Recent developments in compound semiconductor radiation imaging detectors

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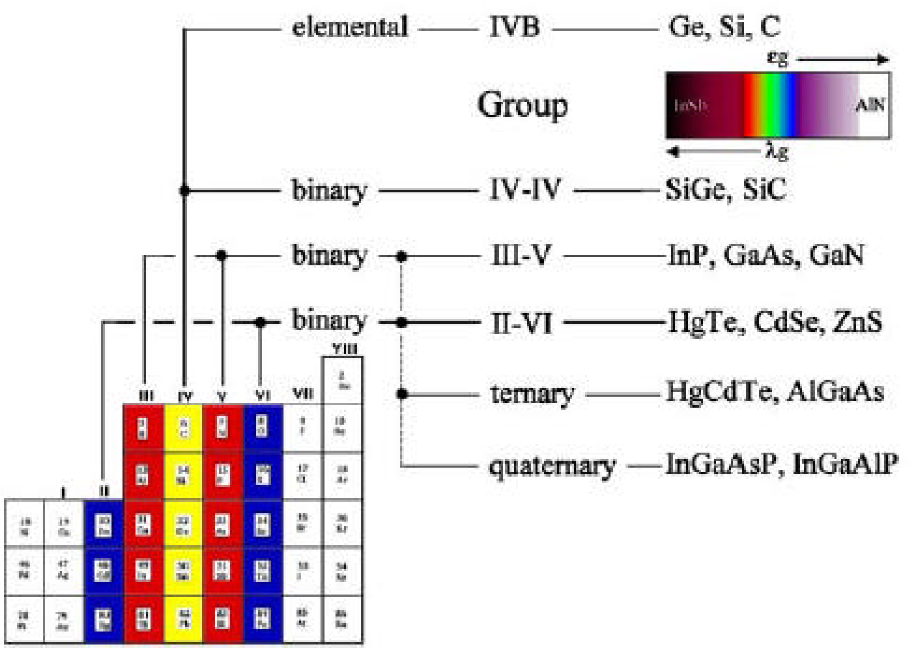
Introduction

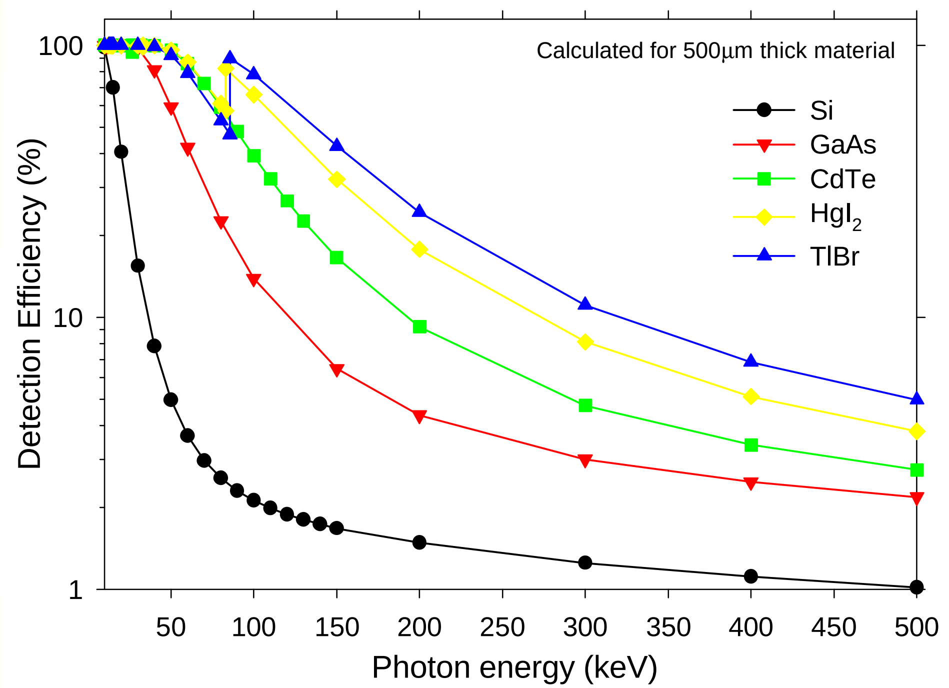
A review of recent developments in semiconductor detector materials and technology for X-ray and gamma imaging:

- Commercially available or near-market materials:
  - status of CdZnTe/CdTe
  - summary of best spectroscopic results from other materials
  - INTEGRAL/SWIFT – imaging detectors in space
- New developments in large-area thick film materials:
  - polycrystalline and epitaxial CdZnTe/CdTe thick films
  - Heavy element (Z≥80) thick films (Hg, Tl, Pb, Bi)
- Future materials – latest results from promising candidate materials:
  - CdMnTe
  - GaN
  - Synthetic diamond
- Conclusion

Commercially available or near-market materials

Commercially available material continues to be predominately CdZnTe, plus CdTe and GaAs.



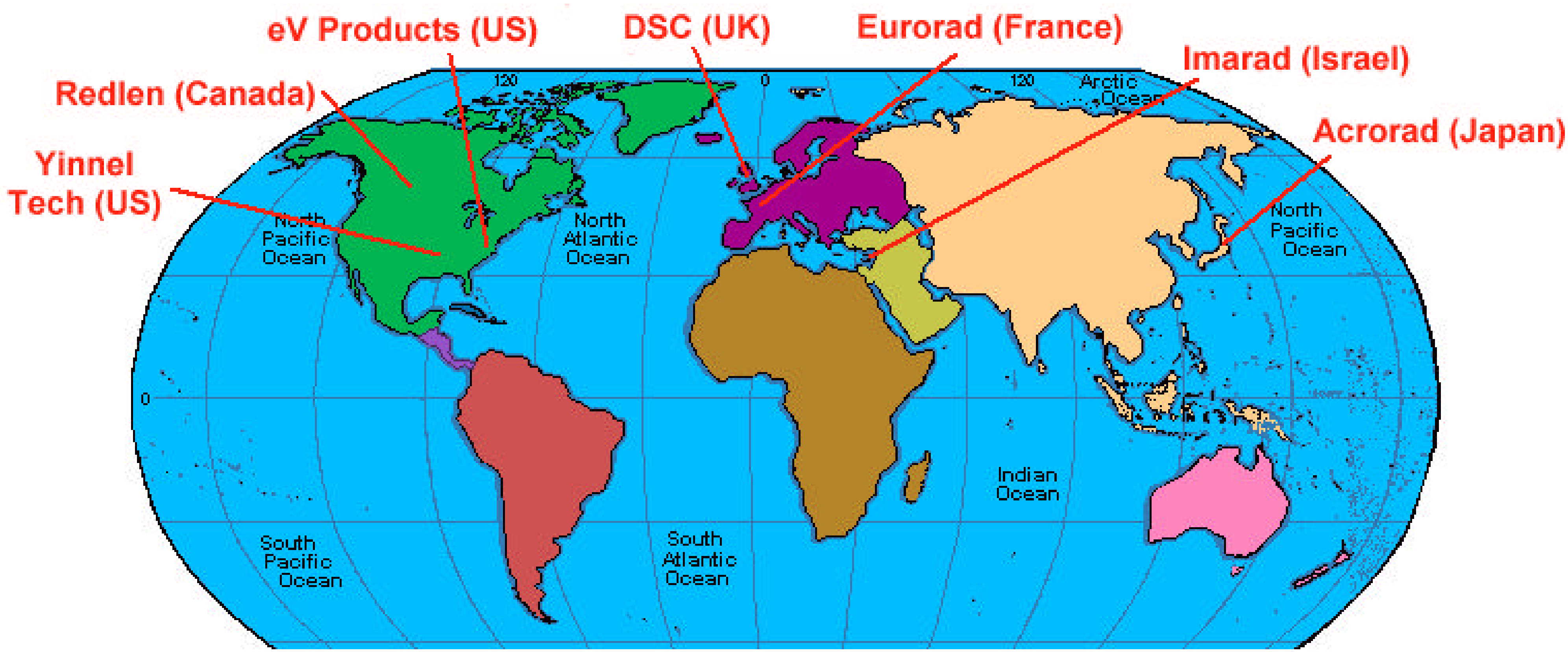


- II-VI materials CdTe and CdZnTe cover a suitable range of band gaps:
  1.44 eV (CdTe), 1.57 eV (CdZnTe, 10% Zn), 1.64 eV (CdZnTe, 20% Zn)
- Resistivity of CdZnTe is higher than CdTe ⇒ lower dark current, higher spectroscopic resolution
- Poor hole transport requires electron-sensitive detector geometries

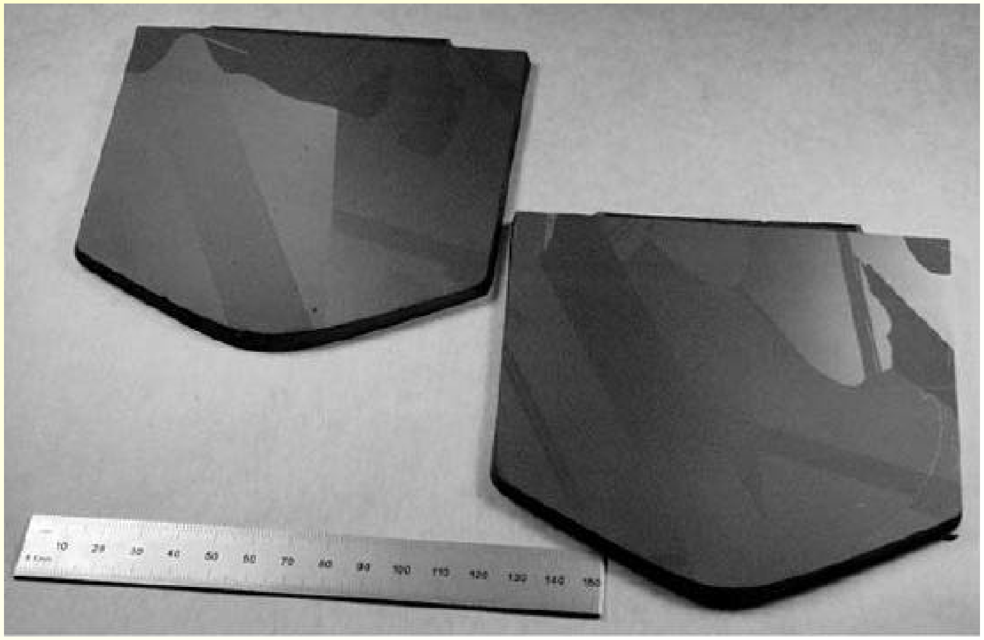
Commercial suppliers of CdTe/CdZnTe

eV Products continues to be the lead supplier of CdZnTe, grown using various Bridgman techniques:

- High Pressure Bridgman (HPB): 1992
- High Pressure Gradient Freeze (HPGF): 1998
- High Pressure Electro-Dynamic Gradient (HP-EDG): 2000
  - Electronic heating control, stationary crucible/heater
  - Reduced thermal stress, less cracking, better single crystal material



CdZnTe ingots grown by HP-EDG

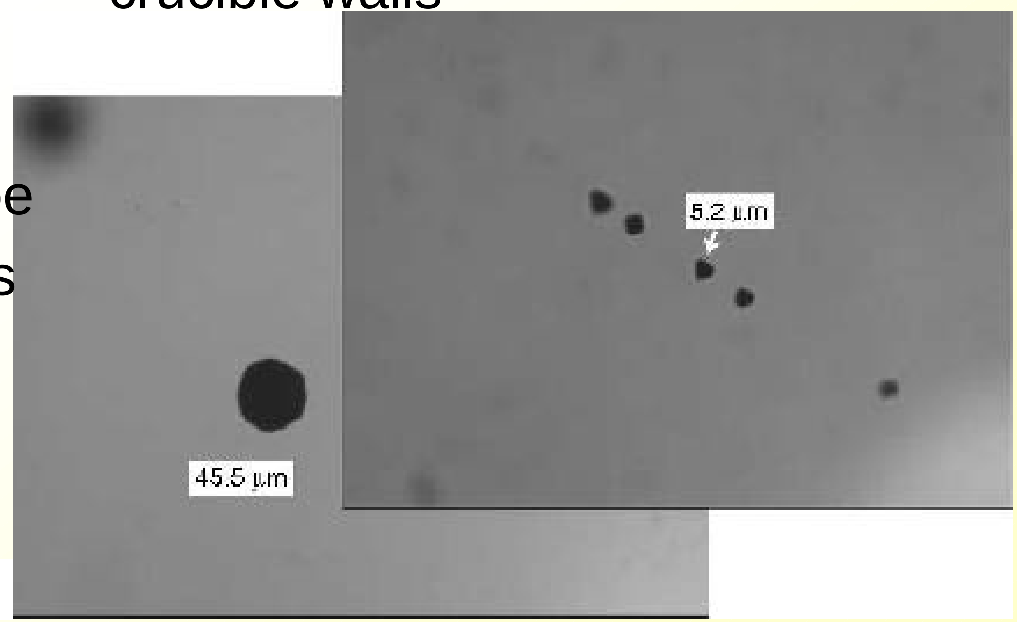


Latest published results from eV Products show 10kg crystals, 140mm (5.5 inch) diameter:

- No cracking
- Large-grain polycrystalline, with improved single-crystal yield
- Reduced concentration of twins
- Secondary grain nucleation on crucible walls

IR microscopy used to assess Te inclusions, formed from Te-rich melt:

- Mainly triangular or polyhedron shape
- Often located along grain boundaries and
- Te inclusions act as trapping sites, over a large range

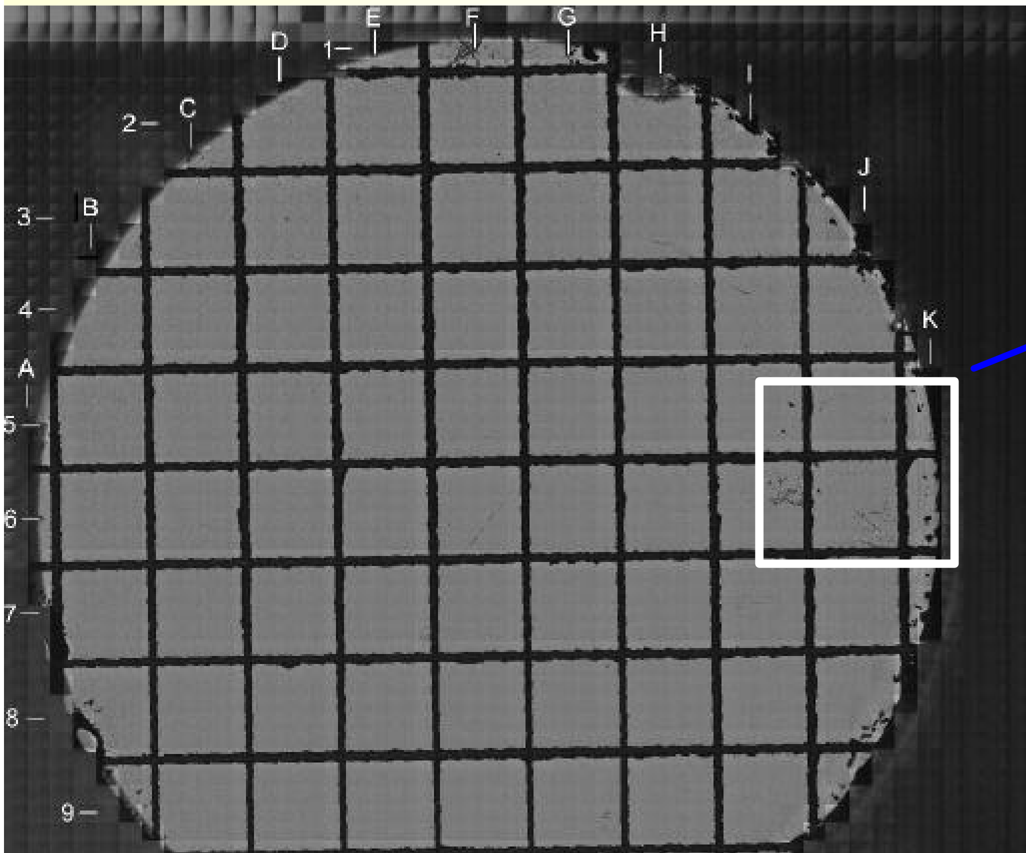


C. Szeles et al, J. Electronic Materials, 33 (2004) 742-751

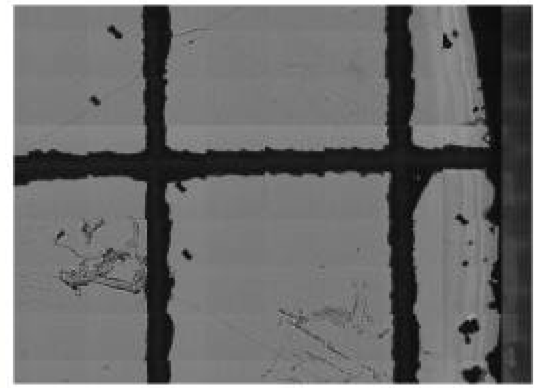
Tellurium inclusions in CdTe

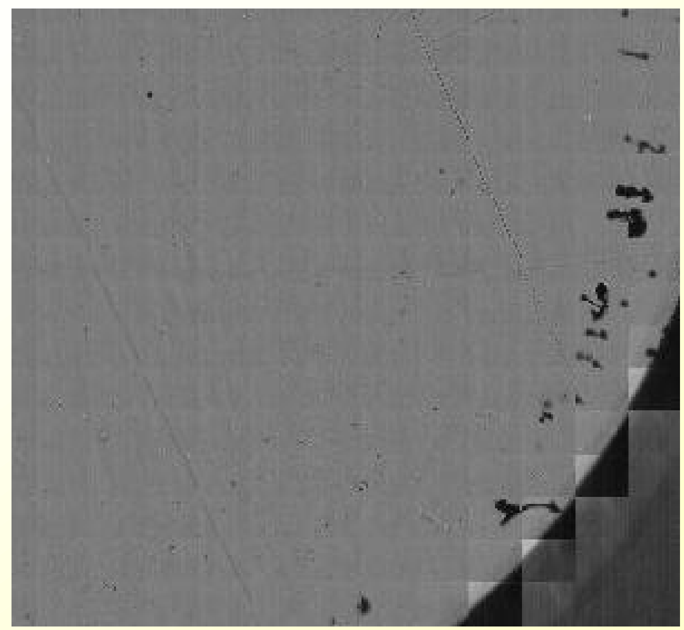
CdTe has also been studied using IR microscopy – presence of Te inclusions causes:

