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Improved CZT for Gamma Detection

Ralph B. James

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Brookhaven National Laboratory
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Radiation Detector and Nonproliferation Research and Development

U.S. Department of Energy
National Nuclear Security Administration
Defense Nuclear Nonproliferation Research and Development Program

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National Nuclear Security Administration
Defense Nuclear Nonproliferation

Defense Nuclear Nonproliferation Research & Development

Nuclear Weapons and Material Security Portfolio Review

WMS 2015

Improved CZT for Gamma Detection

Ralph B. James

Brookhaven National Laboratory

March 19, 2015

UNLIMITED DISTRIBUTION

Name/Org: **Peter Vanier** Date: **March 16, 2015**

Introduction

Spectrum of opinions about CZT detectors (very conflicting)

Will never work, too inefficient, …

CZT DETECTORS

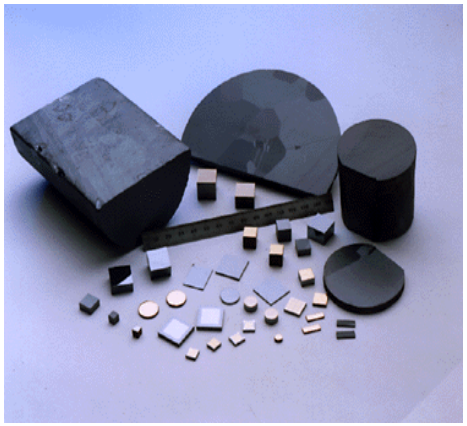
Everything is done, useful in the field, mission accomplished

Problems from a User Perspective

- **Energy resolution**: Most nonproliferation applications desire excellent energy resolution, and a 0.5% FWHM would be a huge improvement over scintillators
- **Efficiency**: Ability to make measurements quickly is an imperative for most scenarios
- **Cost**: Concerns over procurement costs will probably grow – not shrink

Advantages of Wide Band-gap Semiconductors

- Device fabrication utilizes photolithographic patterning for high spectroscopic and positional information
- Sub-millimeter spatial resolution can be achieved for imaging devices and directional detectors
- Ideally suitable for high-speed fully digital signal processing – required for extracting all possible information from every photon



Photon Processing vs. Photon Counting Detectors

Extract maximum possible information for every photon (interaction points, electron cloud distribution, timing, E field, and trapping to reconstruct relevant detector attributes for users)

- Choose a configuration (electrodes or detectors) to generate the parameters needed to describe every photon interaction event
- Develop an accurate likelihood model based on all readout channels for each photon-processing attribute, accounting for noise (super list-mode)
- Consider if calibration methods are needed to determine unknown system characteristics that influence the likelihood model (e.g., quantum efficiency, gain, Fano factor, trapping parameters)
- Collect all detector-relevant values that correspond to each photon event with maximum precision
- Use data and likelihood models for real-time calculation of estimates for all relevant photon-detector attributes
- Use ML estimates for detailed reconstruction of charge transport, energy spectra, photon directions, images, timing and other parameters

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Project Goal and Objectives

National Nuclear Security Administration
Defense Nuclear Nonproliferation

Project Goal: Advance state of the art for spectroscopic and imaging detectors for nonproliferation uses

Objectives

1) **Develop a fundamental understanding of defects limiting the performance of large-volume CZT detectors and fix them**
2) **Increase the detector thickness by 2x, the electron mobility-lifetime product by 2-3x, and reduce the cost of detector-grade crystals by 2-5x**
3) **Demonstrate principles, hardware and software to extract maximum possible information from every photon and use concept to increase the acceptance of usable materials**
4) **Provide rapid feedback to collaborators and vendors, and translate our technologies into commercial practice**
5) **Initiate a plan for continuing R&D capabilities after sunset**

Peer-reviewed Publications

Overview: Dissemination and Project Themes

- **Publications (since 2014 WMS review meeting): 26**

Themes: *Spectroscopic and Imaging Response (5), Defects and Trapping (4), Impurities/Purification/Dopants (5), Annealing (6), Contacts and Surface Treatments (6)*

- **Presentations (since 2014 WMS review meeting): 37 (4 invited talks)**

- **Patent Applications (since 2014 WMS review meeting): 2**

- **Awards: 2014 R&D 100 Award, 2014 RTSD Scientist Award, 2015 Award of Scientific Excellence in Solid-State Physics, and 2014 Long Island Technology Hall of Fame Inductee**

Collaborating Institutions

University Collaborators: Alabama A&M Univ., Stony Brook Univ., Univ. of Michigan, Univ. of Illinois, Univ. of Tennessee, Fisk Univ., Rensselaer Polytechnic Institute, Penn State Univ., Univ. of Madrid, Univ. of Alabama, Univ. of Texas at El Paso, SUNY Binghamton, Korea Univ., Charles Univ., Univ. of Freiburg, Univ. of Bologna, IMEM and Chernivtsi National Univ.

Industrial Collaborators: Redlen, Kromek, FLIR, KIPT, SureScan, Institute of Semiconductor Crystals

Student Training in Current Funding Period

Name	Institution	Name	Institution
Matthew Petryk	SUNY Binghamton	Václav Dedic	Charles Univ.
Nicholas Kivi	Univ. of Tennessee	Samuel Uba	Alabama A&M Univ.
Dominick Raimondi	Alabama A&M Univ.	Diedre Hodges	Univ. of Texas at El Paso
Megan Wart	Rensselaer Polytechnic Institute	Stephen Babalola	Alabama A&M Univ.
Tsige Zergaw	Alabama A&M Univ.	Luis Ocampo	Penn State Univ.

