HIGH-PURITY GERMANIUM: DETECTOR FABRICATION AND PERFORMANCE*

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Summary

The availability of germanium with less than 10^{10} acceptors/cm^3 has enabled us to fabricate and study the performance of germanium detectors of substantially larger volume than has hitherto been possible except by using lithium-drifting techniques. Detectors having a sensitive volume of up to 25 cm^3 have been made. These detectors have a lithium diffused n^+ contact and a metal barrier non-injecting back contact, and are totally depleted. The energy resolution of these detectors is similar to that of equivalent sized lithium-drifted detectors, but the high-purity detectors are easier to fabricate, and present considerably fewer handling problems.

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Introduction

During the past year we have seen high-purity germanium detectors advance from being an interesting plaything to the point where they are on the verge of competing directly, and successfully, against lithium-drifted germanium detectors.

Since lithium drifting is not necessary, these high-purity germanium detectors have the fundamental advantages of being simpler and quicker to fabricate, and they can be stored at room temperature without deterioration. These factors facilitate the construction of large area mosaics of detectors, or thick stacks suitable for measurements on high-energy particles.

Over the past year we have made a total of almost 100 detectors from 15 of the germanium crystals grown by W. L. Hansen at Lawrence Berkeley Laboratory.[1] The purpose of this paper is to report our experience with various fabrication methods, general handling problems and detector performance. The performance of several detectors will be presented to illustrate the patterns observed in this study.

Detector Fabrication

Nearly all the high-purity germanium crystals grown in our laboratory have been p-type. Consequently, almost all our detector development work has been devoted to working with p-type material, and this paper will be exclusively concerned with p-type detectors. Theoretically, the choice of typeness is irrelevant as far as the ability to deplete a planar diode is concerned. However, p-type

material appears to be easier to work with because n^+ contacts that will support high fields are more readily made than p^+ contacts--similar conclusions have been reached elsewhere.[2]

All the crystals have been grown in the [100] direction by the Czochralski method, and the detectors made from these crystals have been depleted parallel to the growth direction.

Three different planar detector shapes have been used:

a) Full area of the slice.

b) Deep groove cut into the wafer from the n^+ contact side to within about 1 mm of the p^+ contact.

c) Shallow groove, usually 1 mm deep, cut through the n^+ contact--these devices are then mounted in a guard-ring configuration with the outer region connected to a dc potential nearly equal to that of the central region.

Contacts

Initially the same techniques used for making contacts on lithium-drifted germanium detectors were used for high-purity germanium.[3,4] The n^+ contact was formed by lithium evaporation onto a lapped surface followed by a six minute diffusion at 400°C, and the metal barrier contact consisted of a gold evaporation onto an etched surface.

Although a number of good detectors were made using the Au contact, it would not consistently sustain a large overvoltage. Since development of a contact that would consistently allow the application of a significant overvoltage was imperative if high-purity germanium detectors were to become practical, we then tried a Cr evaporation.

This proved to be an improvement, and most of our detectors have a Cr back. We later tried a few Pt evaporations; these detectors all withstood very high overvoltage (> 1000 V), and additional Pt backs will be tried when electron-gun evaporating facilities are available. Recently we have also worked with a Pd-Ge back that appears to be very promising, especially in regard to producing an extremely thin entrance window. An Al diffused p^+ contact has also shown promise.

The lithium-diffusion temperature has now been reduced to 330°C because of an apparent anomalously deep lithium-diffusion tail that extends the n^+-p junction far into the germanium. When lithium is diffused at > 350°C, the effect of the diffusion tail is large in germanium having $\approx 10^{10}$ acceptors/cm^3 or less. The following case history illustrates the situation. A 1 cm thick detector, 155-10.5, from crystal #155 that contained 1 x 10^{10} acceptors/cm^3 as determined by a resistivity measurement, was made using a 375°C diffusion. Our standard measurements include placing a ^{207}Bi source inside the vacuum cryostat to determine the depletion voltage by observing the conversion-electron lines. For this detector we first observed the 976 keV electrons at a reverse bias of only 9 V! However, the capacity-voltage curve did not become flat until about 500 V, and the slope of the curve was much steeper than the expected $V^{-1/2}$ relationship. Furthermore, the effective detector volume, as measured by the intensity of γ-ray lines, continued to increase as the bias was increased up to at least 350 V. This result indicates that the n-p junction had been formed very near the back contact, and that the detector was depleting back toward the n^+ contact--in essence, we were working with n-type germanium after the lithium-diffusion. If this detector, and others

whose story is similar, were compensated by deep levels, usually ascribed to Cu diffusion, the resultant spectrometer would be expected to be extremely poor because of charge trapping. But with a bias of only 400 V this detector produced a resolution of less than 2 keV on the 1173 keV γ-ray of ^{60}Co. Furthermore, Cu acts as an acceptor in germanium, and it could only increase the residual acceptor concentration in the material.

