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SNM Movement Detection/Radiation Sensors and Advanced Materials Portfolio Review
Development of Improved CZT for Gamma-Ray Spectrometers and Imaging Arrays

Ralph B. James

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SNM Movement Detection / Radiation Sensors and Advanced Materials Portfolio Review

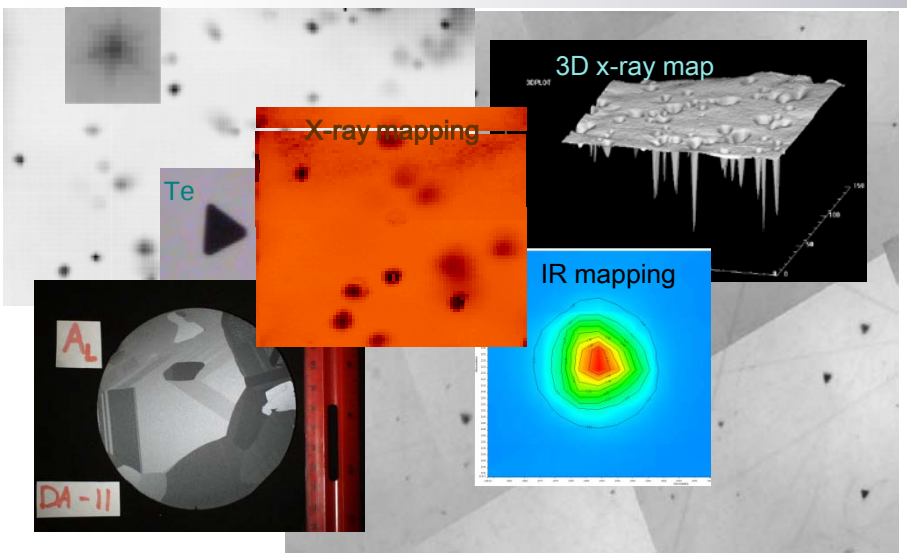
RadSensing2009

Development of Improved CZT for Gamma-Ray Spectrometers and Imaging Arrays

June 4, 2009

Ralph B. James
BNL

Research Team



On-Site Student Support: Fisk University (2), University of Michigan, Cornell University, Tennessee Technological University, Idaho State University, and Vanderbilt University

<u>Project started October 2008</u>

Budget: FY09 - $774K
Budget Spent/Committed: $378K
Budget Remaining: $396K

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Industrial collaborators: H. Chen, Redlen, Vancouver, BC, Canada; C. Szeles, eV Products, Saxonburg, PA; L. Li, Yinnel Tech, South Bend, IN; K. Mandel, EIC Labs, Boston, MA.

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University of Freiburg:
M. Fiederle and A. Fauler

Idaho State University:
R. Gul, R. Rodriguez, K. Keeter

Charles University:
J. Franc, V. Babentsov, R. Grill, E. Belas, M. Bugar, and P. Hoschl

BNL:
A. E. Bolotnikov, G. S. Camarda, Y. Cui, A. Hossain, A. Hossain, A. E. Bolotnikov, G. S. Camarda, H. K. Kim, Y. Cui, Z. Zhong, W. Yao, G. De Geronimo, J. Fried, P. O'Connor, Z. Li, and R. B. James

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Goal and Objectives

Goal: Resolve the material show-stoppers limiting the production of high-energy-resolution (0.5% FWHM at 662 keV), large-volume gamma detectors for nonproliferation applications

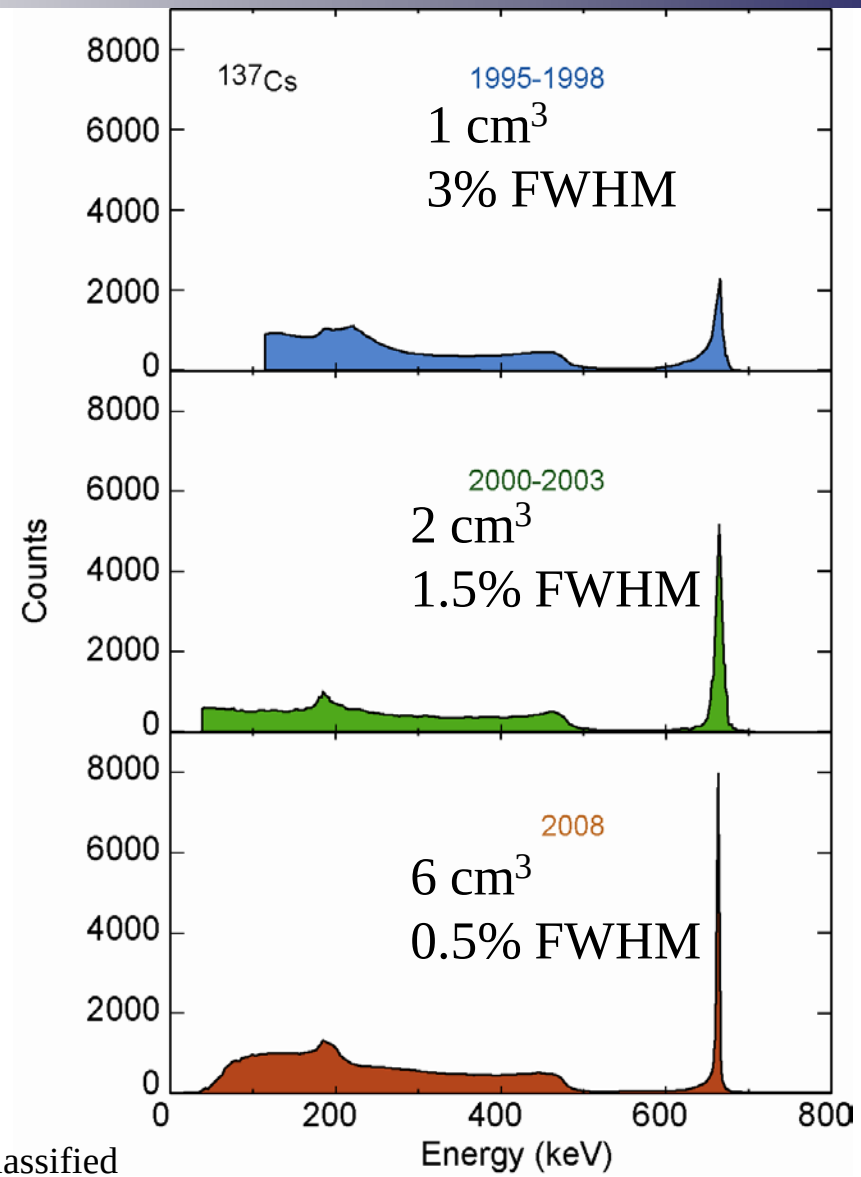
1) **Understand the material properties limiting the performance of large-volume (20x20x15 mm^3) CZT detectors**

2) **Provide rapid feedback to crystal growers and, in conjunction with suppliers, conduct growth experiments to improve CZT detector performance and yield as measured by energy resolution, efficiency and stability**

3) **Develop efficient, non-destructive techniques to screen the quality of CZT crystals for selecting detector-grade blanks**

Steady advances from year-to-year: Next phase could be game changing

CZT is best candidate material today for room-temperature, field-portable semiconductor radiation detectors. With sustained rate of performance improvements over the next 5 years, the deployment of CZT gamma-detector technology is poised for rapid expansion.



Challenge: Availability of high-performance detectors is seriously restricted due to material deficiencies

Approach: Fix the Material through Feedback-and-Improvement Process

(1) Crystal Growth
- THM, HPB and conventional Bridgman

(2) Characterize Range of Crystals
- DLTS, TCT, PL, EPDs, XRD, PCD, TEM, and IR transmission

(3) Detector Fabrication
- Surface polishing, etching, contact deposition and passivation

(4) Detector Evaluation
I-V, μ_e, $(\mu\tau)_e$, internal E fields, energy spectra, and high-resolution x-ray response mapping

(5) Correlations between Material and Device Properties

(6) Understanding of Performance-Limiting Defects

Modified Manufacturing Processes

Four material problems found to limit many detector applications:

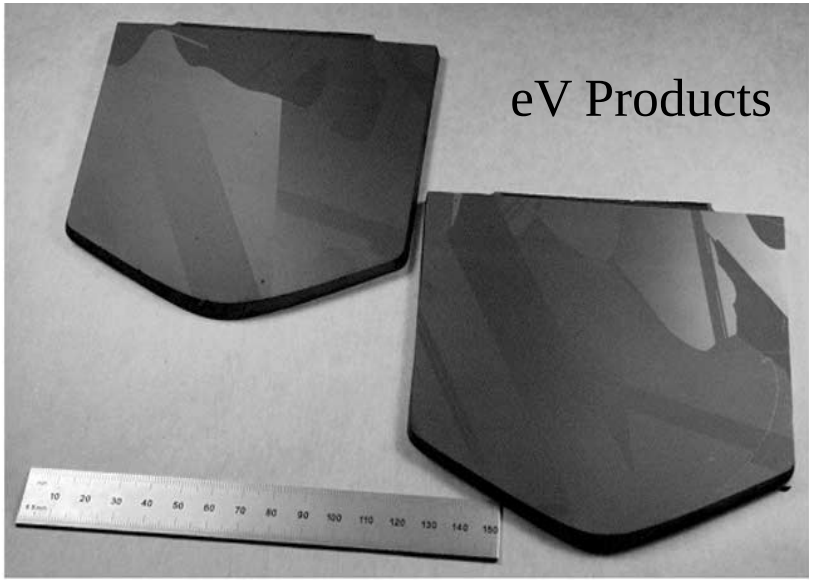
(1) Multiple crystals (e.g., grain boundaries and twins)
(2) Te-rich inclusions and precipitates
(3) Dislocations
(4) Reproducible manufacturing processes to produce high yields and acceptable costs

Supporting Researchers:

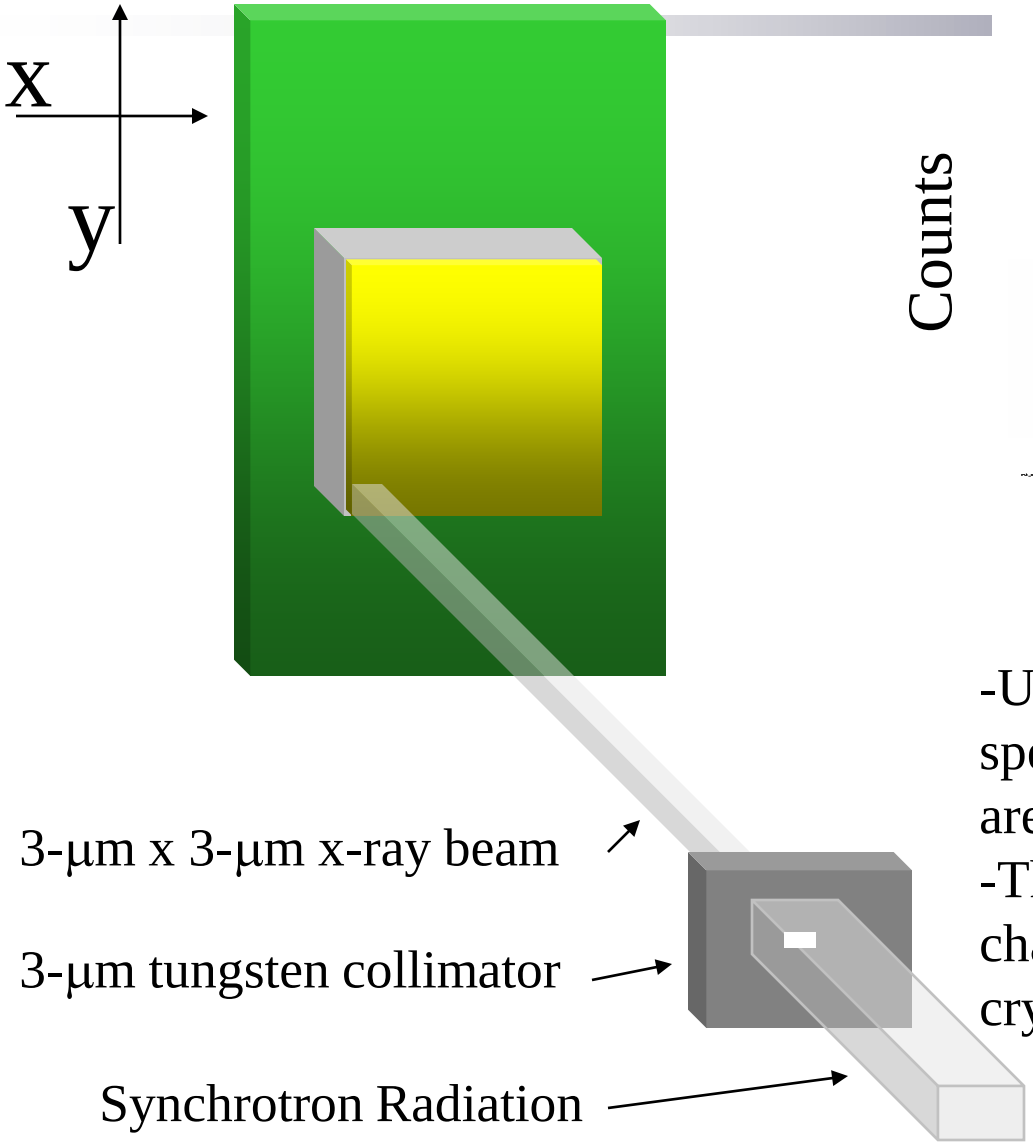
(1) eV Products and Redlen (Canada) – High Priority

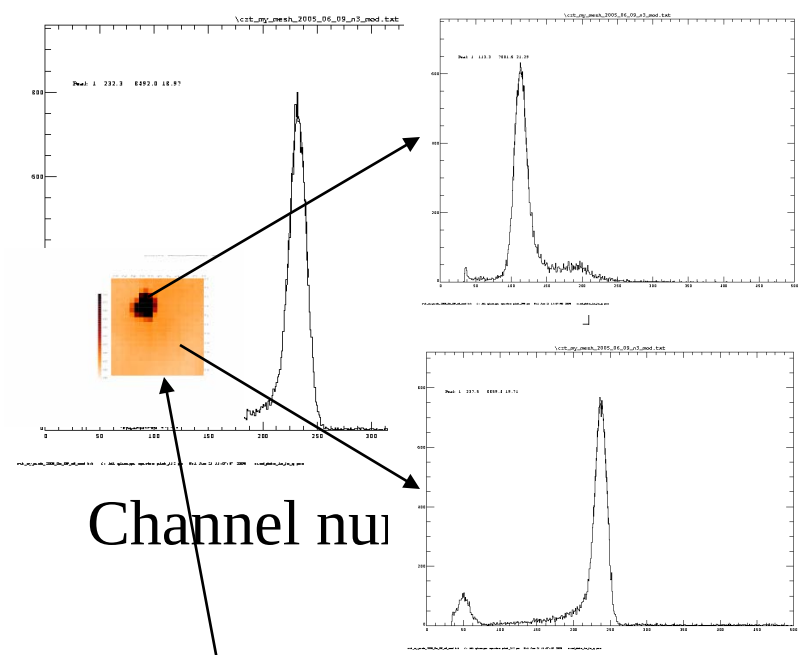
(2) Yinnel Tech*, RMD, EIC Labs, Orbotech Medical (Israel), Eurorad (France), CIEMAT (Spain), IMEM-CNR (Italy), Acrorad (Japan), and Univ. of Freiburg (Germany) – Lower Priority

Example of CZT wafers with multiple grain boundaries and twins.