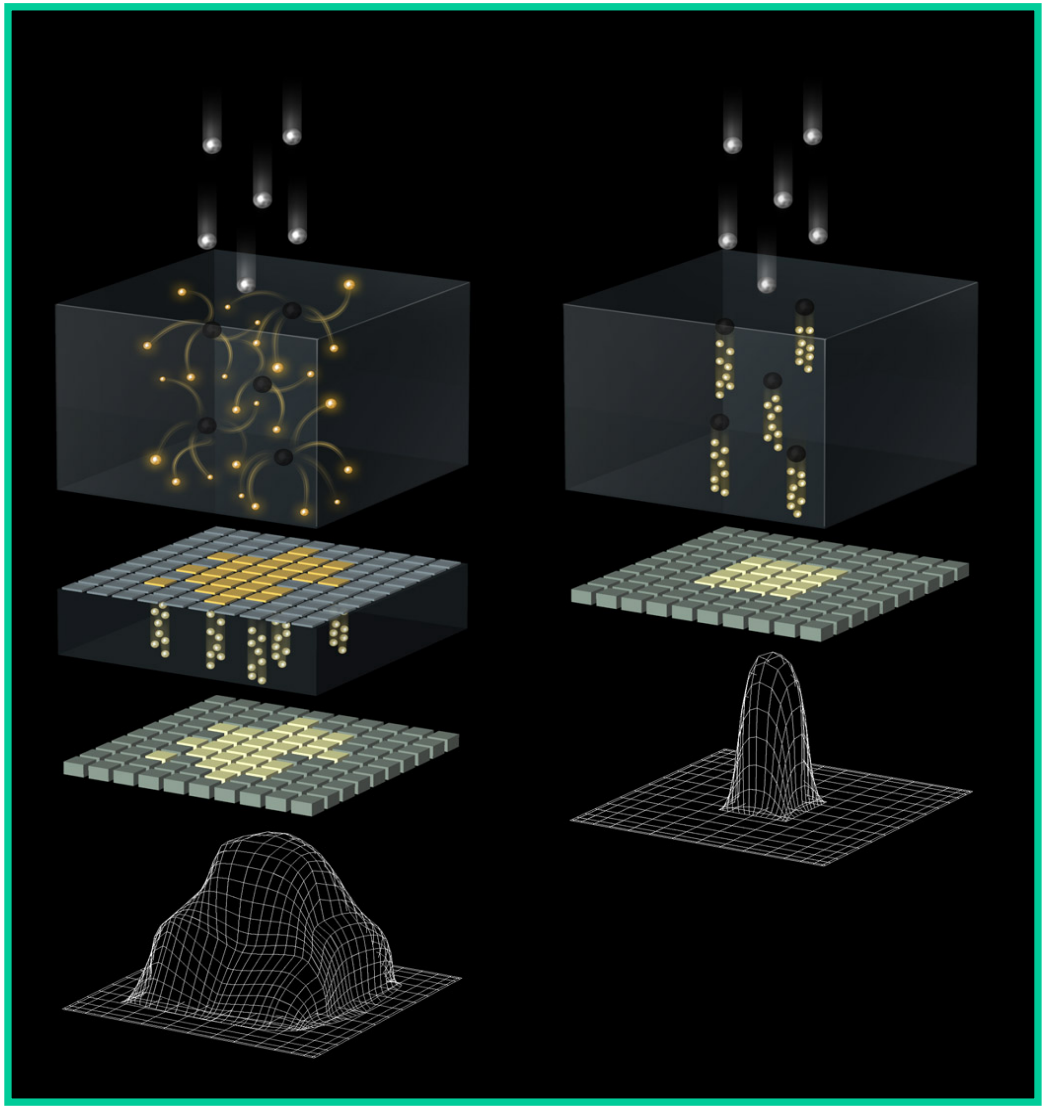
Interaction points can be precisely determined in 3D for CZT detectors

High energy- and spatial-resolution are huge advantages for semiconductors

Scintillators – *Indirect* Conversion of Gammas to Electrons

- Spread in electron signals reduces spatial resolution

- Reduced signal degrades energy resolution



Semiconductors – *Direct* Conversion of Gammas to Electrons

- Compactness in electron signals enhances spatial resolution

- Enhanced signal increases energy resolution

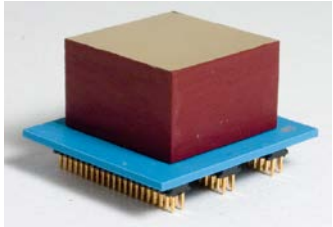
Detectors have limited capability to correct response non-uniformities and rely on high-quality material

The most competitive detector designs are:

CPG
(the first successful commercial CZT device)

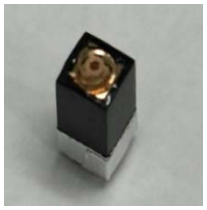
First demonstrated P. Luke

20x20x15 mm^3 3D pixel detector
(the most advanced detector technology today)

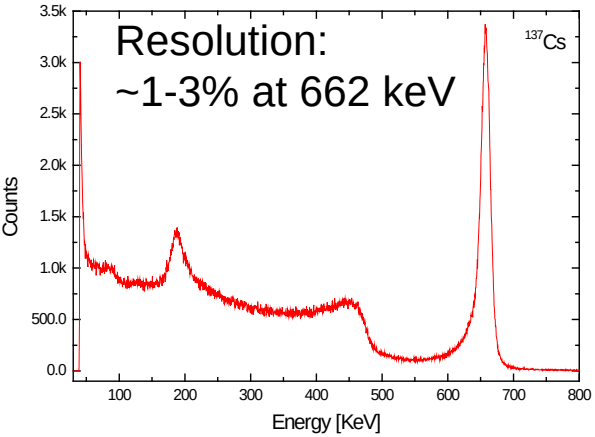


Developed by Z. He et al.

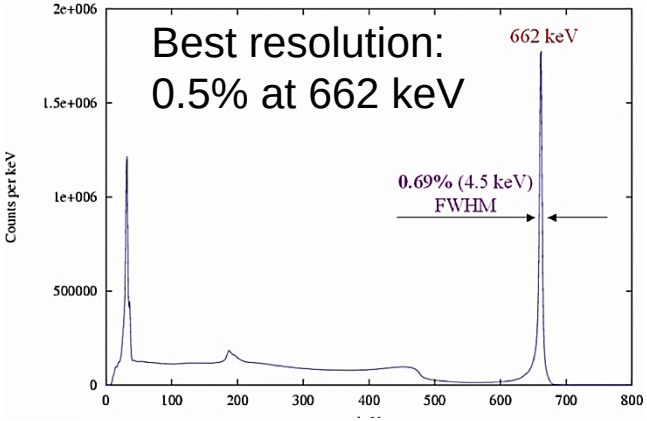
6x6x15 mm^3 virtual Frisch-grid device (new device offering cost reduction



