Low-Temperature Length Change Measurements of Electron-Irradiated Germanium and Silicon*

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Precision measurements of the change in length of high-purity germanium and silicon were made upon 2-Mev electron irradiation and annealing. Two germanium samples were irradiated at maximum temperatures of 365°K and 86°K, respectively. The first sample was irradiated at 33 μa/cm^2 to an integrated flux of 5.4×10^{19} electrons/cm^2. The specific length expansion obtained was $\Delta L/L=(6\pm12)\times10^{-26}$ per 2-Mev electrons/cm^2. The 86°K sample was irradiated at 10 μa/cm^2 to an integrated flux of 3.0×10^{19} electrons/cm^2. The specific length expansion was $(1.5\pm3.9)\times10^{-25}$ per 2-Mev electrons/cm^2. Using the Seitz-Koehler simple theory of displacement ($E_d=30$ ev) and assuming no annealing at the low temperature, we calculate a concentration of displaced pairs equal to 1.6×10^{-3} with a corresponding fractional atomic volume change per interstitial-vacancy of $(f_i+f_v)_{Ge}=0.008\pm0.021$. The silicon sample was initially irradiated at ~50°K to an integrated flux of 8.3×10^{18} electrons/cm^2 and further irradiated at ~115°K to a total integrated flux of 7.9×10^{19} electrons/cm^2 and then annealed to room temperature. The specific length change for both bombardments was $\Delta L/L=(4\pm19)\times10^{-26}$ per 2-Mev electrons/cm^2. The simple theory ($E_d=28$ ev) predicts a concentration of displaced pairs of 4×10^{-3} with a corresponding volume change per interstitial-vacancy of $(f_i+f_v)_{Si}=0.002\pm0.011$. These values are much smaller than would be expected from previous results for deuteron and neutron irradiations of germanium. Calculations of defect cluster size distributions for electron, deuteron, and neutron irradiations of germanium are given and combined with the experimental results to indicate that the volume change per defect is nonlinear and increases as the cluster size increases. The samples were not observed to expand within the presumed error. All measurements did, however, yield a small positive value. The values for Ge and Si are very small compared with the large expansions recently observed for electron-irradiated InSb and GaAs.

I. INTRODUCTION

THE detailed nature of the damage introduced in germanium and silicon crystals by fast-particle irradiation is at present not clear. It is generally agreed that the complexity of the damage increases as one goes from electron to deuteron to neutron bombardments, with the simpler more isolated defects produced by electron irradiation. The purpose of the present experiment was to (1) measure the change in volume of high-purity germanium and silicon upon electron irradiation at low temperatures and above, (2) to determine the specific volume strain per isolated displaced atom in these crystals, (3) and to compare the results of electron bombardment with the previous results for deuteron and neutron bombardments.

The importance of temperature in the determination of the defect configuration was recognized. At sufficiently low temperatures, defects produced on irradiation are not mobile and will not recombine or associate either with each other or impurities[1–3] as they do at higher temperatures. One would then think that the production of primary defects, i.e., defects produced on bombardment, would be larger at a lower rather than at a higher temperature. On the other hand, data have been published[1,2,4] which show that the introduction rate of certain energy levels in electron-irradiated germanium and silicon is larger at a *higher* temperature. This may be explained by the fact that these levels may be attributable to the association of defects with each other or impurities which will take place only at the higher temperature. Such levels, then, would not be related to primary defects. Watkins[1] has definitely established that the silicon A center, whose introduction rate is much larger at room temperature than at low temperature, consists of a vacancy associated with an oxygen atom, and that a vacancy is mobile at room temperature.

The temperature dependence of the introduction rate may also be explained by a model of the displacement process which directly predicts an increase in the production rate of stable primary defects at a higher temperature. The latter has been suggested by Wertheim[5] in a particular model of the potential energy vs distance near the displacement site. Glen,[6] Loferski and Rappaport,[7] and Brown[4] have suggested that the additional vibrational momenta of atoms at the higher temperature may aid displacement.[8] The phenomenon of a higher introduction rate at higher temperature has been observed in electron irradiation by Brown[4] in germanium, and by Wertheim[2] in silicon, measuring conductivity and Hall coefficients. It has also been observed by Bemski[9] and Watkins[1] with spin resonance

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] G. D. Watkins, J. W. Corbett, and R. M. Walker, J. Appl. Phys. **30**, 1198 (1959). G. D. Watkins and J. W. Corbett, Phys. Rev. **121**, 1001 (1961).