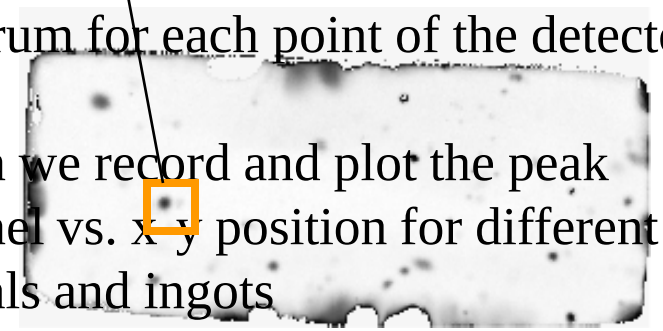
Unique micro-scale detector mapping facility pivotal for understanding large-volume detectors





-Using a MCA we measure the energy spectrum for each point of the detector area

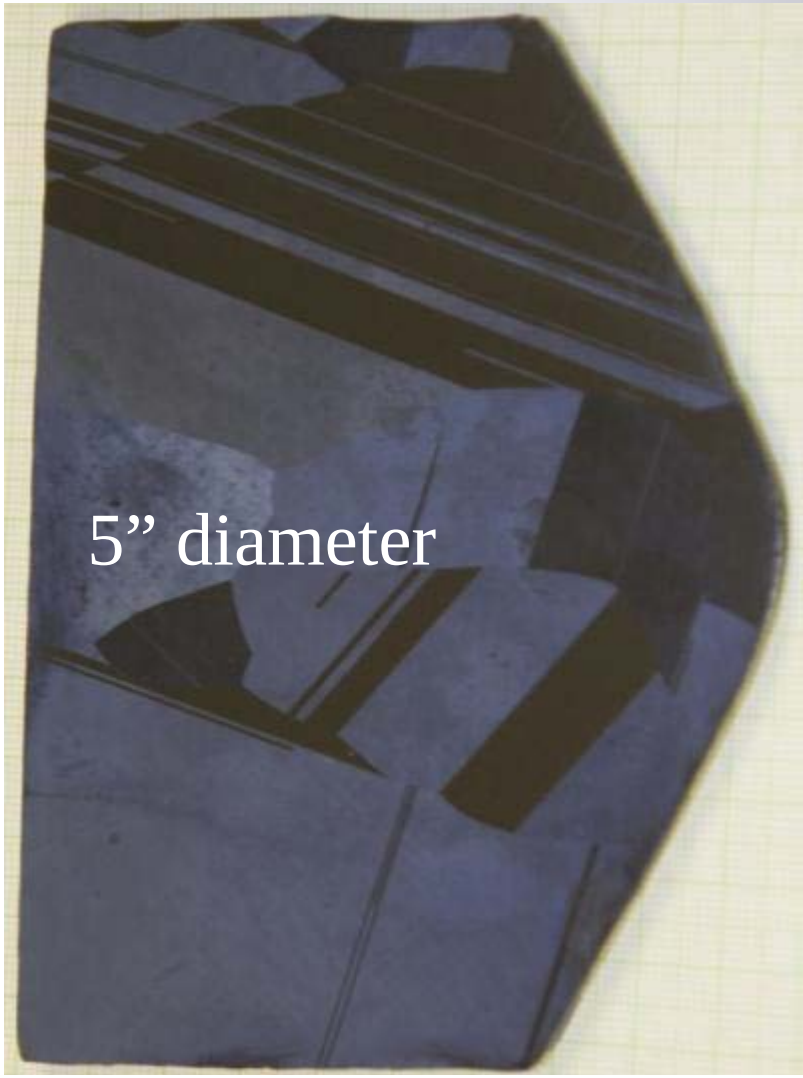
-Then we record and plot the peak channel vs. x-y position for different crystals and ingots



3x3x7mm^3 CZT detector

Challenge #1: Problem of Multiple Crystals

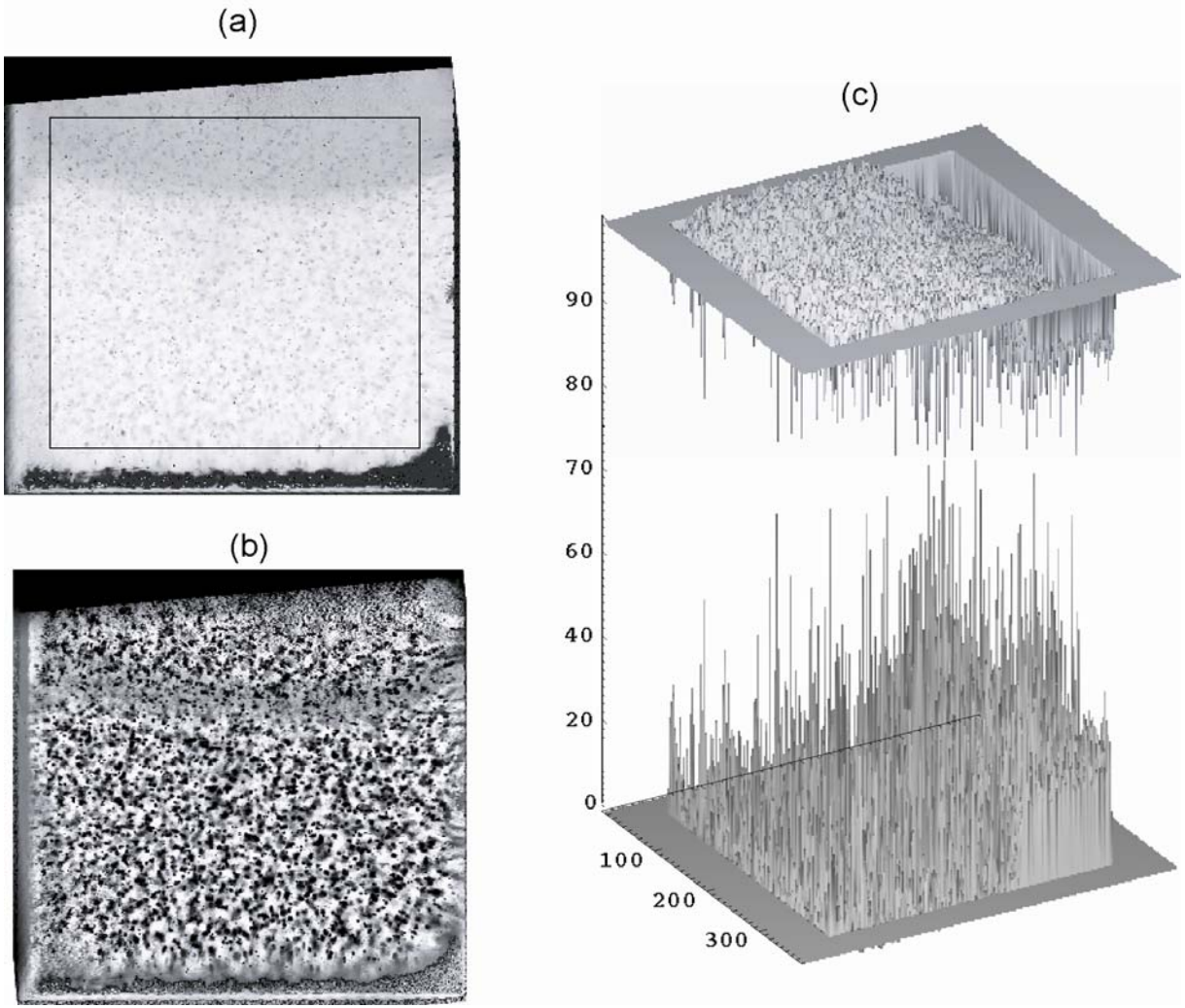
Extended Defects in Commercial CZT Material Lead to Trapping and Non-Uniform Response



- Several types of extended defects can be found in a typical commercial CZT crystal:
  - sub-grain boundaries,
  - twins,
  - Te inclusions,
  - and dislocations - often arranged in dislocation walls or mosaic structures
- Many of these defects are not readily seen with visible or IR microscopes => they are often overlooked by vendors
- Extended defects are less important in thin detectors, but their role increases with device thickness.

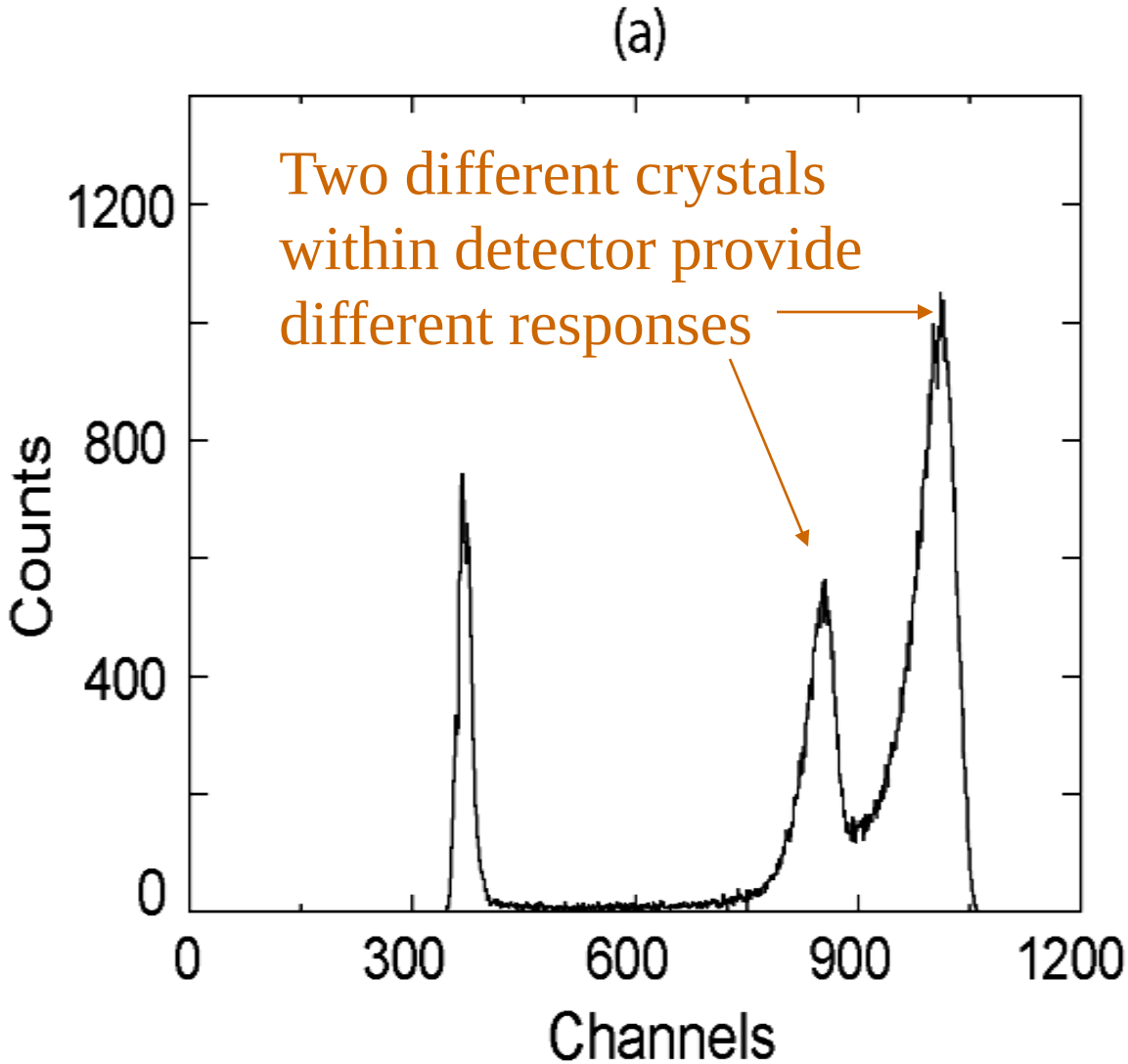
X-ray Response Map for Detector Composed of Two Large CZT Grains