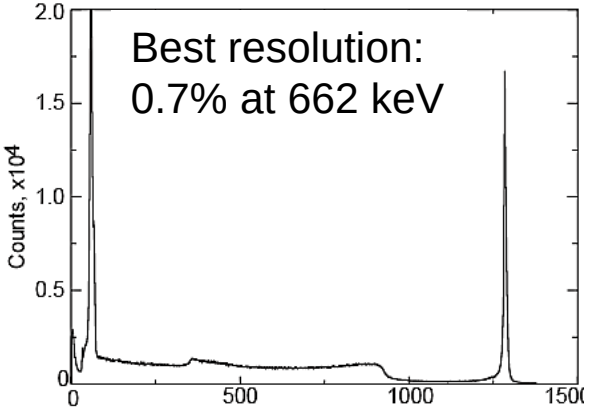
First demonstrated by McGregor and Montémont, and improved by BNL



Drift-time correction only



3D correction but with a coarse spatial resolution



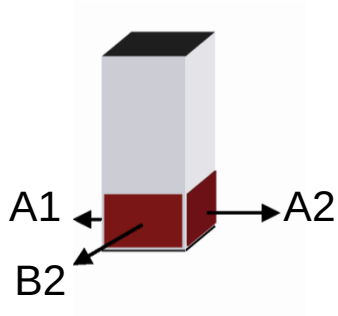
Drift-time correction but with smaller areas

We need detectors with precise knowledge of interaction points!

Improvement in Energy Resolution Observed

Prior design

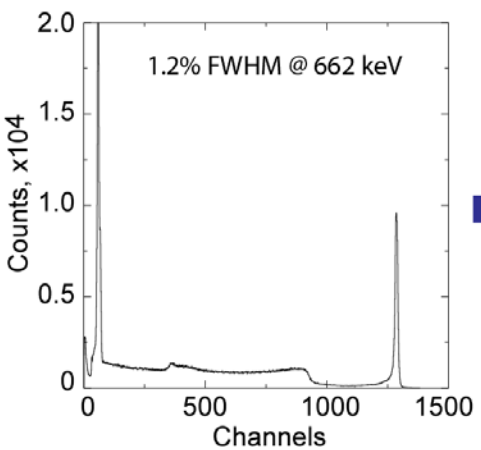
New Design: Position-sensitive Virtual Frisch-grid detector



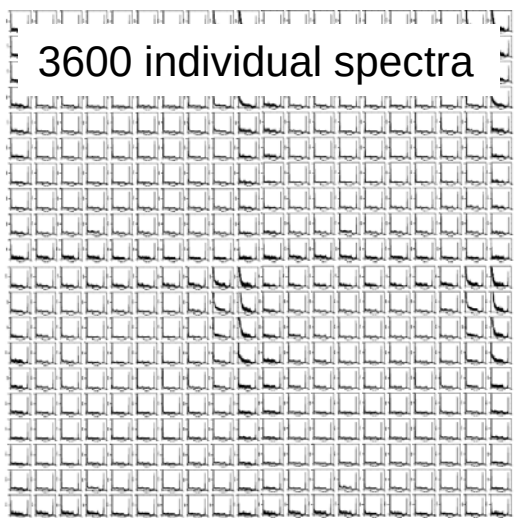
Measured detector variables:
Drift-time (T)
Amplitudes vs. time from four sensing strips (A1, A2, B1 and B2)

Real-time conversion to space with 10^{-6} cm^3 precision:
X~A1/(A1+A2), Y~B1/(B1+B2), and Z~T

Spectral Performance Without Electronic Segmentation

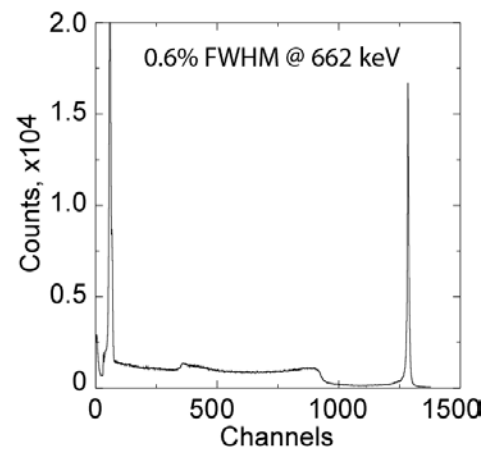


Electronically segment the anode into small sub-pixels



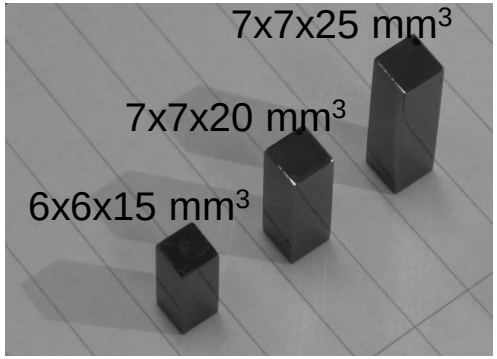
15-mm-thick detector

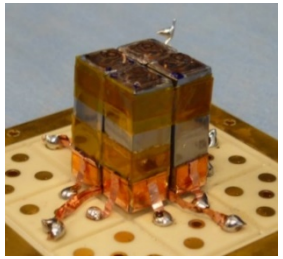
Spectral Performance After Electronic Segmentation



New Design Allows for Correction of Material Defects!