- Non uniform signal response
- Decreased electron drift length, low mobility



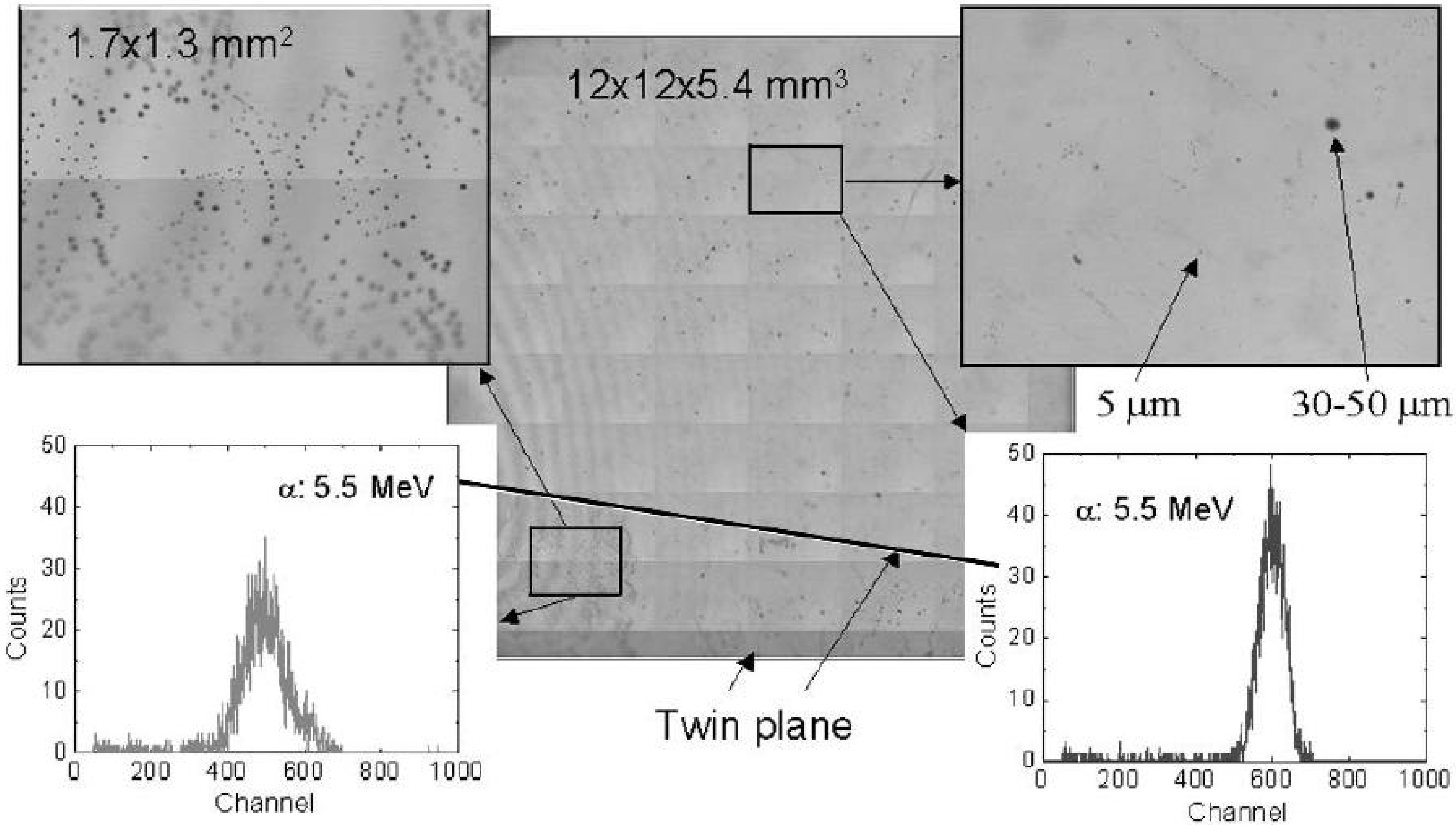
25mm diameter CdTe wafer, scribed with locating grid lines prior to metal deposition





A. Davies, P.J. Sellin et al, IEEE Trans Nucl Sci, in press

Te inclusions in HP-EDG CdZnTe



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C. Szeles et al, J. Electronic Materials, 33 (2004) 742-751

Charge transport performance in CdZnTe

Carrier drift length λ defines the induced charge Q, and hence the spectroscopic performance of the detector:

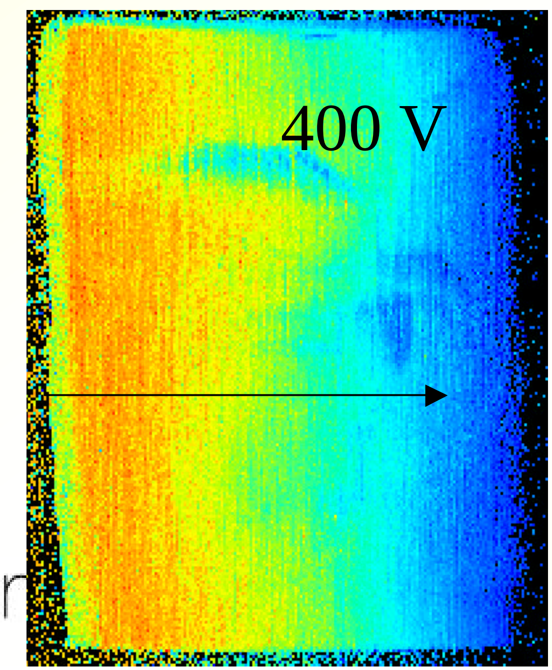
For electrons: $$CCE = \frac{Q}{Q_0} \approx \frac{l_e}{d}\left(1 - \exp\left(\frac{-d}{l_e}\right)\right)$$

HP-EDG material gives $\mu\tau_e$ ~$5\text{x}10^{-3}$ cm^2/V – some of the best values available

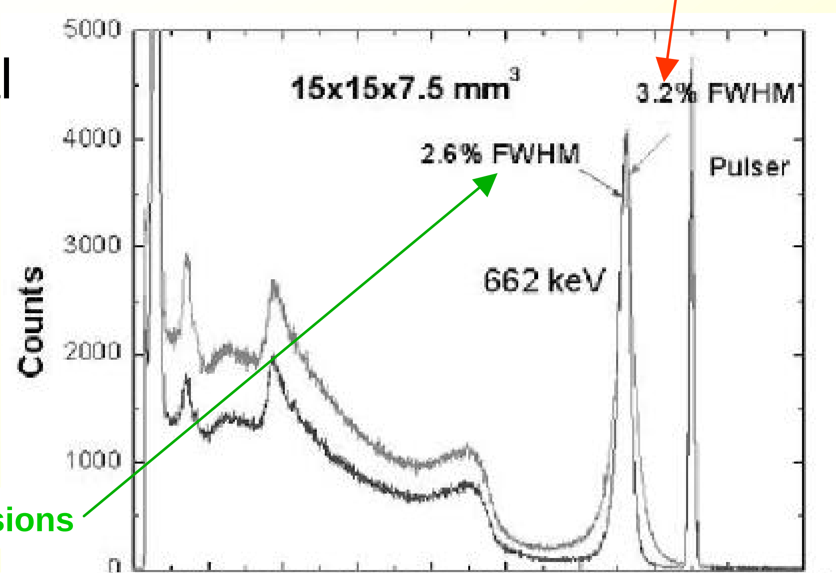
The mobility-lifetime product μτ is often used as a measure of charge transport quality: $l_e = \mu\tau E$

- μτ mapping using focussed MeV proton beams demonstrates depth-dependent gain

cathode

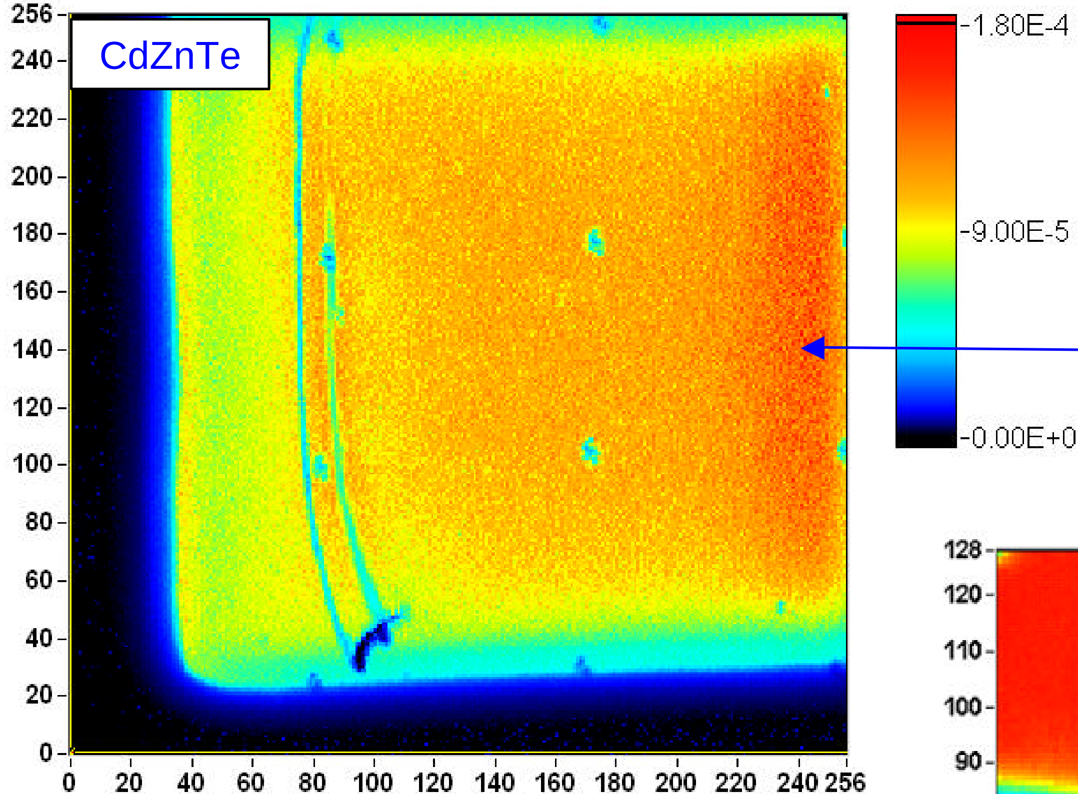


- HP-EDG material shows high non-uniformity of response due to Te inclusion density



A. Lohstroh, P.J. Sellin et al, J Phys Condens Matter 16 (2004) S67-S73
C. Szeles et al, J. Electronic Materials, 33 (2004) 742-751

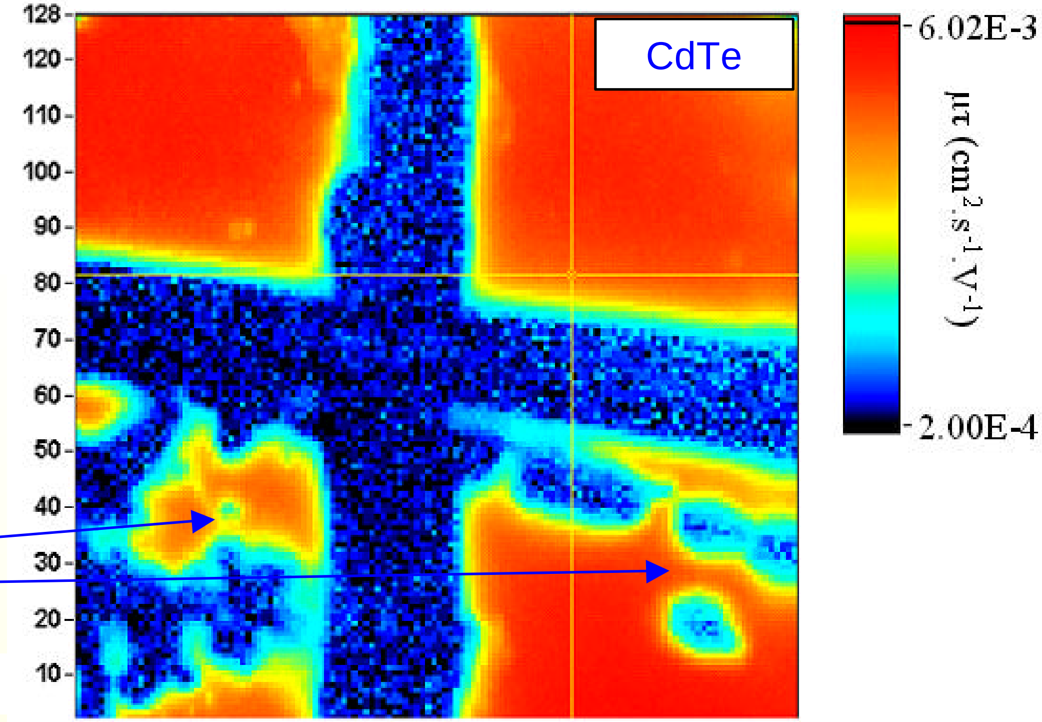
Ion beam mt maps of CdZnTe and CdTe



Map of electron $\mu\tau$ in CdZnTe shows $\mu\tau_e \sim 1x10^{-4}$ cm^2/V

Highly uniform, no evidence of defects in ‘single crystal’ material

Increased $\mu\tau$ at right edge due to beam scanning



CdTe electron $\mu\tau$ map shows $\mu\tau_e \sim 5x10^{-3}$ cm^2/V

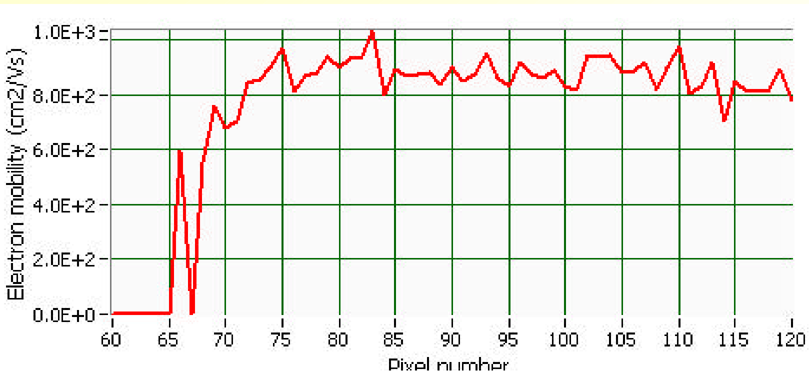
Pixel detector shows problems with contact delamination in lower quadrants

