Lattice Parameter Changes in Deuteron-Irradiated Germanium*

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A lattice expansion of 3×10^{-5} was measured in germanium irradiated by 1.5×10^{17} 9-Mev deuterons/cm^2 at low temperature and annealed to 320°K. Thus residual specimen length expansion (as measured by Vook and Balluffi in similar samples) and lattice parameter expansion are small and nearly equal after annealing to room temperature. Thermal recovery to 380°K occurred parallel to that of macroscopic length changes, but near 380°K the (211) interplanar spacing had contracted about 3×10^{-5} relative to unirradiated crystal. The lattice parameter returned to that of unirradiated crystal at 430°K. No appreciable diffraction line broadening was observed. These results provide confirmatory evidence that structural damage in deuteron-irradiated germanium consists of well-localized centers of dilatation.

STRUCTURAL study of irradiation damage in germanium crystals has been relatively neglected while detailed phenomenological pictures of the changes in electrical properties have been constructed. Clearly, further investigations of both types will be required to elucidate fully the origin and character of these effects, as well as their relation to lattice defects naturally present or occurring under other conditions such as plastic deformation, quenching, and chemical doping. The work reported in the preceding paper[1] (hereafter referred to as I) is an attempt to correlate macroscopic volume and resistivity changes under carefully controlled deuteron irradiation and thermal annealing conditions, and the present measurements of lattice parameter changes of similar samples complement the effects described there. Lattice expansions in germanium and silicon subjected to very heavy neutron bombardment have been reported elsewhere[2] together with a brief outline of other related work.

METHOD

An account of the nature, preparation, irradiation, and thermal treatment below room temperature of the germanium crystals which were used in this investigation appears in I. Briefly, the thin polished samples were irradiated with 6.2×10^{16} deuterons/cm^2 near 25°K, warmed slowly to 308°K, then irradiated with 9.2×10^{16} deuterons/cm^2 near 85°K and warmed slowly to room temperature. Various considerations limited the measurements at temperatures below room temperature to length and resistivity changes, hence the principal x-ray sample (lower half of *ML* of Fig. 1 of I) was removed at 305°K so that parallel annealing studies could be made above this temperature. The measurements were begun at about 320°K because the samples had been stored at room temperature for 10 months after irradiation. X-ray study of a different sample (upper half of *ML* of Fig. 1 of I) was also made at temperatures above 364°K, the limit of the length change measuring apparatus.

A high-angle back-reflection rotating-single-crystal technique was employed for measurements of changes in lattice parameter. The general features of the technique, together with an error analysis, have appeared elsewhere.[3] The observed change in position, x, of the Laue-Bragg line on a film is given by $x=2(\Delta d/d)L\tan\theta$, where the Bragg angle θ (about $82\frac{1}{2}°$ at 30°C) is deduced from the known lattice parameter of germanium[4] and the wavelength of the incident x-rays (Cr$K\alpha_1$), L is the specimen-to-film distance (853 mm) and $(\Delta d/d)$ is the relative change in the (422) interplanar spacing. A gaseous helium path reduced the necessary exposure time to 20 minutes for 40′ angular rotation of the sample. Measurements were made near room temperature and the lattice parameter changes measured relative to an unirradiated homologous sample maintained as a comparison standard and subjected to identical annealing treatment.

The precision was somewhat improved over that of previous work[3] by the use of a narrower x-ray collimating slit (angular divergence 0.21°) and can be judged from Fig. 1 which shows thermal expansion of a standard sample. The linear thermal expansion coefficient was found to be $(5.7\pm0.1)\times10^{-6}$ per degree

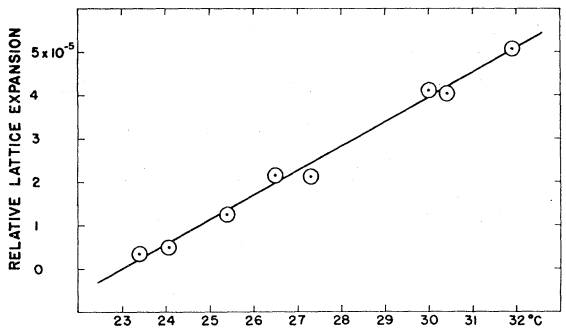

FIG. 1. Linear thermal expansion of germanium.

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[1] F. L. Vook and R. W. Balluffi, preceding paper [Phys. Rev. **113**, 62 (1959)].

[2] M. C. Wittels, J. Appl. Phys. **28**, 921 (1957).

