Effects of Gamma Radiation on Germanium

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High-purity n- and p-type samples of Ge have been exposed to γ rays from a 900-curie Co^{60} source. The extrinsic electron concentration of n-type material decreases at a rate only $\sim 10^{-3}$ of that for fast-neutron irradiation. Two electron trapping levels which are apparently identical to those produced by fast neutrons have been observed. Extended exposure converts n-type material to p-type, indicating the presence of an acceptor level at ~ 0.26 ev above the valence band. This energy is somewhat larger than that found in material irradiated by fast neutrons. Interstitial clustering and nonuniform defect distribution are believed to lower the energy levels produced by fast neutrons; hence energetic photons apparently produce randomly distributed defects throughout the volume of the specimen. Exposure of p-type material causes a decrease in extrinsic hole concentration with a removal rate much less than that for n-type Ge. The cross section for atomic displacements obtained from these results is $\sim 1.5 \times 10^{-26}$ cm^2. This value is in reasonable agreement with the calculated cross section expected for the Compton electrons and photoelectrons produced by Co^{60} γ-ray absorption in Ge.

INTRODUCTION

IT is well known that fast-particle bombardment of solids is capable of altering to a marked degree certain of their physical properties. These so-called radiation effects result from the production of lattice defects by collisions of the nucleons with lattice atoms and the influence of the defects on the defect-sensitive physical properties.[1]

Recently Dugdale[2] has presented evidence that high-energy photons (Co^{60} γ rays) enhance the ordering rate of disordered Cu_3Au in much the same manner but to a considerably less degree than that resulting from fast-neutron bombardment.[3] He explains this behavior as arising from the generation of atomic displacements by energetic Compton electrons and photoelectrons which result from γ-ray absorption in the alloy. These resultant defects enhance the diffusion rate and the rate of ordering is thereby increased.

We report here the results of studies of the effect of γ radiation on the electrical properties of Ge. This material was chosen because of its very great sensitivity to fast-particle bombardment. In high-purity specimens, whose extrinsic carrier concentrations are in the neighborhood of 10^{13} cm^{-3}, the presence of as few as 5×10^{11} displaced atoms per cm^3 may be detected in good precision from Hall coefficient and resistivity measurements. Five years ago, at the suggestion of K. Lark-Horovitz, we attempted to observe γ-ray effects in Ge but the purity of available material at that time was too poor and the intensity of the Co^{60} source used was too low to yield positive results.

RESULTS

Both n- and p-type specimens cut in the form of Hall plates ($10\times 3\times 1$ mm) were exposed to γ rays (1.17 and 1.33 Mev in equal intensity) from a 900-curie Co^{60} source. The temperature of exposure was $\sim 20°$C. Before irradiation and after subsequent exposures the resistivity ρ and Hall coefficient R were measured as a function of temperature from liquid nitrogen to room temperature. The photon flux was estimated from both geometrical considerations and water decomposition measurements,[4] the two methods yielding values in satisfactory agreement with each other.

Exposure of n-type Ge to γ rays decreases the extrinsic electron concentration at an initial rate of $\sim 1.4 \times 10^{-3}$ electron per photon at liquid nitrogen temperature (a rate only about 10^{-3} as great as for fast neutrons). In a high-purity specimen (2×10^{13} electrons/cm^3) rapid saturation is apparent, but in a more impure sample (7×10^{14} electrons/cm^3) the effect is approximately linear out to an exposure of 5×10^{18} photons/cm^2. The high-purity specimen has been converted to p-type by γ radiation, and the results obtained are summarized in Fig. 1 in which log R is plotted against reciprocal temperature after several of the exposures. Curve I, obtained before exposure, exhibits the expected behavior for a high purity specimen. After 1.6×10^{16} photons/cm^2, Curve II exhibits the existence of two electron-trapping levels, a deep one which is completely occupied throughout the entire extrinsic range and a shallow state which is ionized at the higher temperature but completely occupied at low temperature. The shallow level is responsible for the step in the extrinsic portion of Curve II. A closer examination of curves of this type in a range where the concentrations of exposure-produced levels are not sufficient to remove all of the chemical donor electrons reveals that the two levels are produced in approximately equal concentration. After additional bombardment, the shallow level controls the temperature dependence of the extrinsic electron concentration, as shown by Curve III. The slope of this curve indicates that the shallow level

[1] See, for example, G. H. Kinchin and R. S. Pease, Repts. Progr. in Phys. 18, 1 (1955).
[2] R. A. Dugdale, *Report of Bristol Conference on Defects in Crystalline Solids* (The Physical Society, London, 1955), p. 246.
[3] T. H. Blewitt and R. R. Coltman, Phys. Rev. 85, 384 (1952); Acta Metallurgica 2, 549 (1954).
[4] We are indebted to C. J. Hochanadel of this laboratory for this calibration.