A. Davies, P.J. Sellin et al, IEEE Trans Nucl Sci, in press

Amplitude and drift time maps in CdTe

Zoom into corner of CdTe sample – uniform region, $\tau_e \sim 5.0\ \mu s$

V = 150V, d = 8mm,
Þ m_e ~ 850 cm^2/Vs



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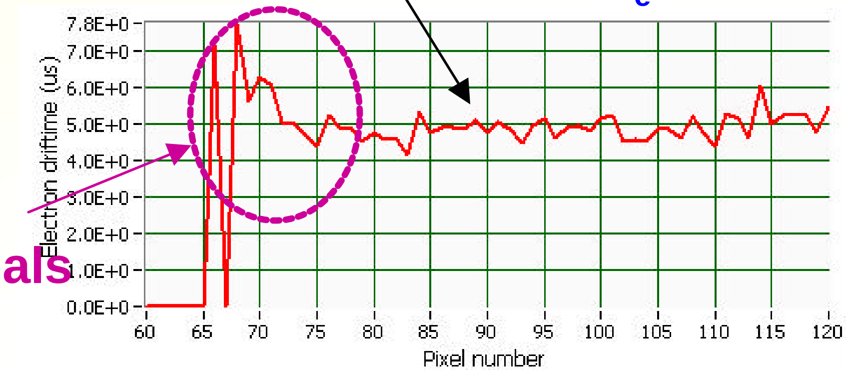
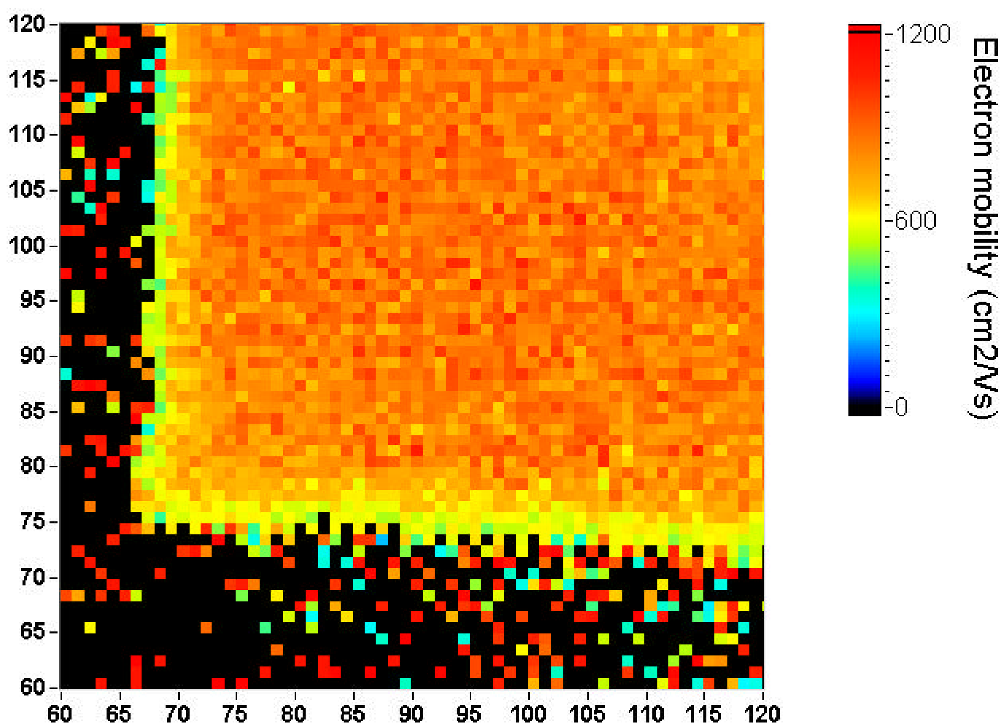
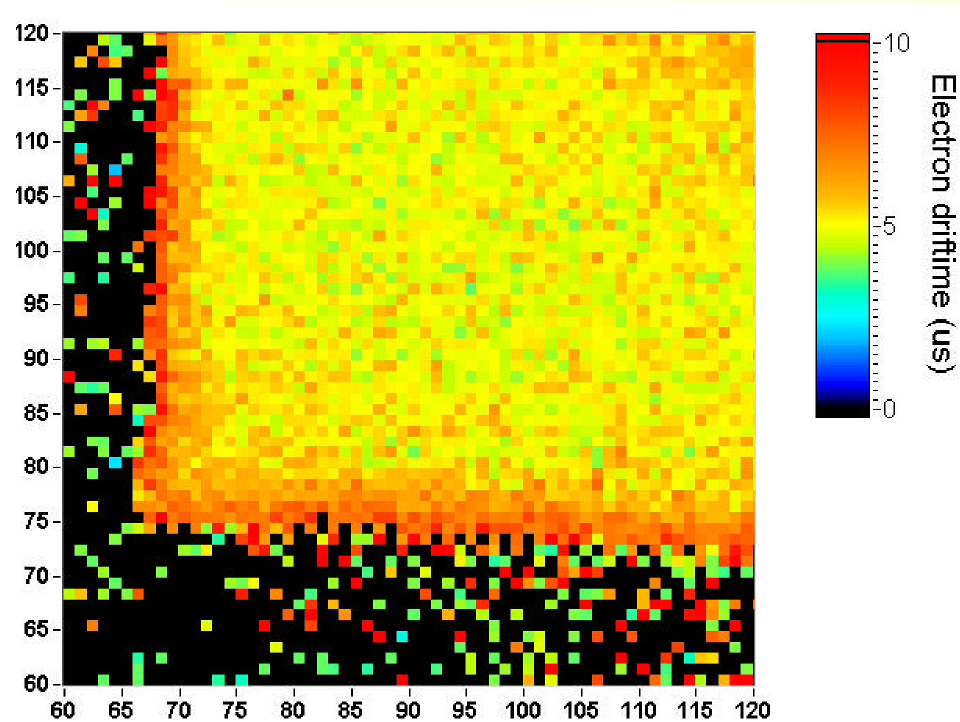


Electron mobility



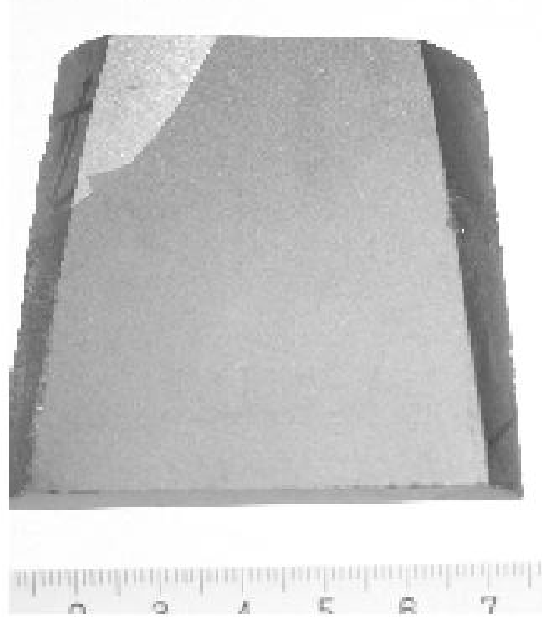
Electron drift time



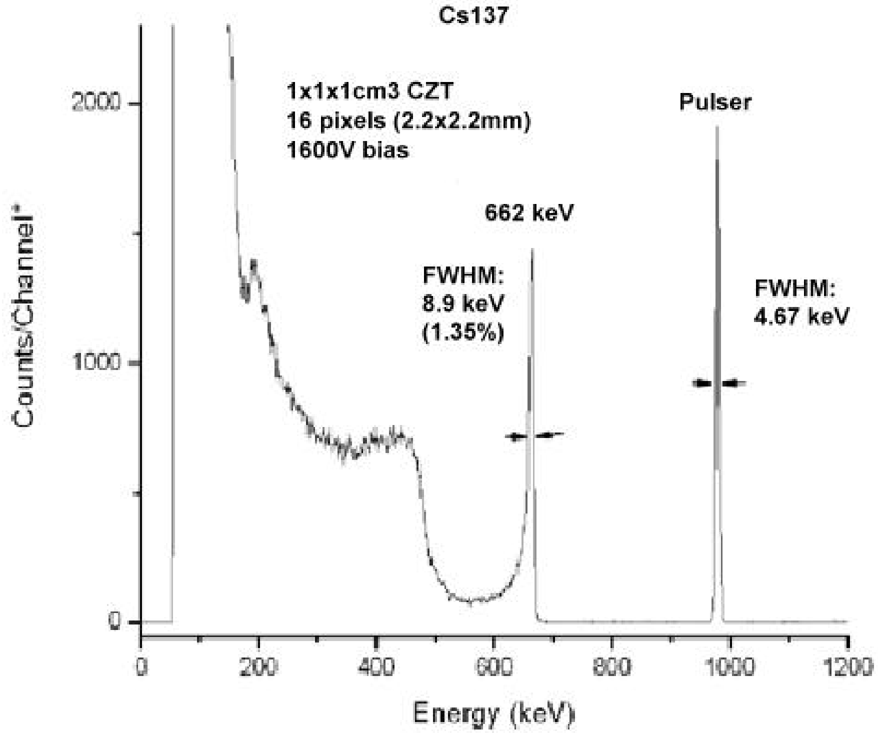
CZT grown by Modified Vertical Bridgman – Yinnel Tech

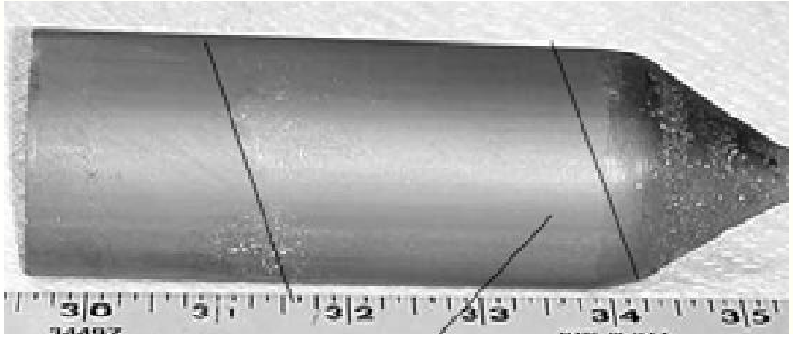
Modified Vertical Bridgman (MVD) CZT has been produced by Yinnel Tech

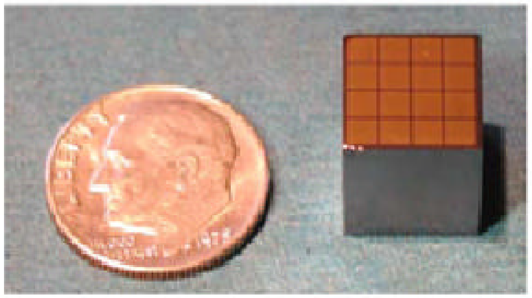
- wafers of large single-crystal areas are claimed, with excellent charge transport
- High resistivity $\rho=3\times10^{11}\Omega$cm, and $\mu\tau_e=1.8\times10^{-2}$ cm^2/V



4x4 pixellated devices have shown very good resolution
$\Rightarrow$ 1.35% FWHM at 662 keV

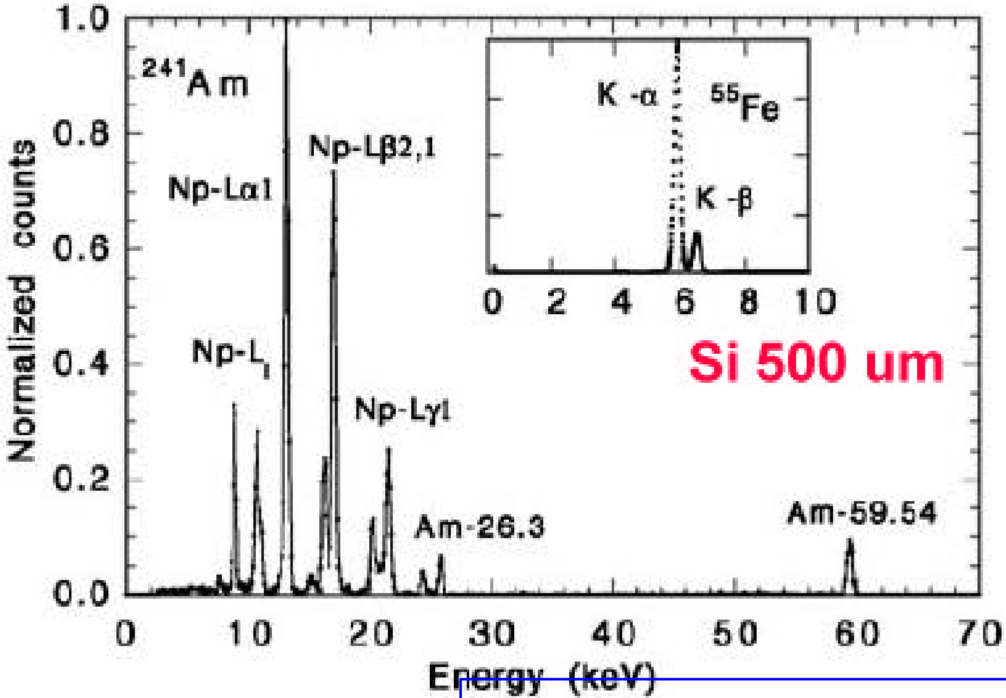


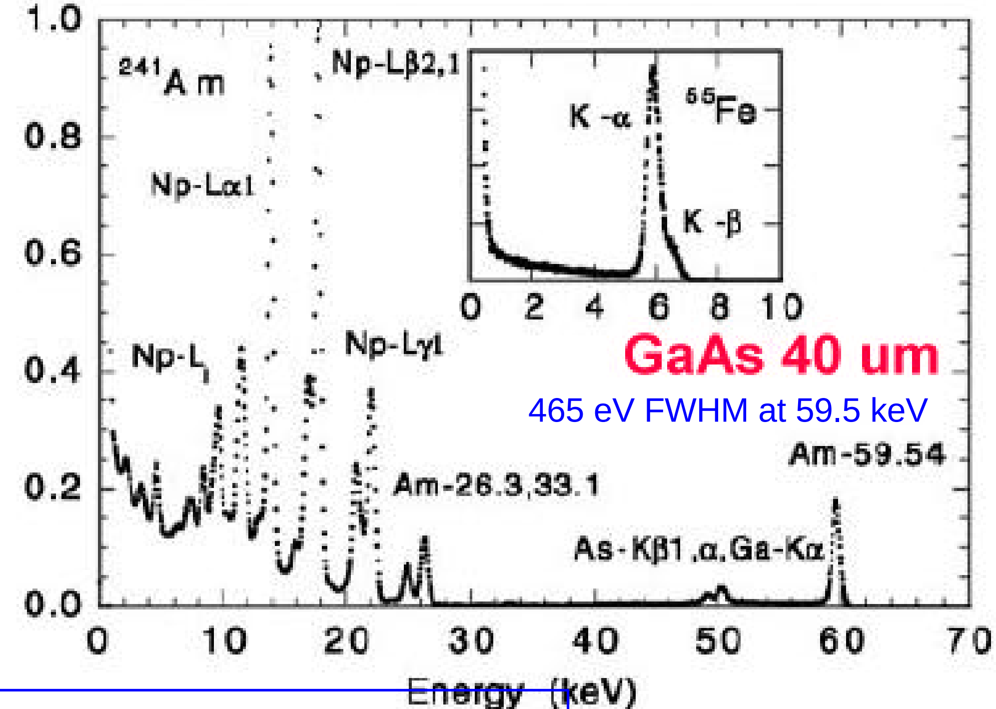


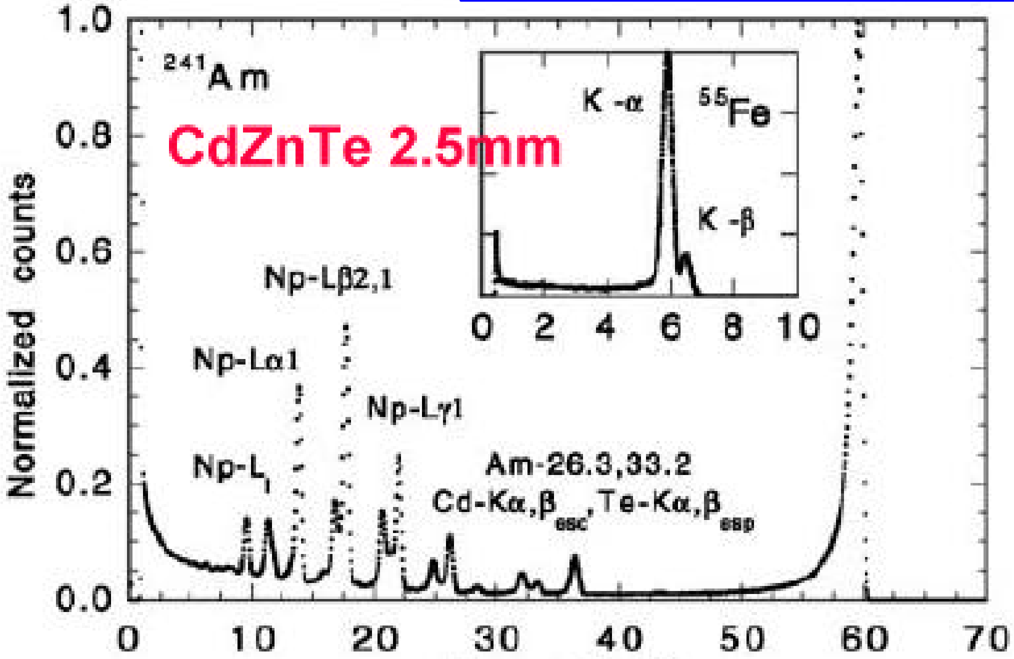


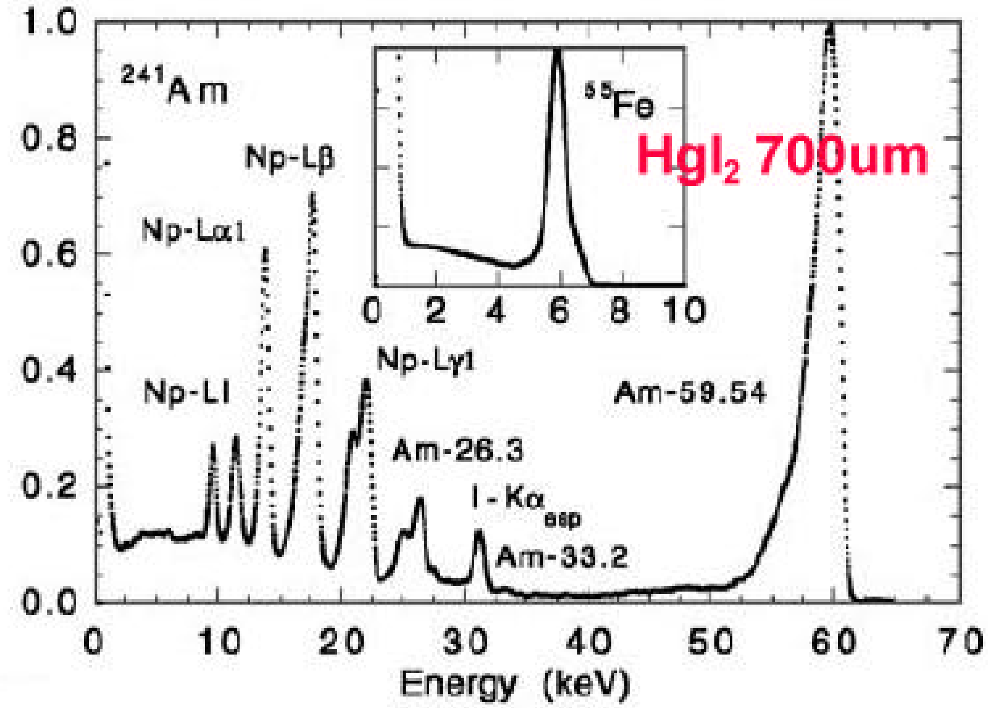
L. Li et al, Proc. Proc of IEEE Nuclear Science symposium, Rome 2004