10x10x2 mm^3

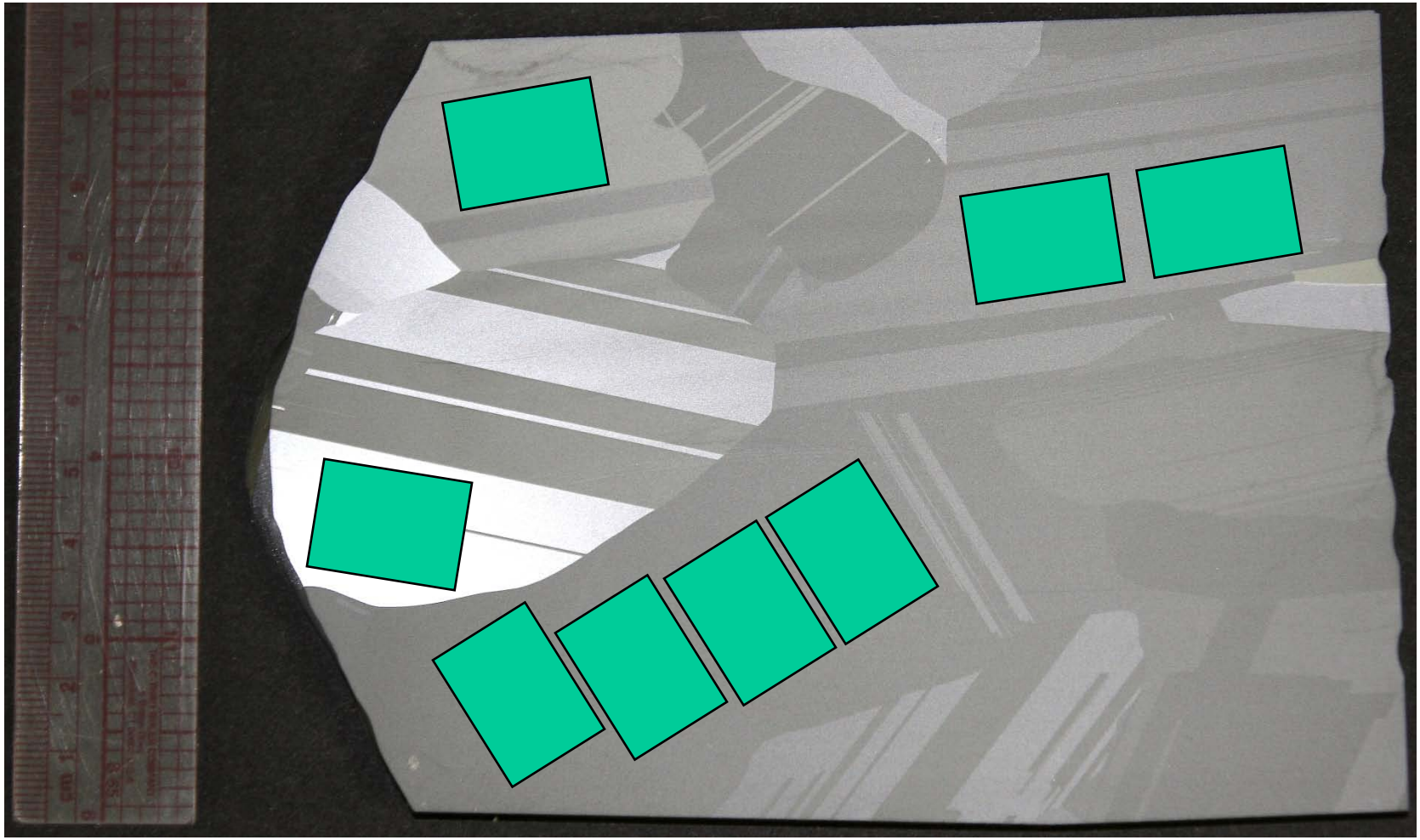


Pulse-height maps taken with 10-micron spatial resolution

Grain boundaries are the zero-order problem: Gamma spectrometers require single-crystal CZT



Single crystals are necessary but not sufficient: Even poor ingots possess *many* single crystals for 20x20x15 mm^3 detectors

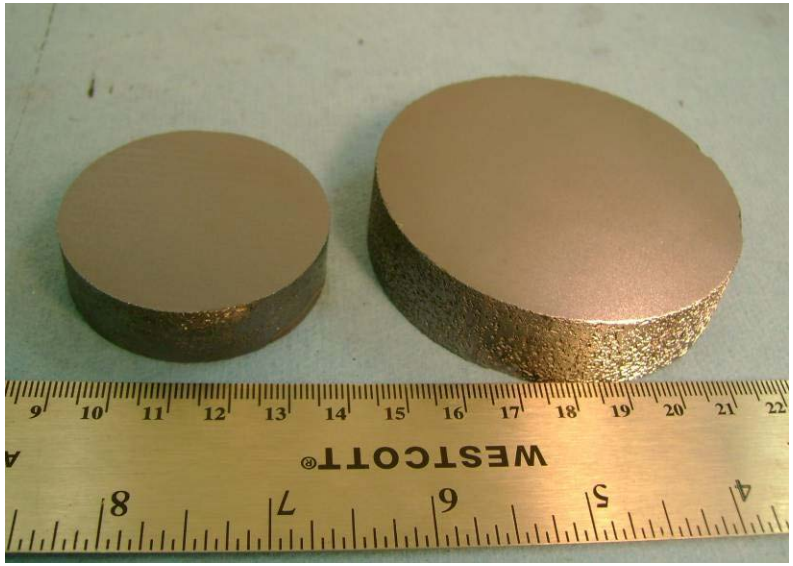


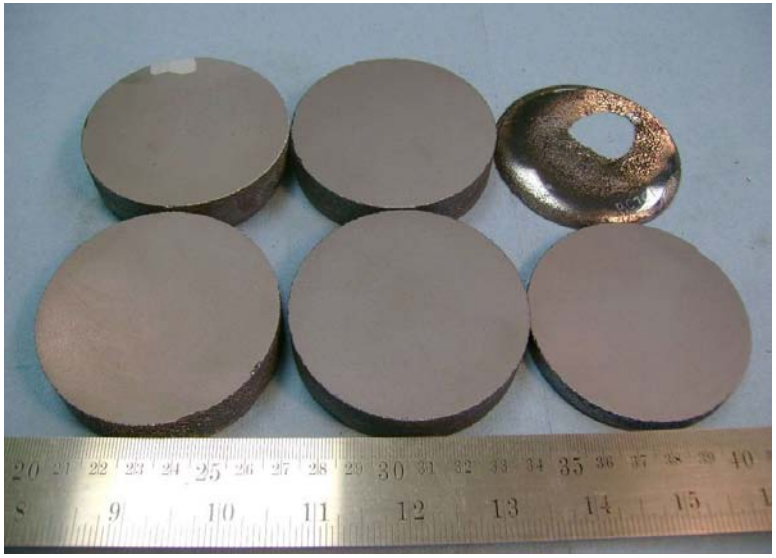
- Grain boundaries are *not* limiting availability of 3-D devices, but do affect cost.
- Moreover, grain boundaries are easily identified.

Redlen now able to grow bigger and thicker single crystals (THM)

Redlen's 50-mm diameter CZT ingot

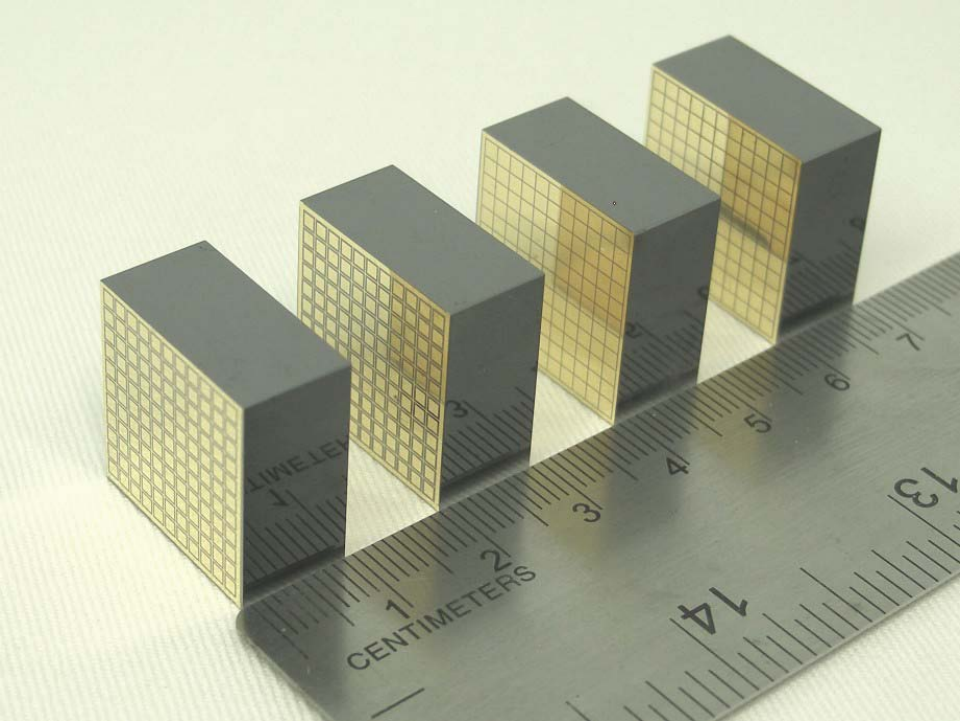
Redlen's new 75-mm diameter ingot

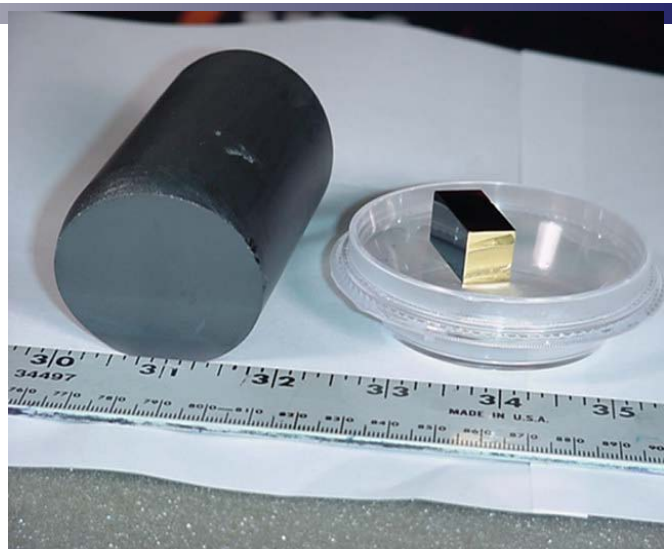


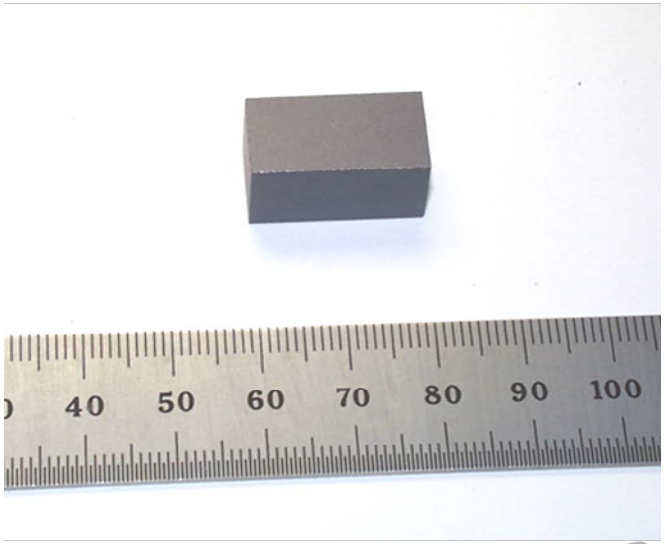


Major jump in growth of large single crystals also achieved using conventional Bridgman

Today, it is possible to grow large volume single crystals up to hundreds of cubic centimeters.



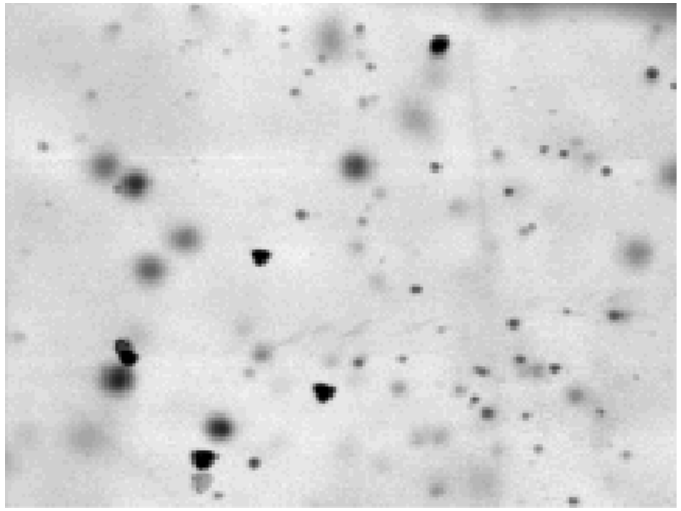




Despite ability to produce large-volume single crystals, high-energy-resolution (<0.7% FWHM) CZT detectors are rare.

Reason: High-resolution mapping data reveal non-uniformity in the charge transport that cannot be fixed electronically

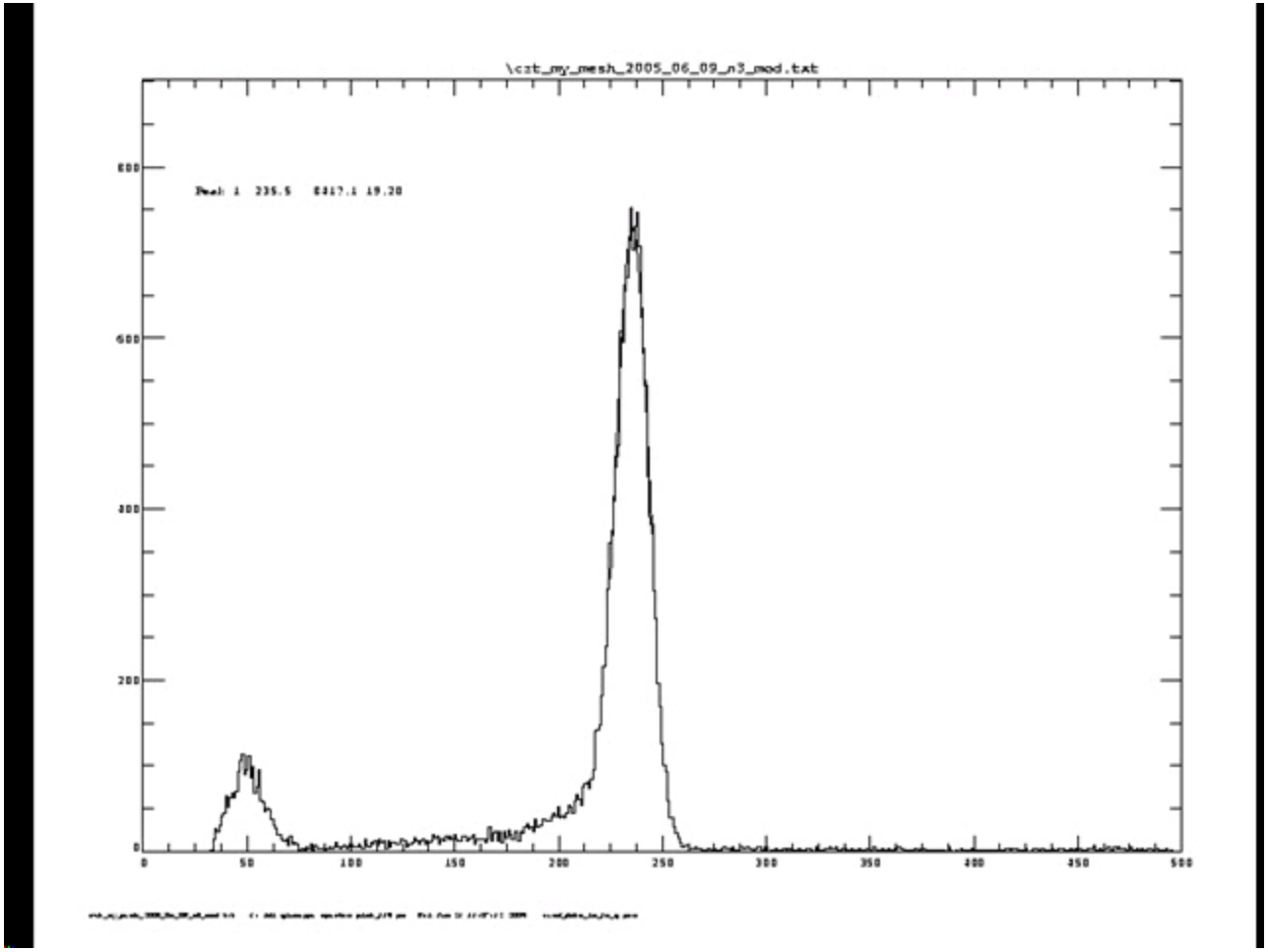
1.0x1.5x2.0 mm^3

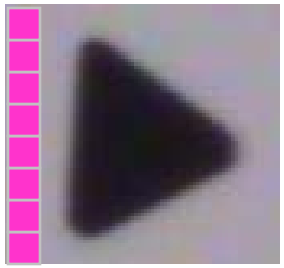


Charge-collection efficiency map

Correlation of material and device properties confirms that degraded regions are caused the presence of tellurium-rich secondary phases in the crystals