[2] G. K. Wertheim and D. N. E. Buchanan, J. Appl. Phys. **30**, 1232 (1959).

[3] W. L. Brown, W. M. Augustyniak, and T. R. Waite, J. Appl. Phys. **30**, 1258 (1959).

[4] W. L. Brown and W. M. Augustyniak, J. Appl. Phys. **30**, 1300 (1959).

[5] G. K. Wertheim, Phys. Rev. **115**, 568 (1959).

[6] J. W. Glen, Phil. Mag. Suppl. **4**, 381 (1955).

[7] J. Loferski and P. Rappaport, Phys. Rev. **111**, 432 (1958).

[8] W. Brown has also suggested the possibility that a high level of ionization in the sample during bombardment aids the displacement process. Data of the introduction rate of acceptors in Ge vs electron beam current indicate a rise for large beam currents. J. Kortwright (private communication) has confirmed a positive linear beam current dependence, but the cause does not appear to be due to ionization. We did not observe such a dependence.

[9] G. Bemski, J. Appl. Phys. **30**, 1195 (1959).

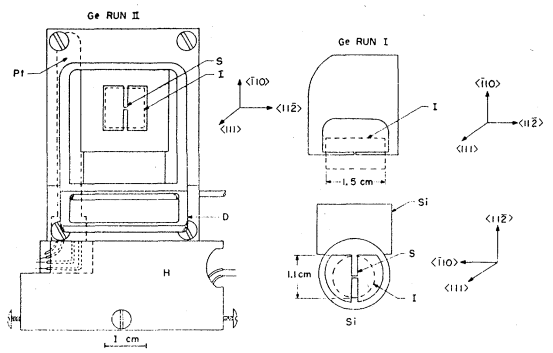

FIG. 1. Sample geometries; *S*, sample arms: *I*, irradiated area: *H*, heater: Pt, platinum resistance thermometer.

in electron-irradiated *n*-type silicon. It has not been observed in deuteron- or neutron-irradiated Ge. This apparently agrees with Brown's observation in germanium that for higher energy electrons around 1 Mev there is no temperature dependence of the introduction rate between 79° and 263°K.

It seems necessary, therefore, in order to resolve the role of temperature in the production of defects from its role in the association of defects, to simultaneously consider purity. Impurities have long been of interest in irradiation damage studies of metals,[10] but their neglected role in the purer semiconductors has increasingly become evident.

In principle then, one of the most desirable experiments would be one of high sensitivity, performed at low temperatures on electron-irradiated high-purity material. At room temperature electrical measurements have high sensitivity. Unfortunately at low temperature, the resistivity of pure Ge and Si becomes too large to measure, restricting electrical studies to impure materials.[11] Even at liquid nitrogen temperatures, impurities of as little as $2\times10^{15}/\text{cm}^3$ affect the annealing of electron-irradiated *n*-type germanium.[3] Trapping of carriers in nonequilibrium states is observed at these temperatures and may be expected to complicate the interpretation of electrical measurements. Measurements of volume changes, while not having the sensitivity of electrical measurements, have been made for deuteron-irradiated copper[12,13] and germanium[14] down to near liquid helium temperature and yield valuable information about the damaged state. Volume changes of neutron-irradiated Ge have been made above room temperature,[15] and generally seem to indicate larger effects than those seen for deuteron irradiation although the concentration of defects is very uncertain due to extensive annealing. No volume change measurements of electron-irradiated semiconductors have yet been published.[16] The present paper presents the results of electron irradiation of high-purity germanium and silicon at temperatures of 86°K and ~50°K, respectively.

[10] T. H. Blewitt, R. R. Coltman, C. E. Klabunde, and T. S. Noggle, J. Appl. Phys. **28**, 639 (1957).
[11] J. W. MacKay and E. E. Klontz, J. Appl. Phys. **30**, 1269 (1961).
[12] R. Vook and C. Wert, Phys. Rev. **109**, 1529 (1958).
[13] R. O. Simmons and R. W. Balluffi, Phys. Rev. **109**, 1142 (1958).
[14] F. Vook and R. W. Balluffi, Phys. Rev. **113**, 62 (1959): **113**, 72 (1959).
[15] M. C. Wittels, J. Appl. Phys. **28**, 921 (1957). M. C. Wittels, Bull. Am. Phys. Soc. **5**, 375 (1960).

II. EXPERIMENTAL

Length change measurements were made on two germanium samples and one silicon sample. Three completely separate irradiation runs were made for the three samples. The experimental techniques used were similar to those described in an earlier paper,[14] and the modifications used in the present work are described below.

