Changes in Conductivity of Germanium Induced by Alpha-Particle Bombardment

W. H. BRATTAIN AND G. L. PEARSON
Bell Telephone Laboratories, Murray Hill, New Jersey
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The bombardment of n-type germanium by alpha-particles from polonium first removes the conducting electrons at the rate of 78 per alpha-particle. After the electrons are gone conducting holes are introduced at the initial rate of 8.6 per alpha-particle. Some of these holes disappear with time at room temperature after bombardment is stopped, leaving only two conducting holes per alpha-particle. This change takes place only to the depth of penetration of the particles, namely 1.9×10^{-3} cm. The distribution of holes with depth is not uniform. The concentration rises from an initial value to a maximum at 1.4×10^{-3} cm depth and then falls to zero. The maximum is about 2.5 times the initial value and the integral under the curve is, of course, two holes per alpha-particle.

I. INTRODUCTION

THE extensive changes undergone by solid materials exposed to radiations of various types have been treated theoretically by Seitz.[1] Lark-Horovitz and his associates[2] have altered the conductivity of germanium by bombardment with nucleons. The mechanism involved was the production of defects or increased p-type conductivity as a result of the germanium atoms being displaced from their normal lattice positions by the incident nucleons. This paper[3] reports quantitative measurements on the change in conductance of an n-type germanium sample during bombardment with alpha-particles from polonium as well as relaxation changes in conductance immediately after stopping the bombardment. An analysis of these data yields values for the rate of change of conduction during the course of the experiment.

A thin slab of material was bombarded on one side and its conductance normal to the direction of bombardment was measured as a function of time. The conductance first decreased linearly, passed through a minimum, and then increased at a diminishing rate as long as the bombardment was continued. The germanium converted from n-type to p-type at the minimum conductance point. After the bombardment was stopped the conductance decreased with time in an asymtotic manner, reaching a final value somewhat larger than the value before bombardment.

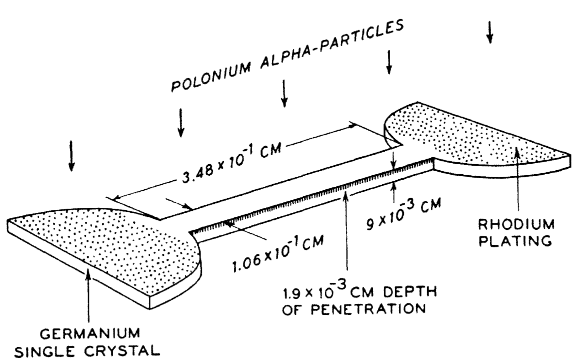

FIG. 1. Germanium single crystal for alpha-particle bombardment experiments.

The interpretation of this experiment is, of course, that the bombardment first removed the conducting electrons through the production of acceptors or traps and after conversion to p-type continuously increased the concentration of acceptors and thus conducting holes. Some of the germanium atoms, after being displaced from their normal lattice positions by alpha-particles, diffused back at room temperature while the remainder were fixed in their new positions. From an analysis of the data we obtain the following quantitative results: (1) at first when the bombarded layer is n-type, electrons are removed from the conduction band at the rate of 78 electrons per alpha-particle, (2) after conversion to p-type conducting holes are introduced at the rate of 8.6 per alpha-particle, and (3) 6.6 of these conducting holes decay with a half-life of 13 hours at room temperature and the remaining 2.0 holes are permanent at this temperature.

After the bombardment and the decay, the distribution in depth of the bombardment produced holes was determined by removing the outer material, a small amount at a time, and measuring the resultant conductance change. It was found that the density of holes is greater near the end of the alpha-particle range than close to the surface, the ratio being 2.5. This is to be expected since it is similar to that found for alpha-particle ionization as a function of range in gases. The total range of polonium alpha-particles in germanium was measured as 1.9×10^{-3} cm.