Single element planar contact detectors









A. Owens et al, Proc. SPIE 4851 (2003) 1059

CdTe and CdZnTe in space: INTEGRAL and SWIFT

IBIS is the gamma ray imager on INTEGRAL:

- fine angular resolution imaging (12 arcmin FWHM),
- spectral sensitivity, wide energy range (15 keV - 10 MeV)
- 16384 elements of 4x4x2mm CdTe, plus 4096 CsI, covering 3100 cm^2

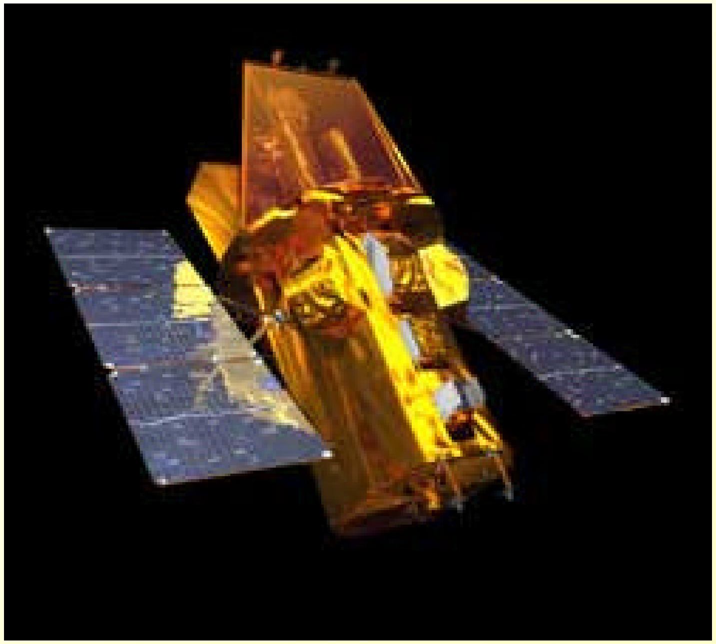
SWIFT Burst Alert Telescope (BAT) produces a first image within 10 seconds of the event trigger

- large imaging range (15-150 keV) using CZT, with additional response up to 500 keV
- 32768 elements of 4x4x2mm CZT, forming an array detector 1.2 x 0.6 m

INTEGRAL launched October 2002

See talk by F. Lebrun, Tuesday afternoon: "Semiconductor detectors for soft gamma-ray astrophysics"

SWIFT launched November 2004



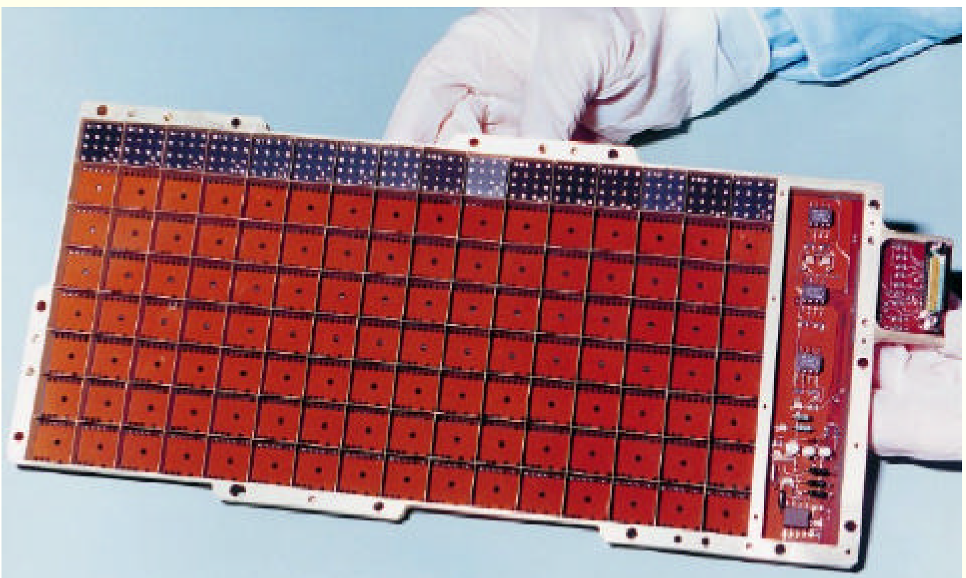
Imaging detector modules

INTEGRAL CdTe detector array:

2 parallel planes of pixels separated by 90 mm:

- ❒ top layer uses 16384 CdTe pixels, covering 2600 cm^2, each 4x4x2 mm ⇒ low energy gammas
- ❒ second layer uses 4096 CsI scintillators covering 3100 cm^2, each 9x9x30 mm

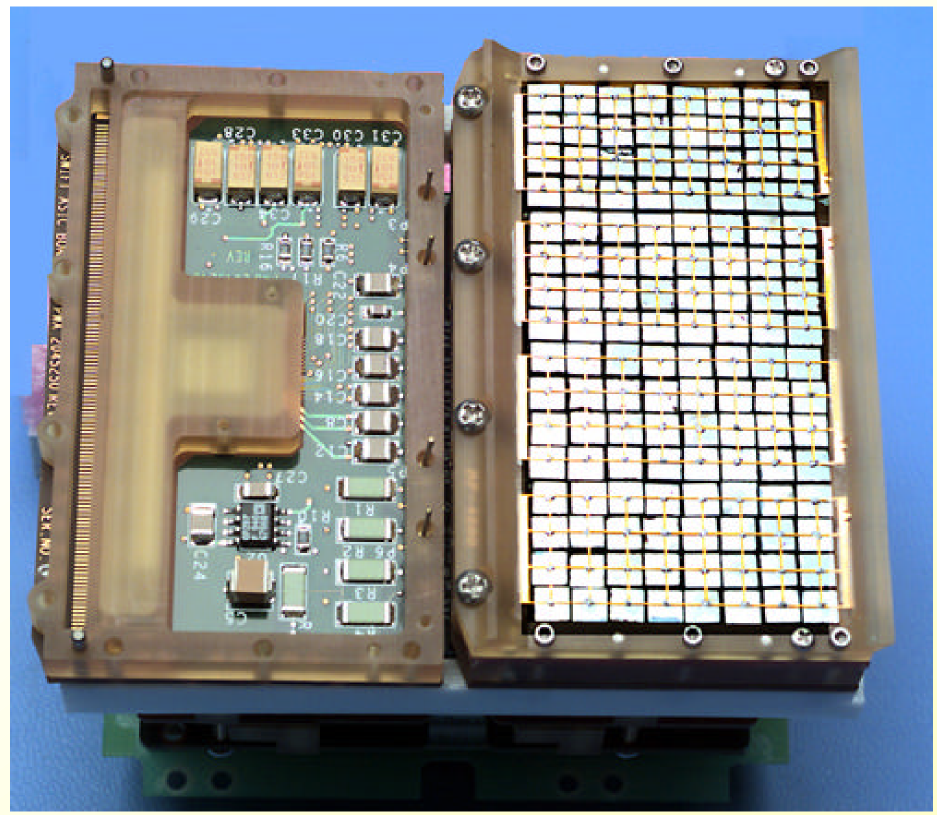
⇒ high-energy gamma rays.



SWIFT CZT detector array:

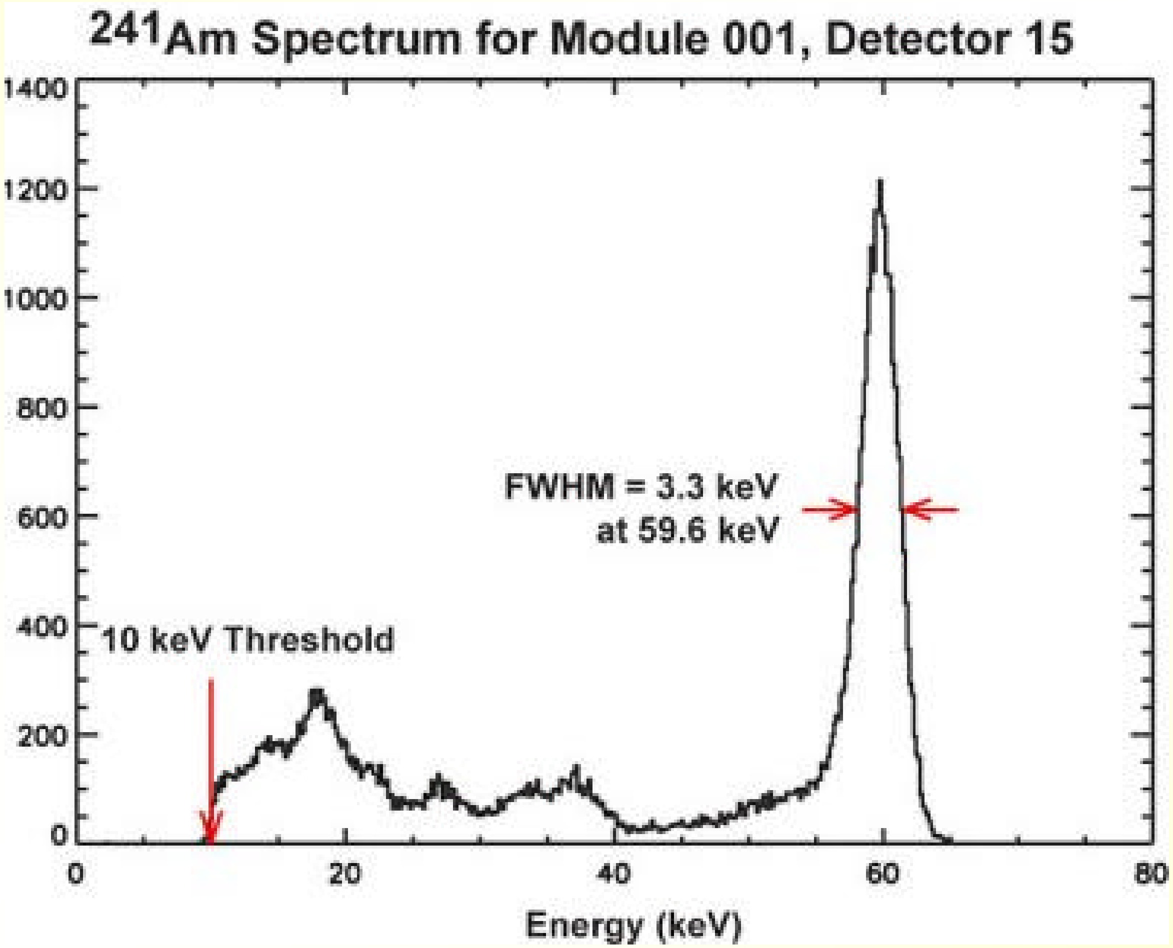
❒ Contains 32768 elements of 4x4x2mm CZT, forming an array detector 1.2 x 0.6 m

❒ The coded aperture mask is ~54,000 lead tiles!

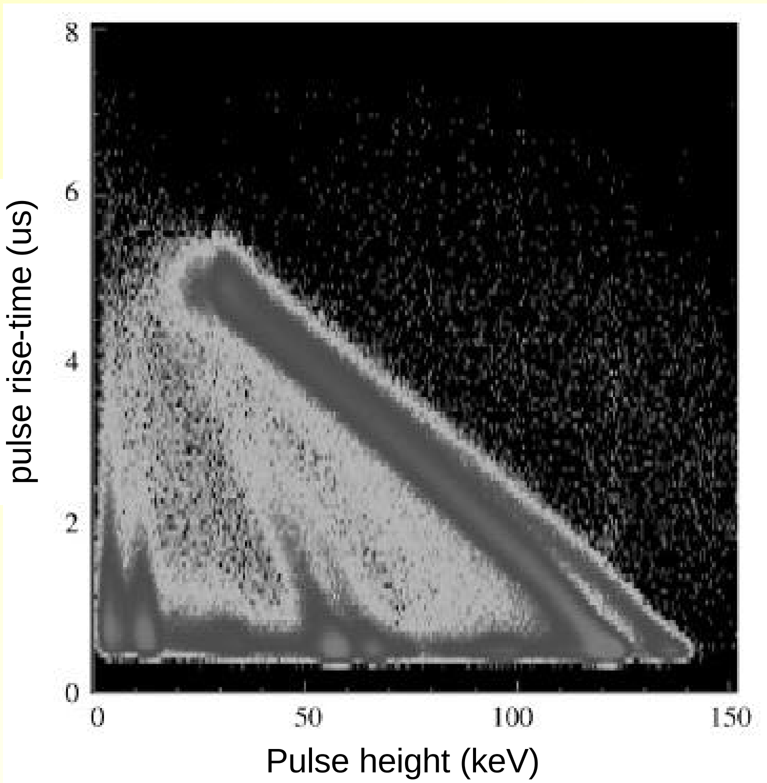


CZT detector performance

The typical performance of a single CZT module is 3.3 keV FWHM at 60 keV (5.5% FWHM):



The background event rate in the CZT array is ~10 kHz



INTEGRAL CdTe spectroscopy

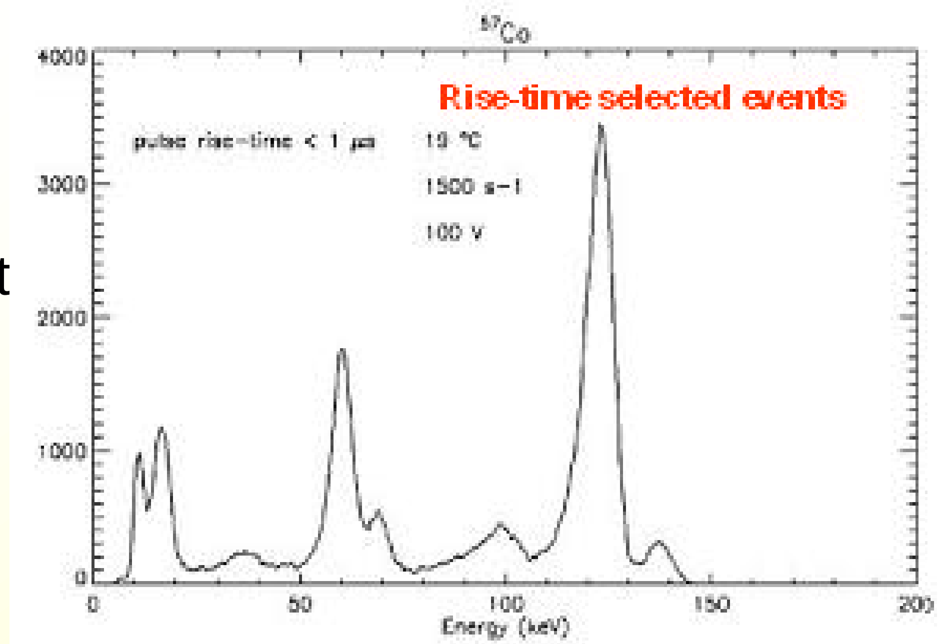
Pulse rise time correction applied to 2mm thick CdTe at 100V:

- uses simultaneous pulse rise time and amplitude measurements
- pulse drift time measures electron drift time to the anode, giving interaction depth
- correction for electron trapping improves total peak efficiency

O. Limousin et al, NIM A504 (2003) 24-37

Rise-time selected CdTe spectrum:

- In CdTe risetime selection is implemented on the ASIC to reject pulses with risetime >1 µs
- CdTe energy resolution is 9.2 keV FWHM at 122 keV (7.5% FWHM)



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Thick film material developments

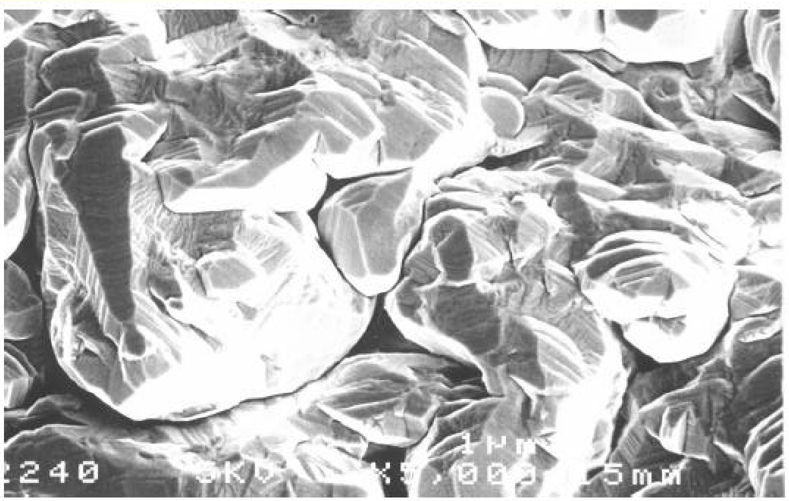
Growth of CdTe/CdZnTe as a large area thick-film has been developed extensively in Japan and Korea:

