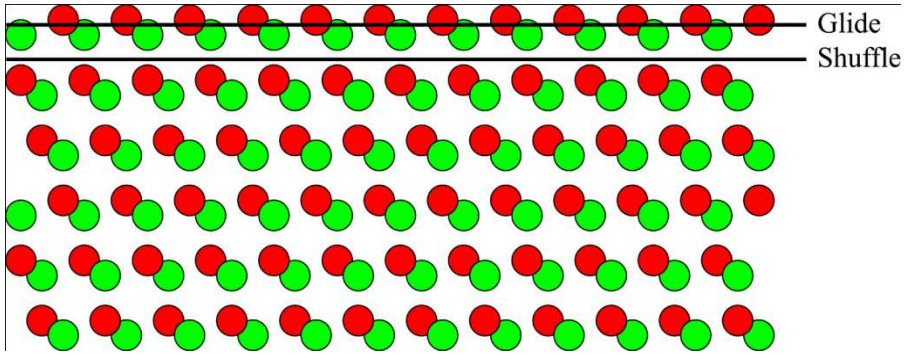








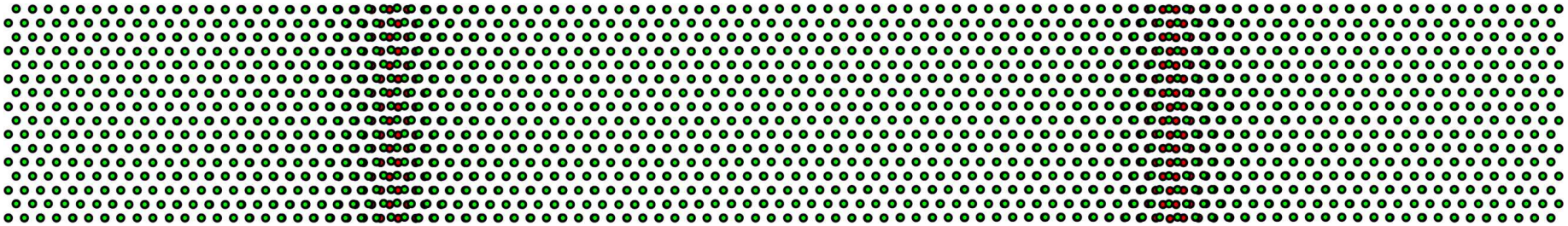
Dislocation Energies / Configurations in CdTe



Dislocation line energies (eV/Å)

edge		screw	
shuffle	glide	shuffle	glide
1.66	> 4.0	0.83	> 3.8

Edge shuffle dislocation configurations (top view of two planes)



