HP-Ge Crystal Growth at USD

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May 14, 2024

This work is done under the collaboration PIRE-GEMADARC. It was sponsored by NSF OISE-1743790, NSF PHYS 2310027, and NSF PHYS 2117774.

CONFERENCE ON SCIENCE AT THE SANFORD UNDERGROUND RESEARCH FACILITY

South Dakota Mines campus, May 14-16, 2024

PIRE GEMADARC

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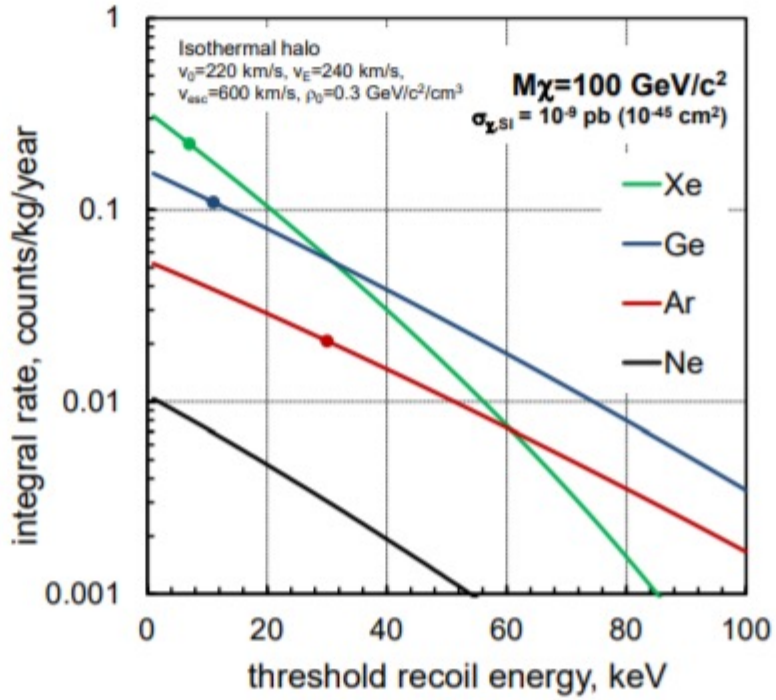
MOTIVATIONS

- Conduct research and development on high-purity germanium crystal (HP-Ge) growth techniques essential for the production of HP-Ge germanium detectors
- Enhance crystal quality by investigating and optimizing the crucial factors affecting the crystal growth process
- Cultivate large HP-Ge crystals, transform them into functional detectors, and conduct in-depth analyses to understand detector behavior, with a particular emphasis on rare event detection

Why Germanium?

- Low energy threshold
- Excellent energy resolution
- Relatively simple cooling systems
- Excellent event/background discrimination power
- For neutrinoless beta decay ^{76}Ge acts as both source and detector with relatively low half time for decay



- More recoil (more energetic interaction) for Ge; more counts for Xe.
- Counts can be increased by increasing detector size.

WIMP Dark Matter Direct Detection
P. Cushman, C. Galbiati, D. N. McKinsey, H. Robertson, T. M. P. Tait

Requirements of Germanium Detectors

- Depletion depth d of a planar detector

$$d = \sqrt{2 \in V_0 / eN}$$

here, V_0 is the applied bias voltage, N is the net impurity concentration in the crystal, e is the charge of, electron, and ∈ is the dielectric constant of Ge

- Net Impurity concentration should be $\sim 10^{10}/cm^3$ for full depletion of about a 1 cm thick detector
- Dislocation density 300-10000$/cm^2$; Otherwise, divacancy-hydrogen complex (V_2H) or dislocation itself will work as charge trapping center (1)
- Homogenous Impurity concentration along the detector
- **Ton-scale Ge-based experiments and large-size crystals are needed to improve the sensitivity and detection efficiency**

1 (DIVACANCY HYDROGEN COMPLEXES IN DISLOCATION -FREE HIGH -PURITY GERMANIUM, E. E. Haller, G. S. Hubbard, W. L. Hansen, and A. Seeger)