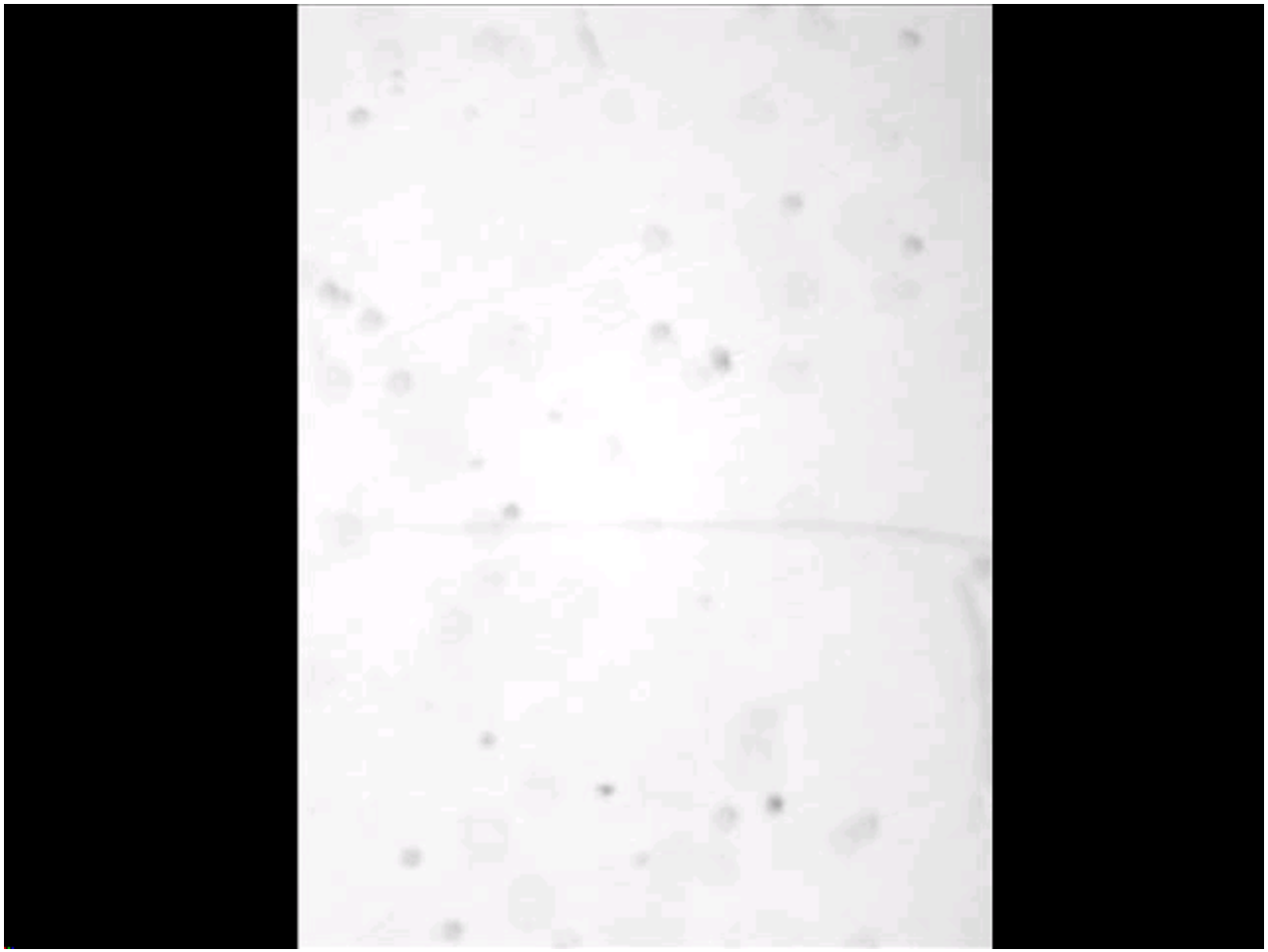
Response map near a Te inclusion





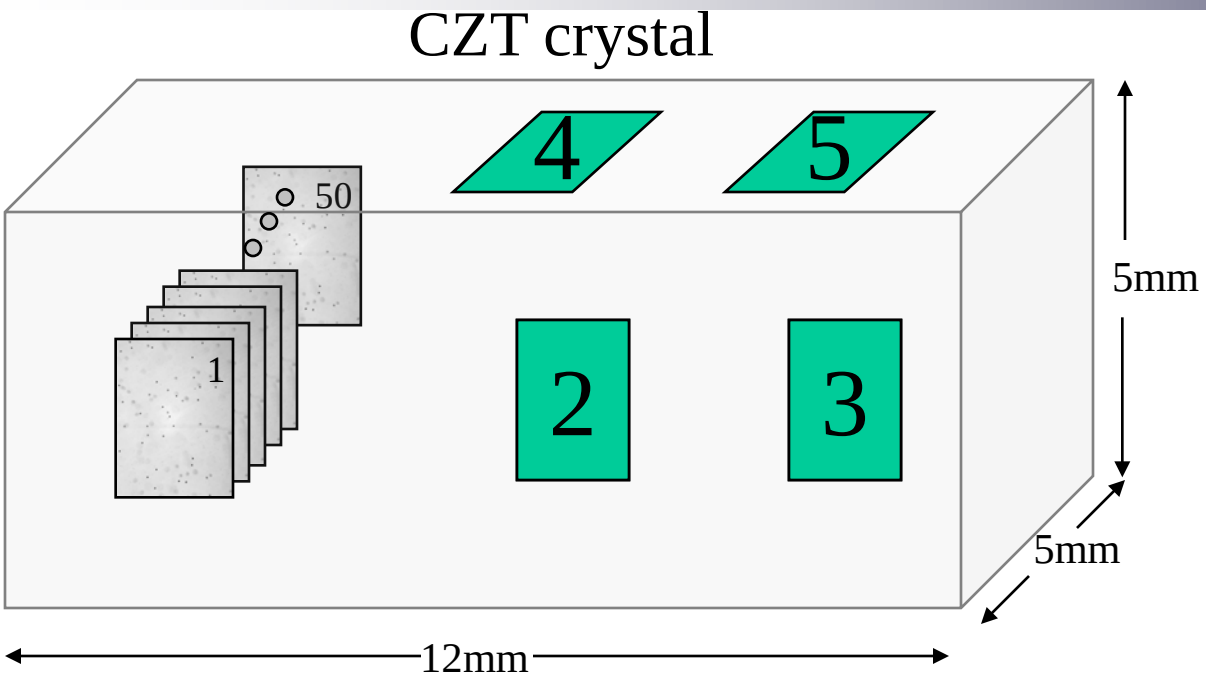
100 µm x 100 µm area
10-µm x 10-µm pinhole
5-µm step size

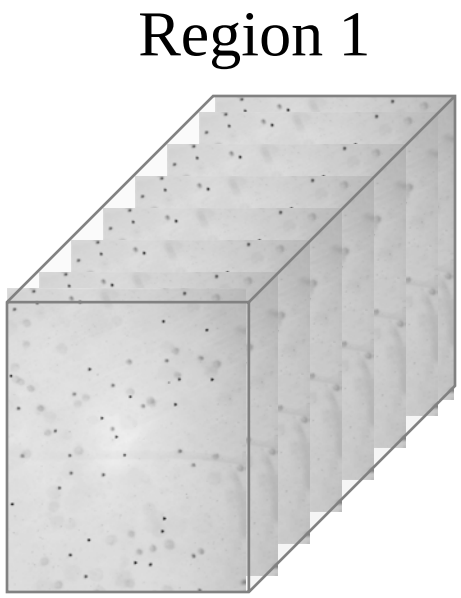
Automated infrared mapping system used to quantify Te inclusions for different growth runs



- Scans in the z (depth) direction with ~100 um steps
- Algorithm applied to accurately record the size and number of Te inclusions in each layer
- Good agreement with manual counting method

IR Transmission Microscopy Technique



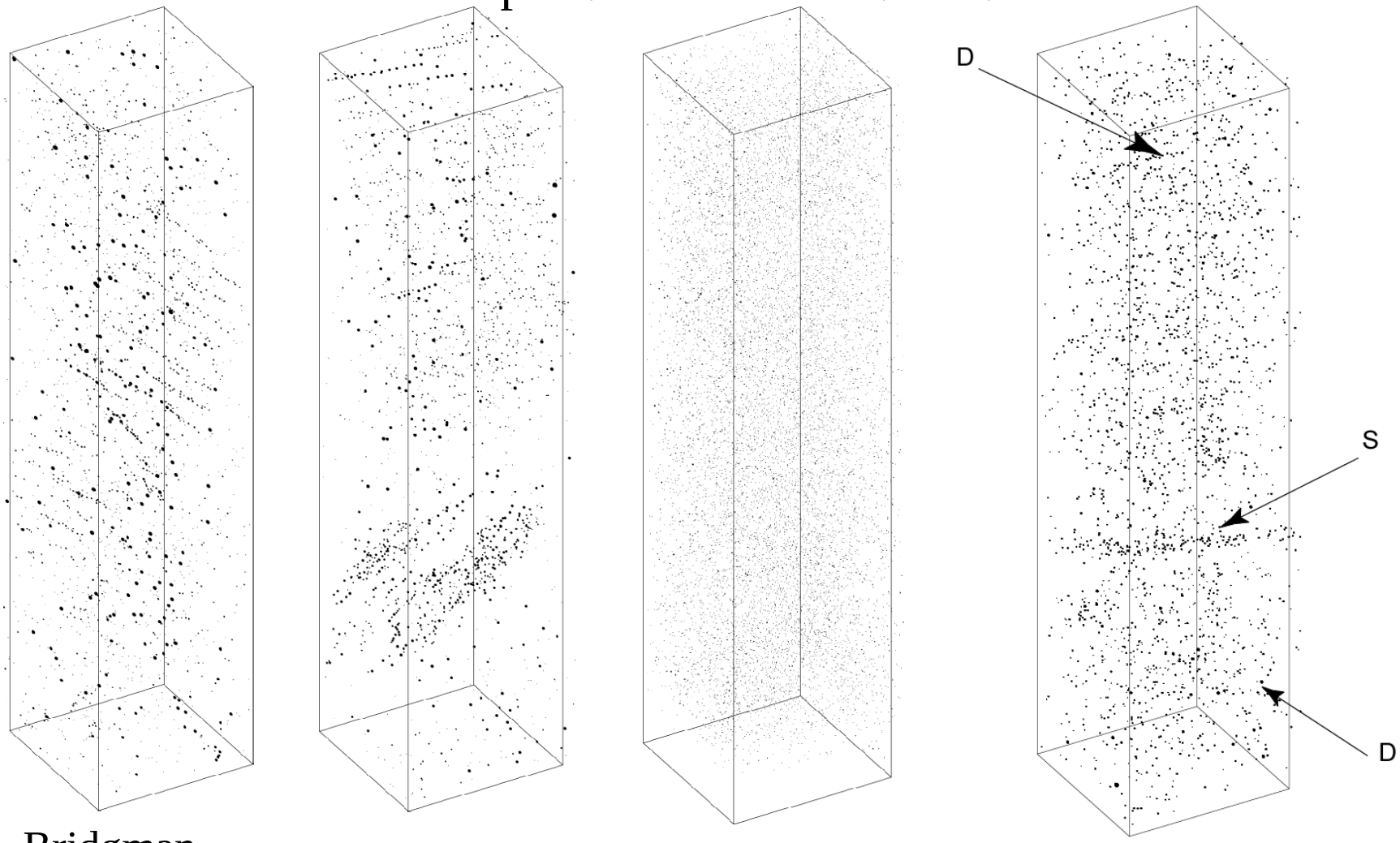


This measurement reveals:

- **Size and density of Te inclusions**
- **Distribution pattern of Te inclusions**
- **Other substantial defects**

Reconstructed 3D distributions of Te secondary phases for different growth methods

