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Imaging of radio-carbon clusters in high-purity germanium using a self-detection scheme

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(Received

Abstract

A new isotope-specific and quantitative technique based on self-detection in a position-sensitive detector has been developed for the imaging of radio-carbon clusters in high-purity Ge. Using a novel position-sensitive detector concept, a one-dimensional spatial resolution of $\sim 200 \mu\text{m}$ FWHM and a sensitivity of $\sim 6 \times 10^7 {}^{14}\text{C}$ atoms per cluster has been obtained. This technique can be applied to other semiconductors which can be used to fabricate self-counting position-sensitive nuclear radiation detectors.

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A new technique for the study of impurity clusters in high-purity semiconductors is introduced in this letter. The advantage of the new technique is that it is isotope specific, highly sensitive, and can be used to determine the bulk distribution of the impurities and their clusters as well as the number of impurity atoms in each cluster. Application to the study of carbon clusters in high-purity Ge will be used to demonstrate the potential of the new technique.

The new method involves the use of a radioactive isotope of an impurity which is introduced into the semiconductor crystal via melt growth, epitaxial growth or other methods. A position-sensitive nuclear radiation detector is fabricated from the crystal. The impurity distribution can be determined in a highly sensitive and quantitative manner by measuring the individual decays of the radioactive impurity atoms inside the detector.

The prerequisites for applying such a technique are 1) that the semiconductor material under study can be used to fabricate a position-sensitive radiation detector, and 2) that the decay products of the radioisotope of the impurity atom have a short range in the host semiconductor (e.g. alpha particles or low energy β -rays). The former requirement is readily met by germanium and silicon which are routinely used in making various kinds of nuclear radiation detectors including position-sensitive ones. Other semiconductors such as CdTe, HgI₂ and epitaxial GaAs have also been exploited as nuclear radiation detectors. Although less developed as detector materials compared to Ge and Si, these compound semiconductors can, in principle, be studied using this technique. In the present study, a novel position-sensitive Ge detector concept has been used. The new concept may be adaptable to other semiconductors as well.

The use of a radioactive tracer technique based on self-counting detectors to study the carbon concentration in high-purity Ge has been reported previously by our group.¹ In that experiment, Ge crystals were grown out of a ^{14}C spiked pyrolytic graphite-coated silica crucible under a hydrogen or a nitrogen atmosphere. A second generation crystal was also grown using one of the hydrogen-grown crystals as charge under standard conditions for producing detector grade materials (bare silica crucible, hydrogen atmosphere). Standard (nonposition-sensitive) nuclear radiation detectors were made from these crystals and the bulk carbon concentration was determined by measuring the count rate of internal β decays. Autoradiography studies using x-ray film have shown that part of the carbon atoms exist in clusters. First generation hydrogen-grown crystals showed a large number of clusters while the second generation crystal contained much less.¹ However, the absolute number density and sizes of the clusters could not be determined quantitatively. The use of position-sensitive detectors is an extension of this earlier technique and has enabled us to quantitatively measure the carbon cluster size and distribution.

The position-sensitive detector used in the present experiment is based on the novel concept of semiconductor drift chambers.² Position information is obtained by measuring carrier drift times. As originally proposed, detector contact segmentations and a means of producing differential potentials on the contact elements are required to create a drift field. A much simplified approach was developed using Ge in which a built-in drift field is automatically produced through the use of material with an impurity concentration gradient.³ A further modification of this idea is used in the present detector which allowed the use of material with uniform impurity concentration.