Material Preparation

Germanium ingots ready for etching

Crucible and glass tubes being cleaned with hydrogen peroxide

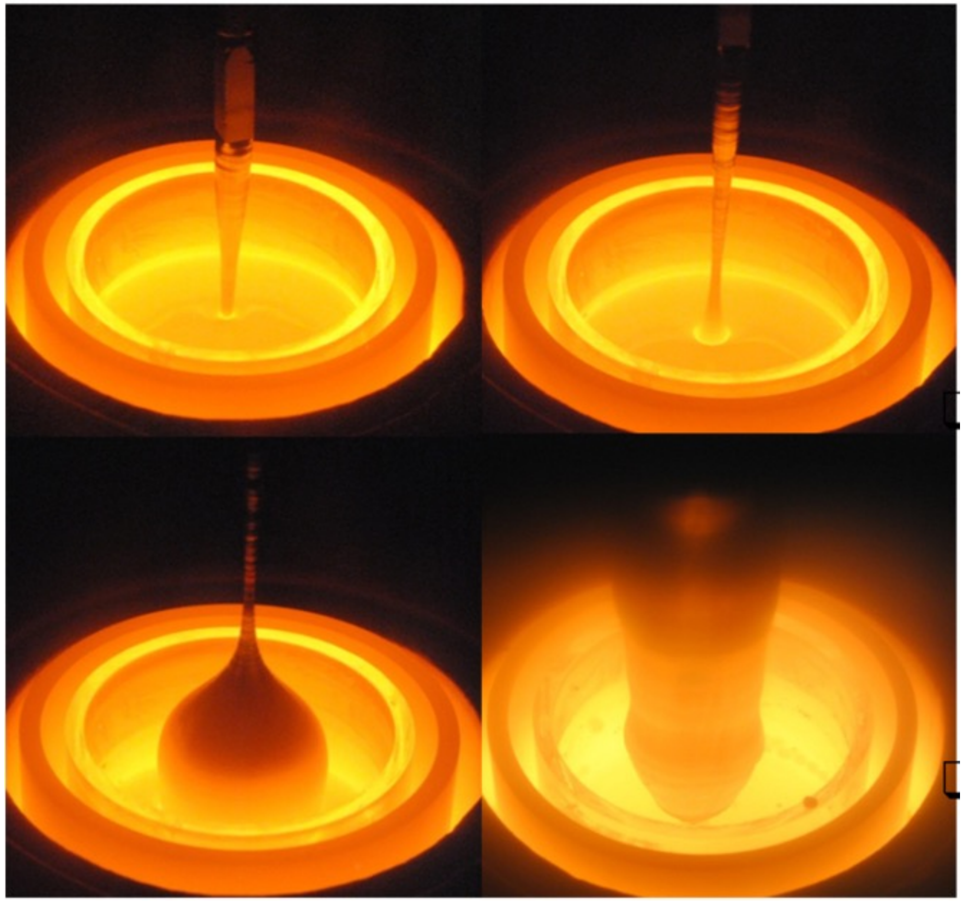
Etching ingots with strong acid,HF: HNO_3 (1:3)

Drying with Nitrogen before loading in the chamber

Procedure

- Seed crystal (100) Ge) is dipped into the molten Ge and withdrawn slowly, while maintaining the temperature of melt just above the melting point.

- The rate of crystal withdrawal (pull speed) and temperature (applied power) of melt are adjusted to control the growth of the crystal.



Diameter Control

$$D = \sqrt{\frac{\frac{4dw}{dt}}{p\pi\rho_s + (\frac{dw}{dt}.\rho_s)/R^2\rho_L}}$$