Some Examples: CZT volume: 1x1.5x5 mm^3

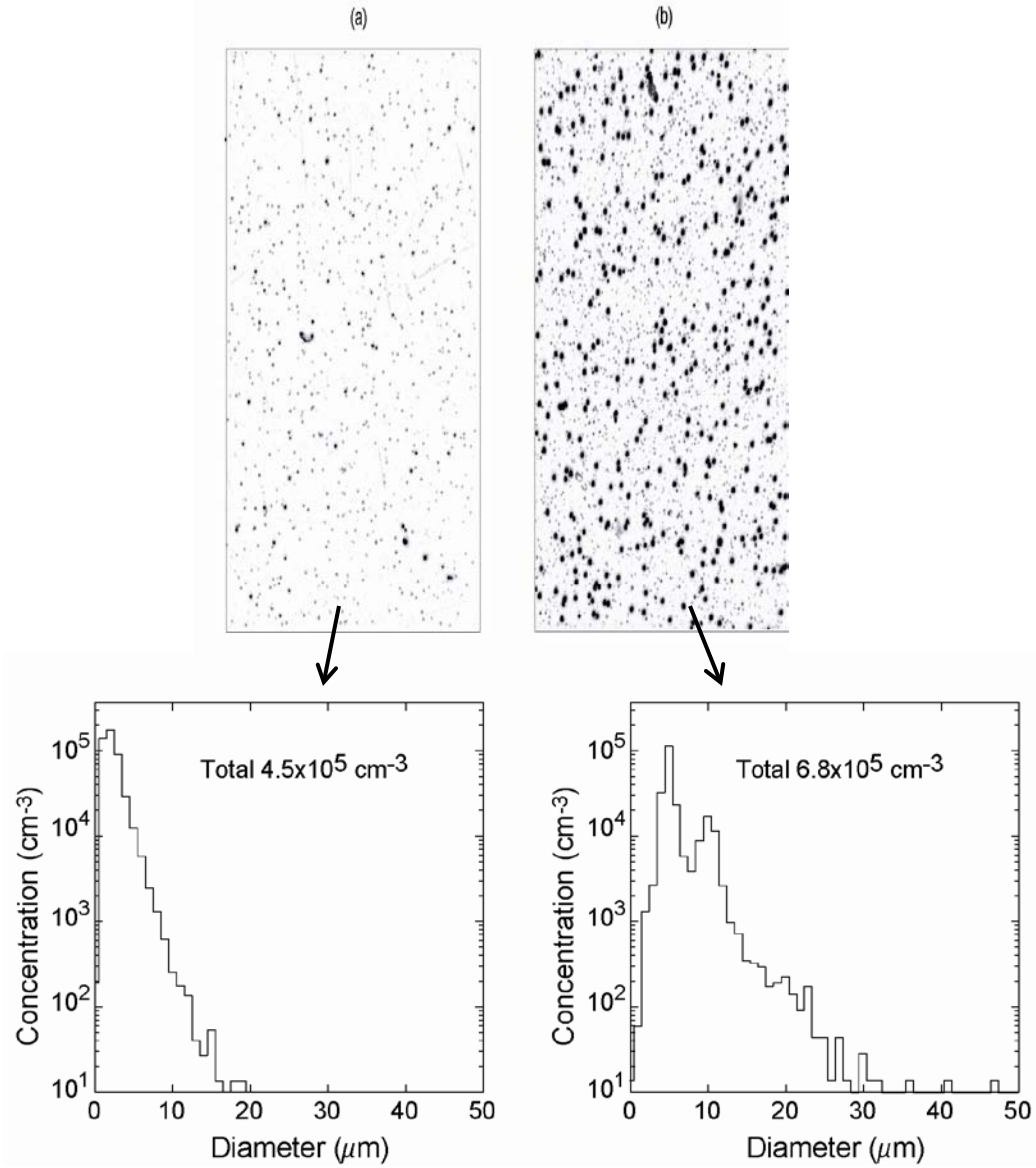


Bridgman | Bridgman | THM | HPB

Concentration and size of Te-rich secondary phases affected by growth and thermal annealing conditions

Superimposed IR images of 10-μm-thick detectors with small (a) and large (b) sizes and concentrations of Te inclusions. Each image consists of ~100 photographs taken at different depths and "collapsed" on one plane.

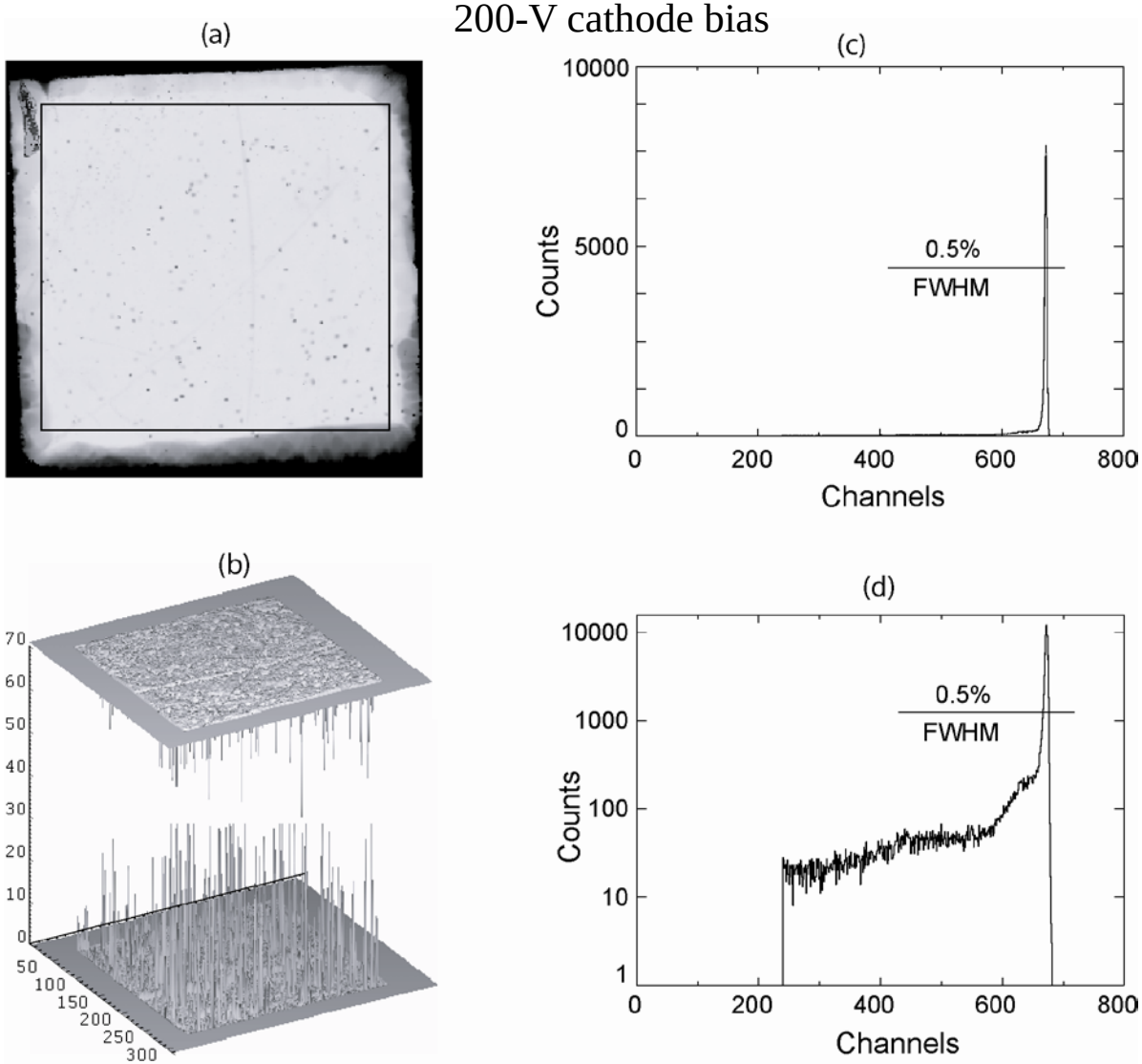
The size distribution of inclusions averaged over the volumes for each crystal.



Detector with low Te inclusions produces extraordinary results (0.5% FWHM with no corrections)

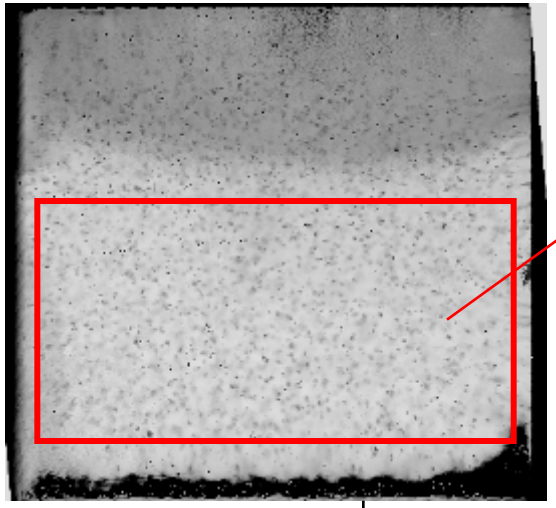
The response map and frequency distribution of the peaks' positions (response function) measured for a 10x10x1.2 mm^3 with a low concentration of Te inclusions.

200-V cathode bias

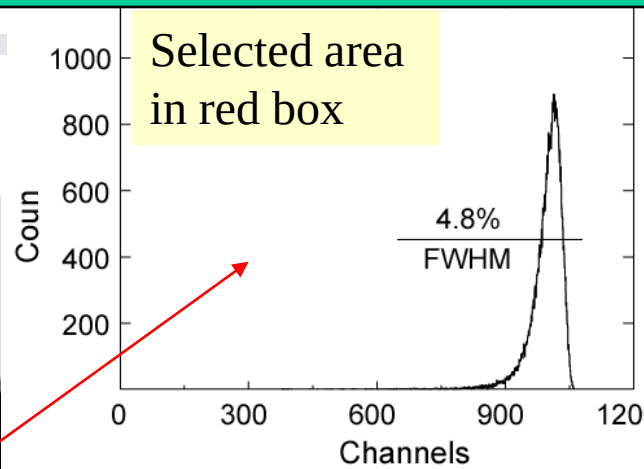


Important discovery: Most effects of non-uniformity can be overcome with high-granularity pixel devices

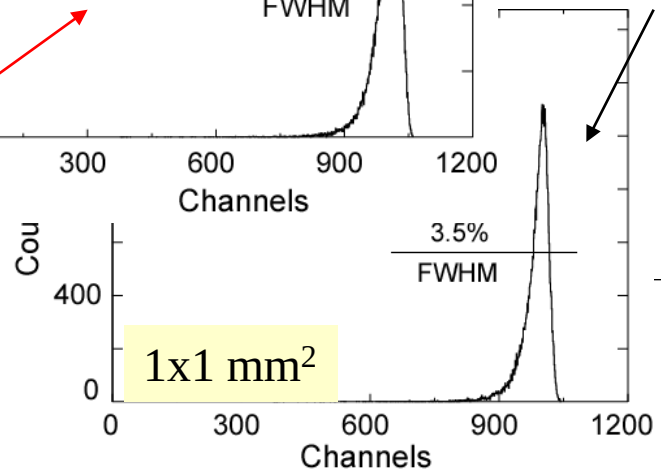
X-ray response map
10x10x10 mm^3

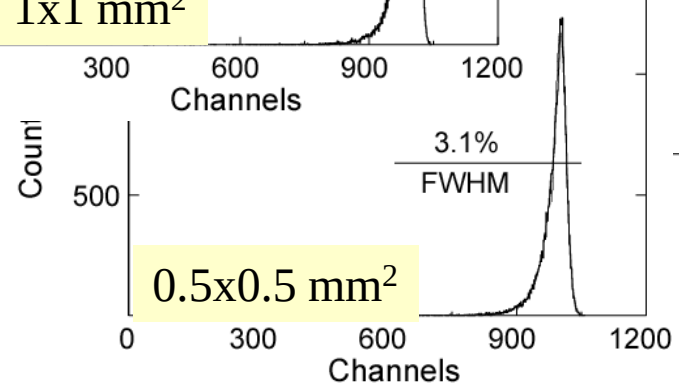


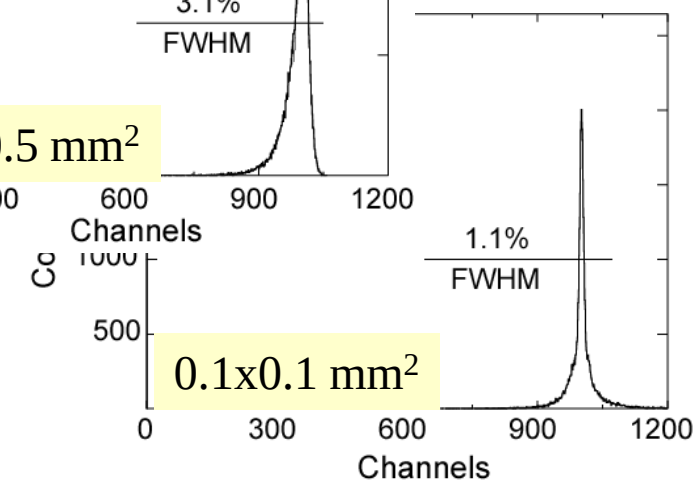
Selected area in red box



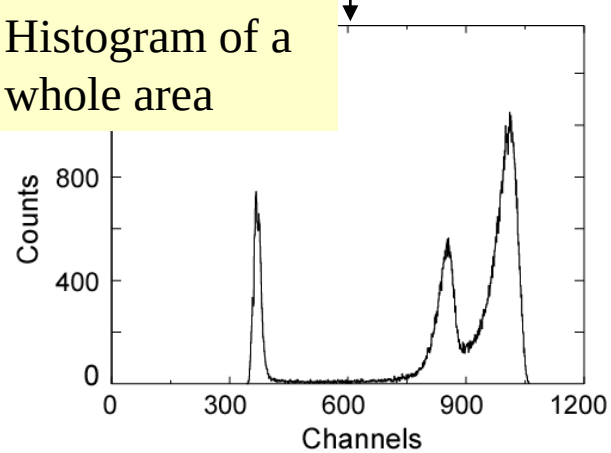
After dividing into small pixels and normalizing responses from each pixel.