1. Apparatus

The irradiations were performed in vacuum using a 30-liter helium cryostat constructed to our specifications by the Superior Air Products Company. The cryostat consists essentially of a liquid helium inner Dewar surrounded by a liquid nitrogen jacket. Liquid helium was used in the inner Dewar for the first part of the silicon run. For the two germanium runs pumped nitrogen (~50°K) was used in the inner Dewar. The samples were soldered to a nickel-plated molybdenum block mounted at the end of the inner Dewar. The block and specimens were in good thermal contact with the inner reservoir and conduction cooled. A copper liquid-nitrogen radiation shield immediately surrounded the specimen block assembly. A set of foils and doors in the nitrogen shield and specimen block admitted the electron beam and allowed the length measurement. The electron beam entered the sample chamber which was bolted to the Van de Graaff accelerator, passed through the specimen, and was stopped in the nitrogen radiation shield which was electrically insulated from the outer manifold thereby forming a Faraday cage. It was possible to measure the electron current during

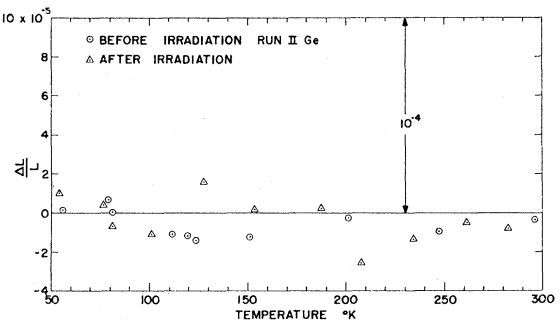

FIG. 2. Unirradiated thermal background curve and annealing, Run II Ge.

[16] Preliminary accounts of the data presented here were given in Bull. Am. Phys. Soc. **5**, 507 (1960) and **6**, 344 (1961); see also Bull. Am. Phys. Soc. **6**, 420 (1961).

bombardment, and the charge accumulated was integrated using an Elcor Model A309A current integrator. The irradiated area was defined by collimating the beam. For the germanium samples the beam was uniformly scanned electromagnetically over the area within the dashed lines in Fig. 1. For the silicon run, which was an earlier experiment, small-angle scattering from a foil in the nitrogen shield was used to spread the beam over the indicated collimated area. This resulted in a less uniform irradiation than in the germanium runs. The average irradiation flux quoted for the silicon case was found by measuring the total current through the collimated area and mathematically integrating the distribution over the subtended sample area using calculations for the small-angle scattering. The irradiations in the germanium runs were uniform, and no corrections were necessary.

The bombardment temperatures quoted are to be interpreted as the maximum sample temperatures at the tips of the specimen. Since the length measurements were made to a fraction of a wavelength of visible light, the samples had to be kept stress free. It was, therefore, impossible to attach a thermocouple to the sample. The temperatures on bombardment of the silicon sample were calculated using the measured values of the beam current, incident beam energy, sample block temperature, sample geometry, published values for the thermal conductivity of silicon, and the energy loss of the electrons passing through the sample. The block temperature was measured by a platinum resistance thermometer imbedded in the sample block. The temperatures for the germanium runs were obtained by measuring the tip temperature with a 0.002-in. copper-constantan thermocouple soldered to the sample tip after the length measurements had been taken.

2. Sample Preparation and Orientation

Single-crystal high-purity undoped material was used for both the germanium and silicon irradiations. The germanium samples were cut from a 50 ohm-cm n-type crystal obtained from the Eagle Picher Company. The

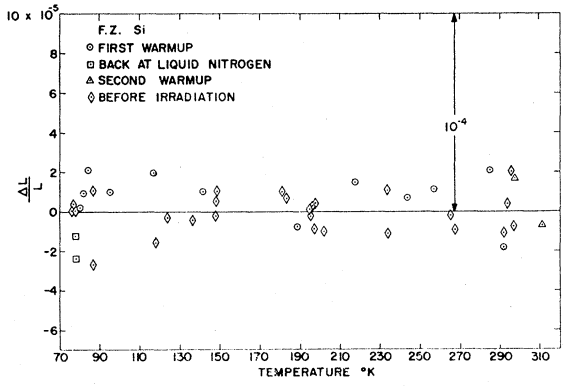

FIG. 3. Unirradiated thermal background curve and annealing, silicon.