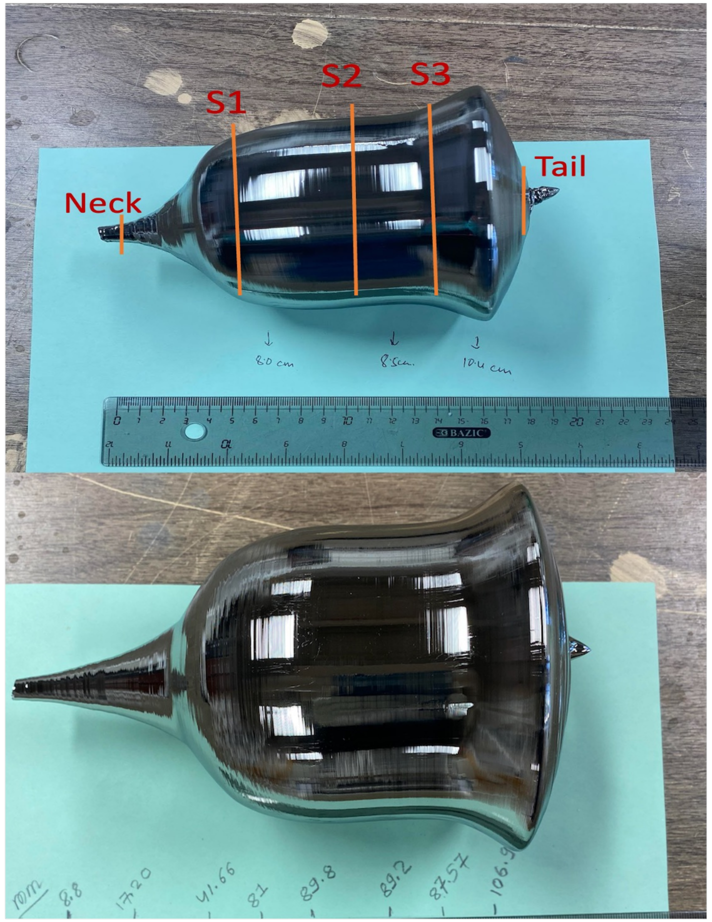
$\frac{dw}{dt}$ = growth rate

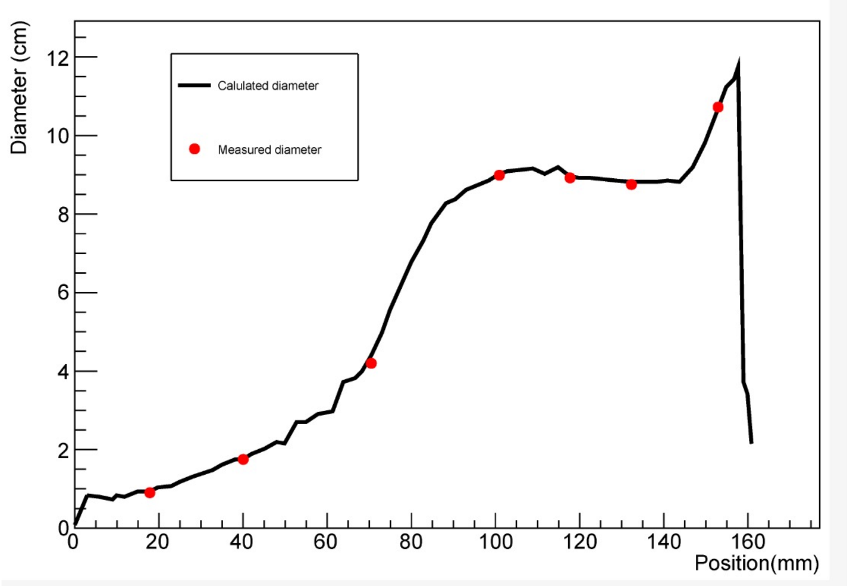
ρ_s = density of solid germanium

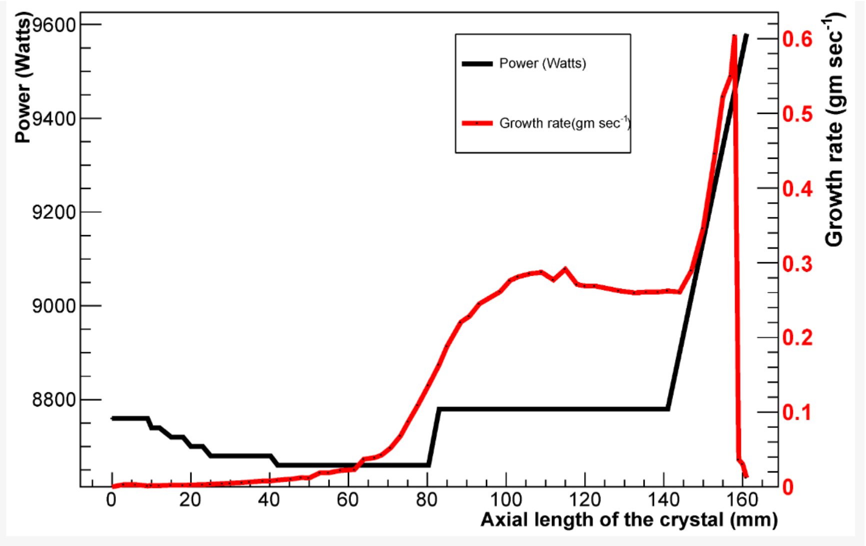
ρ_L = density of liquid germanium

R = radius of the crucible

p= pull rate







Impurity Segregation

position	Net Impurity (/cm^3)	Resistivity (Ω cm)	Mobility (cm^2/Vs)
Neck	1.01×10^{11}	5.40×10^{2}	3.24×10^{4}
S1	6.31×10^{10}	3.74×10^{3}	3.63×10^{4}
S2	1.55×10^{10}	4.92×10^{3}	3.48×10^{4}
S3	-1.13×10^{11}	3.80×10^{2}	2.14×10^{4}
Tail	-1.04×10^{13}	6.32×10^{1}	1.41×10^{4}

$$C(g_i) = C_B(g_i) + C_{Al}(g_i) + C_{Ga}(g_i) + \mathrm{C_P}(\mathrm{g_i})$$

Wang, G.; Amman, M.; Mei, H.; Mei, D.; Irmscher, K.; Guan, Y.; Yang, G. Crystal growth and detector performance of large size high-purity Ge crystals. *Mater. Sci. Semicond. Process.* 2022, *39*, 54–60.