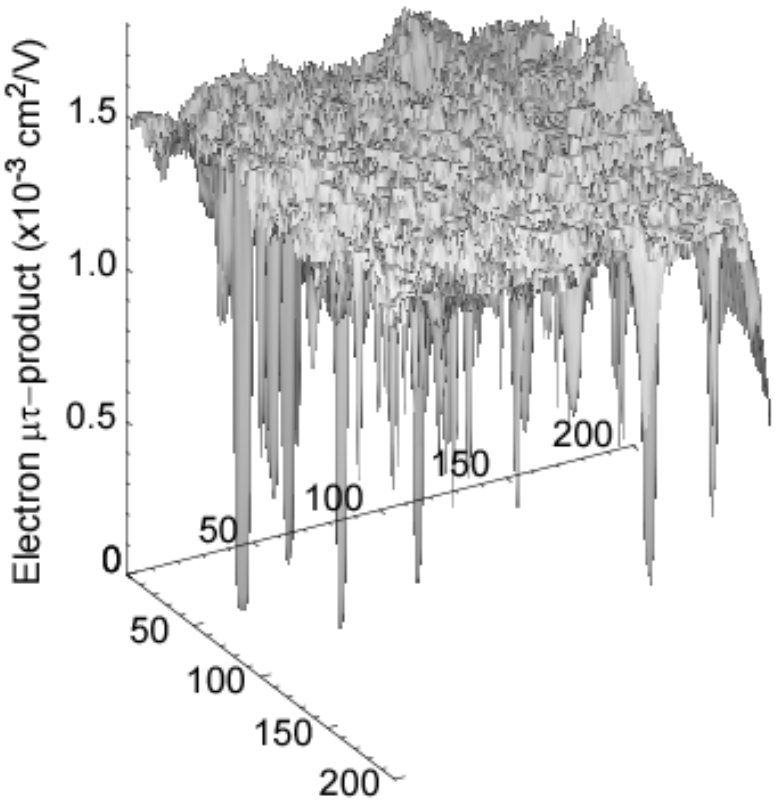


Histogram of a whole area

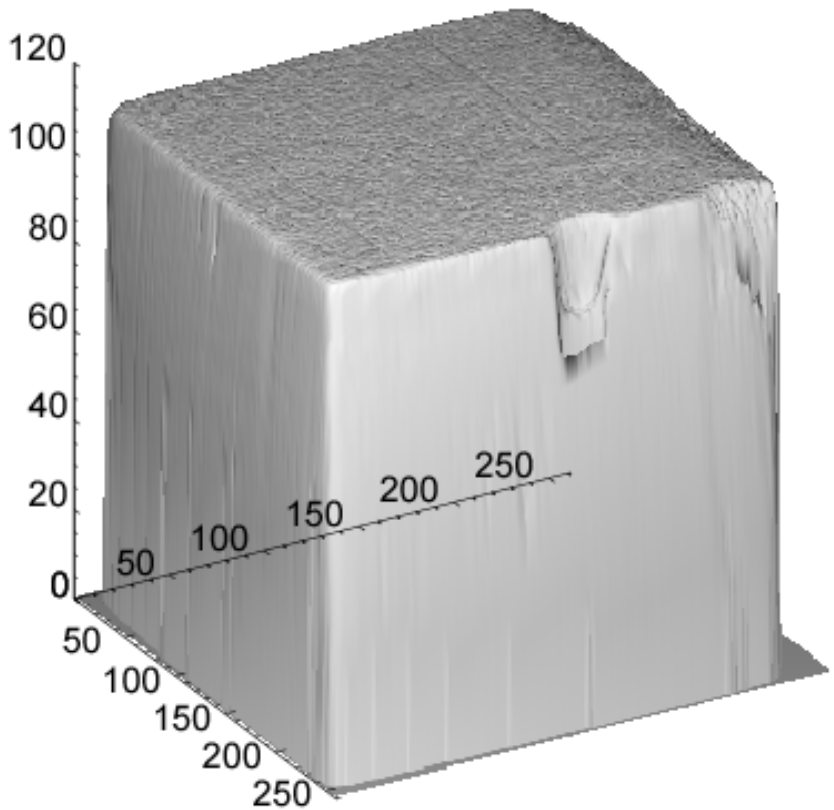


Data reveal new approach to resolve problem of low availability for detector-grade crystals

Electron μτ map (No correction)



Uniform charge collection after correction



2x1.5 mm^2 area; Spot size 10x10 μm^2

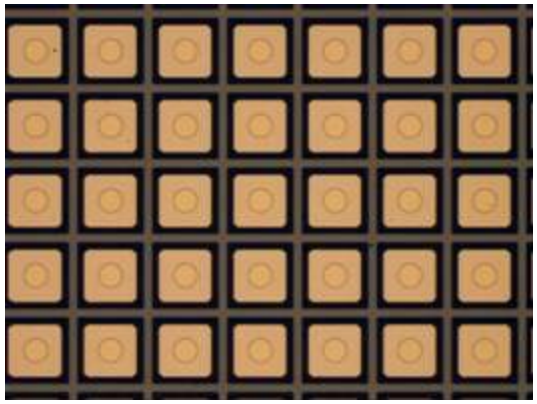
Energy resolution up to 0.3% FWHM expected

- ***Even on relatively low-quality CZT!!!***

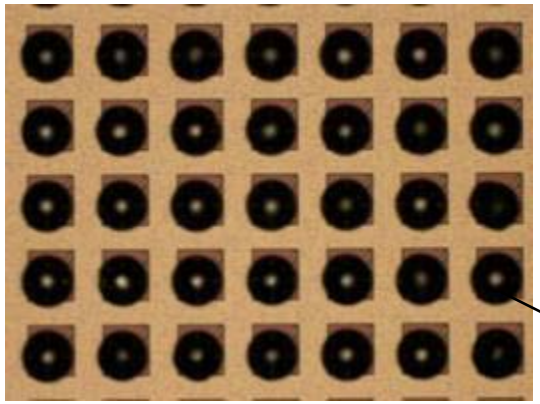
Pixel dimensions with 100-μm size achieved

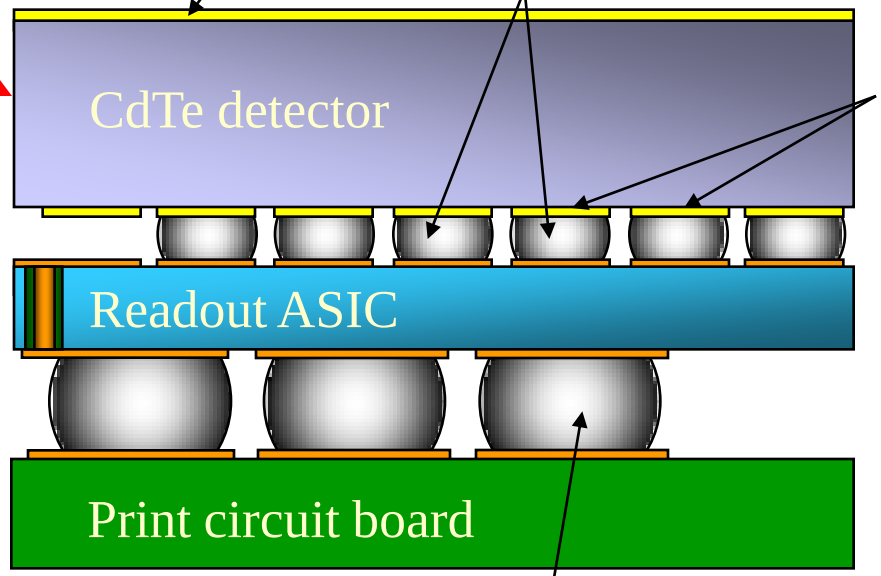
Demonstrated for CdTe in current-mode operation (i.e., no charge-loss correction for pixels)

Pixelized CdTe detector



CMOS Readout ASIC





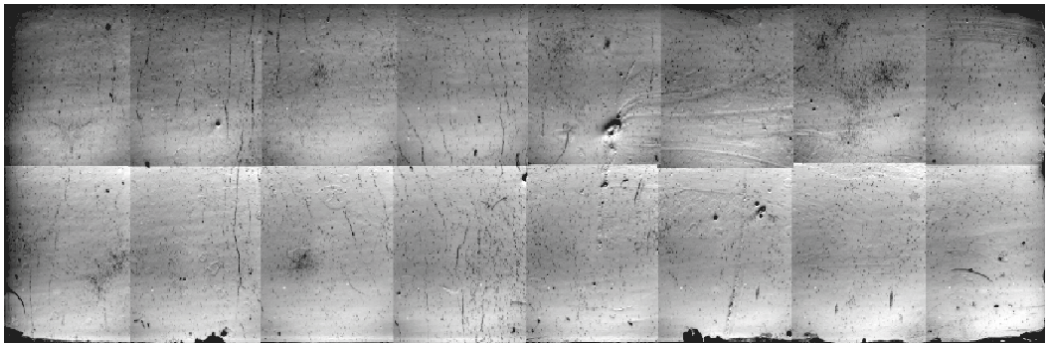
Bumps

Credit ACRORAD

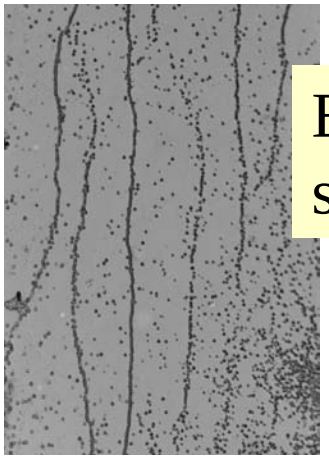
Challenge #3: Problem of charge loss by dislocations and effects on the internal electric field

Charge losses caused by dislocations

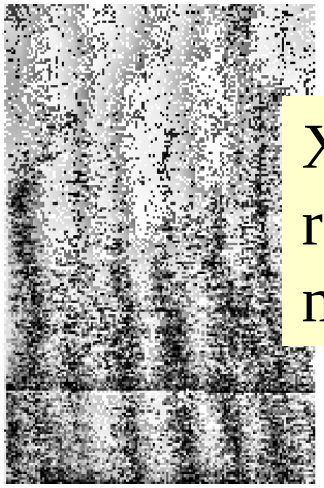
Photograph of the etched surface of a 5x5x12 mm^3. Etch pits form linear patterns on the surface.



Magnified areas



Etched surface

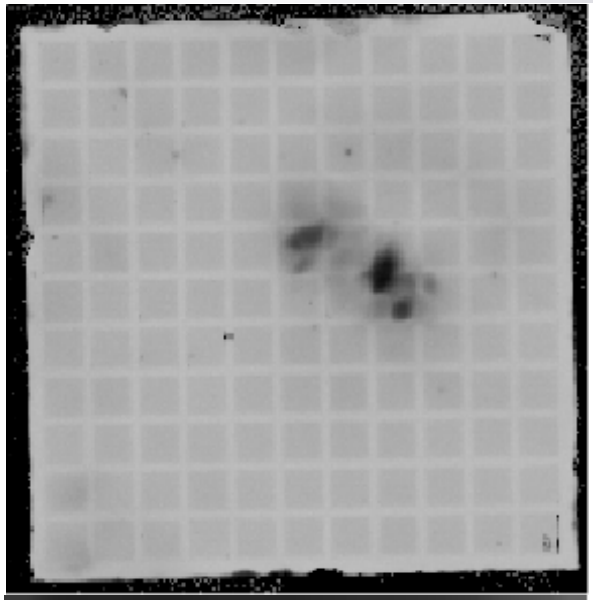


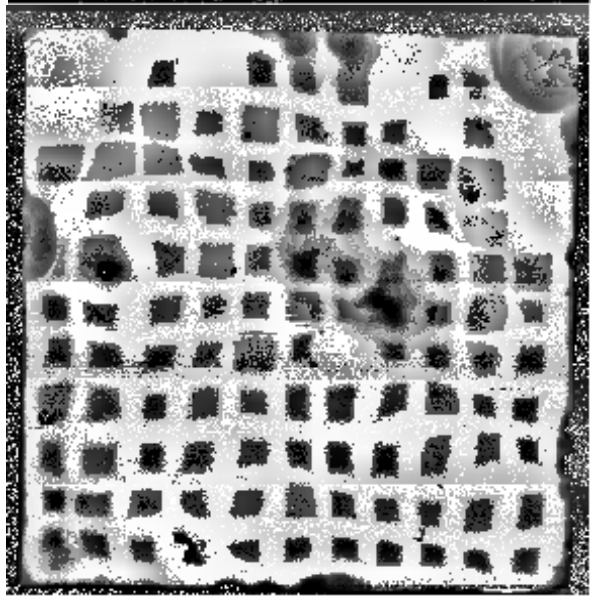
X-ray response map

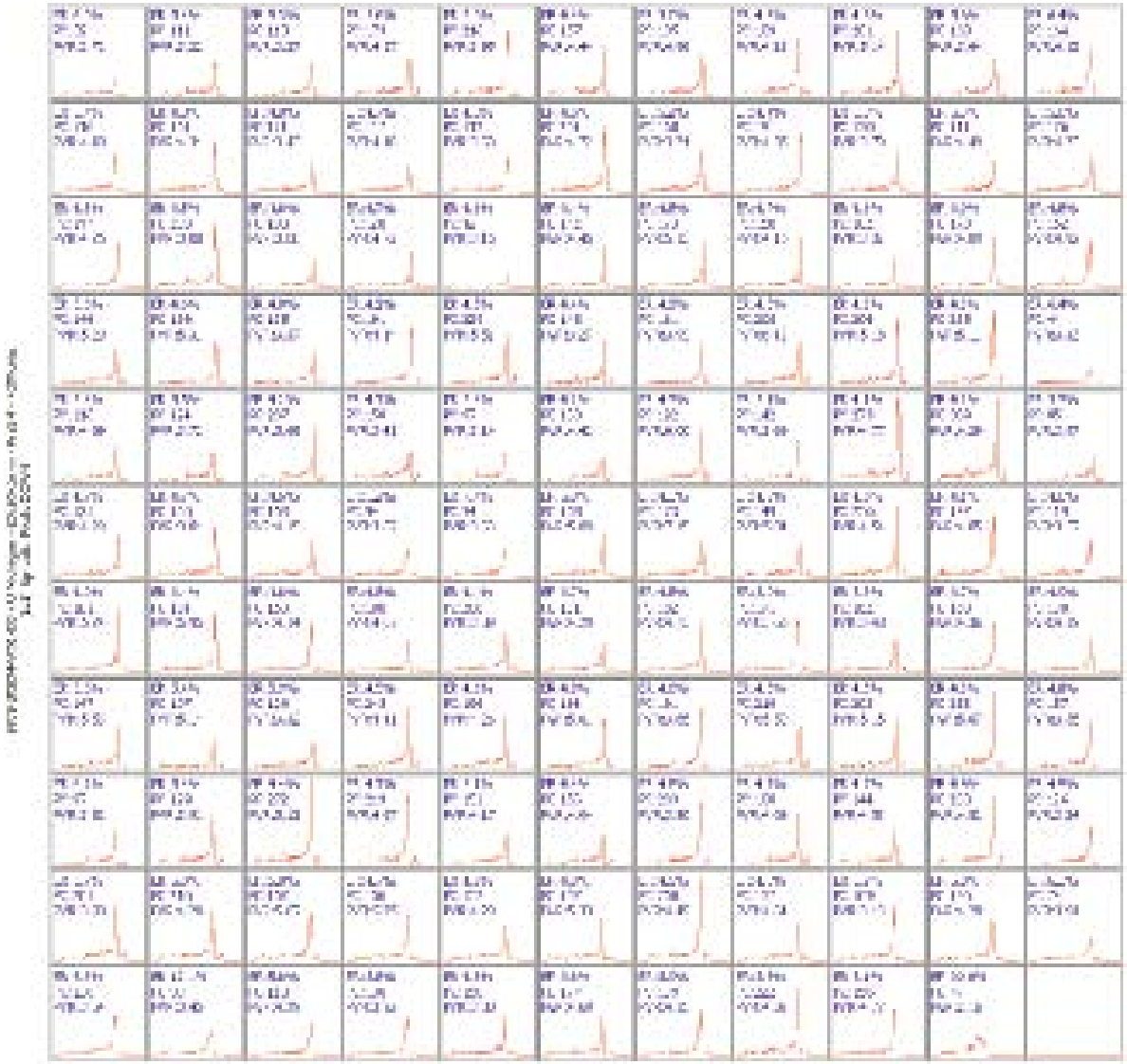
- X-ray map correlates with the etch-pit patterns.
- Dislocation walls trap charge carriers.
- Charge loss is small, but their effects reduce energy resolution.