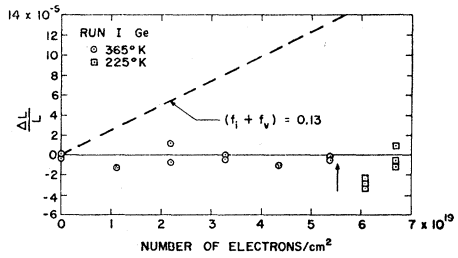

FIG. 4. Run I Ge bombardment $T \leq 365°K$.

silicon sample was cut from a 4600 ohm-cm p-type (6 parts per billion residual boron) vacuum floating zone crystal obtained from Merck and Company.

The samples were ground and polished plane and parallel to thin slabs. The germanium and silicon samples were ground and polished to 0.037 cm and 0.025 cm, respectively. The sample geometries and crystallographic orientations are shown in Fig. 1. All the bombardments were made in the ⟨111⟩ direction and the length measurements were made in directions perpendicular to this.

3. Length Measurement

The change in length was observed directly by measuring changes in distance across a 0.015-cm gap cut transversely through the midpoint of the irradiated region of the sample. In this arrangement the two halves are free to expand, and the change in length may be measured by observing only the narrow gap region. The optical technique described previously[14] was adopted. This method consists of photographing the gap region through a Gaertner creep test microscope having a long working distance. The optical system used here had a magnification of 100×. Distances on the photographic plates were measured by means of an x-y coordinate comparator. This experimental method allowed length measurements to $<\pm 2\times 10^{-5}$ cm on virtually stress-free crystals.

In order to minimize any temperature dependence of the length measurements, the peculiar geometries of the samples were adopted. The smallness of the gap and the yoke arrangement reduced the thermal background below experimental error. The unirradiated thermal background curves are shown in Figs. 2 and 3.

III. RESULTS

1. Germanium

Precision measurements of the change in length of high-purity single-crystal germanium were made on irradiation and annealing. Two samples were irradiated with 2-Mev electrons at temperatures $\leq 365°K$ and $\leq 86°K$ designated Run I and Run II, respectively. The data are shown in Figs. 4 and 5. The sample in Run I was irradiated at 33 $\mu a/cm^2$ to an integrated flux of 5.4×10^{19} electrons/cm^2. The specific length

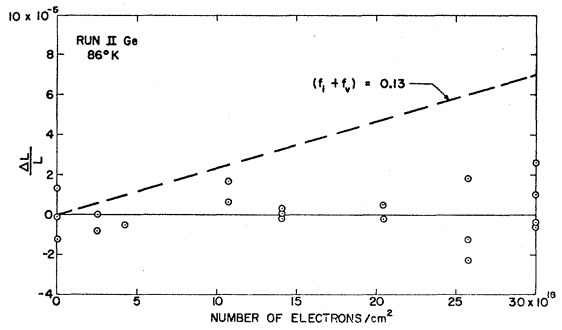

FIG. 5. Run II Ge bombardment $T \leq 86°K$.

expansion obtained by a least squares fit to the data was $\Delta L/L = (6 \pm 12) \times 10^{-26}$ per 2-Mev electrons/cm².

The sample in Run II was irradiated at 10 μa/cm² to an integrated flux of 3.0×10^{19} electrons/cm². The specific length expansion obtained was $(1.5 \pm 3.9) \times 10^{-25}$ per 2-Mev electrons/cm².

Despite the large bombardments given to the samples they were not observed to expand within experimental error. To make this result more vivid one can make the following statement in the case of Run II. A one-centimeter sample of germanium was irradiated for 140 hours at 10 μa/cm² below 86°K with 2-Mev electrons, and its length remained constant to within one-fifth of a wavelength of visible light!

After the Run II bombardment the sample was annealed to room temperature with a constant heating rate of 10°K per hour. Continual length measurements were made during the warmup and are plotted in Fig. 2. After reaching room temperature the specimens were cooled back to liquid nitrogen for low-temperature measurements. No regions of annealing were detected in the warmup.

2. Silicon

Similar measurements were made of the length of a high-purity vacuum-floating-zone silicon sample. The sample was initially irradiated at ~50°K with 2-Mev electrons to an integrated flux of 8.3×10^{18} electrons/cm² and further irradiated at ~115°K to a total integrated flux of 7.9×10^{19} electrons/cm² and then annealed to room temperature. The measurements are plotted in Fig. 6. The specific length expansion for both bombardments obtained by a least squares fit of the bombardment curve was $\Delta L/L = (4 \pm 19) \times 10^{-26}$ per 2-Mev electrons/cm².