$C(g_i)$ = Net impurity concentration at position g_i
$C_B(g_i)$= Impurity concentration of **boron** at position g_i
$C_{Al}(g_i)$ = Impurity concentration of **aluminium** at position g_i
$\mathrm{C_P}(\mathrm{g_i})$ = Impurity concentration of **phosphorous** at position g_i
$C_{Ga}(g_i)$ = Impurity concentration of **galium** at position g_i
Scheil's equation

$$C = C_0 k_{eff}(1-g)^{K_{eff}-1}$$

Where, g = the solidified fraction at different positions: 0.005 at the neck, 0.15 at S1, 0.45 at S2, 0.75 at S3, and 0.995 at the tail.

k_{eff} = effective seggregation coefficient = $k_0 + (1-k_O)\exp\left(-\frac{f\delta}{D}\right)$

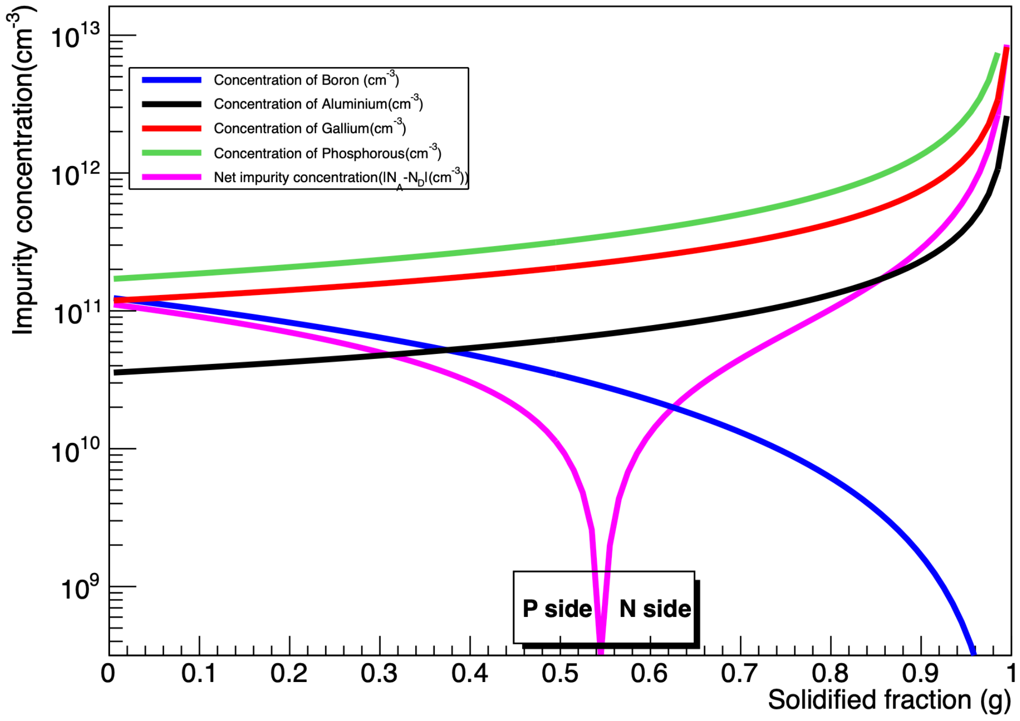
Where, k_0 is equlibrium seggrgation coefficient, f= growth rate , D = diffusion coefficient of impurity in germanium melt.