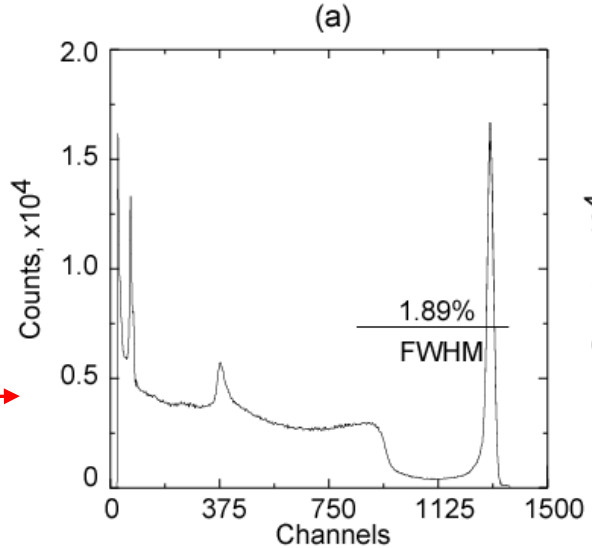
Improvement in Efficiency



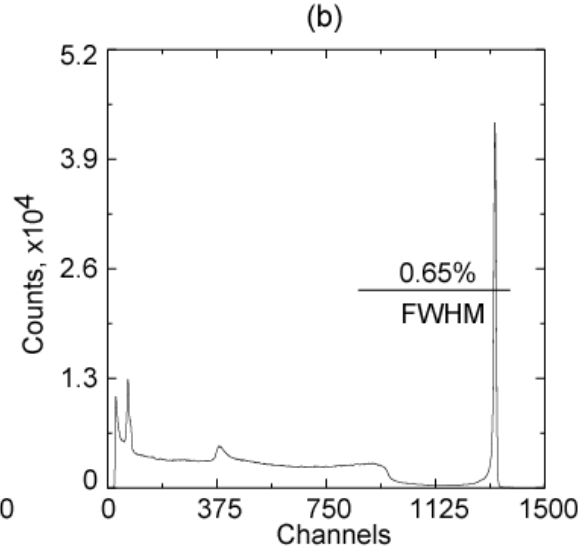


20-mm thick →

Only depth-interaction points

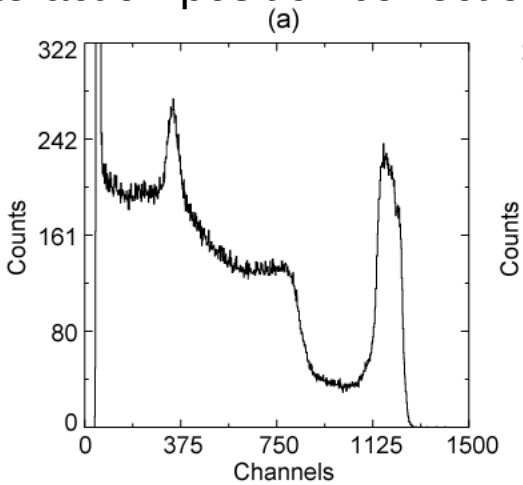


With X-Y-Z information

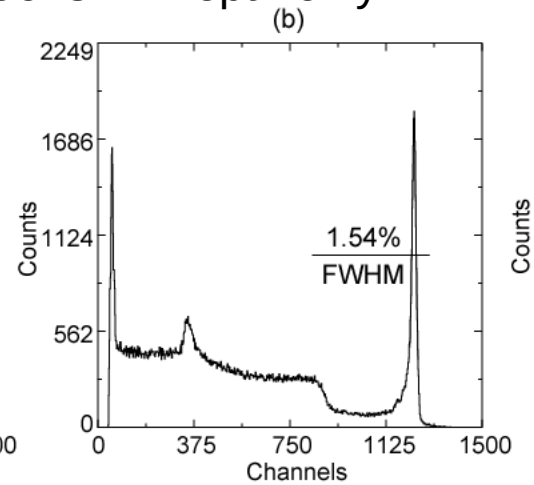


25-mm thick →

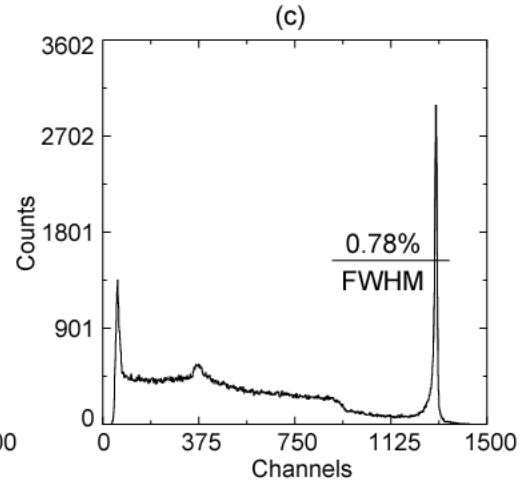
No interaction-position corrections



Depth only

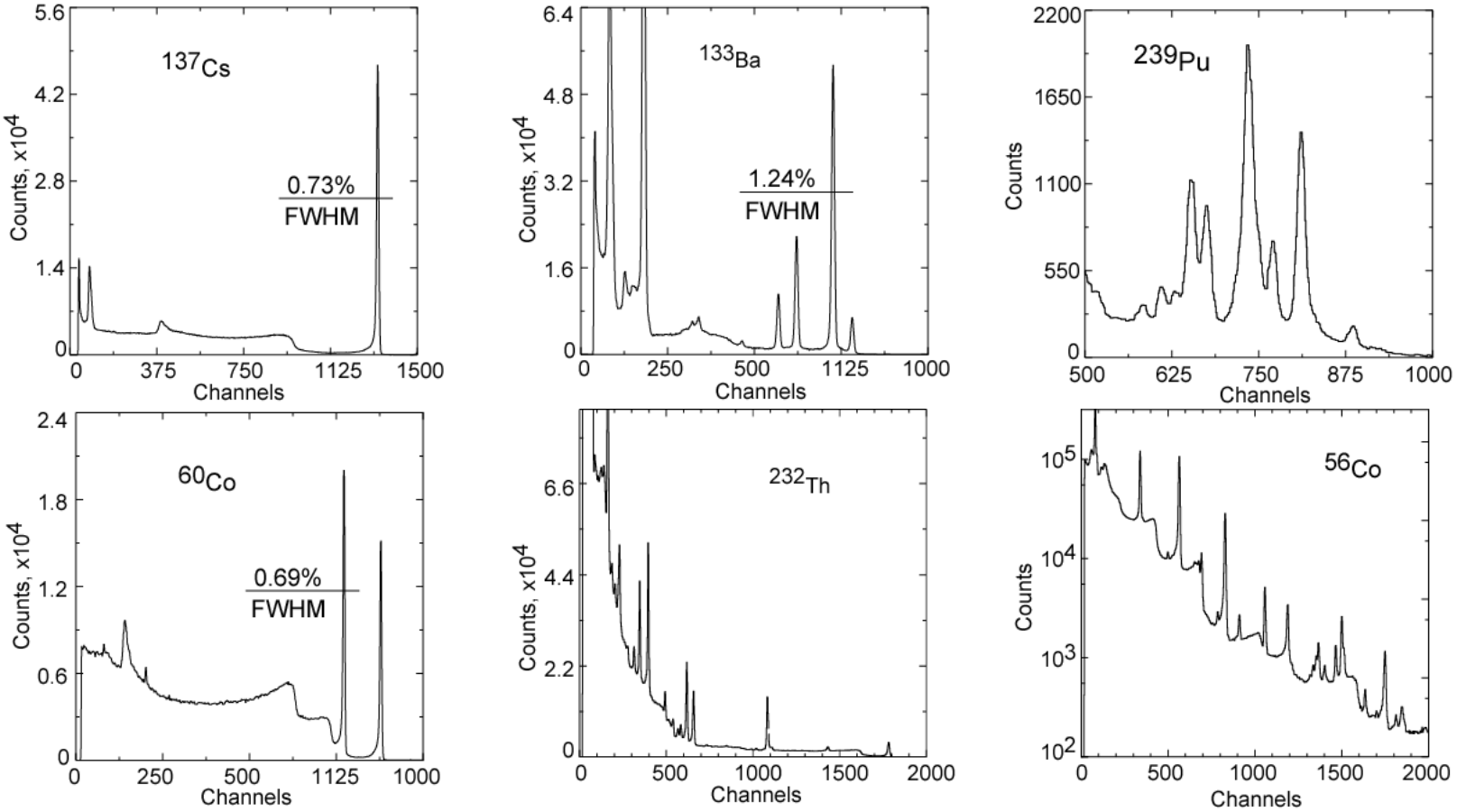