After bombardment the sample was annealed and measured on a warmup to room temperature. After reaching room temperature the sample was again cooled to liquid nitrogen and measured. No regions of annealing were detected.

IV. DISCUSSION

The major results of these measurements are the following: the strikingly small effects observed, the smallness of the lattice distortions around an interstitial and a vacancy that these effects in turn imply, and the apparent contradiction between the deuteron irradiation results[14] and these electron irradiation results, and the implications and possible solution to this difference.

1. Experimental Observations

Vook and Balluffi[14] irradiated high-purity germanium with deuterons of average energy 10.2 Mev on two bombardments at 25°K and 85°K. The data plotted in Fig. 7 show that the specific length expansion for

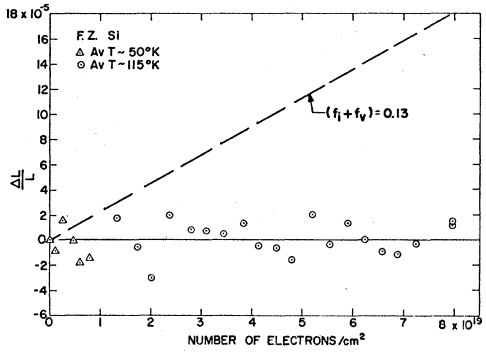

FIG. 6. Silicon bombardment.

both bombardments was $\Delta L/L = (1.5 \pm 0.3) \times 10^{-21}$/(deuteron/cm²). The annealing after the irradiation plotted in Fig. 8 shows a gradual recovery of the expansion which was observable only after warming above 200°K.

Wittels[15] has irradiated high-purity germanium at the Oak Ridge Low Intensity Test Reactor at approximately 85°C to fast-neutron fluxes of up to 4×10^{20} neutrons/cm² examining the crystals by both x-ray diffraction and hydrostatic weighing. He found density reductions as high as 7.7×10^{-3} developed. The density reduction observed in the deuteron case was only 4.5×10^{-4}, and the magnitude of density reductions in the electron case was less than 3×10^{-5}.

In order to determine the volume change per defect, we must now try to combine the experimental observations with a theory of length change as well as a calculation of the number of defects formed per incident particle. Vook and Balluffi[14] considered the structure of deuteron-irradiated germanium, combining length change measurements with small-angle scattering results. Simmons and Balluffi[17,18] have reviewed the application of length and lattice parameter measurements to atomic models of the defects in damaged crystals. We wish to specifically consider the case of

[17] R. O. Simmons and R. W. Balluffi, J. Appl. Phys. **30**, 1249 (1959).

[18] R. W. Balluffi and R. O. Simmons, J. Appl. Phys. **31**, 2284 (1960).

clustered point defects. Eshelby[19] and Simmons and Balluffi[17] have shown that at large distances a cluster of arbitrary shape behaves like a spherically symmetric center of dilatation of the same strength. The strength of a cluster containing n distributed defects is n times the strength of an individual defect *provided* there is no overlap of the nonelastically strained cores of the point defects. In cases where such overlap occurs the strength of the centers of dilatation is not determined by simple addition. As Simmons and Balluffi point out for the case of a divacancy, for example, there are two types of effects. The first is caused by the fact that one nearest neighbor of each component vacancy is missing. The second arises because the defect is no longer cubically symmetric. However, when the clusters are uniformly distributed throughout the specimen the over-all dilatation should be uniform and isotropic. Here, however, the strength of a cluster of n defects will *not* be n times the strength of an isolated defect. As yet no theoretical estimates of the nonlinearity of volume dilatation vs cluster size have been made.[20] We believe that the electron, deuteron, and possibly neutron length change measurements provide the first experimental evidence which bears on this point.

2. Calculations

Let Ω be the atomic volume of a crystal equal to $1/n_0$, where n_0 is the number of atoms/cc for the crystal.

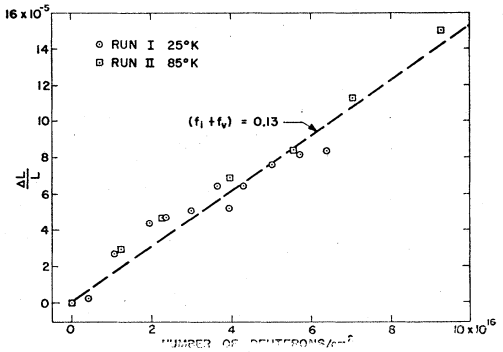

FIG. 7. Deuteron bombardment of Ge.