$\delta = 1.6D^{\frac{1}{3}}v^{\frac{1}{6}}w^{-\frac{1}{2}}$, v =kinematic viscoity, w= rotation rate of crystal

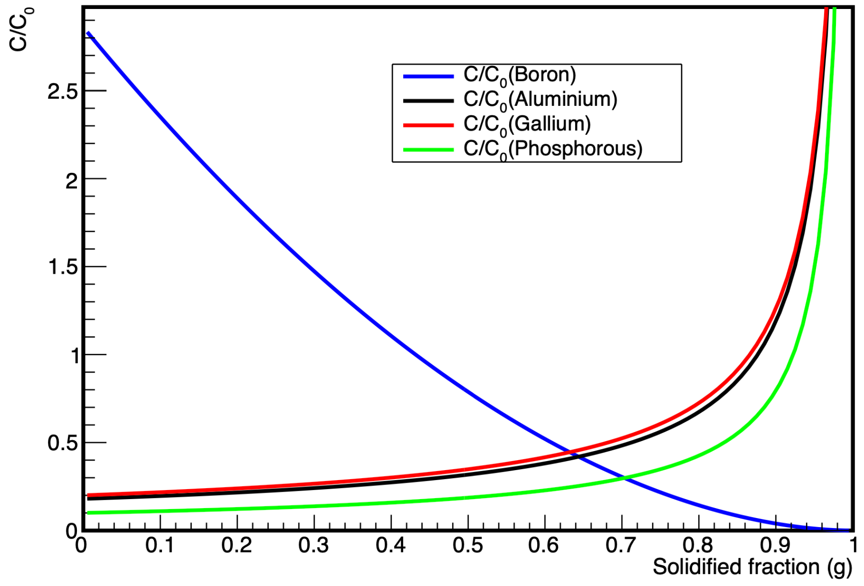
Bhattarai S, Mei D, Budhathoki N, Dong K, Warren A. Investigating Influential Parameters for High-Purity Germanium Crystal Growth. *Crystals*. 2024; 14(2):177. https://doi.org/10.3390/cryst14020177

Results

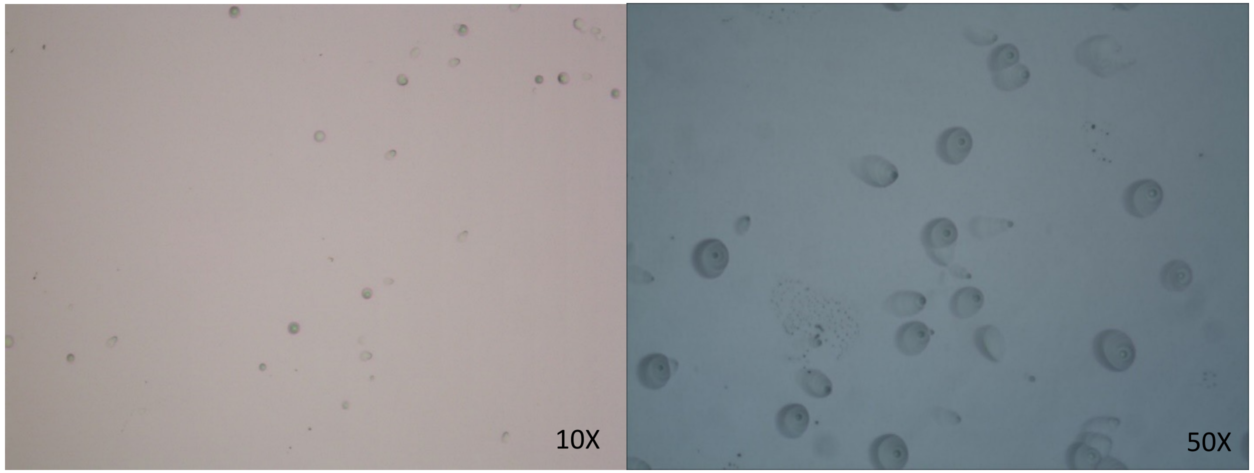
$$2.84C_{0B} + 0.19C_{0Al} + 0.2C_{0Ga} - 0.10C_{0P} = 1.01 \times 10^{11},$$
$$2.12C_{0B} + 0.20C_{0Al} + 0.21C_{0Ga} - 0.11C_{0P} = 6.31 \times 10^{10},$$
$$0.22C_{0B} + 0.58C_{0Al} + 0.6C_{0Ga} - 0.35C_{0P} = -1.13 \times 10^{11},$$
$$0.00014C_{0B} + 13.89C_{0Al} + 13.87C_{0Ga} - 11.86C_{0P} = -1.00 \times 10^{13},$$