X-Y-Z 3D corrections



Detector Cost

Increased efficiency allows detection of energetic gamma rays



All CZT crystals used in this study had been rejected by vendor

Theme: Defects and Trapping (Relationships between Resistivity and Carrier Trapping)

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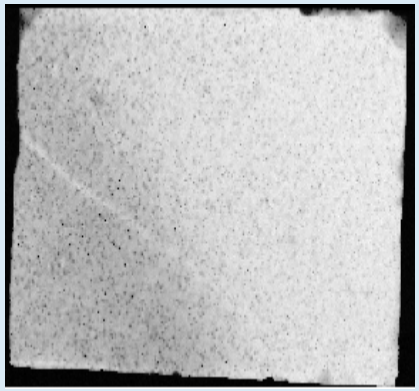
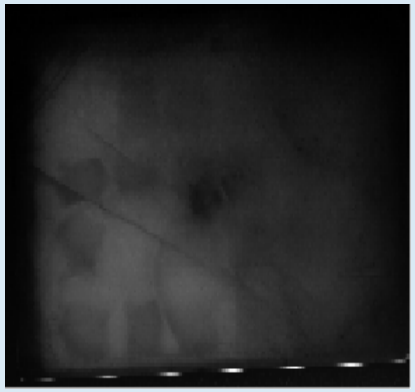
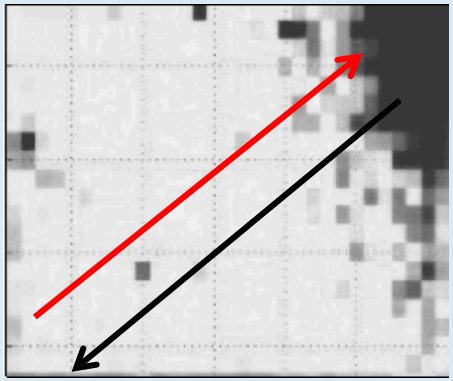
Sample A