To clarify the situation, detectors 155-9 and 155-11.5, both also 1 cm thick, were cut from crystal #155 on either side of 155-10.5. 155-9 was preheated to 400°C for six minutes under typical handling conditions, then cooled prior to a lithium-diffusion at 300°C for six minutes. 155-11.5 underwent a 300°C diffusion for six minutes without any prior heat treatment. Conversion electrons from ^{207}Bi first appeared at 325 V for 155-9, and 250 V for 155-11.5. Although the capacity-voltage curves did not become flat until about 500 V, the slope of the C-V curves for both detectors was precisely $V^{-1/2}$. These detectors thus behaved basically as expected, although it appears that even for a 300° lithium-diffusion the initial junction may be formed at a greater distance into the germanium than predicted on the basis of simple diffusion theory. Furthermore, both detectors provided resolutions on ^{60}Co γ-ray under 2 keV.

Since these two detectors exhibited essentially identical characteristics, the high temperature step by itself cannot account for the unexpected results observed on detectors that had undergone lithium-diffusion at relatively high temperatures (375°C). An anomalously deep lithium-diffusion tail is consistent with these, and other observations, but more study is necessary before any definitive conclusions can be made.

We have yet to see any deterioration of the lithium contact due to lithium precipitation in detectors that have been stored at room temperature for a year. Other laboratories have reported the same experience.[5] The only significant negative aspect of using the lithium contact is its thickness. This effective dead layer prevents the optimum stacking of detectors for counter telescopes.

General Detector Handling

Essentially the same procedures as used for lithium-drifted germanium detectors are employed by us for the high-purity germanium detectors. After the contacts have been made, the contact surfaces are protected with etch-resistant tape, and the exposed surfaces are etched for two minutes in 3:1 HNO_3:HF mixture (7 lbs HNO_3, 1 lb red fuming HNO_3, 2 lbs HF). The etch is quenched in methanol, which is blown off with nitrogen. After removing the tape, the device is mounted in the cryostat. A thin film of indium-gallium eutectic is spread on the n^+ surface to provide a good electrical contact.

Contrary to Llacer,[2] we have found surface state problems to be far less troublesome than for lithium-drifted germanium detectors. In fact nearly every case of high leakage current has been caused by electron injection at the metal barrier contact. Furthermore, the surfaces are surprisingly stable. Following successful testing, some detectors have been stored for periods up to several months at room temperature, then remounted successfully in a cryostat without any additional surface treatment. Apart from examples where the p^+ contact is damaged during handling, we have found no cases of detector degradation. Obviously, temperature cycling alone does not cause detector

degradation, and simply opening the warm cryostat to air, then repumping, has no effect.

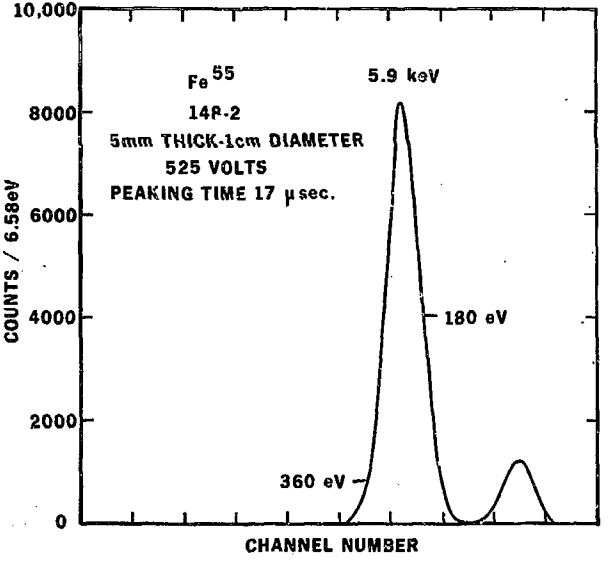
These statements may have to be tempered slightly when discussing ultra-high resolution small-detector spectrometers since a small change in leakage current can be very significant. Our experience with these systems is more limited (about 10 detectors), but voltage-current changes have not been severe, even in the 10^{-13} A region, although re-etching the surface has made a significant improvement on occassion.

Detector Performance

As examples of detector performance we will naturally put forth our most spectacular results, but these results are generally not unique.