Dislocation walls affect internal electric field:

Example for 20x20x15 mm^3 multi-pixel detector

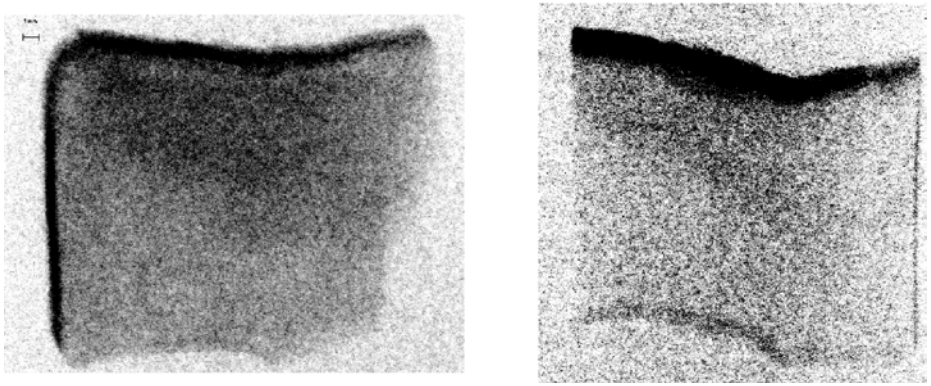




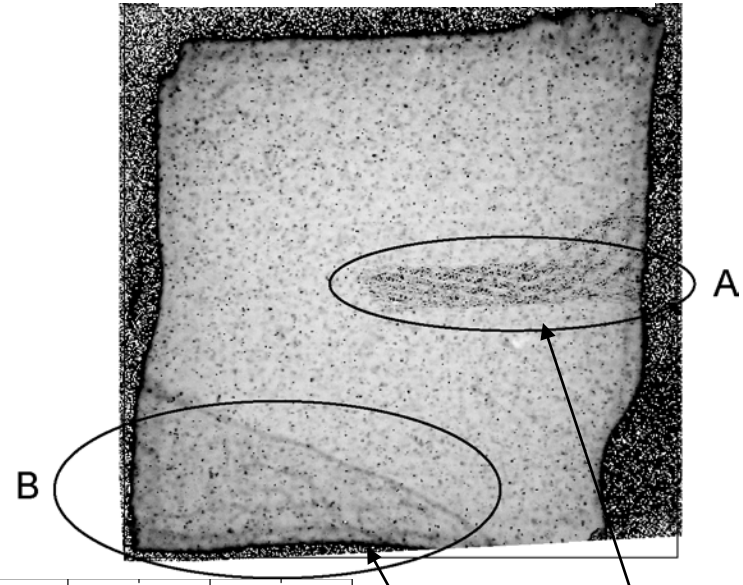


Scanning of a 20x20x15 mm^3 CPG

Diffraction topography map



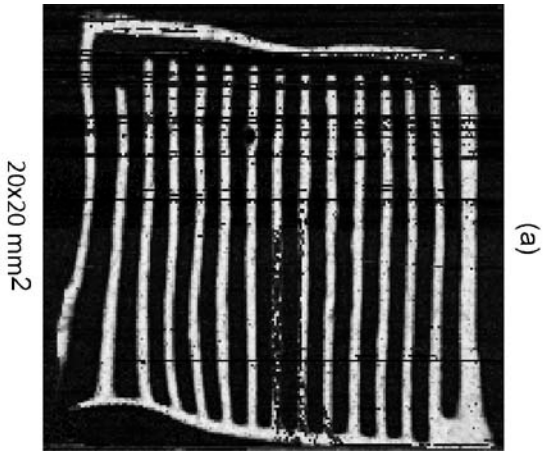
X-ray response map

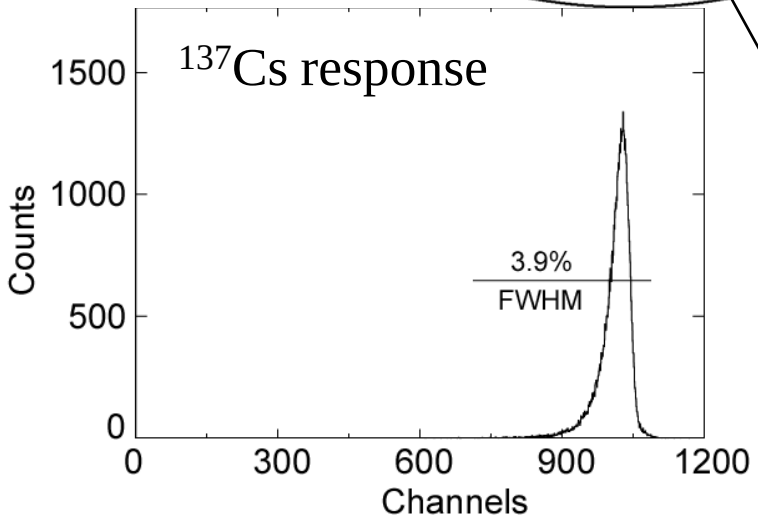


Projection of cathode wire

Twin

E-field is non-uniform



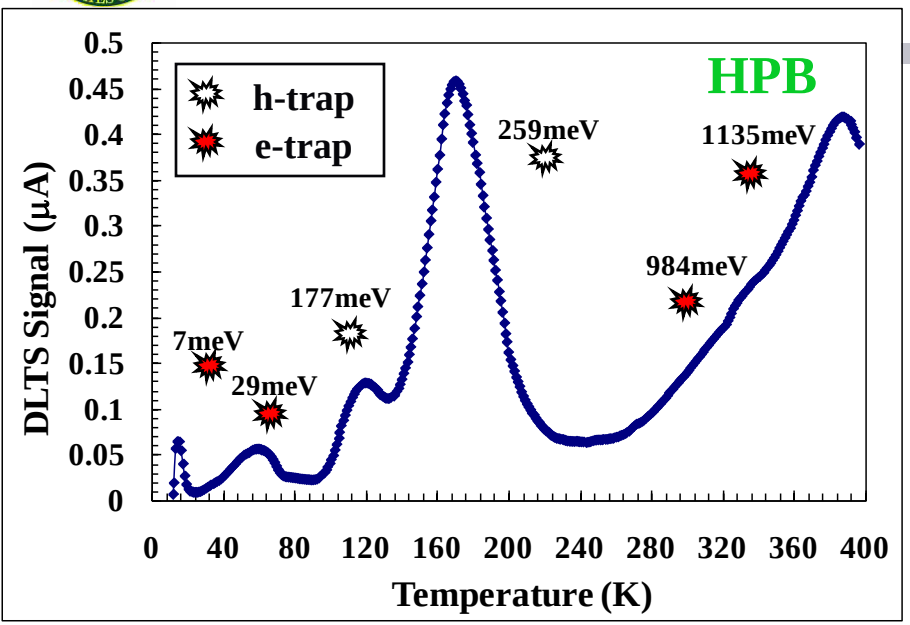


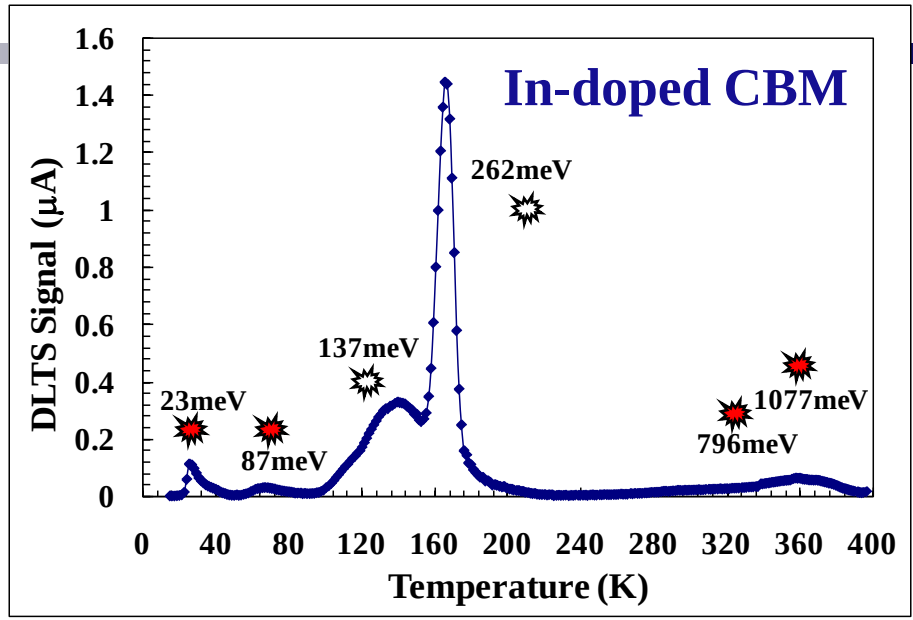
Challenge #4: What is origin of point defects limiting the electron drift lengths?

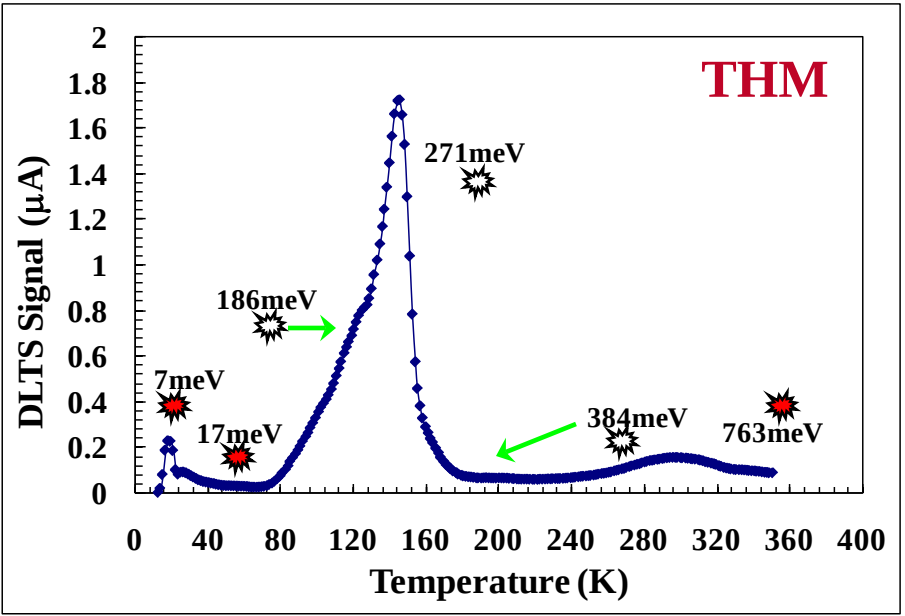
Different growth methods investigated

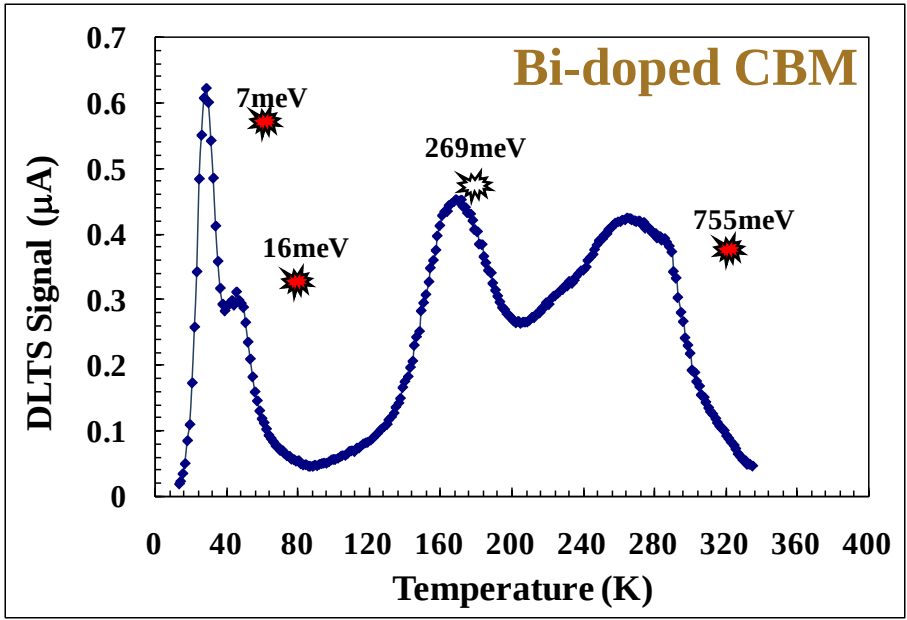
#	Sample	Crystal Growth Technique	Thickness (cm)	Area (cm^2)
1	In-doped CBM	Conventional Bridgman	0.19	0.23
2	Bi-doped CBM	Conventional Bridgman	0.20	0.72
3	HPB	High Pressure Bridgman	0.21	1.02
4	THM	Traveling Heat Method	0.20	0.30