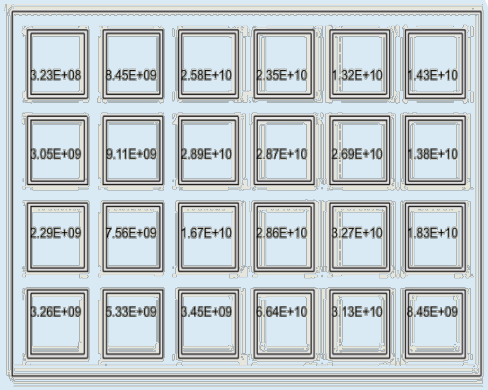
Electron collection map



Hole collection map

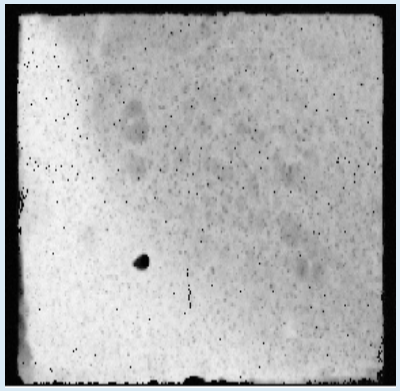


Counting maps at high flux

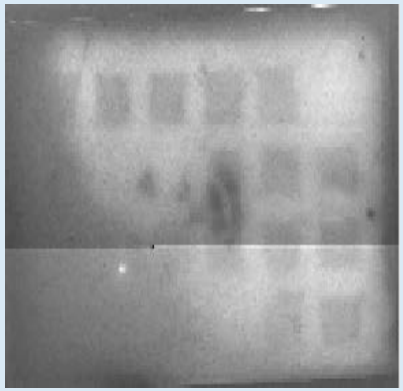


Resistivity Map

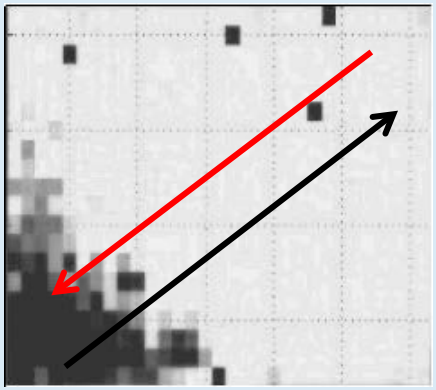
Sample B



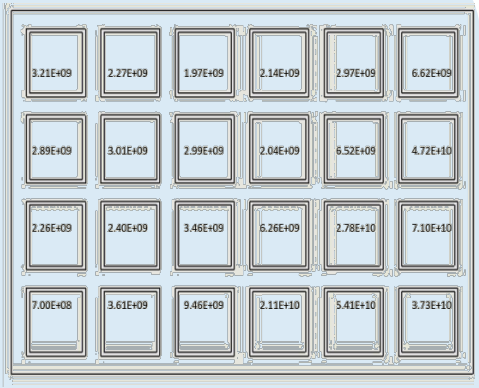
Electron collection map



Hole collection map



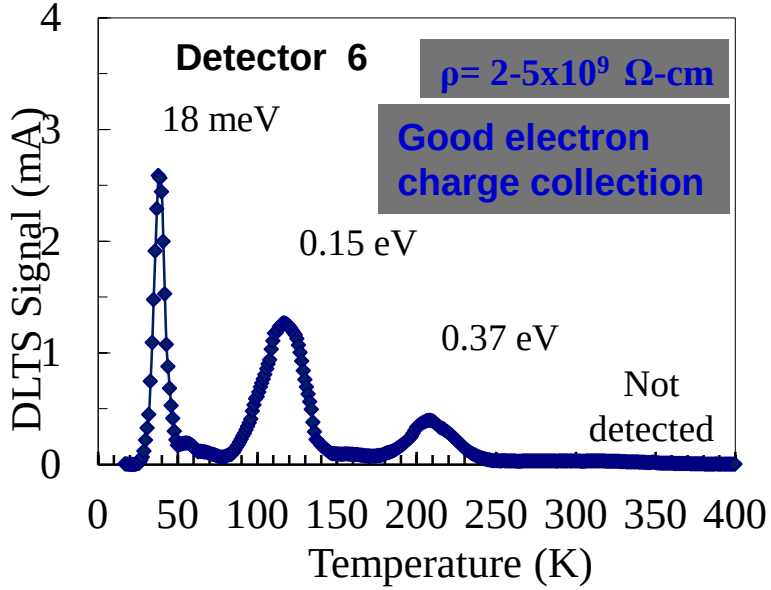
Counting maps at high flux

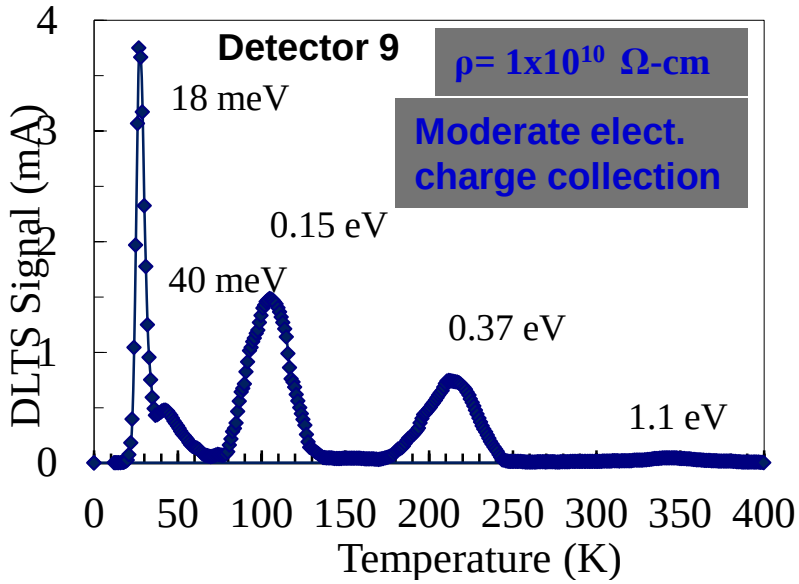


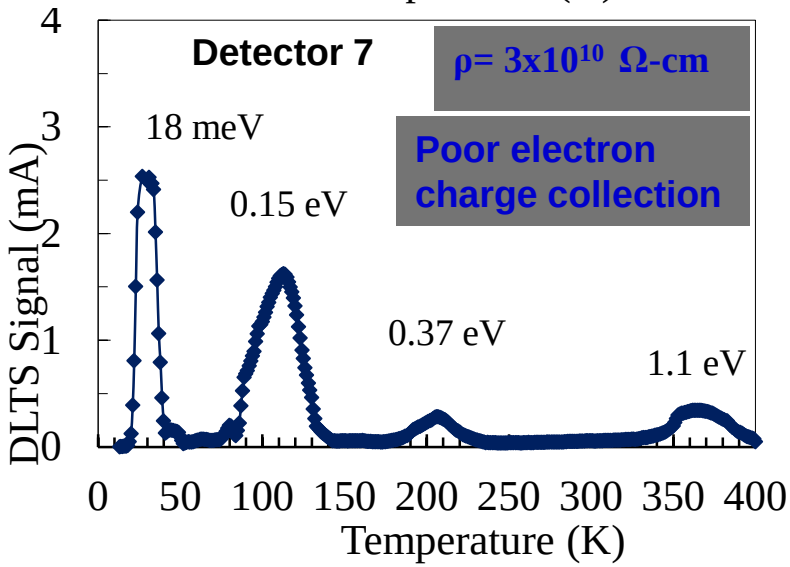
Resistivity Map

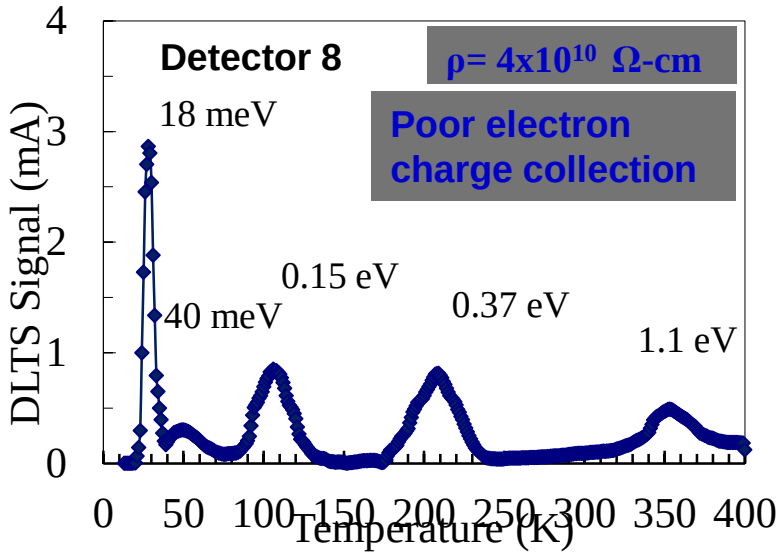
The performances of the detectors correlate with the resistivity maps