II. REMOVAL OF CONDUCTION ELECTRONS IN n-TYPE GERMANIUM

The general conditions of the experiment are shown in Fig. 1. The sample was made from a single crystal[4] of high purity n-type germanium having a conductivity of 0.125 (ohm cm)$^{-1}$. It was in the shape of a thin rectangular plate with the dimensions shown. Ohmic

[1] F. Seitz, Discussions Faraday Soc. No. 5, 271 (1949).
[2] W. E. Johnson and K. Lark-Horovitz, Phys. Rev. **76**, 442 (1949).
[3] A preliminary account of this work was presented by the authors at the American Physical Society Meeting in Oak Ridge (March, 1950), Phys. Rev. **78**, 646 (1950).
[4] G. K. Teal and J. B. Little, Phys. Rev. **78**, 647 (1950).

contact was made to the enlarged ends of the sample with rhodium electroplating.

The alpha-particle source was a square polonium capsule 0.41 cm on a side. The germanium was placed 2 cm away from the source so that the alpha-particles entered the sample at no greater than 12° from the normal. The energy of the particles was 5.3 Mev and the experiment was performed in an evacuated chamber to prevent any reduction in speed of the particles before striking the germanium. The polonium source as calibrated by A. J. Ahearn on April 22, 1949 emitted 1.82×10^{10} alpha-particles per unit solid angle per hour. The actual experiment was performed between March 21 and March 28, 1949, at which time the activity was calculated to be 2.1×10^{10} alpha-particles per solid angle per hour for a polonium half-life of 140 days. This corresponds to 1.9×10^{8} alpha-particles per hour striking the effective portion of the germanium sample.

Figure 2 is a plot of the conductance Y in mhos ($Y=1/R$) *vs.* the time of bombardment for the first eight hours of the experiment. It can be seen that the conductance first decreased linearly at a rate of 7.2×10^{-5} mho per hour, reached a minimum after about three hours and then increased slowly thereafter. A test with a thermoelectric junction showed that the sample changed from n-type to p-type shortly after the minimum in conductance was reached.

A theoretical calculation by Bardeen[5] indicates that 5.3-Mev alpha-particles penetrate germanium to a mean depth of 1.9×10^{-3} cm. A major portion of the sample, as shown in Fig. 1, was therefore not effected and merely contributed a constant conductance in parallel with the bombarded layer. Since parallel conductances are additive in this geometry and since we are primarily interested in conductance changes this contribution was easily handled in the analysis.

The relation between conductance change dY and the change in the total number of conduction electrons

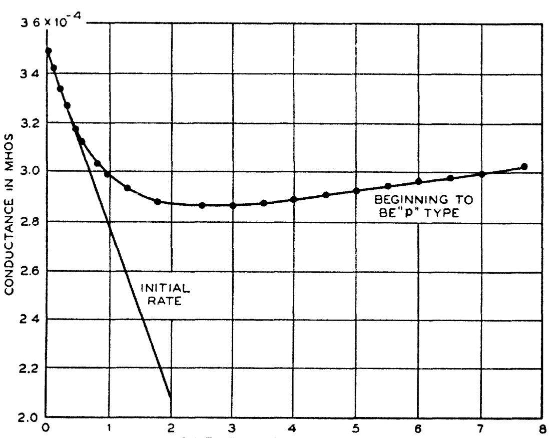

FIG. 2. Conductance *vs.* time characteristic for initial eight hours of bombardment.

[5] These calculations were based on the data of M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 245 (1937).

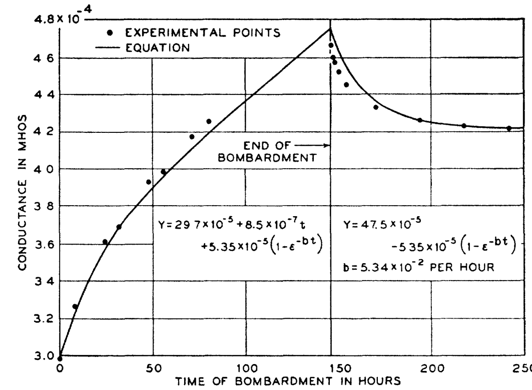

FIG. 3. Conductance *vs.* time characteristic for final 150 hours of bombardment and 100 additional hours of relaxation.

dN_e is

$$dN_e=l^2dY/e\mu_e=2.07\times 10^{14}dY, \tag{1}$$

where l is the length of the sample, e the electronic charge (1.59×10^{-19} coulomb), and μ_e the drift mobility of electrons in germanium[6] which has the value 3600 cm^2/volts sec. The similar relation for conduction holes dN_h is

$$dN_h=l^2dY/e\mu_h=4.45\times 10^{14}dY, \tag{2}$$

where μ_h the drift mobility of holes[6] is 1700 cm^2/volt sec.