Let $f_{i1}\Omega$ be the volume change due to the dilatational field of one interstitial. Let $f_{i2}2\Omega$ be the volume change for a cluster of two interstitials. Let $f_{in}n\Omega$ be the volume change for a cluster of n interstitials. Let $f_{v1}\Omega$, $f_{v2}2\Omega$, and $f_{vn}n\Omega$ be the corresponding volume changes for one, two, and n vacancies, respectively. Let C_{in} and C_{vn} be the concentrations of clusters of interstitials and vacancies of size n. We now analyze the total volume change by considering an entire cluster as a center of dilatation. We also assume an isotropic distribution so that even with an overlap of nonelastically strained cores of point defects we will get an isotropic dilatation. The volume change then becomes

$$\frac{\Delta V}{V}=3\frac{\Delta L}{L}=\sum_n nC_{in}(f_{in}-1)+nC_{vn}(f_{vn}+1). \quad (1)$$

If we now assume that there are equal numbers of vacancy and interstitial clusters of size n present, i.e., $C_{in}=C_{vn}=C_n$, we obtain

$$3\frac{\Delta L}{L}=\sum_n nC_n(f_{in}+f_{vn}). \quad (2)$$

To determine the quantities $(f_{in}+f_{vn})$ we now must calculate the distribution of cluster sizes C_n for the various particle irradiations. We have made displacement calculations for germanium using the simple theory of displacement,[21] similar to those made by Koehler[22] for copper. We have made these calculations specifically for 2-Mev electrons, 10.2-Mev deuterons, and 1-Mev neutrons. In all the calculations for germanium we have assumed a step function displacement probability with a threshold for displacement equal to $E_d=30$ ev. This is felt to be reasonable for incident particles with energies much greater than threshold.[23] Recent theoretical calculations[24] as well as experimental data[4,25] seem to indicate that there exists a curve giving the probability of displacement vs the energy transferred. For the case of germanium the experimental data indicate that some displacements can take place with 12–14 ev transferred to the struck atom. However, if one used a constant displacement energy with this lower value, introduction rate curves show that the theory would then greatly overestimate the number of displacements.

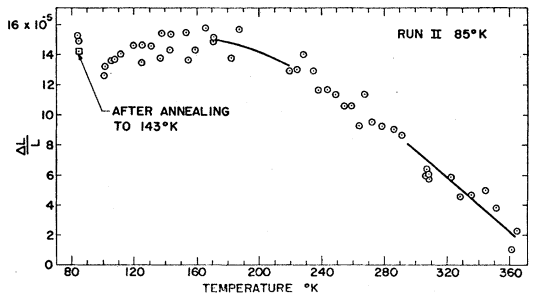

FIG. 8. Annealing of deuteron bombardment of Ge.

[19] J. D. Eshelby, Proc. Roy. Soc. (London) **A241**, 376 (1957).

[20] G. Vineyard (private communication) has calculated volume changes for interstitials, di-interstitials, vacancies, di-vacancies, and tri-vacancies in copper. These calculations do not indicate a greater relaxation per defect for the associated defects than the isolated defects in the close-packed metal copper. [Discussions Faraday Soc. (to be published).]

[21] F. Seitz and J. S. Koehler, *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2, p. 378.

[22] J. S. Koehler, Discussions Faraday Soc. (to be published).

[23] J. H. Cahn, J. Appl. Phys. **30**, 1310 (1959).

[24] J. B. Gibson, A. N. Goland, M. Milgram, and G. H. Vineyard, Phys. Rev. **120**, 1229 (1960).

[25] J. J. Loferski and P. Rappaport, J. Appl. Phys. **30**, 1296 (1959).

The maximum energy a fast nuclear particle can transfer to a lattice atom is

$$E_{2m}=\frac{4M_1M_2}{(M_1+M_2)^2}E_1, \tag{3}$$

where the subscript 1 refers to the incident nuclear particle and 2 to the struck lattice atom. Nuclear particles make close Coulomb encounters with the nuclei of lattice atoms and are scattered by Rutherford scattering with a cross section for energy transfer greater than E_2 of

$$\sigma_2=\frac{4\pi M_1}{M_2}(z_1z_2)^2\frac{R^2a_0^2}{E_1E_2}, \tag{4}$$

where R and a_0 are the Rydberg constant and the Bohr radius, respectively. For 2-Mev electrons the maximum energy transmitted is