Resistivity and Trapping are Related to 1.1-eV Deep Defect



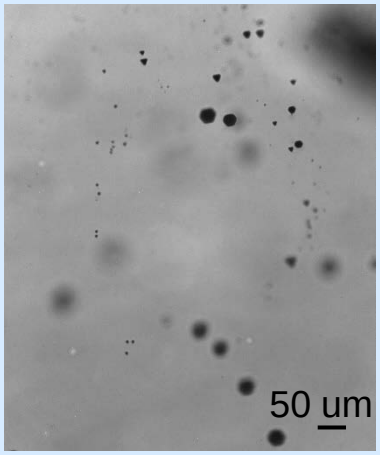




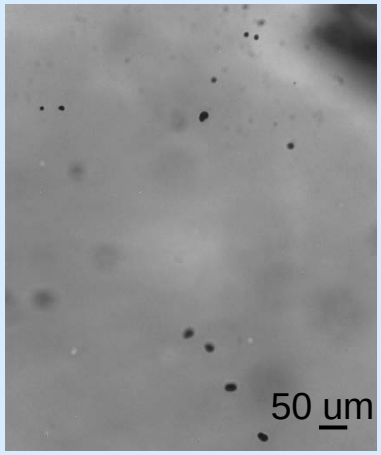


Theme: Post-growth Annealing

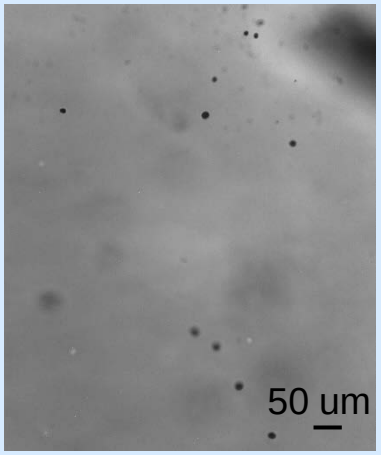
Track the reduction efficiency of Te inclusions during thermal annealing



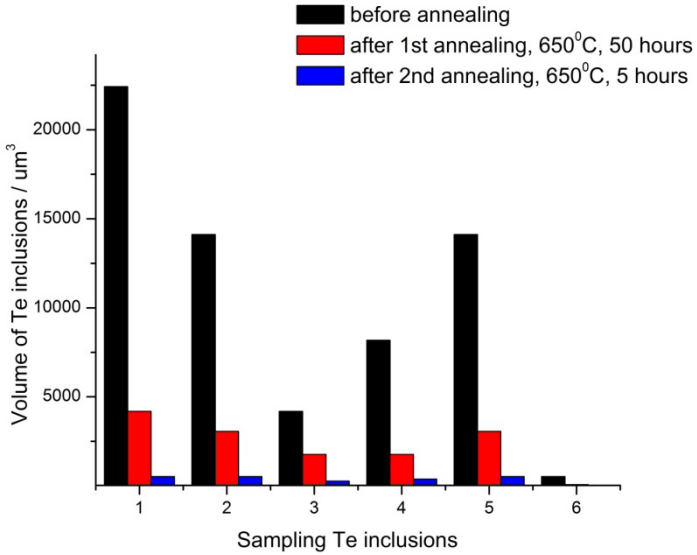
Before annealing



1st annealing
650 ℃, 50 hours



2nd annealing
650 ℃, 5 hours



Average reduction in volume of Te inclusions is ~ 96%

Note: All Te inclusions of < 5 μm disappear or become smaller than 1 μm (infrared microscope resolution) after annealing

Theme: Dopants, Purification and Impurities

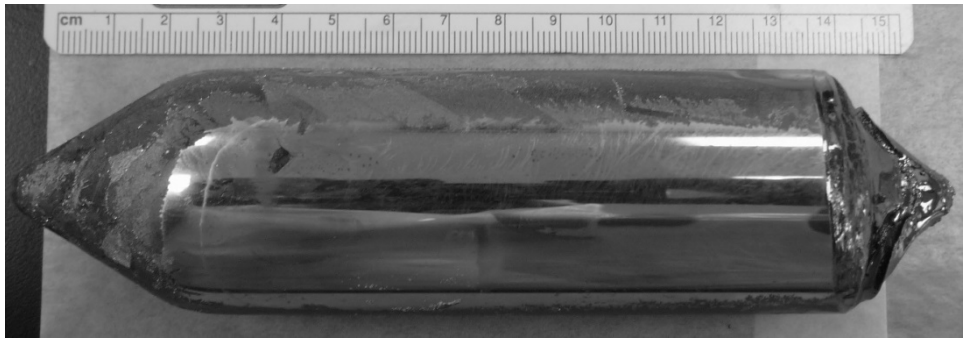
Example: Intentional introduction of Se to CZT matrix

- 6N-purity CdZnTe and CdSe were taken for the initial charge
- The compound was synthesized and purified in-house by sublimation
- Indium was also added as a dopant to increase the electrical resistivity

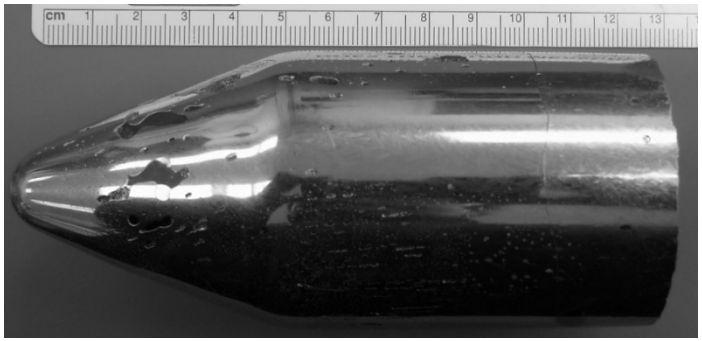
Crystals were grown by the vertical Bridgman technique and THM with nominal Se concentrations of a few % or less.

i) THM growth: Growth temperature ~ 850 ^{0}C and growth rate ~3 mm/day

ii) Vertical Bridgman growth runs were carried out at a temperature slightly higher than the melting point (1092 ^{0}C) and at a growth rate of ~3 cm/day



Bridgman-grown ingot (40-mm diameter)



THM-grown ingot (52-mm diameter)

Impurity analyses by GDMS

Comparison for crystals grown by Bridgman and THM

Elements	Conc. (ppb atomic) THM-grown crystal	Conc. (ppb atomic) Bridgman-grown crystal
Na	10	210
Mg	49	49
Al	39	44
Si	970	2600
S	150	290
Fe Ni	85 100	63 12
Zn	160	180
Pb	28	34

The purity is slightly higher for THM-grown sample or comparable to BM. Ni and Fe were found to be higher in THM-grown ingots; these impurities might have been introduced from the Te solvent.