The initial rate of conductance decrease as shown in Fig. 2 is $dY/dt=7.2\times 10^{-5}$ mho per hour. The rate of arrival of alpha-particles as determined by the source calibrated is $dN_\alpha/dt=1.9\times 10^{8}$ per hour. Substituting these values in Eq. (1) we find that

$$dN_e/dN_\alpha=78 \text{ electrons per alpha-particle.}$$

Thus in the initial stage of the bombardment each alpha-particle impact removed, on the average, 78 conduction electrons from the n-type germanium sample.

This part of the experiment yields data from which the approximate depth of penetration of the alpha-particles can be calculated. The conductance Y is given by

$$Y=\sigma wd/l, \tag{3}$$

where σ is the electrical conductivity and l, w, and d are the sample length, width, and thickness, respectively.

$$\Delta Y=wd\Delta\sigma/l, \tag{4}$$

has the value 6.3×10^{-5} mho as determined from the initial value and the minimum value of Fig. 2. $\Delta\sigma$ is the difference between the initial conductivity (0.125 mho/cm) and that of intrinsic germanium at 296°K (0.014 mho/cm) or 0.11 mho/cm. If the effect of the bombardment is non-uniform the left-hand side of Eq. (4) will be less than the right so that

$$d\geqq l\Delta Y/w\Delta\sigma. \tag{5}$$

[6] Pearson, Haynes, and Shockley, Phys. Rev. **78**, 295 (1950).

TABLE I. Resistance R and conductance Y of the bombarded germanium sample after etching layers of thickness x from its surface.

x (cm)	R (ohms)	Y (mhos)	Y (corrected) (mhos)
0	2411	4.15×10^{-4}	4.15×10^{-4}
3.50×10^{-4}	2502	3.99	3.99
7.00	2673	3.75	3.79
10.50	2897	3.45	3.52
14.00	3270	3.06	3.13
17.50	3740	2.67	2.76
21.00	3940	2.54	2.65

Substitution of the numerical values in this equation gives $d\geqq 1.83\times10^{-3}$ cm, which is consistent with Bardeen's calculated value of 1.9×10^{-3} cm. In another experiment on a similarly bombarded sample the thickness of the converted p-type layer was found to be 2.0×10^{-3} cm by testing with a thermoelectric junction.

The final calculation to be made from the data of Fig. 2 is the average rate of formation of traps plus acceptors near the filled band up to the time at which p-type conductivity definitely appears which we take as four hours. The total number of conduction electrons at the start of the experiment can be determined from the conductivity relation,

$$\sigma = ne\mu_e, \tag{6}$$

where n is the number of conduction electrons per cm^3. Substitution of the numerical values gives $n=2.17\times10^{14}$ and $N_e=n\times\text{volume}=1.5\times10^{10}$ conduction electrons so that

$$dN_{(T+A)}/dN_\alpha=(1.5\times10^{10})/(4\times1.9\times10^8)=19.$$

III. PRODUCTION AND RELAXATION OF CONDUCTION HOLES IN p-TYPE GERMANIUM

After the data shown in Fig. 2 were obtained the alpha-particle bombardment was interrupted for 16 hours. During this period the conductance of the sample decreased from 3.02×10^{-4} to 2.97×10^{-4} mho. A second experiment was then performed in which the conductance of the sample was measured as a function of time during additional bombardment of the p-type layer for 150 hours after which bombardment was stopped and the conductance followed for a relaxation period of 100 hours. These data are plotted as experimental points in Fig. 3. We see that during bombardment the conductance Y increases rapidly at first and then at a slower rate. Finally, after the bombardment is stopped, the conductance decreases asymptotically with time and approaches a final value of 4.2×10^{-4} mho which is somewhat larger than the original value before bombardment.