Figure 1a shows schematically the geometry of the detector. It has a planar $n^+ - p - n^+$ structure which is tapered toward one end, away from the p^+

contact. The opposing n^+ contacts are held at the same DC potential. With reverse bias applied between them and the p^+ contact, depletion starts from both n^+ contacts and moves toward the midplane of the detector. The potential distribution inside the detector at full depletion is shown in Fig. 1b. For a linearly tapered structure with local thickness $L(x) = L_0 - kx$, the resulting drift field along the detector midplane is:

$$E_d(x) = \frac{dV_w(x)}{dx} = -\frac{eN}{4\epsilon} k(L_0 - kx)$$

where $L(x)$ is the local thickness of the detector, N is the net electrically active impurity concentration, e is the electronic charge and ϵ is the permittivity. After electron-hole pairs are generated inside the detector by an ionizing event, there exists a first phase of rapid carrier collection as the electrons move to one n^+ contact and the holes move toward the midplane. As the holes approach the midplane, they change direction along the field lines and start to drift slowly along the midplane to the p^+ contact under the relatively weak transverse drift field, $E_d(x)$. The drift time for carriers originated at position x in the midplane of the detector is:

$$\tau_d(x) = \int_0^x \frac{dx'}{\mu |E_d(x')|} = \frac{4\epsilon}{\mu e N k^2} \ln \frac{L_0}{L_0 - kx}$$

where μ is the carrier mobility. Figure 1c illustrates qualitatively the development of electric charge signals at the three contacts by the movement of the carriers. The initial, rapid charge collection gives rise to prompt signals at the two n^+ contacts. As the holes move along the midplane, a negligibly small signal is induced because the moving charges are effectively shielded by the n^+ contacts. Upon close approach to the p^+ contact, the induced charges rise sharply leading to signal pulses on each of the contacts. The time difference between the prompt pulses and the delayed pulses is the

carrier drift time. It can be used to compute the position of the interaction. In practice, since the drift time is sensitive to variations in the impurity concentration along the detector and in the carrier mobility which has a strong dependence on temperature, a calibration can be used to accurately determine the actual drift time versus position relationship. The total charge induced at the p^+ contact is proportional to the energy deposited in the detector by the radiation. For n-type material, the above description is still valid except the contacts and the carrier types are exchanged. In this case it is the electrons' drift along the detector midplane which is used to determine the position.

A drift chamber with this geometry has been fabricated from a p-type slice of the second generation ^{14}C -doped Ge crystal which has a net acceptor concentration of $5 \times 10^{10} \text{ cm}^{-3}$. The detector is 3 cm long, 2 cm wide and 1 mm to 3 mm thick. N^+ contacts were formed by Li diffusion and the side electrode was a Pd Schottky barrier. During operation, the detector is placed in a cryostat and cooled by liquid nitrogen. Three charge-sensitive amplifiers are connected to the three contacts. Their input junction field effect transistors and feedback components are mounted near the detector and are cooled for best noise performance. The prompt and delayed signals from the amplifiers trigger a time-to-amplitude converter (TAC) whose output is fed to a pulse height analyzer (PHA) to obtain time spectra. Energy signals from the side contact amplifier can be used to gate the TAC so that only signals within a certain selected energy window are accepted. Since the polarity of the prompt signals depends on which side of the detector the carriers are produced, signals from the two halves of the detector can be distinguished and examined separately. The detector is fully depleted at 70 V and is operated at 120 V. The slight over voltage will not affect the internal drift field significantly except near

the side contact where the drifting carriers will be collected more rapidly thus decreasing the rise time and improving timing.

Figure 2a shows the time spectrum of a scan along the detector (i.e., parallel to the drift direction) at 2 mm intervals using a collimated ^{241}Am 60 keV γ -ray source. The result demonstrates the position-sensitive properties of the detector and serves as a position vs drift time calibration. The width of the peaks are dominated by the source spot size which is ~ 0.5 mm in diameter. Scanning the full area of the detector indicated small dead regions near the bare sides; this is attributed to surface channel effects. Also, due to the low drift field, carrier trapping effects are dramatically revealed. The energy signal amplitude decreases as the drift distance increases, first gradually then abruptly near the far end where no measurement of the drift time can be made. The total effective detector area is limited to a width of 1.3 cm with a maximum drift distance of 2.2 cm.