$$E_{2m}=(2mE_1/M_2E_0)(E_1+2E_0), \tag{5}$$

where m is the rest mass of the electron and E_0 is the rest energy, 0.511 Mev. The cross section is a complex expression given in reference 21. For a 1-Mev neutron the following expression was used:

$$E_{2m}=4M_2E_1/(1+M_2)^2. \tag{6}$$

If the primary displaced atom is given energy much greater than E_d, it can in turn displace other lattice atoms, called secondary displaced atoms. We consider here a cluster of defects to be produced on bombardment and to consist of the primary and associated secondary displaced atoms as interstitials leaving behind a cluster of an equal number of vacancies. Using the Harrison and Seitz[26] or Neufeld and Snyder[27] method of calculating the number of secondaries, one obtains the total number of displaced atoms resulting from a collision which transmits energy E_2 to the primary: $n(E_2)=0.561E_2/E_d$, $E_2>E_d$.

Following Koehler and using the differential cross section, one can calculate the fraction of the damage which is associated with clusters in a range of size or which is in turn associated with a primary whose energy lies between two limits. The fraction is

$$f(T_1,T_2)=\int_{T_1}^{T_2}n(E_2)d\sigma(E_2)\Big/\int_{E_d}^{E_{2m}}n(E_2)d\sigma(E_2).$$

In the case of deuteron irradiation one finds:

$$f(T_1,T_2)=\ln(T_2/T_1)/\ln(E_{2m}/E_d).$$

For the neutron case of constant energy E_1 one finds:

$$f(T_1,T_2)=\int_{T1}^{T_2 \text{ or } T_m}\frac{0.561E_2}{E_d}\sigma_s\frac{dE_2}{T_m}\Big/\int_{Ed}^{T_m}\frac{0.561E_2}{E_d}\sigma_s\frac{dE_2}{T_m},$$

where the upper limit on the integration is the smaller of T_2 or T_m.

$$f(T_1,T_2)=[E_2^2]_{T_1}^{T_2 \text{ or } T_m}/(T_m^2-E_d^2).$$

TABLE I. Displacement calculations for germanium. $E_d=30$ ev; $\Phi=10^{17}$ particles/cm².

	2-Mev e	10.2-Mev d	1-Mev n
Maximum energy given lattice atom, E_{2m}	176 ev	1.065 Mev	5.36×10^4 ev
$\bar{\nu}$, Average number of secondaries/primary	1.325	5.98	501
Size of cluster containing the median displaced atom	1.355	103	700
$\nu_{\max}$	3.3	2.0×10^4	1.0×10^3
Fraction of atoms displaced (Seitz-Harrison calc.)	5.4×10^{-6}	3.5×10^{-3}	2.07×10^{-4}
Fraction of atoms in clusters			
1–10	1.000	0.275	0.000
10–10^2	0	0.220	0.010
10^2–10^3	0	0.220	0.987
10^3–10^4	0	0.220	0.003
above 10^4	0	0.065	0.000

The results of the above calculations are given in Table I. It should be noted that the larger clusters contain a large fraction of the displaced atoms for both the deuteron and neutron cases. Two other quantities of interest are the average number of secondaries per primary, i.e., the average cluster size $\bar{\nu}$, and the cluster containing the median displaced atom. By the cluster containing the median displaced atom we mean the cluster size for which half the displaced atoms are in clusters of smaller size, and half are in clusters of larger size. In general the size of the cluster containing the median displaced atom is larger than the average cluster size. It should be mentioned that in the case of pile neutrons if one takes into account the neutron energy spectrum, the average cluster size is smaller than that for a 1-Mev neutron, but the cluster containing the median displaced atom is larger than the corresponding one for a 1-Mev neutron.

We now consider Eq. (2) and apply these calculations to the length change measurements for electron and deuteron bombardments using the simple theory in both cases to estimate the number of displaced atoms. The case for the electron irradiation is particularly simple because of the limited nature of the distribution. We may reasonably replace the summation over n by the average cluster size. From the experimental data we then obtain $(f_{i1.3}+f_{v1.3})_{\text{Ge}}=0.008\pm0.021$. Similarly for silicon ($E_d=28$ ev) we obtain $(f_{i1.8}+f_{v1.8})_{\text{Si}}=0.002\pm0.011$.

The analysis for the deuteron case is more complicated because of the wide spread in the distribution of cluster sizes. The volume dilatation per defect given by the experimental data is $(f_{i6-100}+f_{v6-100})_{\text{Ge}}=0.13\pm0.02$. The subscripts indicate that the cluster size which gives this average dilatation is not completely determined.