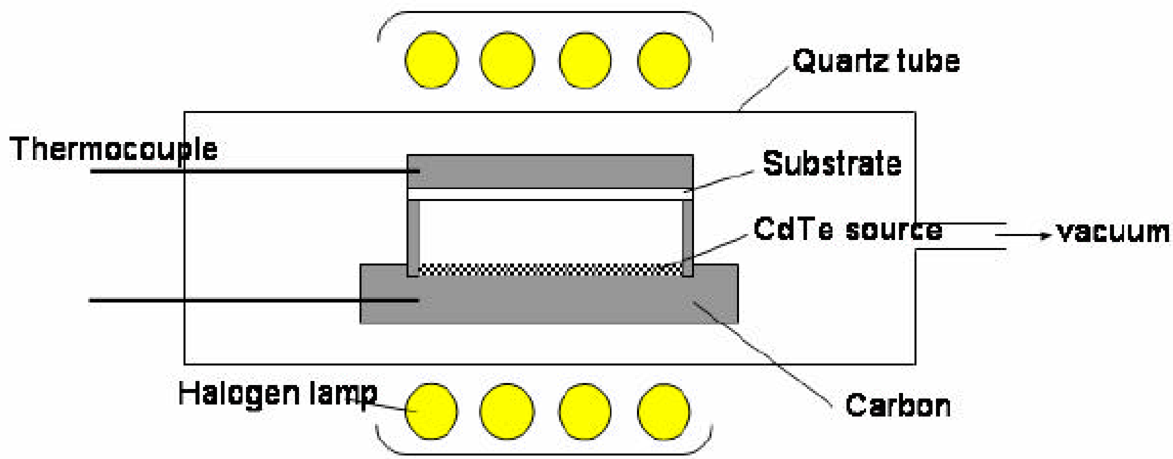
- Thick films deposited onto active substrates are required for medical imaging
  - avoids flip-chip bonding required for single-crystal wafers
- Polycrystalline films suffer from poor charge transport and low spectroscopic performance – grain boundaries act as:
  - sites for electron-hole recombination and trapping
  - charge scattering which reduces the mobility
  - local regions of disturbance to the applied electric field
- Recent results from:
  - Nagayoshi (Tokyo) - polycrystalline CdTe by Close Space Sublimation
  - Park (Seoul) – thermally evaporated polycrystalline CdZnTe
  - Tokuda (Shimadzu Co, Kyoto) – poly CdTe/CdZnTe by CSS
  - Niraula (Nagoya) – epitaxial CdTe films by Vapour Phase Epitaxy
- 59 keV photopeak observed from epitaxial CdTe material

Large area CdTe by Close-Space sublimation

Close space sublimation (CSS) is a growth method capable of large area thick-film growth – recently applied to CdTe layers >300μm thick

- CSS growth is one of the fastest thick-film techniques: 5-10 μm/min
- Grown on quartz or glass substrate – thick film peels off to provide a free-standing ‘wafer’ – temperature 440 – 550 °C
- Lightly p-type films with resistivity ~10^{10} Ωcm
- Typical crystallite sizes ~10 μm



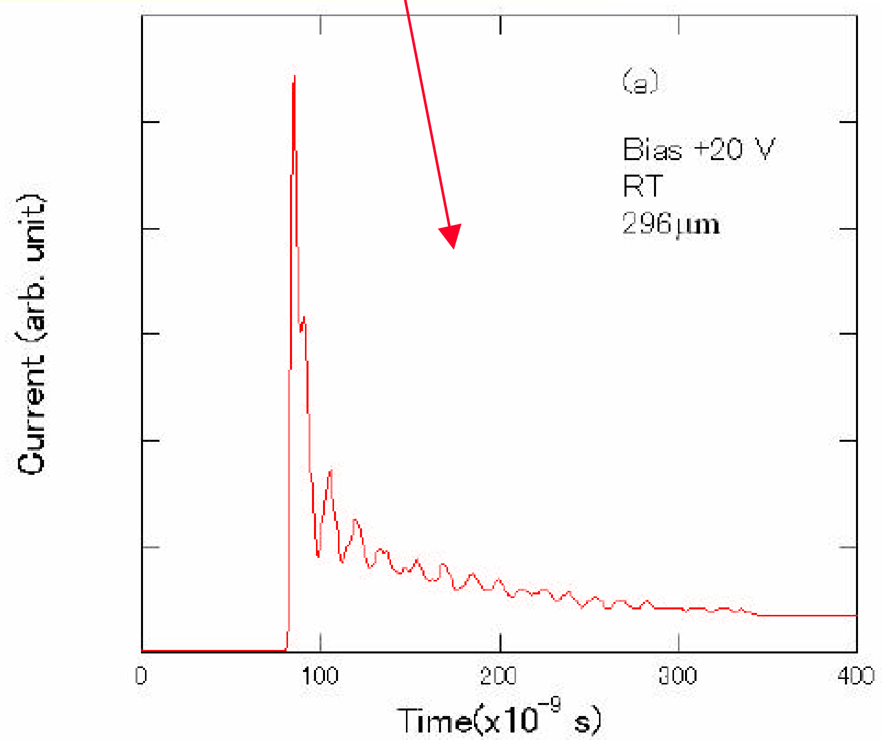


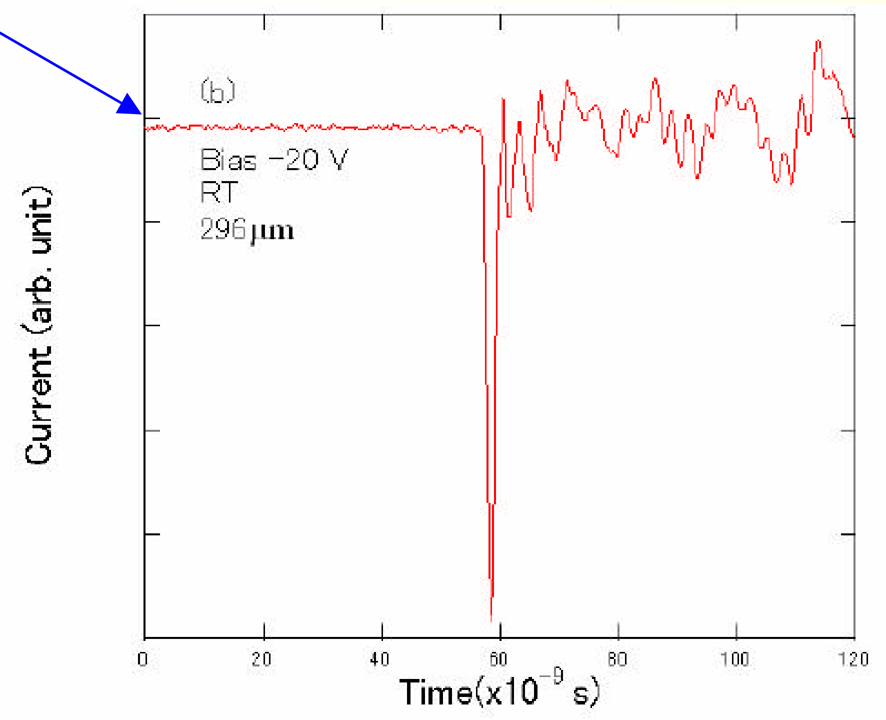
H. Nagayoshi, IEEE Trans Nucl Sci, in press

Photoresponse of CSS CdTe

Time of flight response using a 300ps nitrogen laser showed short carrier lifetimes:

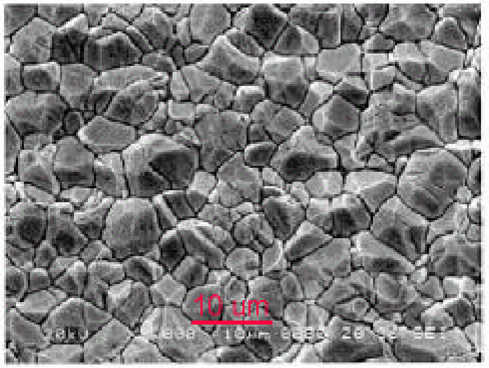
- For anode irradiation (hole transport) a prompt peak of <10ns is observed, followed by thermalisation: ⇒ shallow hole traps
- For cathode irradiation short lifetimes are also observed, with no thermal component: ⇒ deep electron traps
- No radiation response yet reported –$\mu\tau$ values still very small



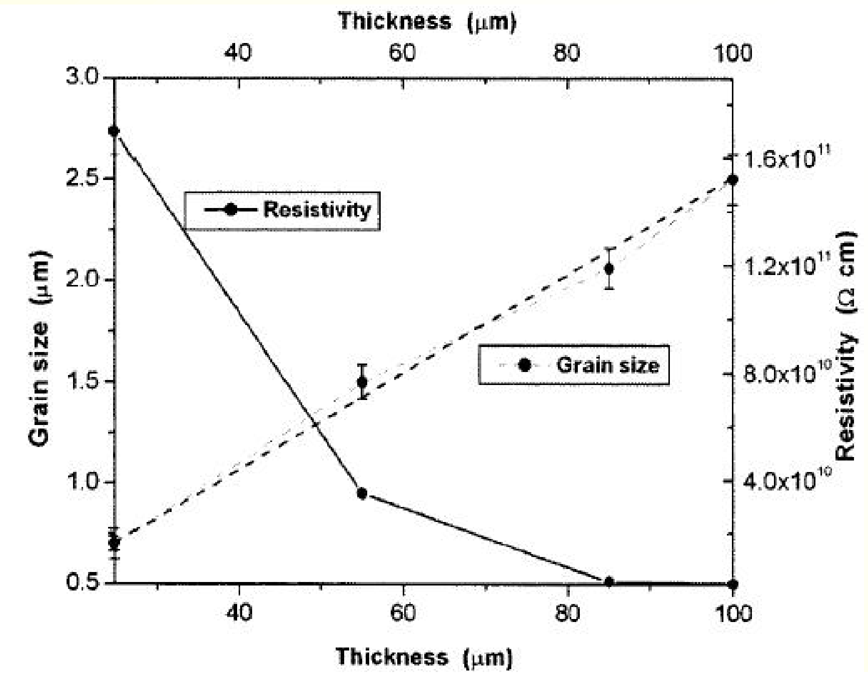


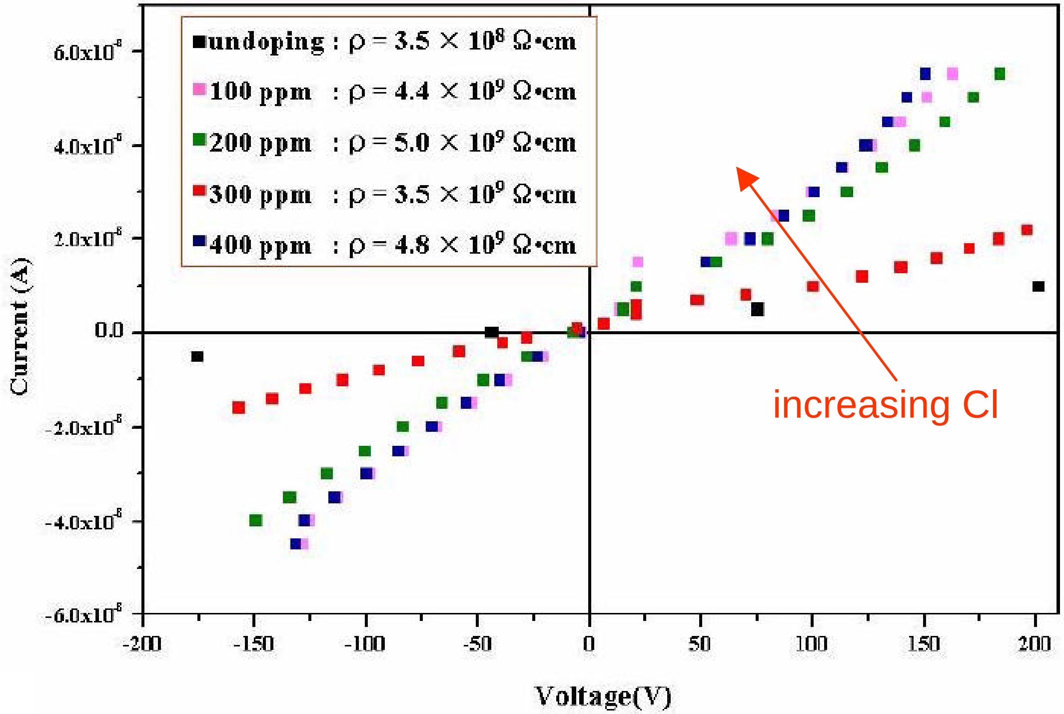
Polycrystalline CdZnTe grown by thermal evaporation

CdZnTe films have been grown by thermal evaporation, to 100 μm thickness:



- ❒ Using a source temperature of 700°C CdZnTe is evaporated onto an ITO/glass substrate at <200°C – compatible with TFT arrays
- ❒ Slower deposition rate than for CSS method, <2 μm/hr
- ❒ Grain sizes are ~2 μm, with $\rho = 10^9$ to 10^{11} Ωcm
- ❒ Chlorine doping used to increase resistivity for large grain sizes





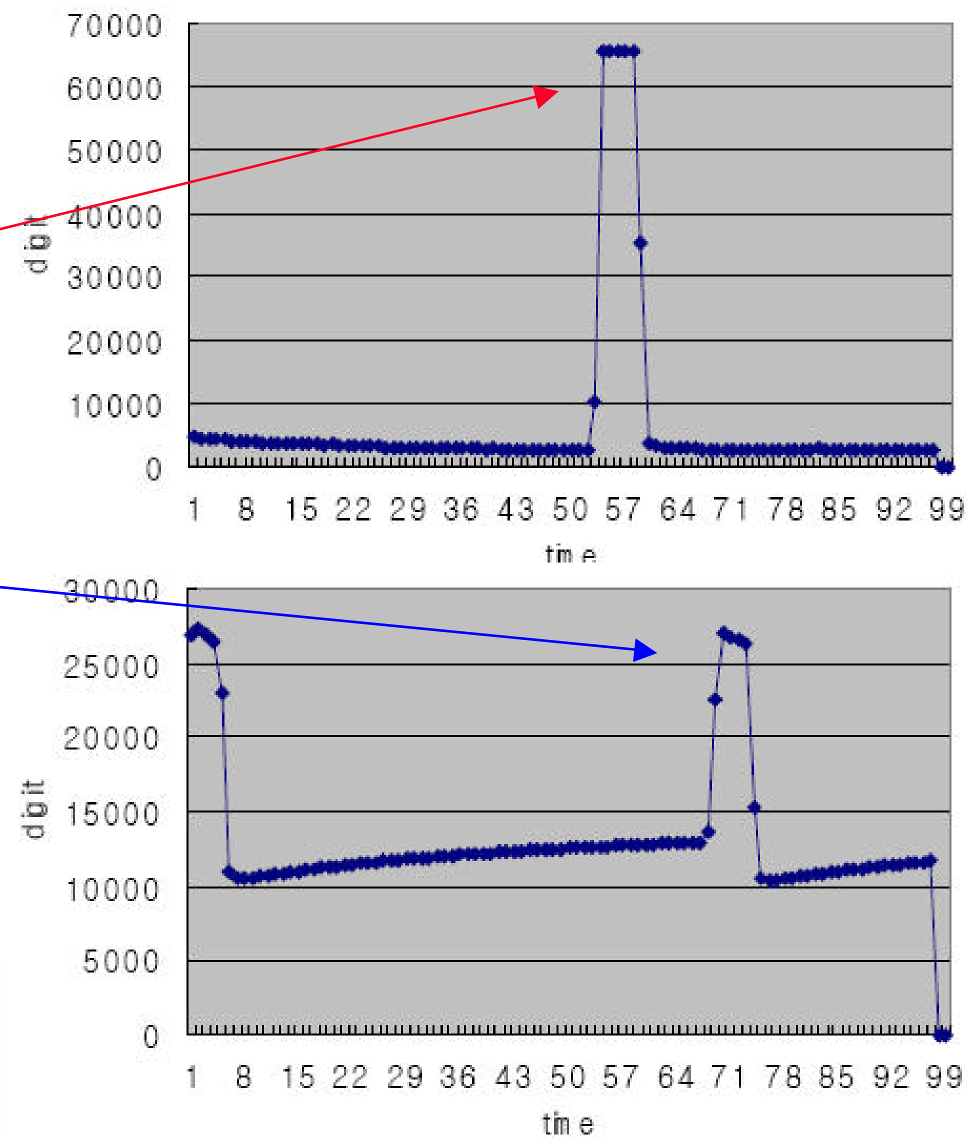
J.S. Kwon, Physica Status Solidii b 229 (2002) 1097-1101

X-ray response of polycrystalline CdZnTe

X-ray response of poly CdZnTe measured using a 65 kV_p X-ray tube at 7.5 mA

Measuring DC photocurrents:

- **Single crystal CdZnTe:** signal amplitude saturated at 65,536 adc units
- **Polycrystalline CdZnTe:** signal amplitude ~14,000 adc units
- Polycrystalline material showed significant dark current and response to ambient light
- Non-stable dark current suggests thermal de-trapping of deep levels
- No single-pulse sensitivity demonstrated yet