lies ~0.2 ev below the conduction band edge. These findings are virtually identical with those for fast-neutron bombardment of *n*-type Ge.[5]

Further exposure converts the specimen to *p*-type and the slope indicates the presence of an acceptor (presumably the deep electron trap) at ~0.26 ev above the valence band edge. This acceptor ionization energy is somewhat larger than that estimated for fast-neutron bombardment (~0.18 ev) but the difference may arise from the nonuniform distribution of defects and a tendency for clustering of interstitials which is indicated for reactor-irradiated specimens.[5,6] This suggestion is supported by the low-temperature annealing behavior of the γ-irradiated specimen. After 90 min at 120°C, the sample of Fig. 1 showed a marked decrease in Hall coefficient toward low temperature and the slope of the curve indicates an acceptor ionization energy of ~0.16 ev.

These results suggest that γ radiation indeed produces lattice defects in Ge. Moreover, the correspondence of these states with those of neutron-bombarded specimens and the apparent absence of complications expected from clustering and nonrandom distribution of defects would strongly suggest that individual interstitial-vacancy pairs are uniformly produced throughout the volume of the specimen.

The behavior of *p*-type Ge, though not so clear-cut as high-purity *n*-type specimens, is consistent with expectation based on the *n*-type results. Exposure decreases the hole concentration over the entire extrinsic range for a specimen whose initial concentration is $\sim 1\times 10^{13}$ cm^{-3} and the decrease saturates after prolonged exposure. The rate of carrier removal and the total decrease after saturation is considerably smaller than for *n*-type Ge. In view of the large ionization energy of the bombardment produced acceptor level and the expected presence of hole traps,[6] a hole concentration decrease is expected in this temperature range (77 to 200°K). After appreciable saturation, the hole concentration was considerably greater in magnitude and possessed a much smaller temperature dependence than the converted *n*-type specimen (Curve V, Fig. 1).

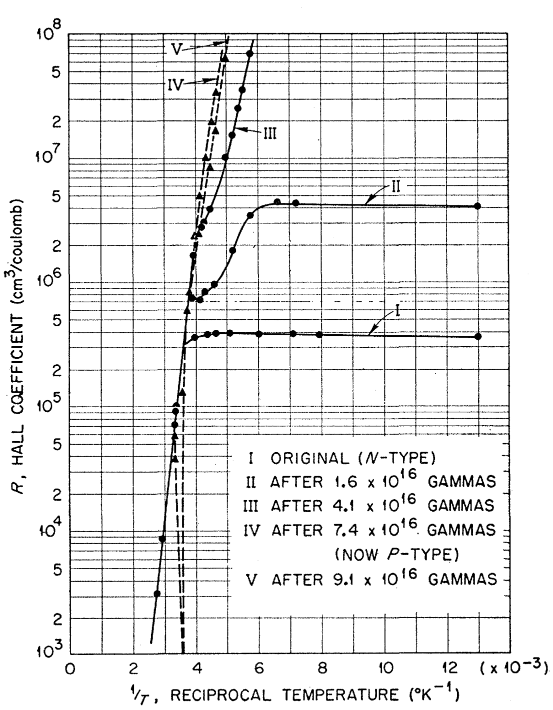

FIG. 1. Log of Hall coefficient *vs* reciprocal temperature of initially high-purity *n*-type Ge after successive exposures to Co^{60} γ radiation.

DISCUSSION

If it is assumed that two electrons, one for each type of trap, are removed at liquid nitrogen temperature for each displaced atom, the cross section σ_γ for displacing atoms by Co^{60} photons in Ge may be readily obtained from the initial rate of electron removal. The value of σ_γ has been found to be $\sim 1.5\times 10^{-26}$ cm^2. The observed σ_γ is evidently the sum of the contributions resulting from the various γ-ray absorption processes. Since at these photon energies (1.17 and 1.33 Mev) pair production is negligible and since the cross section for direct interaction of the γ ray with the nucleus is very small, the effect must be caused primarily by the Compton and the photoelectric processes.[2] It should be mentioned that the mechanisms suggested by Seitz[7] and Varley[8] to account for photon generation of defects in ionic crystals do not seem applicable to Ge. Evaporation of defects from dislocations by an exciton energy-transfer process [7] does not seem probable because of the small exciton energies in Ge. Moreover, Varley's mechanism, which depends on the ejection of a multiply ionized anion from its lattice site by the crystal potential, is not expected to operate in a homopolar, covalent crystal.