Less Defects Affecting Charge Collection by Se Doping

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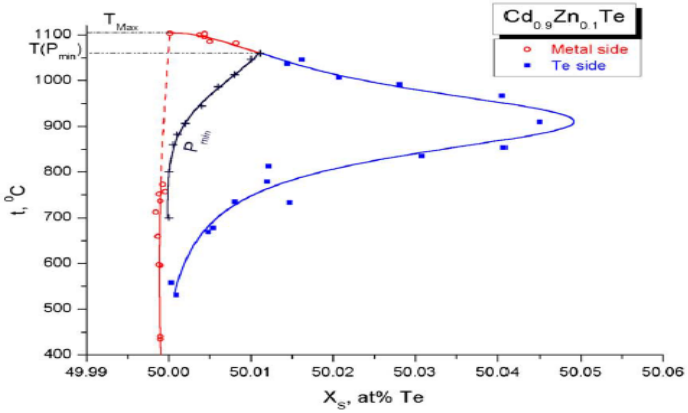
1. Secondary phases such as Te inclusions/precipitates

Typical CZT	Average Conc.	~ 2-7x10^5 cm^{-3}
5-10% Se	Average Conc.	~ 3-7x10^4 cm^{-3}

The lower distribution (size and concentration) of inclusions should have no adverse effect on detector performance.

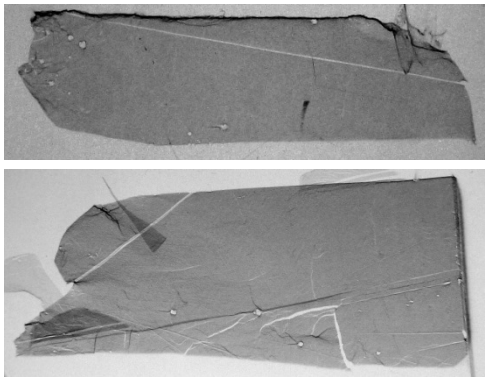


T-X projection of $Cd_{0.9}Zn_{0.1}Te$ solidus

2. Sub-grain boundaries and their networks

Standard CZT: High concentration of sub-grain boundaries and their networks

5% Se: Very few sub-grain boundaries, and free from sub-grain boundary networks



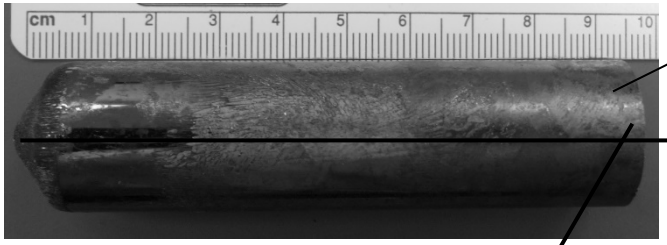
X-ray diffraction topography

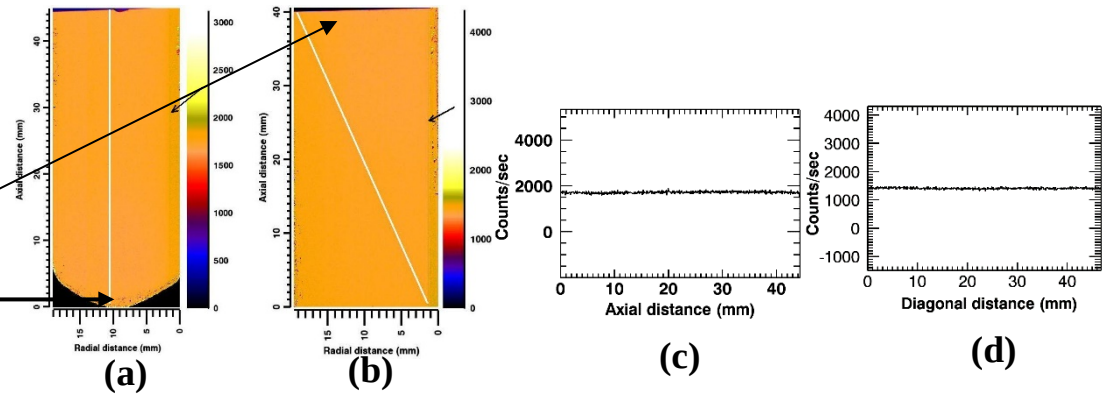
Very high compositional uniformity, absence of sub-grain boundary network, and one order of magnitude lower concentration of Te inclusions are very encouraging for the future of CZTS detectors.

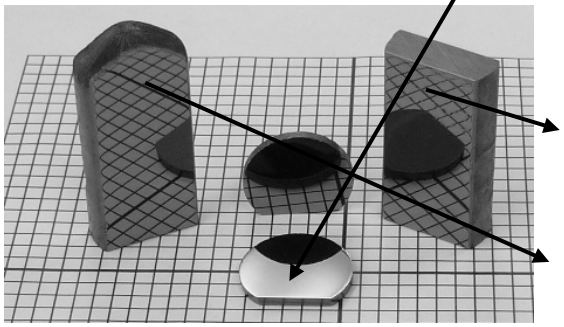
Compositional homogeneity

Synchrotron measurements show that the Se concentration is highly uniform along the ingot, and the segregation coefficient of Se is ~1.00.

Ingot grown by vertical Bridgman







Upper part of ingot

Lower part of ingot

Wafers cut across the growth direction from the top of the ingot and along the length

High resolution X-ray fluorescence elemental map of Se across the whole wafer cut along the length of the ingot. The line profile of Se K_α c) of the lower part along the length, and d) of the upper part of the ingot along the diagonal, as measured along the white lines in a) and b), respectively.

What do we see as the next innovations for CZT Base Program?

- Extend thickness of CZT detectors to 30 mm (this year) and even larger in FY16
- More complete understanding and utilization of photon-processing detectors with different electrode and sensing-pad configurations, appropriate for large pixels and varied detector form-factors
- Demonstrate high-spatial-resolution imaging using precise positional information by photon processing ($< 10^{-6}$ cm^3)
- Transfer technology for new detector designs to lower the costs by relaxing the acceptance criteria for CZT crystals, since there will no longer be a need to find "perfect" crystals
- Complete first 3D mapping of internal electric field in detectors and its relationship to extended defects and charge-transport nonuniformities
- Bring the knowledge gained on Se as a dopant to full fruition (CZTS) to advance state of the art for room-temperature semiconductor detectors
- Continue implementation of our transition plan for dedicated synchrotron beam-lines at ALS and APS facilities, and then bring facilities back to BNL once the NSLS II is ready (Q4 of FY17).

Special Thanks to DNN R&D WMS Team

Questions?