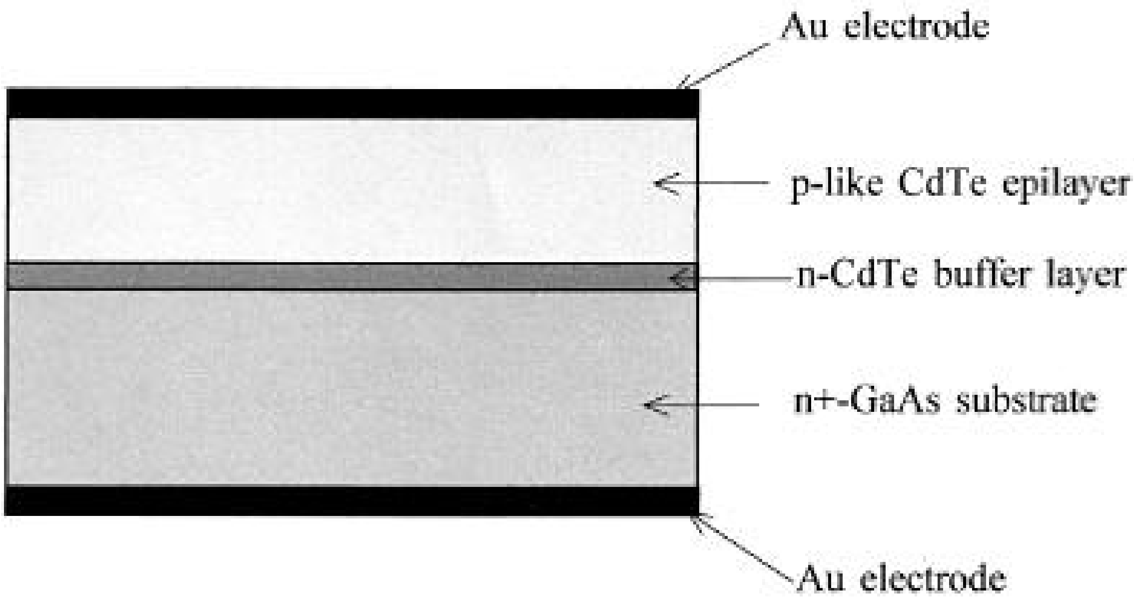


S.J. Park et al, IEEE Trans Nucl Sci, in press

Large-area epitaxial CdTe grown by MOVPE

Metal-organic vapor-phase epitaxy (MOVPE) is capable of growing large-area epitaxial thick films, eg. up to 200 μm thick

- MOVPE growth of CdTe or CdZnTe on GaAs or Si substrates, produces uniform mono-crystals
- GaAs substrates provide a good lattice match and strong adhesion



- Iodine-doped buffer layer grown onto substrate (10^{17} cm^{-3})

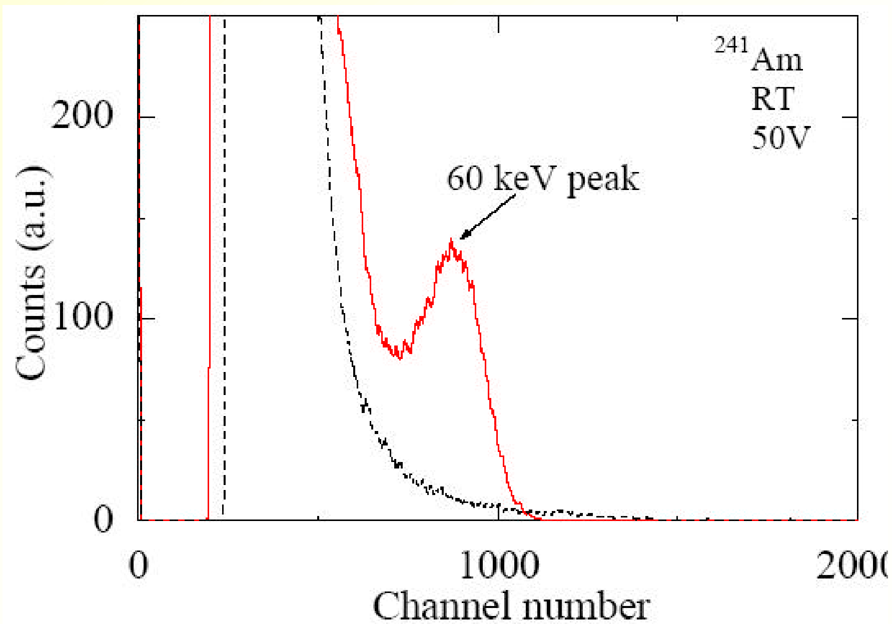
Prevents Ga diffusion into epitaxial CdTe layer

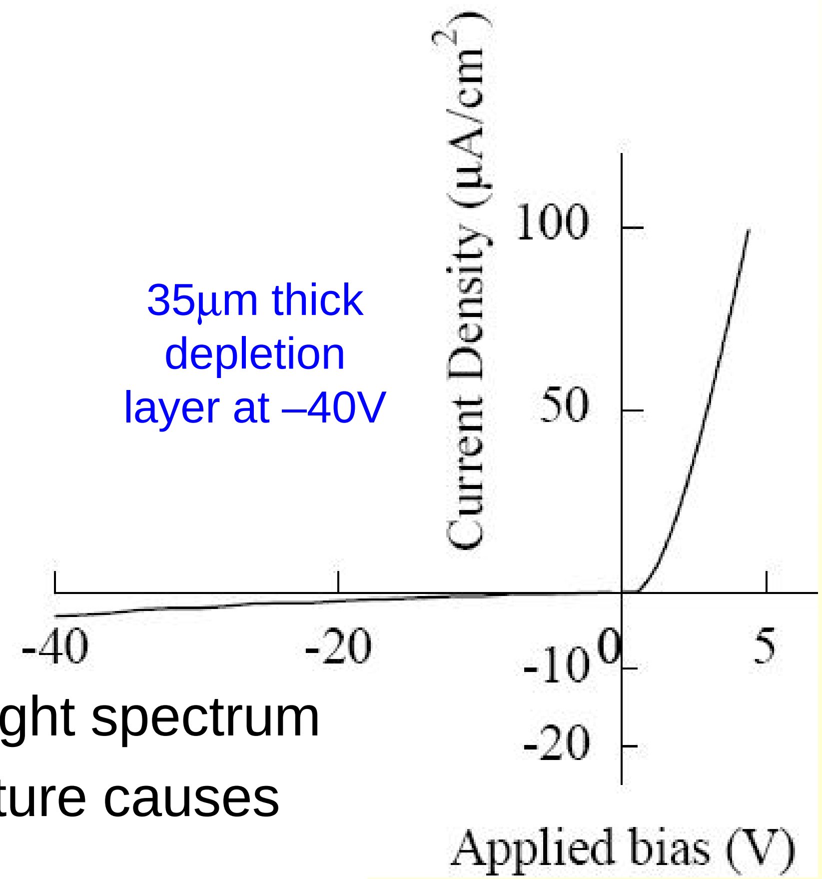
- undoped p-type epitaxial CdTe layer grown at 415-560 °C
- rectifying p-n junction formed at the CdTe/GaAs interface

M. Niraula et al, J. Elec Mat 34 (2005) 1-5

Dark current and spectroscopy performance

- 100µm thick CdTe layer
- IV shows good rectification, ⇒ reverse current ~3x10^{-6} A/cm^2
- CV measurements show carrier concentration of ~10^{14} cm^{-3}





- resolved 59 keV photopeak in pulse height spectrum
- Large leakage current at room temperature causes high noise level in the spectrum
- Adjustment of buffer layer thickness, and use of guard electrodes, required to reduce current

High-Z polycrystalline materials (Hg, Tl, Pb, Bi)

Polycrystalline thick film high-Z (Z≥80) materials have been extensively studied for X-ray imaging applications:

Material	Z	density	mobility	E_G	resistivity
Iodides:		g/cm^3	cm^2/Vs	eV	Ωcm
HgI_2	80/53	6.4	50	2.1	10^{13}
PbI_2	82/53	6.2	53	2.5	10^{12}
BiI_3	83/53	5.8	48	1.7	10^{12}
Bromides:					
TlBr	81/35	7.6	75	2.7	10^{12}
PbBr	82/35	-	-	2.5-3.1	-
Oxides:					
PbO	82/8	9.5	-	1.9	-

The iodide and bromide families have many suitable candidates:

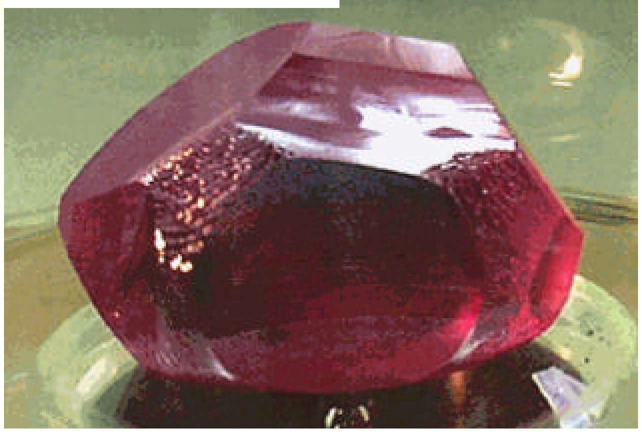
- Detailed studies of HgI_2 and PBI_2 have been carried out
- HgI_2 shows superior dark current and charge transport properties
- Promising results from TlBr, also as single crystal material

Polycrystalline Mercuric Iodide

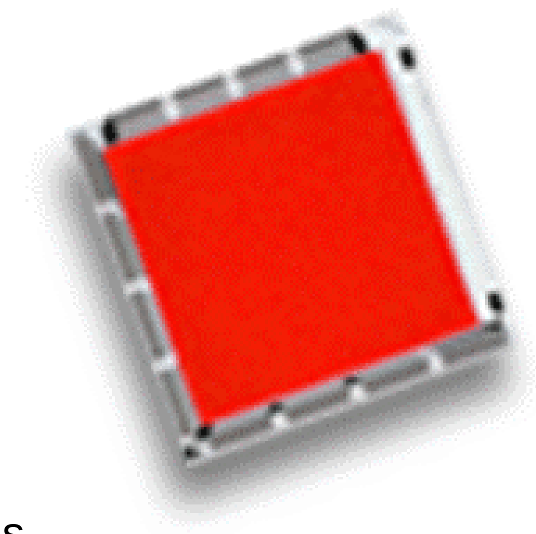
Polycrystalline HgI_2 is a material receiving new interest – provides a thick-film X-ray Photoconductor coating for Thin Film Transistor (TFT) arrays:

- Extremely high X-ray Sensitivity
- Direct Conversion - no scintillators required
- Large area thick film technology (physical vapour deposition, or polymer binder) – compatible with TFT arrays for flat panel digital X-ray imaging detectors

single crystal HgI_2



www.realtimeradiography.com



Application areas:

- Fluoroscopic and Conventional Radiography modes
- CT, security and industrial applications

Paul Sellin, Centre for Nuclear and Radiation Physics

Crystalline quality of HgI_2 films

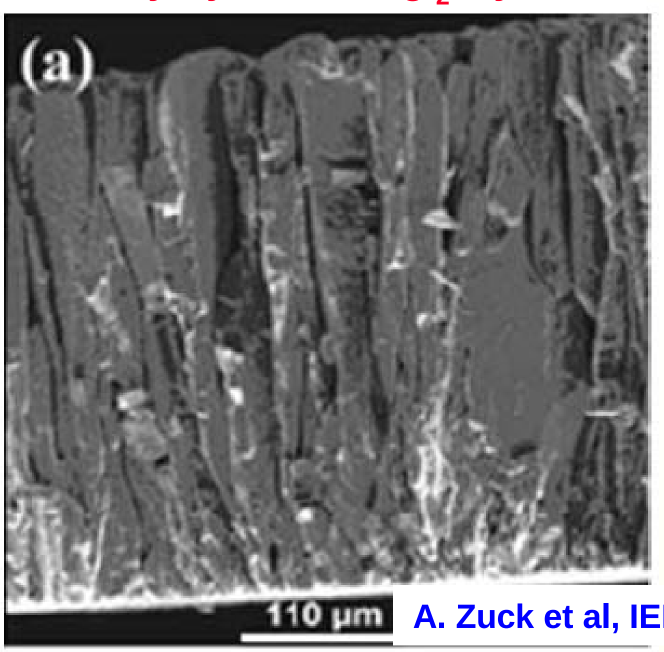
Very high quality films, grown by Real-Time Radiography Inc

Columnar structure, typically 80µm long, growing from the substrate surface

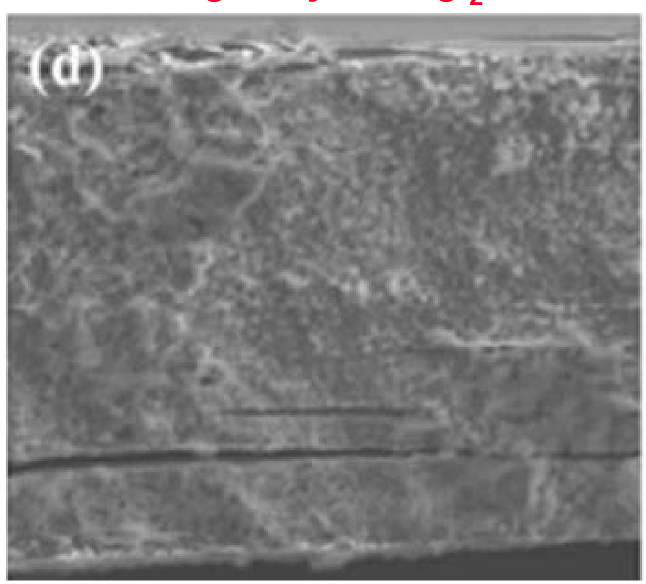
Well-defined alpha pulses show no significant charge trapping, and mobility values comparable with single crystals:

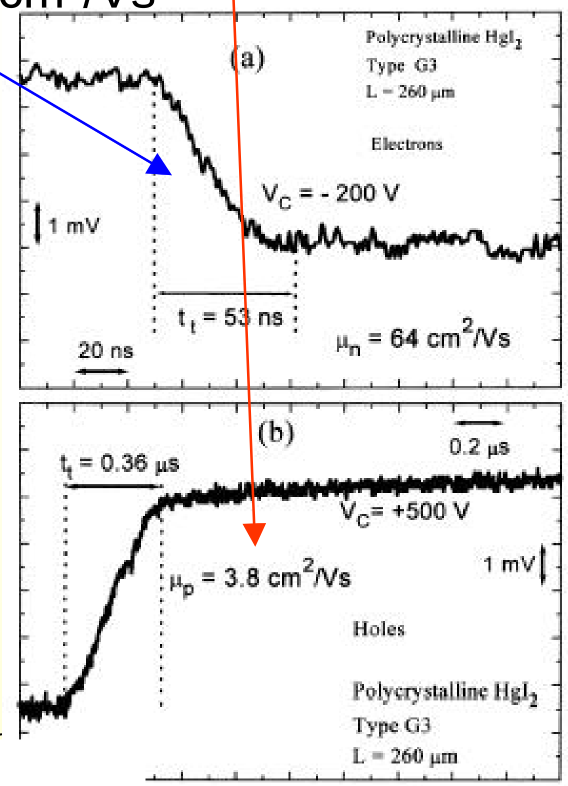
- best polycrystalline values: μ_e ~87 cm^2/Vs and μ_h ~4 cm^2/Vs
- typical single crystal: μ_e ~93 cm^2/Vs and μ_h ~5 cm^2/Vs