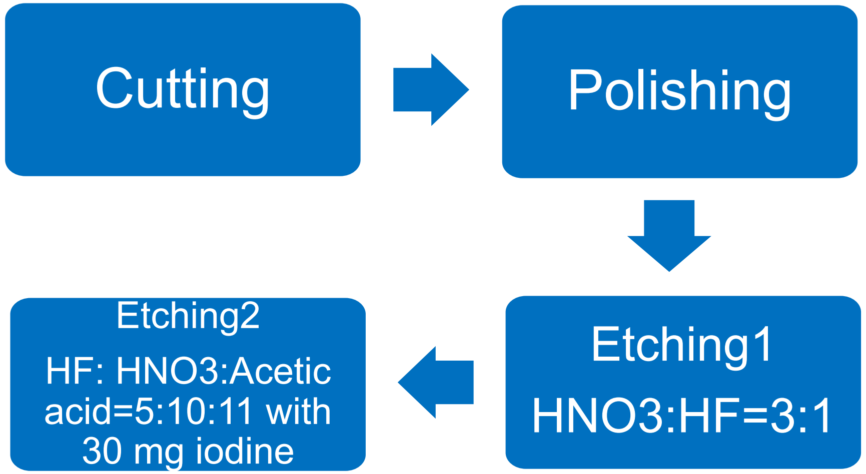


The initial concentration (C_0) of B, Al, Ga and P for this crystal were found to be $3.87 \times 10^{10}/cm^3$, $2.21 \times 10^{11}/cm^3$, $7.13 \times 10^{11}/cm^3$ and $1.89 \times 10^{12}/cm^3$ respectively.