It was found that these results could be explained if it were assumed that two processes were going on: One a production of holes proportional to time of bombardment, or number of alpha-particles, and another process in which the mechanism of hole production was proportional to the number of alpha-particles plus the additional assumption that this mechanism of hole production was decaying with time at a rate proportional to the number of holes present. Expressed in terms of conductance, one then has the two equations.

$$dY_1/dt=A \tag{7}$$

$$dY_2/dt=K-bY. \tag{8}$$

Integrating these two equations and adding the conductances we get:

$$Y=Y_1+Y_2=Y_0+At+(K/b)(1-\epsilon^{-bt}) \tag{9}$$

for the equation of the curve during bombardment in Fig. 3. An easy way to determine the constants for Eq. (9) is to differentiate this equation and note that

$$(dY/dt)-A=K\epsilon^{-bt}. \tag{10}$$

The circles in Fig. 4 show the experimental data treated in this manner, indicating that the assumptions are reasonably well justified.

The critical test is whether the decay, after the bombardment is stopped, follows the equation:

$$Y=Y_0'+(K/b)\epsilon^{-bt}, \tag{11}$$

or

$$dY/dt=-K\epsilon^{-bt} \tag{12}$$

with, of course, the same values of K and b as determined for Eq. (10). The solid points in Fig. 4 show the experimental data obtained during the relaxation period treated according to Eq. (12) and it is seen that the results before and after bombardment do agree reasonably well. Using the constants determined from Fig. 4, namely $b=5.34\times10^{-2}$, $K=2.8\times10^{-6}$, and $A=8.5\times10^{-7}$ and also $Y_0=2.97\times10^{-4}$, the solid curve in Fig. 3 has been drawn using Eq. (9) during bombardment and

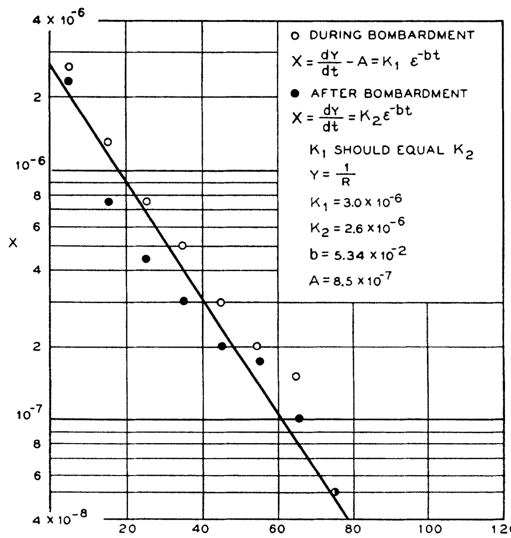

FIG. 4. The time rate of change of conductance during and after bombardment.

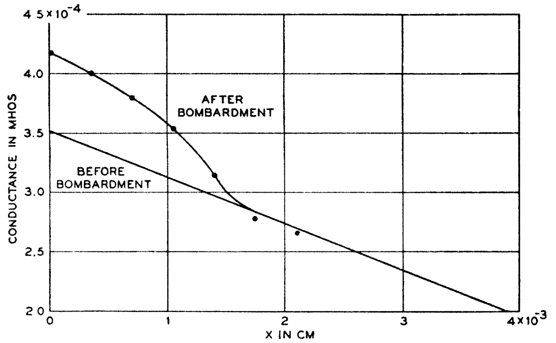

FIG. 5. Conductance of sample as successive layers are etched from the bombarded surface.

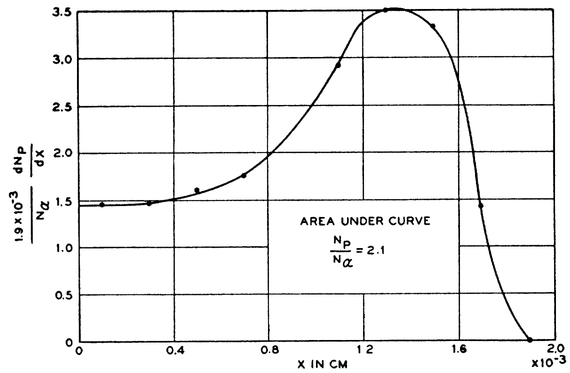

FIG. 6. Holes per alpha-particle per length of range as a function of the depth x.