Polycrystalline HgI_2 layer



Single crystal HgI_2

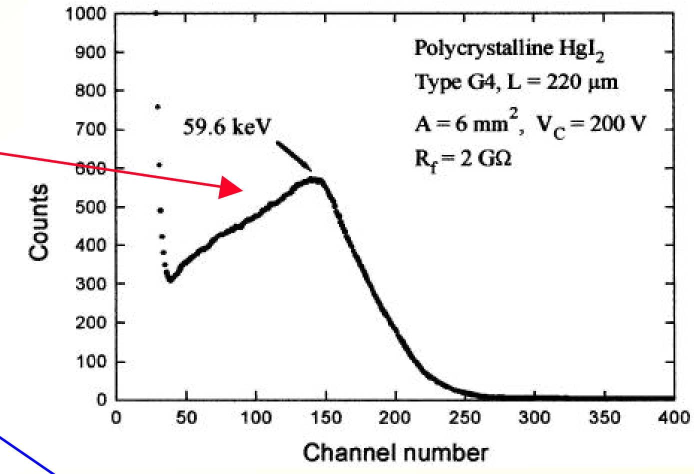


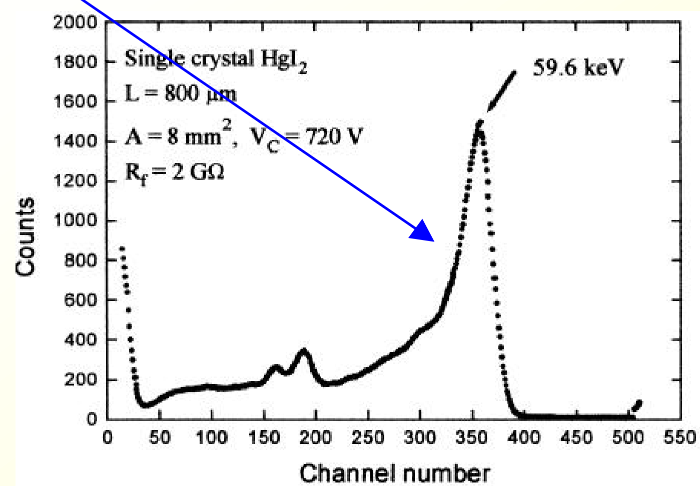


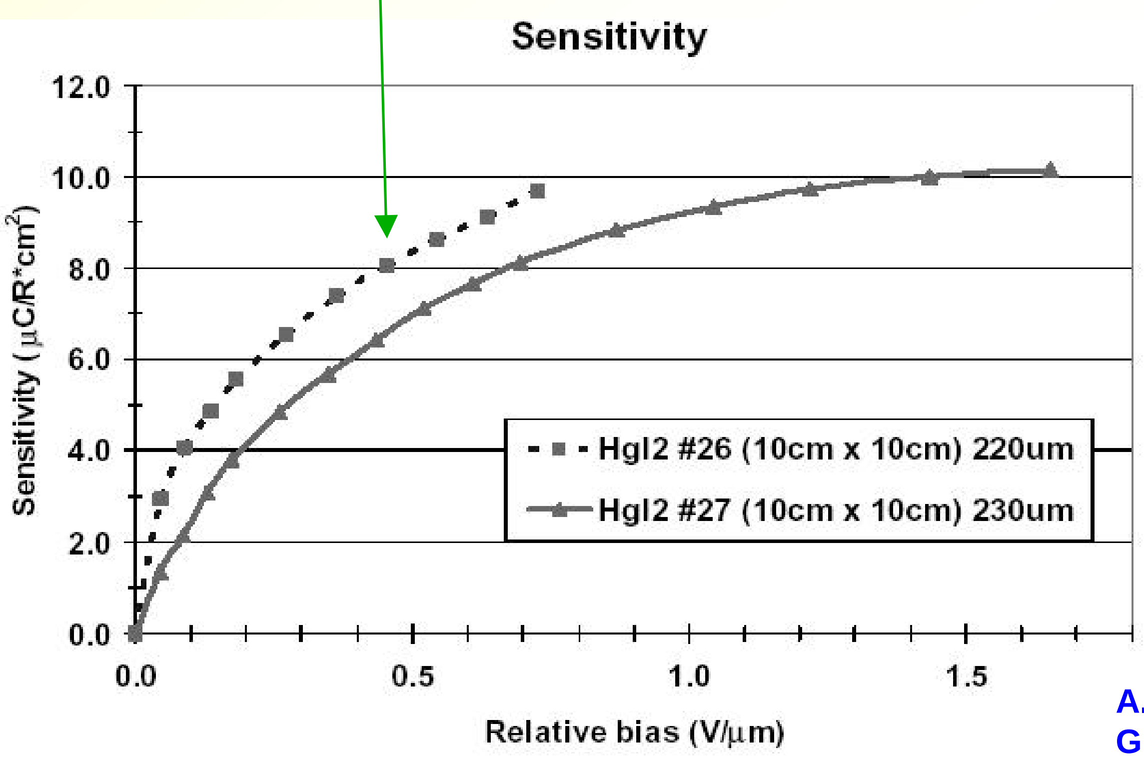
A. Zuck et al, IEEE Trans Nucl Sci 51 (2004) 1250-1255

Radiation response of HgI_2

- Best **polycrystalline HgI_2 film** alpha particle response shows a broad full-energy peak
- Not as good as **single crystal HgI_2**
- Low dark current, 24 pA/mm² @ 0.7 V/µm
- **High sensitivity,** up to 10 µCi/Rcm⁻² without early saturation





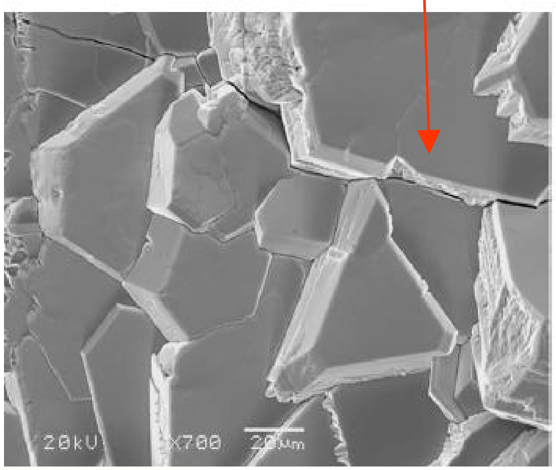


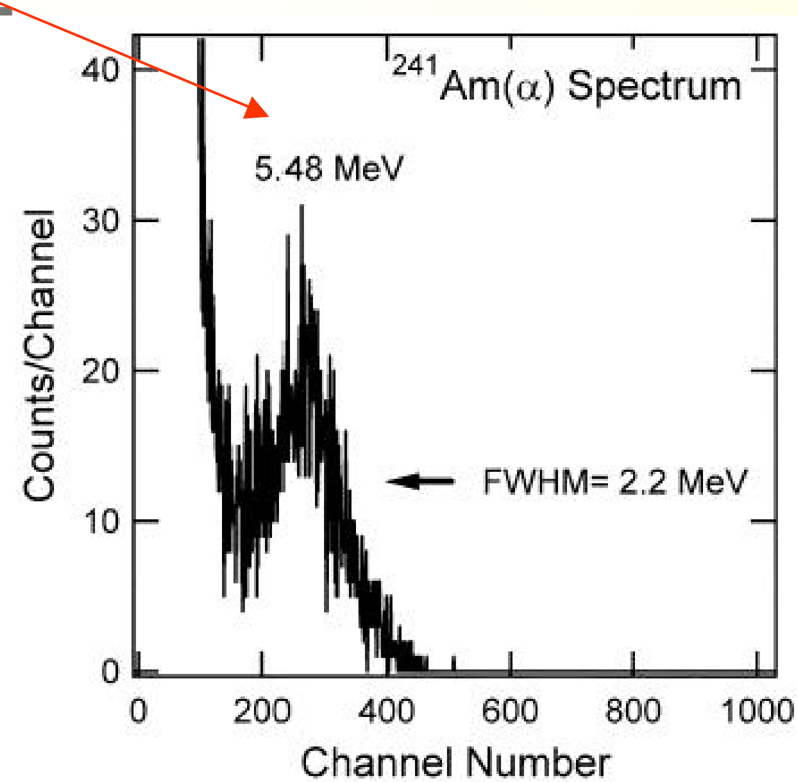
A. Zuck et al, IEEE Trans Nucl Sci 51 (2004) 1250-1255
G. Zentai et al, Proc SPIE-MI (2004) 5368-23

Bismuth Tri-iodide

BiI_3 offers a good range of properties for X-ray detection, with Z_{Bi}=83 and Z_I=53. Detector response has been reported from:

- Single crystal BiI_3 grown by Bridgman methods
- BiI_3 platelets by vapour deposition (~10-20mm^2, 50-80µm thick)
- Polycrystalline growth at low temperature (150°C) with ~50µm crystallites and film thickness of up to 130µm





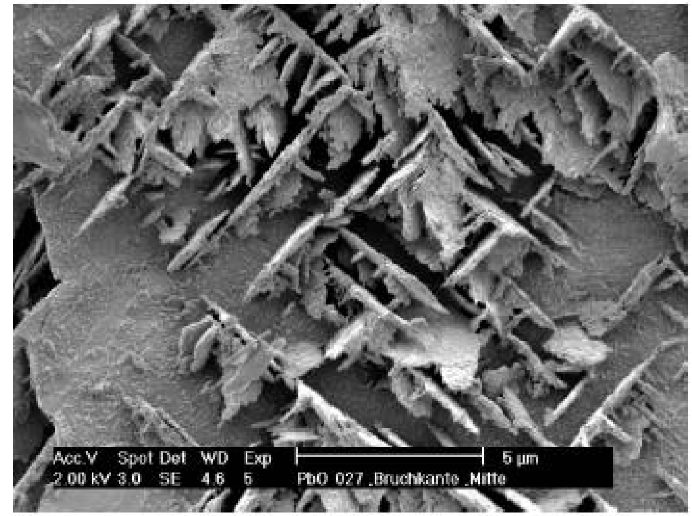
- Charge transport worse than HgI_2, PbI_2: μ_e ~10 cm^2/Vs

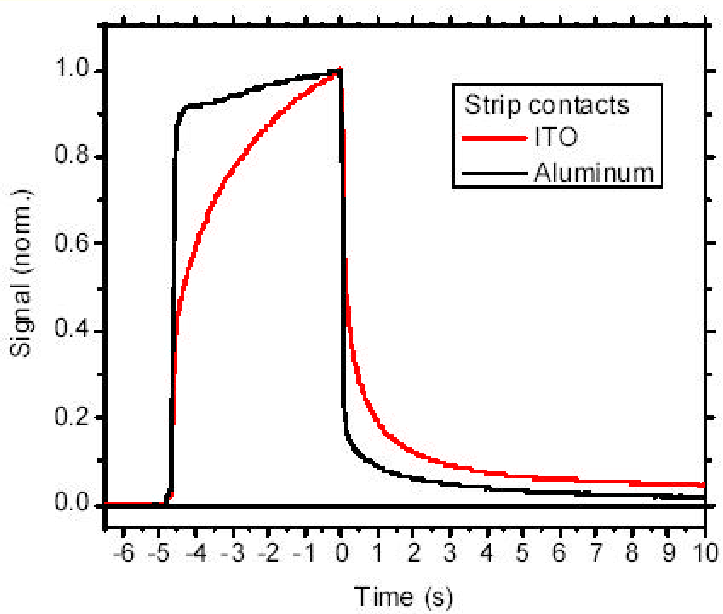
M. Matsumoto et al, IEEE Trans Nucl Sci, 49 (2002) 2517-2520
L. Fornaro et al, IEEE Trans Nucl Sci, 51 (2004) 96-100, 2461-2465

Lead Oxide films

Thick film polycrystalline PbO films have been studied by Philips Research:

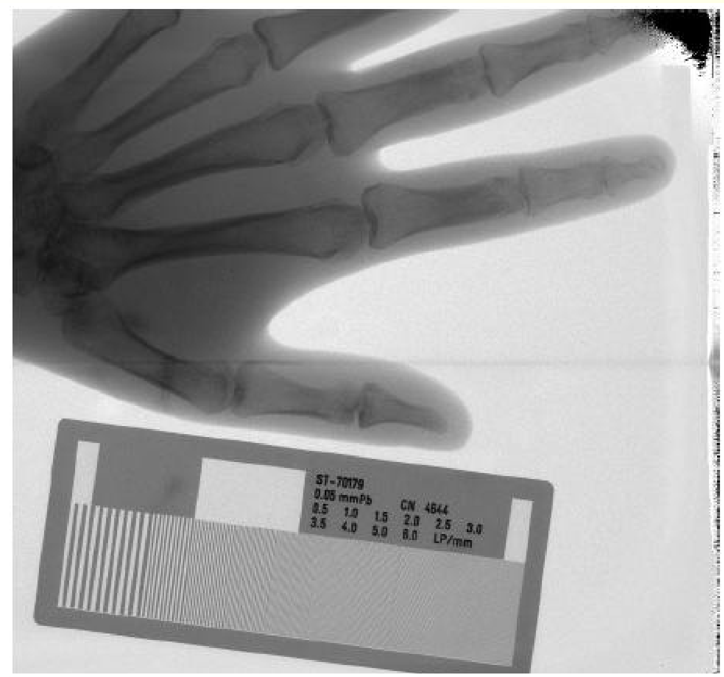
- Thermal evaporation process (100°C) for 25x25cm films, with 300µm thickness
- Thin platelet structure, 50% porous
- Low charge transport ($\mu\tau_e \sim 4\times10^{-7}$ cm^2/V) but low dark current ~200 pA/mm^2
- X-ray temporal response dependent on contact structure





PbO prototype imager uses 18x20cm film on 960x1080 TFT pixel matrix

160µm thick PbO film, 70kVp X-rays



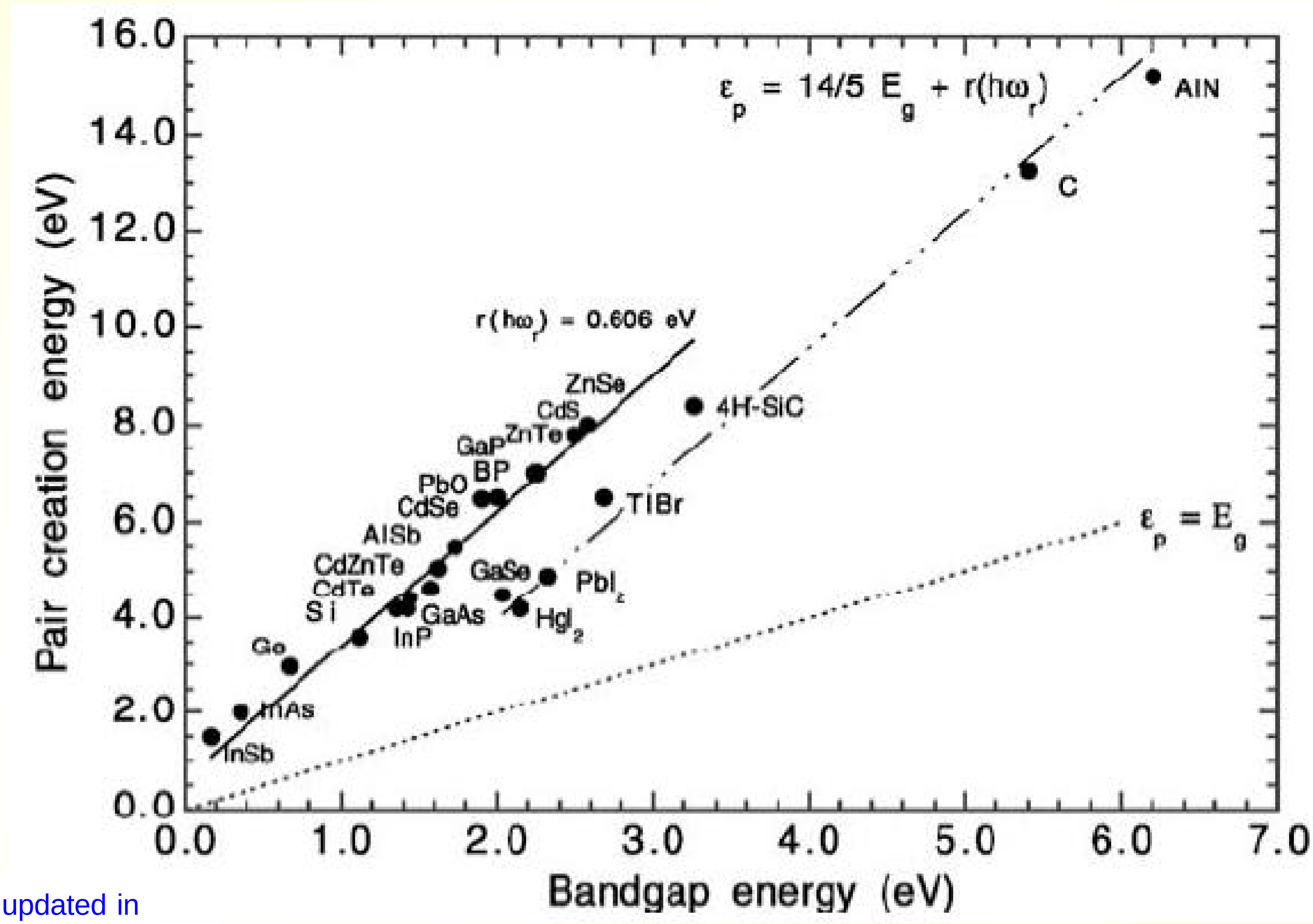
M. Simon et al, IEEE Trans Nucl Sci, in press

New crystalline materials

The Klein chart shows the relationship between **ehp creation energy** and **bandgap**:

Here we consider 3 possible new materials:

1. CdMnTe
2. GaN
3. Diamond



CA Klein, JAP 4 (1968) 2029, updated in A Owens et al, NIM A531 (2004) 18-37

CdMnTe – a future alternative to CdZnTe?