Figures 2b and 2c are time spectra obtained with the detector in a self-counting mode to detect internal ^{14}C β decays. Counting time was 4.6 days each. This time has to be contrasted to typical one-month exposures with x-ray film. An energy window of 30 - 100 keV was used to include the main portion of the β continuum which has an end point energy of 155 keV¹ and cut down the contributions from background radiation. The two spectra correspond to the two halves of the detector. The peaks in the spectra are evidently due to local concentrations of ^{14}C , i.e., carbon clusters. Spectra obtained with the energy window set above the end-point energy showed only a continuum distribution arising from background radiation and no peaks are detected. Since the average range of the maximum energy β 's in Ge is < 0.1 mm, the ~ 0.2 mm full-width-half-maximum (FWHM) of the peaks represents the position resolution of the system for radiation in this energy range. A total of 50 clusters or a

number density of $\sim 130 \text{ cm}^{-3}$ are detected. The number of counts under each peak is directly related to the number of ^{14}C atoms in a particular cluster. From this number, the total number of carbon atoms in each cluster was calculated knowing the half-life of ^{14}C and the ratio of C to ^{14}C (11.3:1) which was used in our crystal growth experiments. The fraction of the β energy spectrum falling within the electronically determined energy window was 58% and was used in the computation of the number of the carbon atoms in each cluster. The result is summarized in Fig. 3 which shows the distribution of the number of carbon atoms in each cluster along the detector. For multiple peaks that are not fully resolved, estimates were made of the ratio of the components and the contributing clusters' carbon contents. The number of clusters with carbon content lying in intervals of 5×10^9 atoms is also plotted showing the carbon content distribution. The smallest carbon content in a cluster that can be unambiguously recognized from the spectra is $\sim 4 \times 10^9$ atoms. In order to lower the detection limit, the number of peaks in the time spectrum has to be reduced so that they are well separated and small peaks can be readily seen. To accomplish this, the p^+ contact was divided into three segments with a 4 mm long middle section. With the outside sections grounded, only the middle 4 mm strip of the detector contribution counts. Figure 2d is the resulting spectrum accumulated over a period of 24 days. No cluster is seen with a carbon content $< 2 \times 10^9$ atoms down to the new limit of $\sim 7 \times 10^8$ atoms ($\sim 6 \times 10^7$ ^{14}C atoms). The continuum of the spectra is mainly due to dispersed and presumably atomically dissolved carbon. The decrease in count rate toward longer drift times is due to the tapered detector structure and the increase of drift time per unit distance toward the far end of the detector. Contributions from background radiation, which constitute only a small fraction of the continuum, are subtracted assuming that the background is constant within and outside the

^{14}C β energy distribution. This gives a total carbon concentration of $1.4 \times 10^{13} \text{ cm}^{-3}$ which agrees with the value obtained previously for this crystal¹. The ratio of carbon atoms in clusters to dispersed carbon is $\sim 1:5$.

In summary, a new technique for studying impurity clusters in semiconductors has been introduced. The technique has been applied successfully to the study of carbon clusters in a high-purity germanium crystal. The position-sensitive detector described is of a novel design which can also be applied to a variety of other applications. On the other hand, for semiconductors with a substantially higher concentration of impurities and trapping centers, more conventional types of position-sensitive detectors such as discrete diode arrays read out individually may be more easily realized for use in this application.

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Figure Captions

- FIG. 1. (a) Structure of the semiconductor drift chamber.
(b) Potential distribution inside the detector at full depletion.
(c) Schematic representation of the signal currents and the integrated charges induced at the three contacts arising from the movement of electrons ($-Q$) and holes ($+Q$) deposited at a point inside the detector.
- FIG. 2. Time spectra of (a) a scan along the detector using a collimated ^{241}Am 60 keV γ -ray source at 2 mm intervals, (b) and (c) internal decays in the two halves of the detector, and (d) in a 4 mm wide strip of the detector.
- FIG. 3. Distribution of carbon clusters along the length of the detector (a), and their carbon content distribution (b).

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Fig. 1.

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Fig. 2.

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Fig. 3.

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