Eq. (11) after bombardment. The solid curve does not fit the experimental points perfectly, but the agreement is quite good. It is known from other experiments that the fractional part that decays will increase with temperature and since room temperature during the experiment did fluctuate from about 22°C during the night to 26°C during the day, the agreement is probably as good as can be expected. From the above rate of decay one can calculate that the 16-hour wait before the start of this experiment was fortunately long enough for the sample to be reasonably near equilibrium at the start, otherwise the results would have been more complicated due to the previous bombardment.

From these values of A and K one gets the figures 2.0 holes per alpha-particle for that part which does not decay at room temperature, and 6.6 holes per alpha-particle for the part that decays at room temperature. From the value of b one gets 5.34×10^{-2} per hour per hole for the rate of this decay which corresponds to a half-life of 13 hours. Note that these rates say nothing about the number of acceptors themselves, but give only the rates for the net number of added and/or subtracted carriers or holes.

IV. HOLE CONCENTRATION *VS.* DEPTH

The next experiment was to measure the conductivity of the specimen as a function of thickness and determine the distribution of carriers in the sample as a function of depth after the bombardment and subsequent decay.[7] This was done by etching away successive layers starting from the bombarded side. The numerical results are given in Table I. It was necessary to correct for the reduction in width as both sides as well as the top face were exposed to the etchant. These corrected values of Y *vs.* x are plotted in Fig. 5. In this figure the solid straight line represents what the results of such an experiment would have been before bombardment. The experimental data approach the straight line when the amount removed approaches the penetration depth of the alpha-particles. Of course, the conductivity from $x=0$ to $x=1.9\times10^{-3}$ is now p-type where it was n-type before bombardment. By differentiating the smooth curve drawn through the experimental points and converting from changes in conductivity to changes in number of holes per total number of alpha-particles, one gets the curve in Fig. 6, giving the number of holes per alpha-particle per length of range as a function of depth x. The integral under this curve gives the total number of holes per alpha-particle as 2.1, which agrees reasonably well with the previous value of 2.0 for the part that does not decay. A uniform distribution of holes would have given a curve in Fig. 6 constant and equal to 2 up to 1.9×10^{-3} cm where it would drop to zero. A distribution such as that found here is to be expected, however, since it is of the same general shape as that for alpha-particle ionization in gases.

V. DISCUSSION

The experimental results described here are in qualitative agreement with the earlier study of Lark-Horovitz.[2] In addition our analysis of these data yield accurate quantitative values for a number of important parameters. One of these is the factor 78 for the number of conduction electrons removed by each impinging alpha-particle during the initial part of the experiment when the sample was n-type. This is in good agreement with the theoretical analysis of Seitz[1] which predicts 59 displaced germanium atoms for each impinging alpha-particle, suggesting that each displaced atom removes one conduction electron either by creating an acceptor level or a trap. Seitz's calculations were for 5.0-Mev alpha-particles whereas our polonium source emitted 5.3-Mev particles.

Our quantitative results suggest the following conclusions. At the start electrons are removed at the rate of 78 per alpha-particle. This process can be due to creation of either electron traps or acceptors most anywhere in the energy gap between the filled band and the conduction band. Such traps or acceptors would lower the Fermi level, thus draining out the electrons. In fact, this initial result might be expressed in terms

[7] This experiment was suggested by W. Shockley who predicted the distribution found here.

of the rate of lowering of the Fermi level. When this level has reached the halfway point, the conduction process is intrinsic. Now, however, before appreciable hole conductivity can occur, the Fermi level must be lowered still further to very near the top of the filled band. When this happens all the electrons must be in traps or acceptors near the top of the filled band. From the time at which hole conductivity shows up, about four hours in Fig. 2, one concludes that the rate of formation of this last group of levels is at least 19 per alpha-particle. This is twice the initial value for hole production. One can explain the latter fact several ways. Each alpha-particle produces 19 acceptor levels but they are so far above the filled band that only half of them are ionized to give holes at room temperature could be one explanation. The other possible explanations would involve a combination of electrons traps and acceptors near the top of the filled band or a combination of acceptors and hole traps or some combination of all three. The important point is that the p-type conductivity due to alpha-particle bombardment is not the simple case of the formation of acceptor levels each producing a conduction hole at room temperature.