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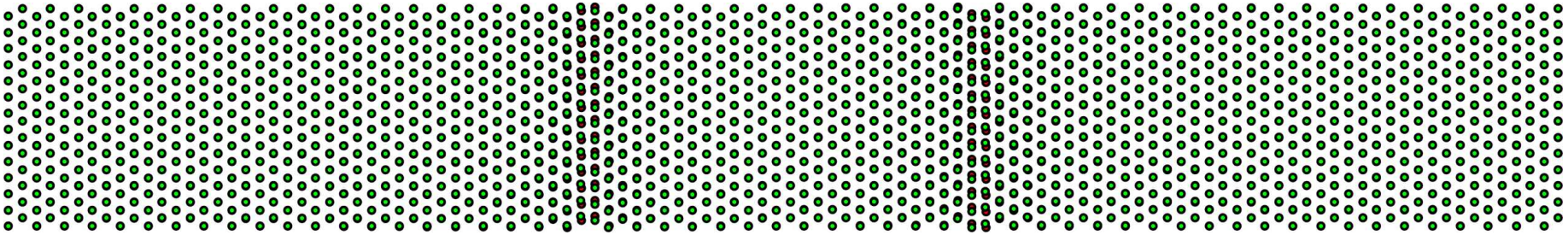
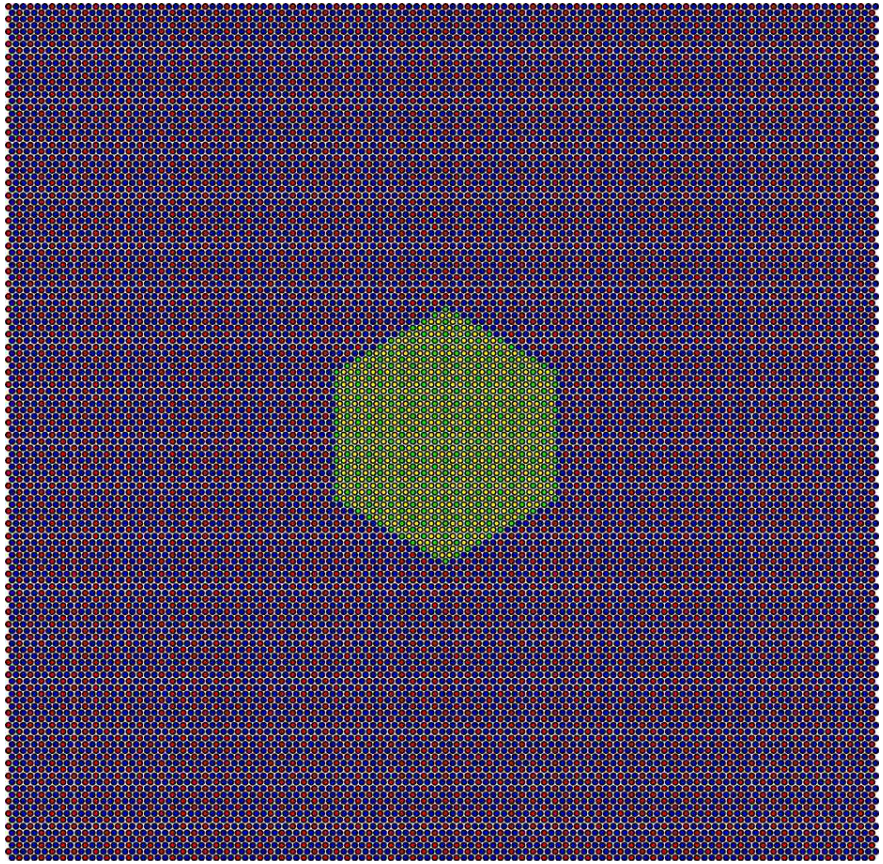
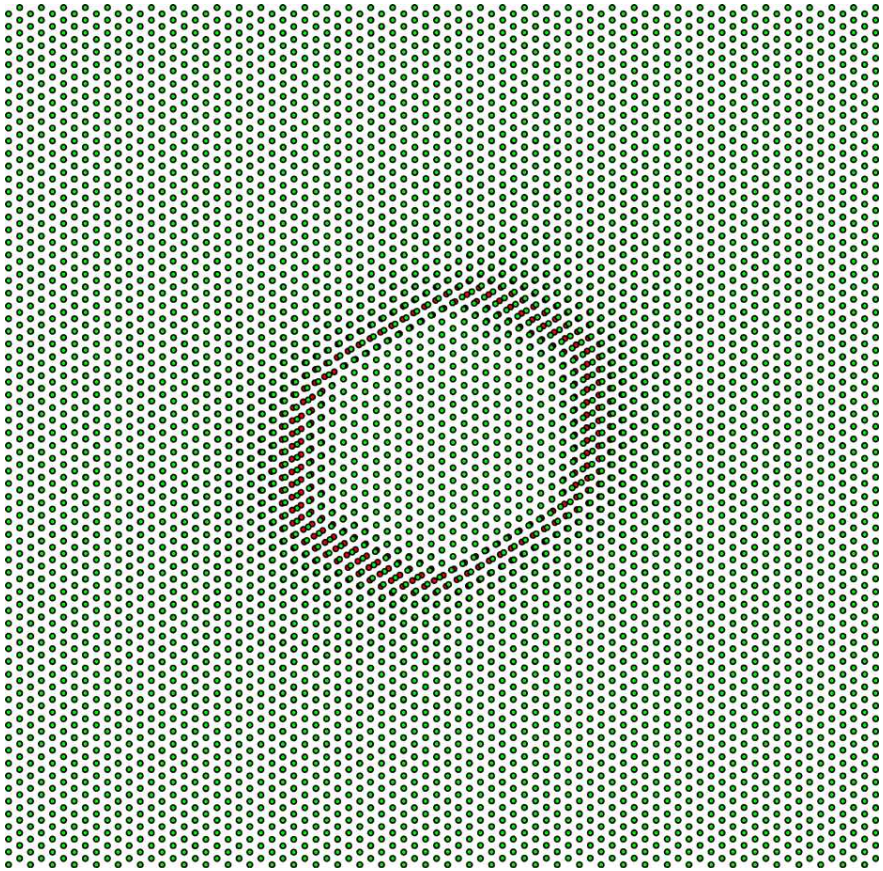


Dislocation Loop Configurations in CdTe

Loop creation



After relaxation



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Future Work for the Remainder of the Project

- **Theoretical Work**
  - Perform quantum-mechanical calculations on lattice constants, cohesive energies, elastic constants, defect energies, and surfaces reconstructions of a variety Zn, ZnTe, CdZn, and CdZnTe phases.
  - Develop BOPs for Zn, ZnTe, CdTe, and CdZnTe systems.
  - Improve algorithm of BOPs calculations in LAMMPS.
  - Perform large scale simulation on various possible dislocation configurations, characteristics, stress field, and interaction with other defects (in particular on precipitate formation).
  - Perform large scale simulations on small scale defect evolution under thermomechanical conditions
- **Experiments**
  - In conjunction with crystal growth work at PNNL, perform SEM CL analysis of dislocations in as-grown CZT samples.
  - Use the input from theoretical work to design improved growth of CZT crystals.

Acknowledgement

This work is supported by the NNSA/DOE Office of Nonproliferation Research and Development, Proliferation Detection Program, Advanced Materials Portfolio, and The National Institute for Nano-Engineering (NINE). Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.