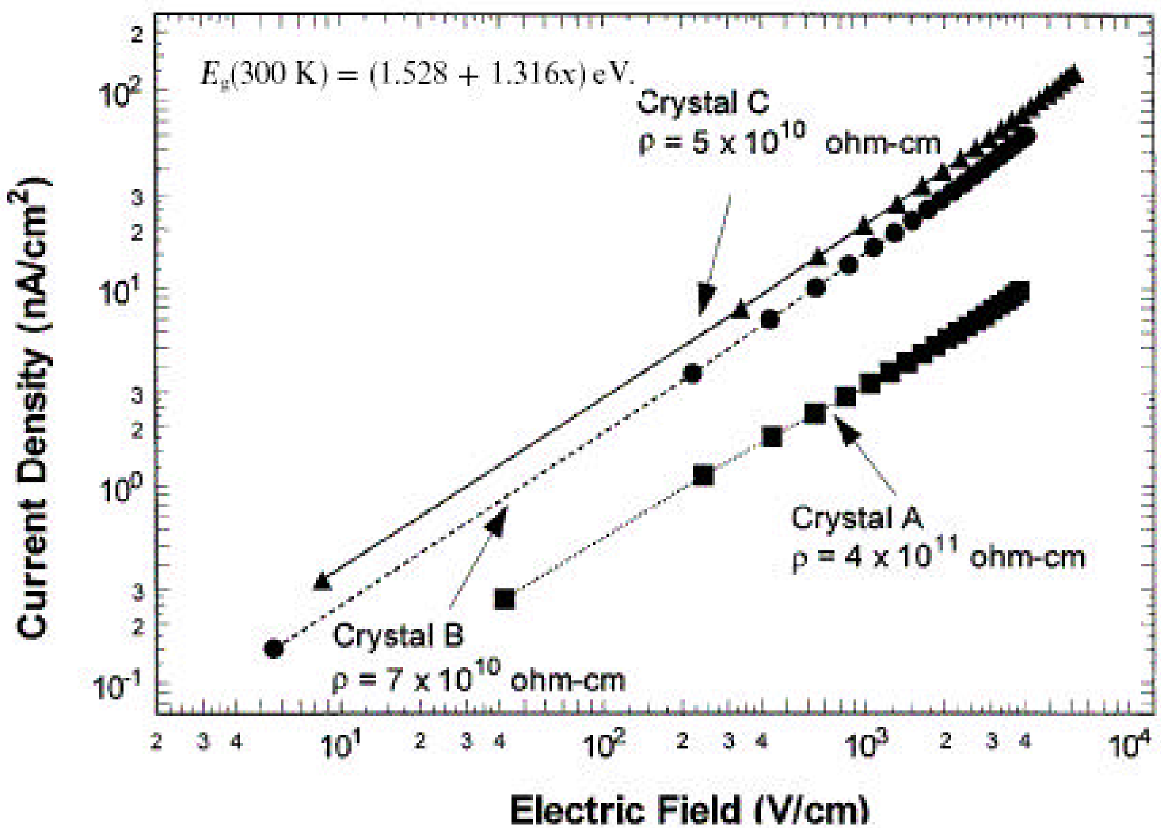
CdMnTe is a ternary alloy similar to CdZnTe – very low segregation coefficient of Mn should produce uniform crystals

• alloying with Mn increases the bandgap twice as fast as Zn (13 meV per % Mn)

• compensation using Vanadium or Indium doping achieves high resistivity

• bandgap values of 1.73 - 2.12 eV (CZT ~ 1.55 eV)

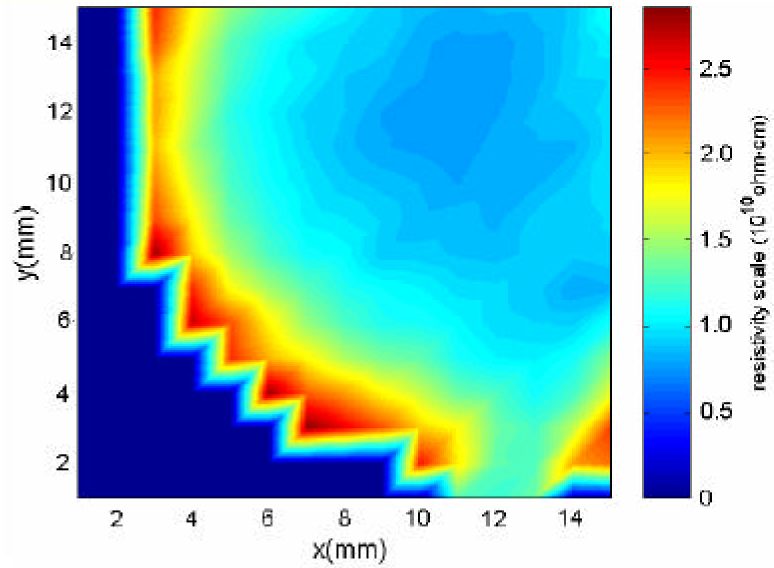
Growth of high resistivity crystals by the Vertical Bridgman technique has been demonstrated

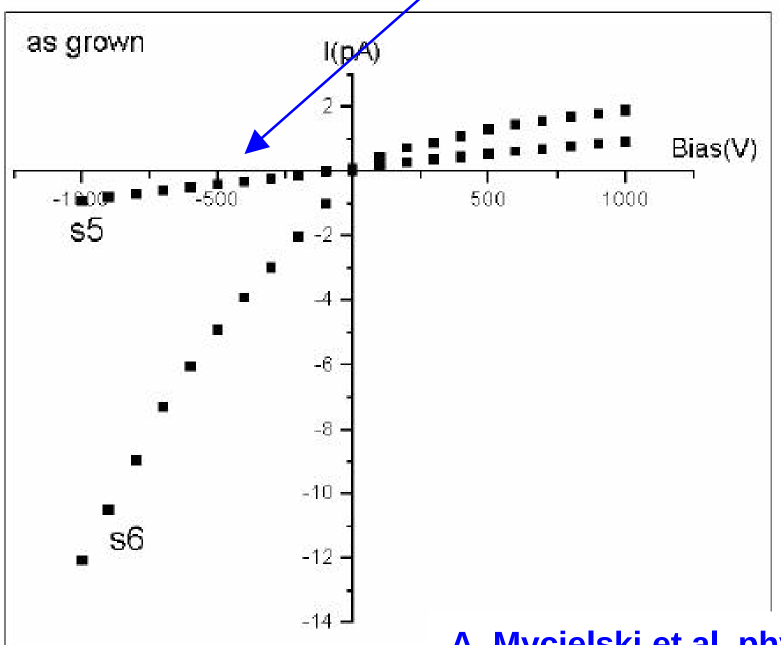


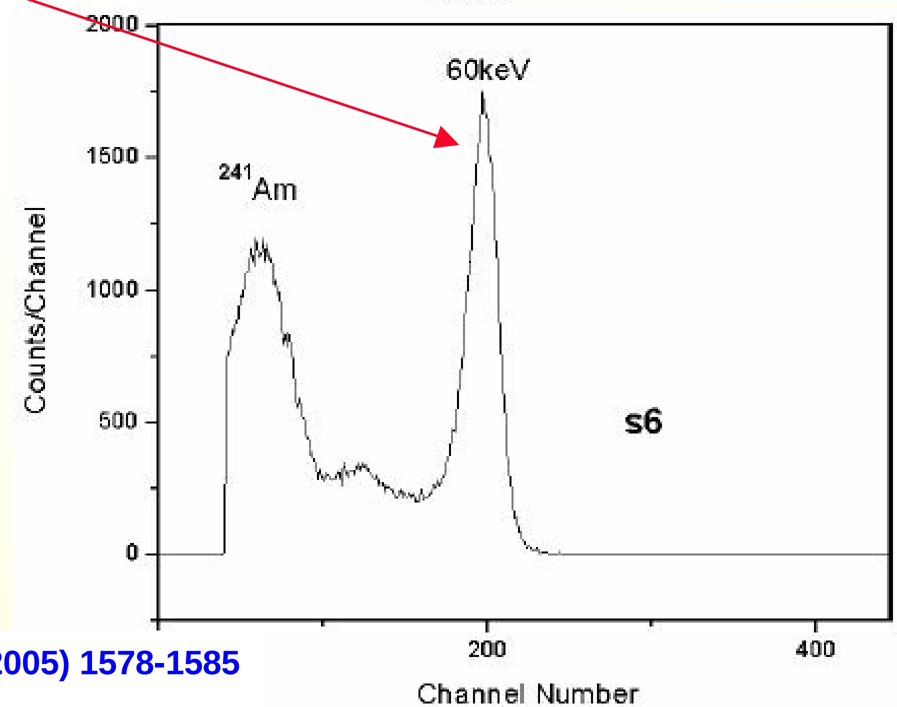
A. Burger et al, JCG 198/199 (1999) 872-876

First results from CdMnTe detectors

- As-grown undoped CdMnTe is p-type: doping with indium has demonstrated $\rho \sim 10^{11} \Omega cm$
- Material quality currently limited by poor "4N" quality of manganese
- Reasonable charge transport observed – $\mu\tau_e \sim 2x10^{-5}\ cm^2/V$
- Resolved photopeak observed at 59 keV







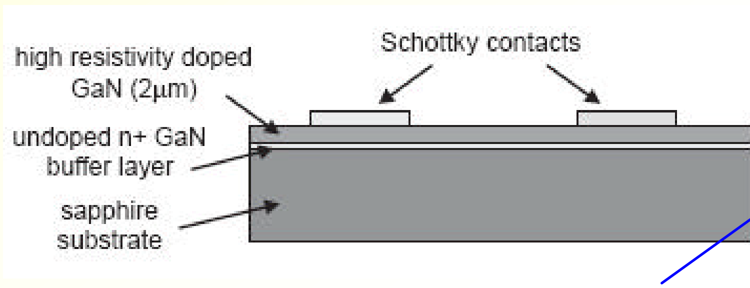
A. Mycielski et al, phys stat sol (c) 2 (2005) 1578-1585

GaN radiation detectors

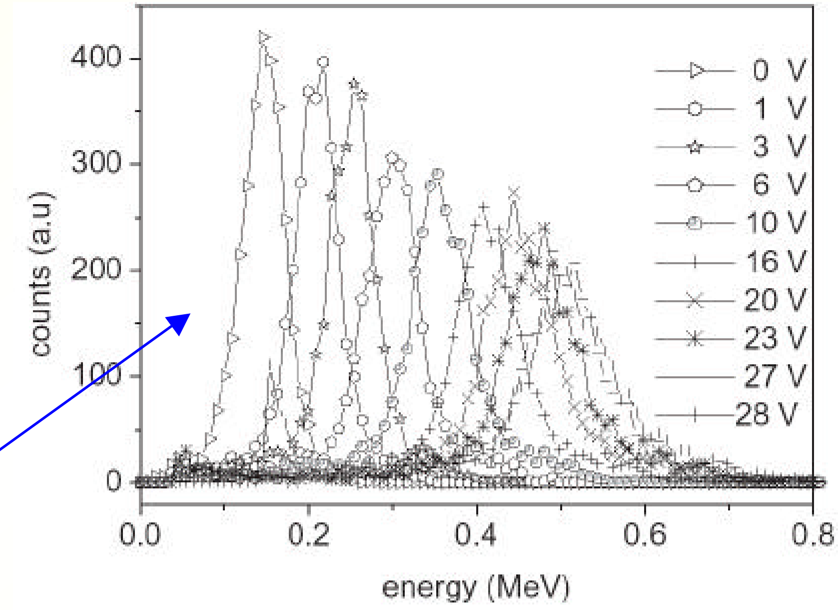
GaN (Z=31/7) is a candidate material for 20-60 keV X-ray detection

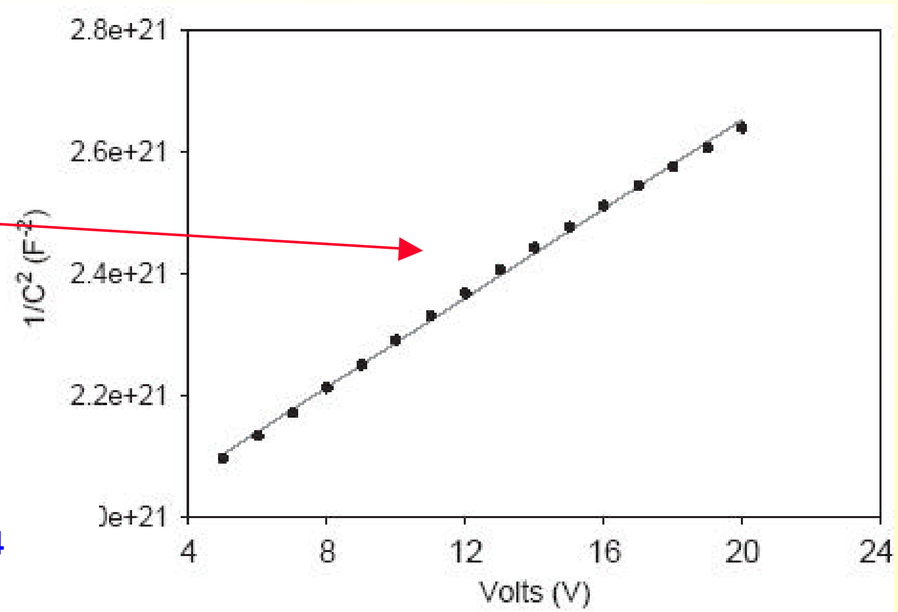
- Band gap is 3.39 eV
- Ehp creation energy is 8.9 eV/ehp

Thin epi-layers are grown onto sapphire:



- Good alpha particle response has been seen in thin (2µm) epi layers
- CV data gives carrier concentration of $1 \times 10^{15} cm^{-3}$
- Free-standing thick (>100µm) GaN layers are now available, grown by High Pressure Vapour Expitaxy



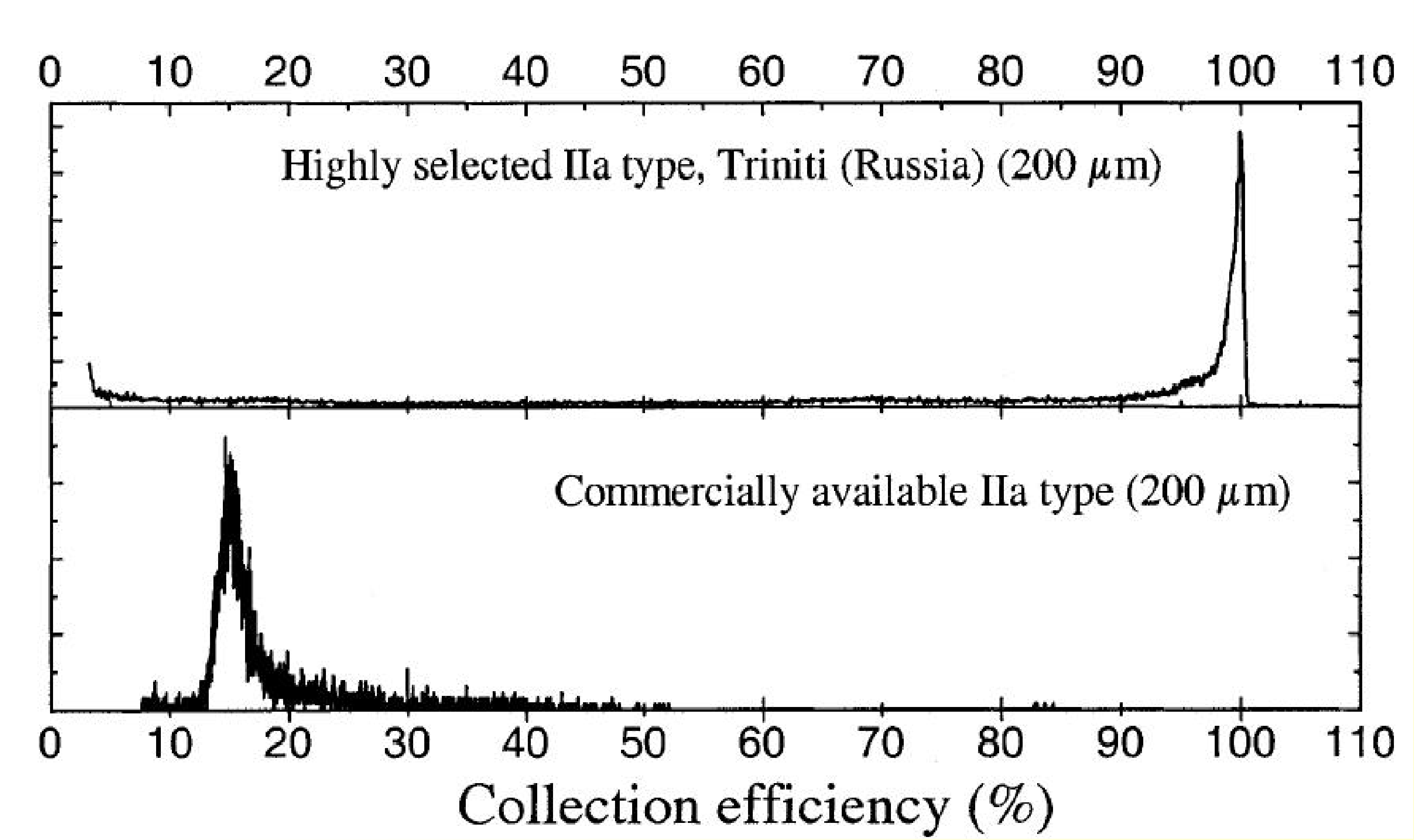


P. Sellin et al, NIM A531 (2004) 82-86
J. Vaitkus et al, NIM A509 (2003) 60-64

UniS Department of Physics University of Surrey

Single-crystal synthetic diamond

Single-crystal natural diamonds have been studied in the past for detector applications – not a viable option.



P. Bergonzo et al, Dia Rel Mat 10 (2001) 631-638

High purity single-crystal synthetic diamond

Companies in the US and UK have recently new growth techniques to fabricate near-perfect single-crystal artificial diamond

Primarily marketed as gem stones, diamond wafers 10x10mm are now available for device applications, with thickness of up to 500μm

Image removed

5x5mm piece of single-crystal synthetic diamond

Photoluminescence image shows real colour:

- HPHT substrate – yellow
- Nitrogen impurities – red
- Dislocations – cyan blue

Single-crystal CVD diamond detectors

Specialist applications of diamond detectors:

- as tissue-equivalent rad-hard detectors, eg megavoltage therapy beams
- detectors for very high temperature, high radiation environments

True single-crystal material removes charge trapping associated with grain boundaries:

- 100% CCE demonstrated from alpha particles
- High mobility ⇒ fast signals
- Radiation hardness tests are in progress

Image removed

Conclusions

- The demand for high-Z semiconductor radiation imaging detectors continues to develop, with potential applications in medical, synchrotron, space and security imaging
- CdZnTe continues to dominate the commercial supply of high-Z materials, with new suppliers of detector-grade material slowly becoing available
- There is a steady improvement in CdZnTe material uniformity, single-crystal volume, and spectroscopic performance, with $\mu\tau_e$ approaching 10^{-2} cm^2/Vs
- There is significant R&D activity in thick film materials, compatible with large-area imaging devices:
  - Polycrystalline and epitaxial CdTe/CdZnTe thick films
  - Various $Z \geq 80$ compounds, with excellent imaging performance demonstrated by HgI_2
- New materials continue to be developed, with examples illustrated from CdMnTe, GaAs and single-crystal synthetic diamond