The authors are indebted to J. Bardeen for assistance with the theory, to G. K. Teal for the preparation of the single crystal germanium sample, and to P. W. Foy and E. G. Dreher who prepared the sample and took many of the data. This research was carried out under the direction of W. Shockley, who made many suggestions during the course of the work.

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The High Energy Neutrons from the Disintegration of C^{13} by Deuterons*

J. E. RICHARDSON**
The Rice Institute, Houston, Texas
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An excitation curve for the 5-Mev neutrons resulting from C^{13} bombarded by deuterons has been obtained for neutrons emitted at $0°\pm 28°$ to the direction of the deuteron beam. The curve covers deuteron energies from 200 kev to 2.1 Mev. Resonances in the yield were observed at deuteron energies of 0.6, 0.9, 1.55, and 1.80 Mev with half-widths of 0.1, 0.4, 0.1, and 0.5 Mev respectively. These correspond to excited states in the intermediate N^{15} nucleus of 16.7, 16.9, 17.47, and 17.69 Mev.

I. INTRODUCTION

Two groups of neutrons from the disintegration of carbon by deuterons were observed by Bonner and Brubaker[1] in 1936 and were attributed to the reaction:

$$C^{13}+H^2\rightarrow(N^{15*})\rightarrow N^{14}+n^1+Q_1. \quad (1)$$

By means of cloud-chamber measurements, they observed Q-values of 1.2 and 5.2 ± 0.4 Mev respectively for the two groups.

As natural carbon contains 99 percent C^{12} and only one percent of the C^{13} isotope, one would expect the relative intensity of neutrons from C^{13} to be very weak when a natural carbon target is bombarded by deuterons. Using such a target and a methane-filled cloud chamber, they observed the relative intensities of the 5.2- and 1.2-Mev groups from C^{13} and the -0.281 ± 0.003-Mev group[2] from C^{12} to be 1, 3, and 300 respectively.

In order to explain the 5-Mev gamma-rays from the disintegration of carbon by deuterons, Bennett, Bonner, Hudspeth, Richards, and Watt[3] looked for a third group of neutrons from reaction (1). They obtained a target which was enriched to 23 percent of C^{13} and found a low-energy group of neutrons with a Q-value of 0.4 ± 0.05 Mev.

A weak group of 50-cm protons from carbon was observed by Bower and Burcham,[4] and independently by Pollard.[5] These protons result from the reaction:

$$C^{13}+H^2\rightarrow(N^{15*})\rightarrow C^{14}+H^1+Q_2. \quad (2)$$

Bennett and Bonner[3] bombarded a target containing 23 percent of C^{13} with 1.00-Mev deuterons and found the Q value of this group of protons to be 6.09 ± 0.2 Mev. Using a target enriched to two percent C^{13}, they obtained an excitation curve for these protons with the counter placed at 90° to the direction of the deuteron beam. The curve showed a maximum at a deuteron energy of 1.55 Mev which may be interpreted as a resonance in the yield. This would indicate an excited state in the intermediate N^{15} nucleus at 17.47 Mev.

This group of workers also studied the 5-Mev gamma-rays and obtained an excitation curve for

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** Now at The Alabama Polytechnic Institute, Auburn, Alabama.
[1] T. W. Bonner and W. M. Brubaker, Phys. Rev. **50**, 308 (1936).
[2] Bonner, Evans, and Hill, Phys. Rev. **75**, 1398 (1949).
[3] Bennett, Bonner, Hudspeth, Richards, and Watt, Phys. Rev. **59**, 781 (1941).
[4] J. C. Bower and W. E. Burcham, Proc. Roy. Soc. **A173**, 379 (1939).
[5] E. Pollard, Phys. Rev. **56**, 1168 (1939).