[26] W. Harrison and F. Seitz, Phys. Rev. **98**, 1530 (1955).
[27] J. Neufeld and W. S. Snyder, Phys. Rev. **99**, 1326 (1955).

3. Conclusions

We see that the volume dilatation per defect for the clustered defects formed in deuteron irradiation is considerably larger than for the isolated defects formed in electron irradiation. For both cases these numbers are small compared to the calculated values of Tewordt[28] for copper. The smallness of the dilatations may be reasonable in view of the open nature of the diamond lattice compared to close-packed copper. As yet no calculations of the volume dilatations of an interstitial and a vacancy have been made for germanium or silicon. It has however been observed for deuteron irradiation that the expansion per deuteron for copper is about twice that for germanium.[12,14,28a]

It is possible that the smallness of the volume dilatation per defect is due to an overestimation of the number of defects produced. We have, however, deliberately tried to underestimate the displacements by using the higher threshold value of 30 ev. Recent stored-energy measurements on copper,[29] combined with calculations[22,28,30,31] of the energy of an interstitial-vacancy pair do indicate that the theory overestimates the fraction of atoms displaced in copper by a factor of 4.3 for deuteron irradiation and 1.75 for electron irradiation. It should be noted that if this is also true for germanium, the difference between electron and deuteron irradiation would be *greater* not smaller even though the dilatation per defect would be increased by these factors. It is therefore felt the ratio of the effects is well established and indicates a striking nonlinearity which increases with the larger clusters. Heretofore calculations have assumed that the dilatation was linear.

If ionization in the sample during irradiation is important and aids displacement as Brown suggests,[8] then, since the ratio of ionization to damage is larger for electron irradiation than for deuteron irradiation, one might expect if anything relatively more defects to be produced in the electron irradiation case. This would tend to further decrease the volume dilatation per defect in the electron case and increase the ratio of the deuteron to electron defect volume. We were not able to detect any difference between the high beam current (i.e., high ionization), high-temperature bombardment (Run I) and the low-beam current, low-temperature bombardment (Run II).

Because of the large total dose[32] and high-sample purity, the defect concentration outweighed the impurity concentration by factors of at least 10^6 to 10^4 for germanium and silicon, respectively, the dominant impurity in the case of silicon being residual oxygen. We therefore are confident that impurities play no role in the interpretation of these data.

If the dilatation observed for deuteron-irradiated germanium is due principally to the defects in clusters, then we conclude that the annealing of the dilatation beginning at 200°K (Fig. 8) probably is associated with the breakup of the larger clusters into smaller clusters and possibly to single and double point defects. It cannot be inferred that interstitial-vacancy recombination does occur near and above 200°K but this is of course not ruled out. What the present electron bombardment results show is that the length change measured for deuteron-irradiated germanium is probably associated with defect clusters. This was not known before.

The neutron irradiation results on germanium are difficult to evaluate since the experiments were performed at a high temperature where considerable annealing would be expected. In spite of this one notes that the density changes are an order of magnitude larger than for the deuteron results below 200°K and considerably larger than the effect remaining after warming to the higher temperature. This suggests the importance of large clusters or displacement spikes in neutron-irradiated germanium, also suggested by the data of Crawford and Cleland[33] and the calculations of Gossick.[34] The large neutron volume change thus agrees with the trend established in the electron and deuteron bombardments.

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[28] L. Tewordt, Phys. Rev. **109**, 61 (1958).

[28a] *Note added in proof.* The diamond structure is not solely responsible for the smallness of the dilatations since recent results by the author[16] show large expansions for electron-irradiated InSb. Results for GaAs (to be published) are similarly large.

[29] T. G. Nilan and A. V. Granato (to be published).

[30] H. B. Huntington, Phys. Rev. **91**, 1092 (1953). H. B. Huntington and F. Seitz, *ibid.* **61**, 315 (1942).

[31] K. H. Bennemann and L. Tewordt, Z. Naturforsch. **15a**, 772 (1960).

[32] The calculated fraction of atoms displaced is 1.6×10^{-3} and 3×10^{-3} for electron-irradiated germanium and silicon, respectively, and 3.5×10^{-3} for deuteron-irradiated germanium.

[33] J. H. Crawford and J. W. Cleland, J. Appl. Phys. **30**, 1204 (1959).

[34] B. Gossick, J. Appl. Phys. **30**, 1214 (1959).