Compton electrons and photoelectrons with energies greater than a threshold value, i.e., sufficient energies to transfer more than the necessary displacement energy E_d to a lattice atom, can displace atoms by Coulombic interaction with the nucleus. In this regard, disordering of solids by γ rays is essentially the same as by electron bombardment[9] except for two factors: (1) a distribution of electron energies results from the Compton process and (2) lattice damage can be produced uniformly throughout a relatively thick specimen by γ-ray bombardment but only within the short electron range by electron bombardment. The first factor renders this method of defect introduction relatively ineffective for precise determinations of E_d, but the second is quite desirable experimentally since this permits introduction

[5] Cleland, Crawford, and Pigg, Phys. Rev. **98**, 1742 (1955).
[6] Cleland, Crawford, and Pigg, Phys. Rev. **99**, 1170 (1955).
[7] F. Seitz, Revs. Modern Phys. **26**, 7 (1954).
[8] J. H. O. Varley, J. Nuclear Energy **1**, 130 (1954).
[9] E. E. Klontz, Phys. Rev. **86**, 643 (1952).

of a uniform distribution of Frenkel defects for bulk property studies without the limitation of sample dimensions by electron range. Kinchin and Pease[1] have suggested an additional process, namely, atomic displacement by recoil during photoelectron ejection. For photon energies involved here however, the electrons are ejected predominantly in the forward direction with respect to the absorbed photon[10] and hence the momentum transferred to the emitting atom is usually so small that the recoil energy is less than the displacement energy in the case of Ge.

The necessary information for calculating the contributions of the Compton electrons and photoelectrons to σ_γ are summarized by Bethe and Ashkin.[11] Using the expression for the electron displacement cross section based on the theory of Mott[12] as modified by McKinley and Feshbach[13] and Dugdale and Green,[14] assuming an E_d of 25 ev, and integrating over both the energy distribution and range of electrons, one obtains a total value of σ_γ for both processes of $\sim 1.3\times 10^{-26}$ cm^2. In view of uncertainties in photon flux and the approximations in the calculation, this value is considered to be in satisfactory agreement with the observed σ_γ. A recent determination of E_d for Ge indicates that 23 ev is perhaps its upper limit.[15] This smaller value of E_d would increase the calculated σ_γ by several percent.

An interesting consequence of the calculation is that, contrary to expectation, the contribution of the photoelectrons is found to be approximately the same as that of the Compton electrons. This results from the fact that the electron displacement cross section increases rapidly with energy and the energy of photoelectrons is essentially that of the absorbed photon whereas the effective Compton electrons have energies ranging from the threshold value to a maximum which is appreciably less than the photon energy (0.54 to 1.12 Mev).

ACKNOWLEDGMENTS

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[10] A. Hedgran and S. Hultberg, Phys. Rev. **94**, 498 (1954).

[11] H. A. Bethe and J. Ashkin, *Experimental Nuclear Physics* edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. 1, p. 166.

[12] N. F. Mott, Proc. Roy. Soc. (London) **A124**, 425 (1929); **135**, 429 (1932).

[13] W. A. McKinley and H. Feshbach, Phys. Rev. **74**, 1759 (1948).

[14] R. A. Dugdale and A. Green, Phil. Mag. **45**, 163 (1954).

[15] J. J. Loferski and P. Rappaport, Phys. Rev. **98**, 1861 (1955).

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Free-Radical Quenching of Positron Lifetimes*

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The annihilation lifetime of positrons stopping in benzene has been measured as a function of the percentage of added free radical, diphenylpicrylhydrazyl. The long component of the complex annihilation curve quenches from 2.67×10^{-9} sec in pure benzene to 5.3×10^{-10} sec at a 3% free radical concentration. An annihilation model following Bell and Graham's assumptions is discussed, postulating the partial formation of bound positron states prior to annihilation. The long lifetime is regarded then as a measure of the conversion rate from triplet to singlet states due to collisions of the positron system with the unpaired electrons of the free radical. The experimental points yield, using such a model, a conversion cross section of $\sigma = 1.18\times 10^{-17}$ cm^2.

INTRODUCTION

THE first detailed study of the lifetimes of positrons in solids and liquids was reported by Bell and Graham.[1] Their result can be summarized as follows:

In metals and crystalline solids, positrons annihilate with a single mean lifetime $\tau_1 = (1.5\text{–}2)\times 10^{-10}$ sec. This short lifetime seems to be almost independent of the particular material used. In some amorphous nonmetals, and in most liquids, about 70% of the two-photon annihilation proceeds via a short lifetime of the order of τ_1 in metals, and the rest via a longer "anomalous" component, τ_2 ($\tau_2 \cong 2\times 10^{-9}$ sec). The τ_2 decay time is sensitive to the thermodynamical parameters of the solid material, such as temperature, phase, and order-disorder.

The exact nature of this anomalous lifetime is still not well understood.

Bell and Graham postulate the formation of bound states (similar to positronium in gases) in order to explain the behavior of the τ_2 component. This lifetime can be regarded then as a measure of the conversion

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† Alfred P. Sloan Research Fellow, 1955–1956.

‡ National Science Foundation Predoctoral Fellow, 1955–1956.

[1] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).