Ultra-High Resolution Systems

Figure 1 presents the ^{55}Fe spectrum obtained with a 5 mm thick, 1 cm diameter detector that depleted at about 375 V. Even without overvoltage the entrance window was extremely thin; there were no more counts below the 5.9 keV line without overvoltage than the negligible number observed at higher voltages. This apparently is due to a built-in field that is characteristic of the Pd-Ge back. If a Au or Cr back had been used on this detector an overvoltage of at least 100 V, probably 200 V, would be necessary before the effective window thickness would have decreased to a minimum, and even then the background would probably be significantly higher than shown in Fig. 1.



XBL 7110-1566

Fig. 1. X-ray energy spectrum from a ^{55}Fe source. There were a negligible number of counts down to about 1 keV, where the spectrum was electronically cut-off.

This detector, mounted in the guard-ring configuration, had an effective capacity of about 4.4 pF. The 180 eV resolution for the 5.9 keV Mn X-ray, obtained with an amplifier peaking time of 17 μsec, is nearly equal to the best resolution obtained with a Si(Li) detector of equivalent area. Since the pulser resolution was 150 eV the effective Fano factor was 0.10. The same Fano factor is determined from the 470 keV resolution obtained with this detector for the 122 keV γ-ray of ^{57}Co.

The low-energy part of an ^{241}Am photon spectrum is shown in Fig. 2. Note the fairly intense Ge X-ray escape peaks. These peaks severely limit the usefulness of Ge spectrometers as analytical tools if photons in the energy region from 11.1 to about 30 keV are present.

Figure 3 shows a spectrum from the first significant physics experiment that we are aware of to use a high-purity germanium detector. A 4 mm thick - 18 mm diameter detector was used to obtain this kaonic X-ray spectrum of Cl. The data were accumulated during three consecutive days of continuous running of the Bevatron. X-rays from hydrogen-like atoms in which the electrons have been replaced by a kaon, a pion or a sigma are observed. This spectrum has a considerably lower background than any previous mesic X-ray spectrum. It is also the first and only spectrum to show a kaonic induced nuclear γ-ray, in addition to X-ray transitions from the three different types of exotic atoms.

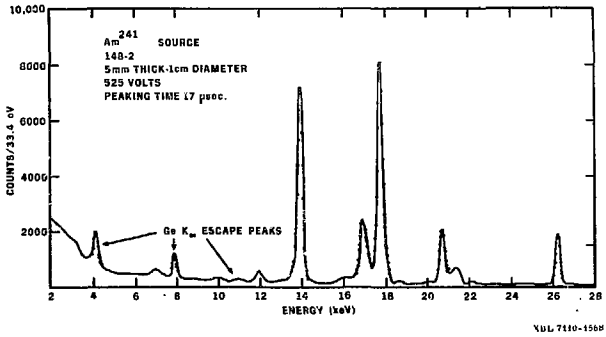


Fig. 2. Low-energy part of an ^{241}Am photon spectrum.

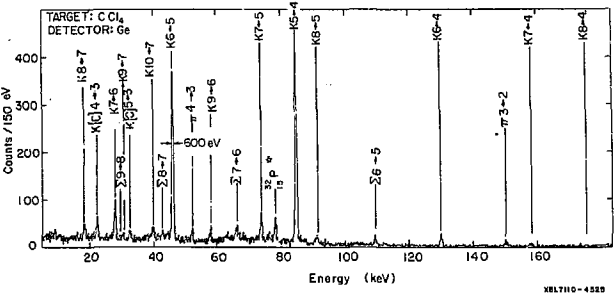


Fig. 3. Kaonic X-ray spectrum of Cl. X-rays from pionic and sigma-hyperonic atoms, and a kaonic induced nuclear γ-ray, were also detected.

Large-Volume Detectors

We have made a number of approximately 7 cm^3 detectors ($\approx$ 3 cm diameter - 1 cm thick). The performance of these devices was essentially equivalent to a good Ge(Li) detector of the same size. As expected, there has been no indication that the best high-purity germanium will provide better or worse resolution than the best lithium-drifted detectors.

The availability of germanium crystals having less than 10^{10} acceptors/cm^3 makes the fabrication of very large volume planar detectors feasible. Figure 4 shows the ^{60}Co spectrum from a 2 cm thick - 2.8 cm diameter detector that has a Pt back. The capacity of this 12 cm^3 detector is only 4.2 pF. Since depletion is reached at 1000 V the acceptor concentration is only 5 x $10^9/cm^3$.

