Large Cryogenic Germanium Detector: Final Report

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Cryogenic Germanium detectors are widely used in high energy physics and astrophysics – for example in direct dark matter searches (such as the Cryogenic Dark Matter Search experiment), or in neutrino-less double-beta decay experiments (such as MAJORANA). Potential future applications include X-ray observations and measurements of coherent neutrino scattering. In all of these fields, increasing the size of individual Ge detectors is critical either to simplify scaling of the total detector target mass to ~1 ton scale (to improve experiment sensitivity) or to increase the solid angle coverage.

The goal of this project was to investigate possible ways of increasing the size of cryogenic Ge detectors. Historically, two parameters played critical roles in determining the maximum size of such detectors:

- 1) Dislocations: density of dislocations (extended deformations in the crystalline lattice) must be kept between 100 and 7000 cm^{-2} in order to optimize the charge collection efficiency of the detector at 77 K temperature. If the dislocation density is higher than $\sim 7000 \text{ cm}^{-2}$, the dislocations trap a substantial fraction of charges and therefore reduce the charge collection efficiency. If the dislocation density is lower than $\sim 100 \text{ cm}^{-2}$ then the density of point defects in the crystal is too high – point defects also act as charge traps and reduce the charge collection efficiency.
- 2) Purity: in order to minimize the effect of charge traps due to impurities and maximize the charge collection efficiency, only high-purity crystals (with impurity levels lower than $10^{12} \text{ atoms/cm}^3$) could be used to fabricate detectors.

These two parameters had a severe impact on the size of Ge detectors. In particular, it is increasingly difficult to maintain the optimal dislocation density as the diameter of the high-purity Ge crystal is increased. Consequently, all Ge detectors used prior to this project had diameters of 3” or smaller.

It is also important to note that the physics of Ge semiconductors at very low temperatures ($\sim 50 \text{ mK}$) is significantly different than at 77 K. In particular, since the charges are “frozen” at such low temperatures, it is possible to neutralize all charge traps by occasionally exposing the detector to infrared photons (while grounding the detector electrodes). This technique was demonstrated on standard Ge detectors with dislocation densities of 100-7000 cm^{-2} (for example, by CDMS), as well as on a small (3cm-diameter) high-purity dislocation-free Ge crystal (test performed in our previous work).

This project identified two possible approaches to increasing the individual cryogenic Ge detector size. The first approach relies on using the existing technology for growing detector-grade (high-purity) germanium crystals of dislocation density 100-7000 cm^{-2} . Interactions with Ge vendors (done within the scope of this project) have revealed that pullers used for growing this type of Ge crystals had the maximum capacity of 4”-diameter, although they typically grow crystals of diameter only up to 3” – at higher diameters it is more difficult to control the thermal gradient, which leads to larger dislocation densities. This option was then pursued by separate

funding: 4" (100mm) Ge crystals were purchased, polished and fabricated into detectors by our SLAC collaborators, and then operated in our Kelvinox 100 cryostat at the University of Minnesota. We demonstrated that these crystals have charge collection efficiency similar to those of the standard 3"-diameter Ge detectors. The 100mm Ge detectors are currently considered the best choice for the next-generation (G2) Ge-based dark matter search experiment (SuperCDMS-SNOlab, to be proposed in the Fall 2013).

The second approach is to consider dislocation-free Ge crystals. The industry has demonstrated that dislocation-free Ge crystals could be grown up to 50 cm in diameter. The difficulty lies in the fact that such crystals are typically used in very different applications (such as windows), and are therefore grown in relatively "dirty" puller environments. Since development of a dedicated new "clean" puller was prohibitively expensive for this project (roughly ~\$1 million), the approach we took was to conduct a sequence of crystal growing tests with one of the vendors (Umicore) to assess whether we could use their existing ("dirty") pullers, suitably modified, to grow dislocation free Ge crystals of sufficient purity. This approach required several deviations from the standard procedures for growing dislocation-free crystals:

- 1) Dislocation-free crystals are typically grown in inert gas environments, while detector-grade Ge must be grown in hydrogen atmosphere for passivation purposes. Due to safety concerns, the 100% hydrogen gas was not an option in the dislocation-free pullers, so we used a mixture of Ar and H₂ (with 2.5% of H₂). Appropriate plumbing modifications were made to introduce this gas into the puller and to properly purify it with a palladium getter.
- 2) Since dislocation-free crystals are typically heavily doped, and are grown in graphite crucibles, considerable care was taken to mitigate potential contamination due to past use of the pullers. In particular, for our growing tests we used new medium purity quartz crucibles (used for growing detector-grade Ge), a new graphite susceptor (which holds the crucible in place, but is not in contact with the Ge melt), and zone-refined charge material.
- 3) In order to open a path toward growing large crystals, we only considered pullers that had the capacity to grow 6"-diameter crystals, although our tests focused on 3"-diameter crystals to minimize the costs.

After the crystals were grown they were characterized with multiple techniques, including the Hall measurement of the density of charge carriers, measurement of the dislocation density, and FTIR measurement of the impurity types and levels.

The first two tests were performed in a high-pressure puller (with support from UC Berkeley collaborators). The first test resulted in high impurity level (2×10^{13} atoms/cm³), and the main cause was suspected to be the gas inside the puller. For the second test we introduced a getter to purify the gas before it was injected into the puller, which resulted in significantly lower impurity levels ($< 10^{12}$ atoms/cm³), sufficient for Ge detector applications. However, the number of dislocations in both tests was rather large, ranging between 3000 cm⁻² and 16000 cm⁻² depending on the location in the crystal. The dislocations were caused by the silica film that was observed to develop on the surface of the melt. In particular, it is believed that oxygen which may be present inside the puller (due to potential leaks from outside) was interacting with Si in the crucible to form the silica.

To mitigate this problem a different, low-pressure puller was tried in tests 3 and 4. The reduced amount of oxygen in this puller indeed resulted in dislocation-free crystals of diameter up to 100mm (4"). However, as this particular puller is more actively used for growing heavily doped crystals, the contamination levels were rather high, reaching 10^{13} atoms/cm³. The FTIR measurements revealed that the dominant contaminant was Ga, likely originating from the contaminated permanent graphite components in the puller. In the last test we attempted to use an even higher-purity gas mixture of Ar and H₂, which did not significantly affect the results.

The results of these four tests with dislocation-free crystals are summarized in Table 1. The conclusions of this part of the project are as follows:

- 1) We have demonstrated that it is possible to grow Ge crystals of sufficient purity in medium-purity quartz crucibles, with graphite susceptor and a 4N mixture of Ar-H₂ gas.
- 2) We have also demonstrated that it is possible to grow dislocation-free Ge crystals in medium-purity quartz crucibles, with graphite susceptor and a 4N mixture of Ar-H₂ gas, and using low pressure pullers.
- 3) We have established that purity of the gas used for the experiments (4N or 6N) is not a limiting factor on the purity of a crystal grown in the low-pressure puller. This leaves the puller itself as the main cause of residual contamination. It is likely that repeated growth cycles in this puller would further reduce the contamination levels, but due to the limited funding such tests are left for future studies.

Table 1: Summary of the tests for growing high-purity dislocation-free Ge crystals.

Test	Puller pressure	Getter	Gas purity	Dislocation Free	Impurity (atoms/cm ³)	Dominant Dopant
1	High	No	4N	No	2×10^{13}	Ga
2	High	Yes	4N	No	7×10^{11}	P
3	Low	Yes	4N	Yes	2×10^{13}	Ga
4	Low	Yes	6N	Yes	3×10^{13}	Ga