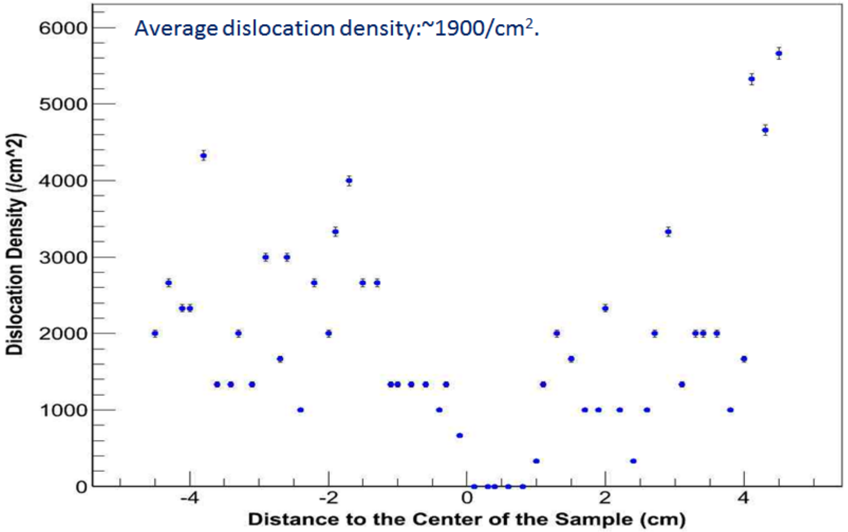


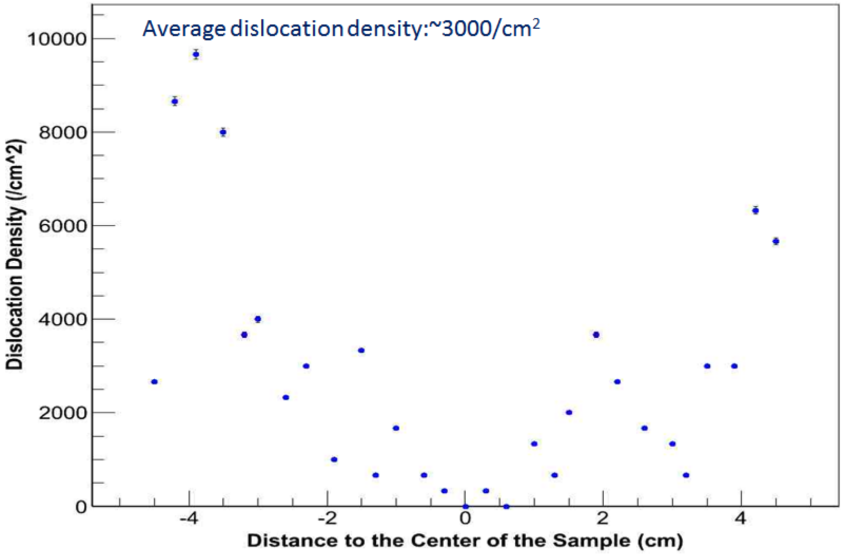
Dislocation Measurement





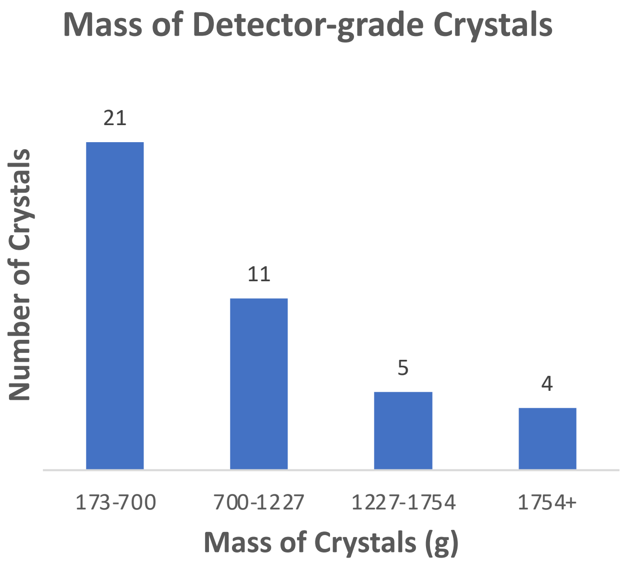
- ➢ Dislocations observation under microscope