The charge collection across such a long distance is excellent in this device. With no overvoltage (1000 V bias) the resolution for the 1173 keV ^{60}Co γ-ray is 2.6 keV, at 1500 V it is 2.0 keV, and at 3500 V it is 1.9 keV --slightly worse than the resolution presented in Fig. 4 at 3000 V because of additional noise.

The largest planar detector we have yet made is 25 cm^3, 2 cm thick - 4.0 cm diameter, from another crystal that had an acceptor concentration of only 5 x $10^9/cm^3$. Although the best resolution obtained with this device for the 1173 keV γ-ray is 2.7 keV at a bias of 2400 V, it is likely that fabrication problems degraded the performance to some degree. At the depletion voltage of 1000 V this detector exhibited considerable charge trapping; the resolution for the 1173 keV ^{60}Co γ-ray is about 5 keV, and the peak is quite asymmetric (FWO.1M$\approx$12 keV).

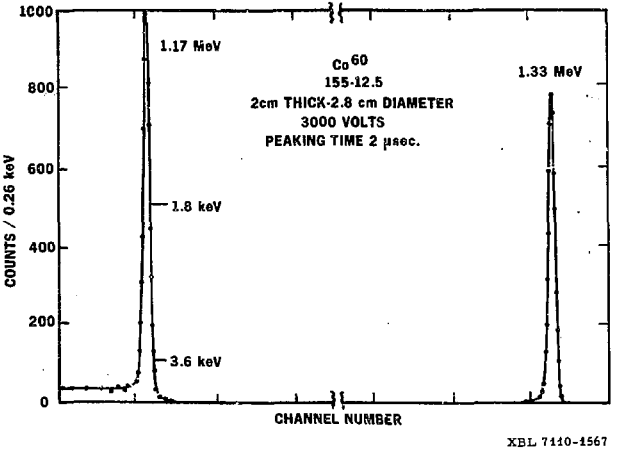


Fig. 4. ^{60}Co energy spectrum.

Charged Particle Detection

In December, 1970 a brief test of stopping 40-MeV protons from the 88-inch cyclotron was made. A proton spectrum obtained by placing a 6 mm thick - 1 x 1 cm area detector directly in the magnetically analyzed beam (protons were not scattered off a target) is presented in Fig. 5. The device was operated in the scattering-chamber vacuum, thus the protons did not pass through a window prior to impinging on the detector. Collimation of the incident protons limited the exposed area to about the center 4 x 6 mm of the detector. The energy spread in the beam itself supposedly was 10 keV. Since the electronic noise was about 7 keV the observed resolution for the entire system of 19 keV is only slightly worse than predicted.[6] Although we have obtained equally good resolution when using a lithium-drifted germanium detector in the same experimental arrangement, high-purity germanium detectors are generally much more practical for use with charged particles because of less handling difficulties.

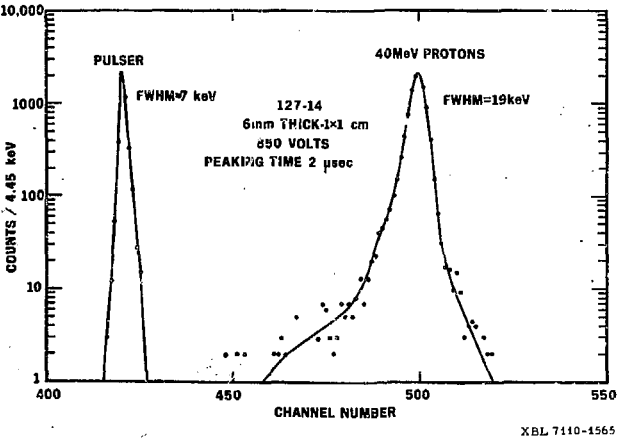


Fig. 5. Energy spectrum of 40-MeV protons.

Discussion

Significant charge trapping has not been observed in any detectors made from crystals grown in pure hydrogen, except in a few cases where fabrication techniques were proven to be at fault. However, detectors made from crystals grown in nitrogen invariably exhibited very severe trapping, and detectors made from a crystal grown in vacuum had significant trapping, while detectors made from crystals shown in forming gas showed slight trapping. Residual oxygen in the crystal-growing environment apparently creates charge trapping centers in the germanium.

Radial scans of several good detectors have been made by directing a collimated beam of conversion electrons from ^{207}Bi onto the p^+ side at different bias voltages. Since we found no variation of the electron peaks along the diameter there can be no significant radial change in the acceptor concentration. This observation is contrary to that reported by Sakai, McMath, and Fowler[7] on detectors made from high-purity germanium crystals grown by R. N. Hall at General Electric.[8] Whether this discrepancy is due to different fabrication techniques or to the different germanium-growing techniques is not known.

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