Energies of Dominant Traps Identified for Different Growth Methods

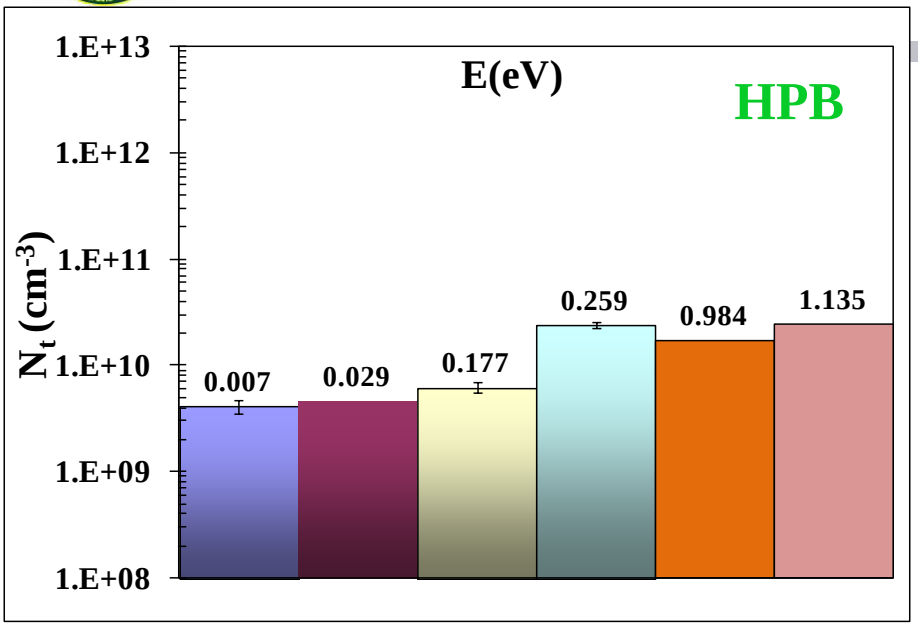


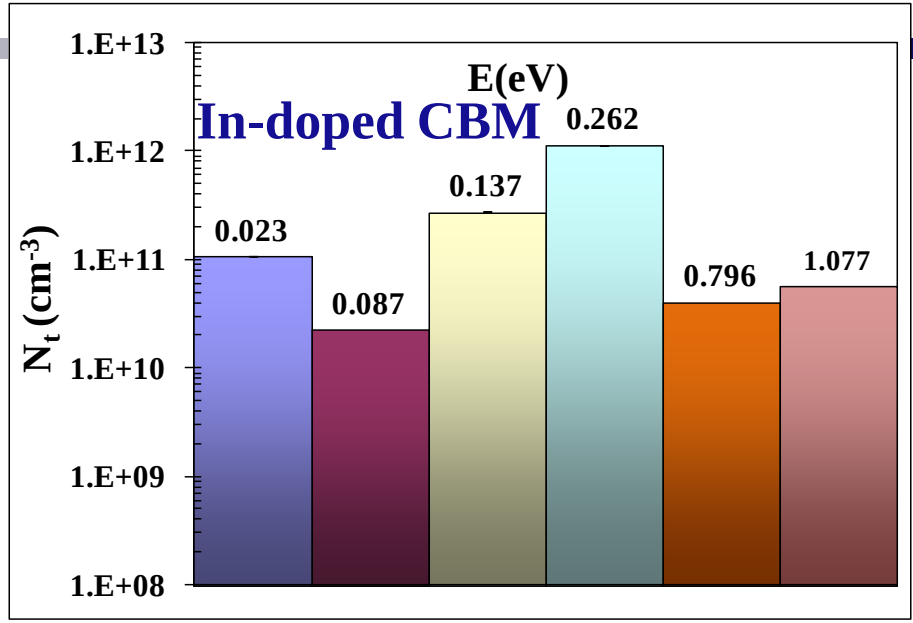


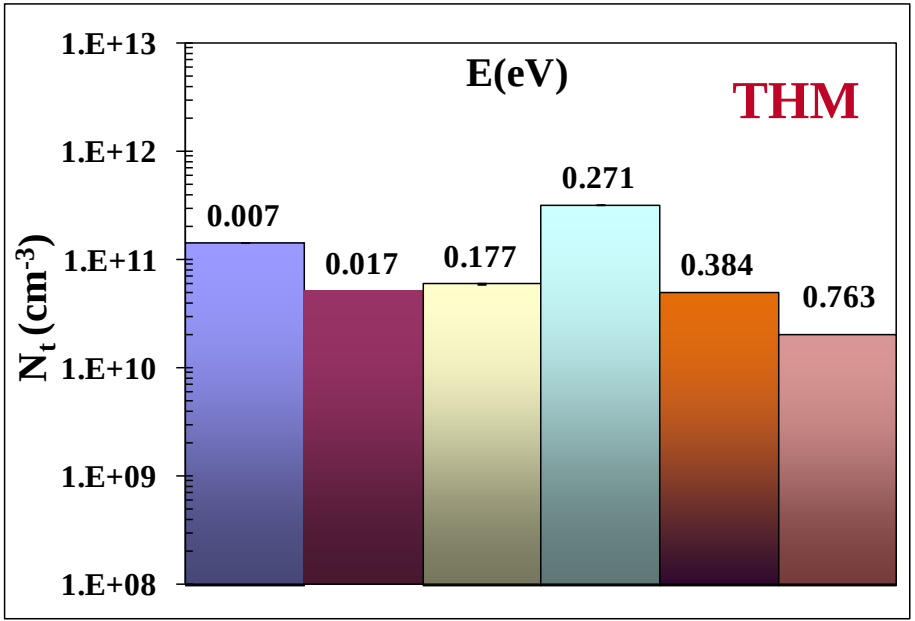


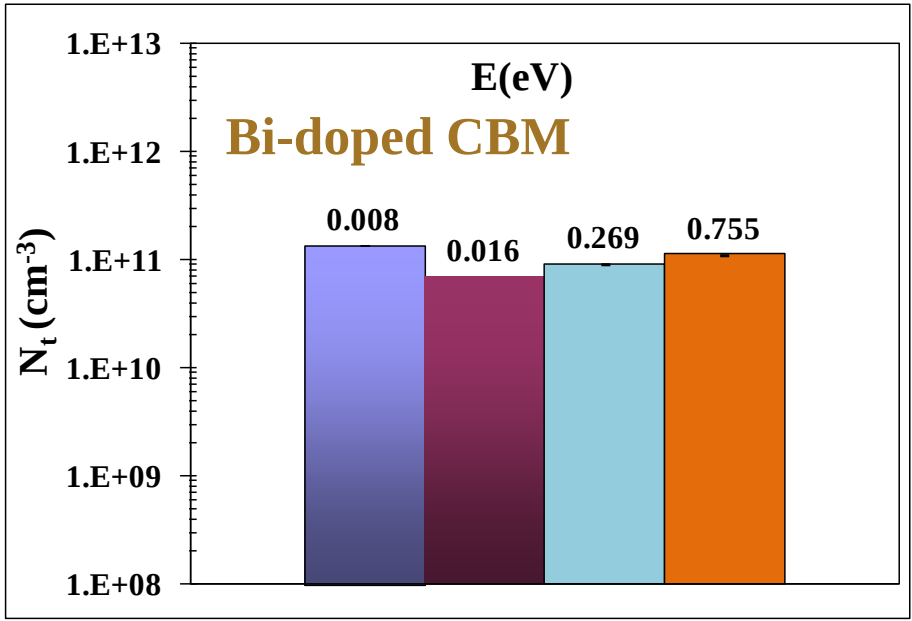


Concentrations of Electrically Active Traps Measured

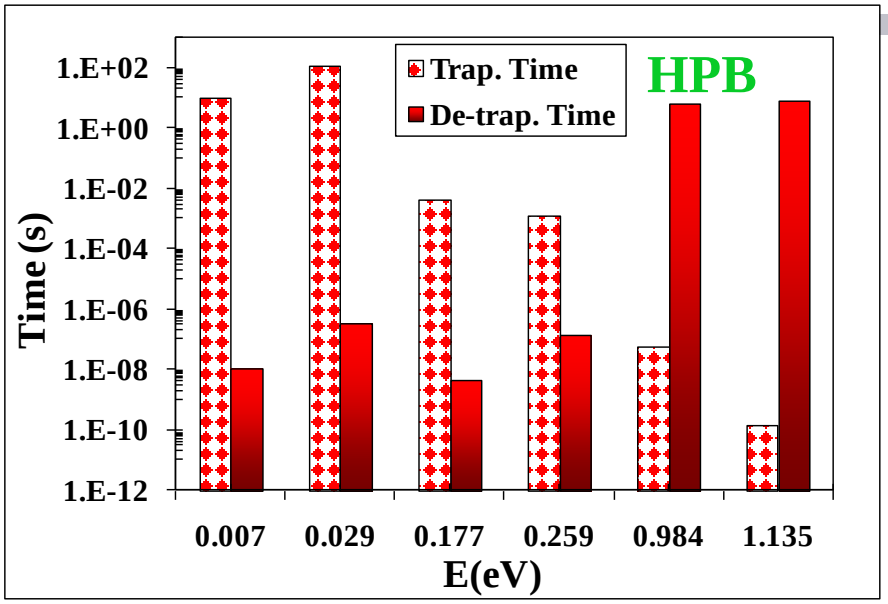


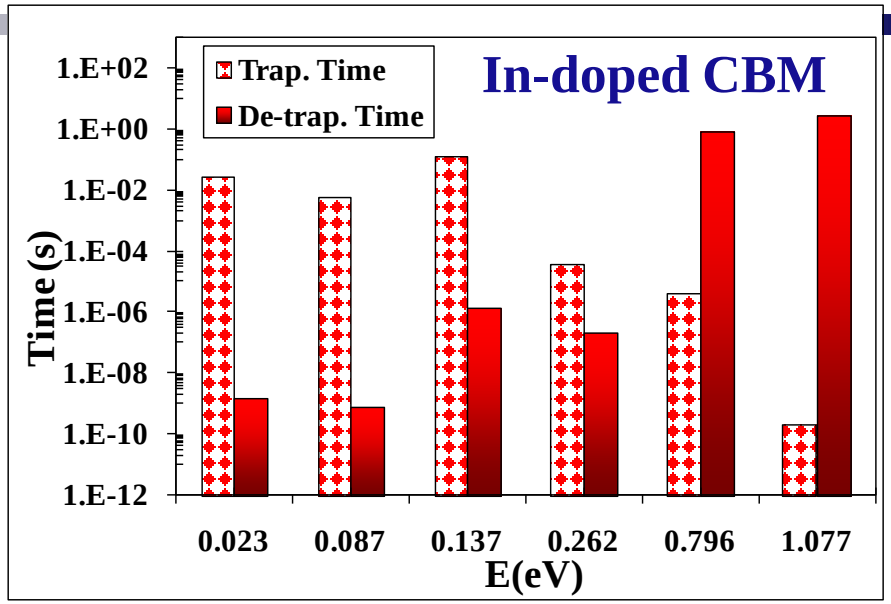


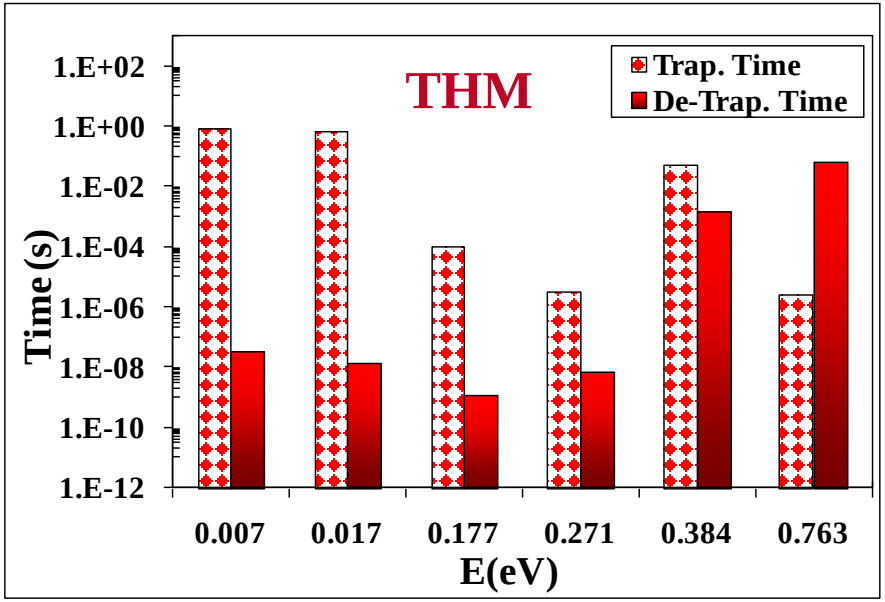


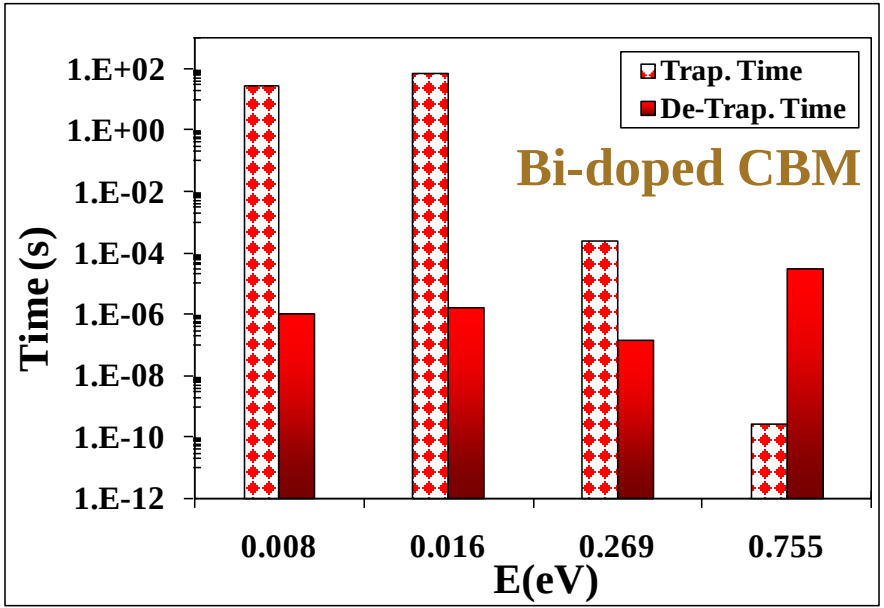


Trapping and De-trapping Times Measured at 295K









Summary of Accomplishments

- Impact of CZT detector technology to nonproliferation is severely limited by the availability of suitable large-volume detector blanks.
- The first-order manufacturing problem is the presence of multiple crystals in ingots: *Great progress on many fronts!*
- For single crystals the primary problem is Te inclusions causing random fluctuations in the electron charge collection. *Project accomplishments have changed investigations around the world!*
- Unique mapping tools allow examination of the effects of dislocations
- Trapping at point defects is simpler than once thought: *Only 3 important traps!*
- Significant gains in understanding the material showstoppers, providing new insights into proposed approaches, and making corrections to the current processes. *Approach is working!*
- Technical details on accomplishments reported in *31 peer-reviewed publications* over the past year.

Future Work

- Continue to relate growth methods to performance, provide feedback on material and device characterizations, and optimize processes
- Systematic experimentation to determine the cause of extended and point defects affecting performance
- Reduce large inclusions by modifying growth conditions (***rotation***, magnetic stirring, ***inverted growth***, superheating the melt, etc.)
- Develop effective post-growth thermal annealing process to remove Te secondary phases while minimizing formation of new defects
- Determine role of trapping by dislocations (and inclusions) on perturbing the internal electric field and charge collection efficiency
- Resolve the problem of point defects and increase the mu-tau products for electrons *and* holes

With crystal growth improvements, CZT will become a disruptive detector technology, replacing traditional materials used for detection and imaging of gamma radiation in many field applications.

Acknowledgement

Thanks to NA22 for financial support!