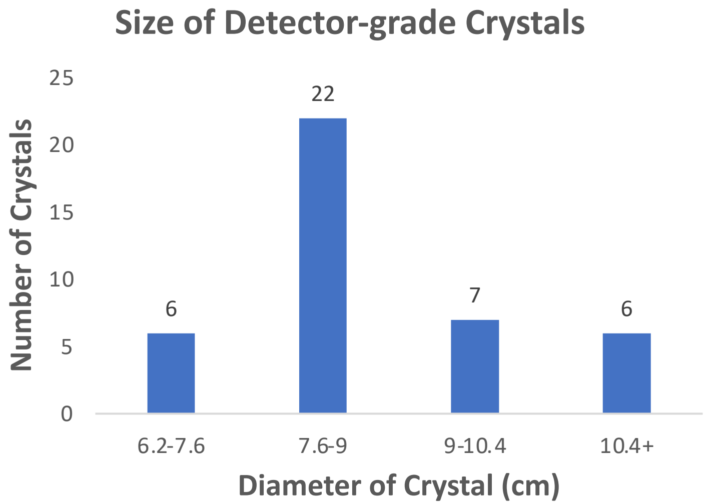




- ➢ Full view of the dislocation distribution on the whole slice

Detector-Grade Crystals Summary

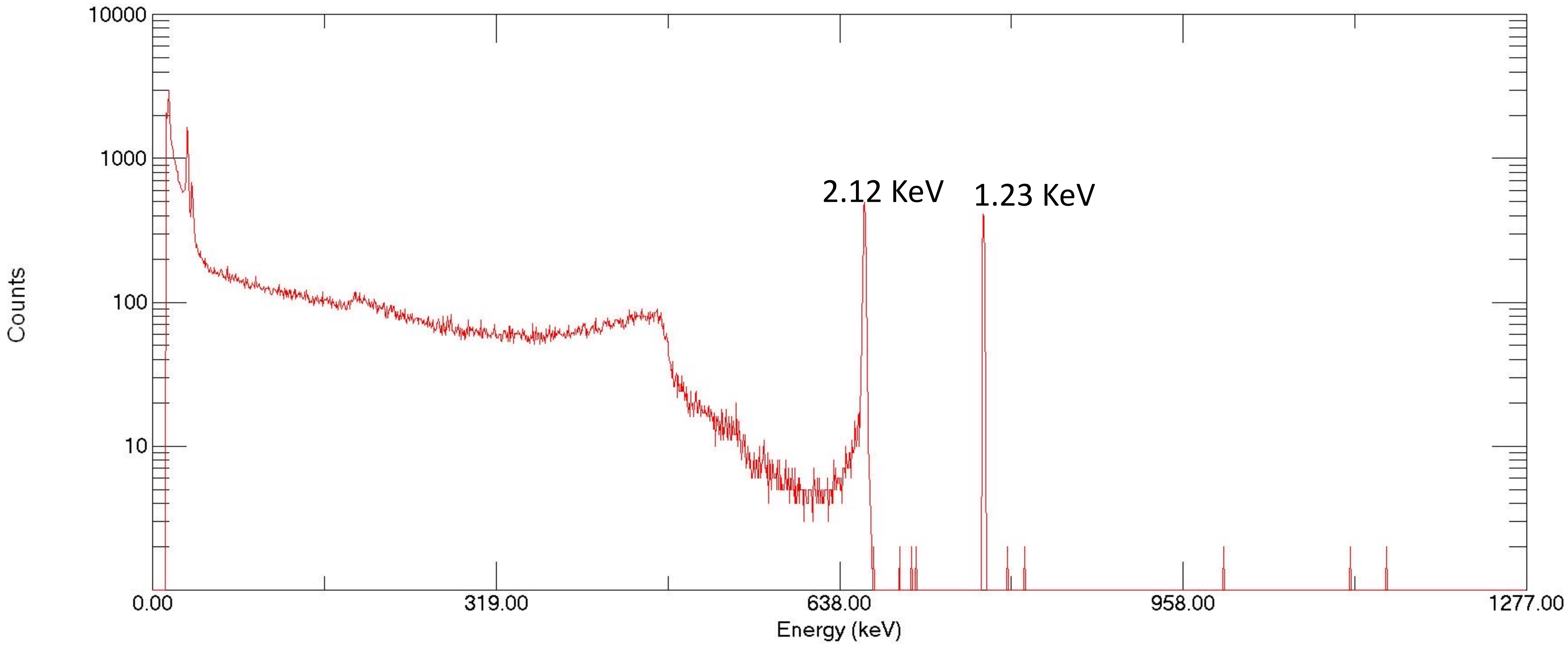




- 36.1kg detector grade crystals at USD:
  - 43 detector grade crystals;
  - Impurity level: $3.77\times10^{9}\sim7\times10^{10}/cm^{3}$.
  - Average diameter: 8.5 cm.
  - Average thickness: 2.5 cm.

- Mass: 2247g
- Diameter: 10cm
- Thickness: 5.4cm
- Impurity Level: $2.06E10^{10}(top)$ $2.68E10^{10}(bottom)$

Energy Spectrum of Cs-137



- Energy spectrum from a Cs-137 source measured with the detector USD-R03. The source was positioned facing the detector top. The bias voltage of -2500 V was applied to the bottom electrical contact on the detector while the signals were measured from the top.

Conclusions

- At USD, we have successfully built a production chain that can purify Ge raw materials, grow detector-grade HP-Ge crystals, and fabricate them into Ge detectors
- A well-controlled segregation method and thermal field are used to control the distribution of impurities during crystal growth to meet the requirements for detector-grade Ge crystals
- We investigated some essential parameters like impurity segregation to improve the quality of the crystal; we will continue to investigate and develop the influential parameters for the entire HP-Ge development chain

Thanks!