[3] R. O. Simmons and R. W. Balluffi, Phys. Rev. **109**, 1142 (1958).

[4] M. E. Straumanis and E. Z. Aka, J. Appl. Phys. **23**, 330 (1952).

near 300°K,[5] and all measured ($\Delta d/d$) values were suitably corrected for differences in sample temperature. The sample was annealed in air for two hours at successively higher temperatures between each series of measurements; the times and temperatures thus simulate the warm-up rate of 10 degrees per hour of I. After each annealing treatment the shape of the (422) crystal reflection curve was measured using a Xe-filled proportional counter of large aperture. In each case, the irradiated sample face examined was that from which the deuterons emerged during irradiation, the deuteron energy there being about 9 Mev.

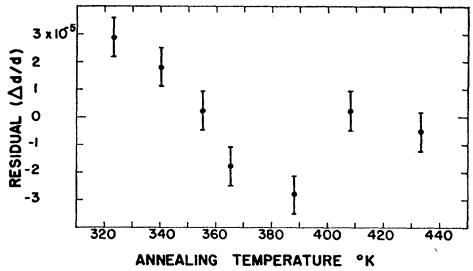

FIG. 2. Thermal recovery of lattice expansion in deuteron-irradiated germanium.

RESULTS

The measured relative changes in (211) interplanar spacing after each anneal are shown in Fig. 2. Each value is the mean of three or more separate measurements and the uncertainty is the combined sample and reference probable errors, usually near ± 7 parts per million. It is clearly apparent that the lattice constant has been increased slightly by irradiation and annealing to 320°K. Further, a comparison with Fig. 7 of I (showing thermal recovery of macroscopic length changes) reveals that while thermal recovery of the lattice parameter in the range 320 to 380°K closely parallels that of the macroscopic length, the residual effect appears to be smaller in magnitude, becoming in fact a slight lattice contraction at the latter temperature. The lattice parameter returns to its normal value after annealing at about 430°K.

It is to be noted that the entire range of the effects reported here is minute. The sensitivity of the x-ray measurements for detecting residual structural changes near room temperature is somewhat greater than that of the length measurements of I, but the apparent disagreement is near the sum of the respective estimated experimental errors. Also, the length and lattice parameter changes were measured along perpendicular directions and these effects, even in unconstrained samples like these,[6] might depend upon the direction of deuteron bombardment.

Curves of crystal (422) diffracted x-ray intensity *versus* crystal angular orientation, taken after each annealing treatment, showed no appreciable change in shape or breadth or any difference from the results obtained from the polished face of the standard sample. The $K\alpha_1$ line width at half-maximum intensity remained at 16′, more than 75% of which was contributed by spectral and instrumental broadening.

The general character of the x-ray effects reported here is consistent with the presence in the irradiated material of well-localized centers of dilatation.[7] In the absence of detailed calculations of crystal lattice relaxation around possible defect models, however, no numerical comparison can be made at present with the predictions of irradiation displacement theory. The absence of appreciable line broadening indicates that no systematic long-range strains have been introduced, and the apparent rough correspondence of the small length and lattice parameter changes makes improbable the existence of gross mutually compensating effects leaving only moderate dimensional changes. The apparent decrease of lattice spacing below the normal value is perhaps not too surprising in such a loosely packed solid and suggests that the annihilation of defects during thermal recovery may not be merely a simple process such as interstitial-vacancy recombination but may involve more complex structural interactions. Observation of lattice expansions in heavily neutron-irradiated samples[2] do not necessarily contradict the present results in which a lattice contraction appears centered near 380°K because of the rather different irradiation conditions in the two cases and also because the neutron-irradiation temperatures of about 310 and 410°K straddle the present minimum.

Further x-ray studies of this type carried out at low temperature might resolve the question of possible structural annealing below[8] 85°K and determine whether the length and lattice parameter changes during irradiation and annealing are congruent.

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[5] This agrees closely with a recent interferometer value obtained by D. F. Gibbons [Phys. Rev. **112**, 136 (1958)] which is 5.75×10^{-6} at 300°K, but is appreciably less than the value of 5.92×10^{-6} reported in reference 4 for material of lower purity.

[6] Work on GaSb and InSb [U. Gonser and B. Okkerse, Phys. Rev. **105**, 757 (1957); **109**, 663 (1958); D. Kleitman and H. J. Yearian, Phys. Rev. **108**, 901 (1957)], where expansion effects may be one or two orders of magnitude larger, has not yet given any quantitative comparison for those materials. In such experiments very complex constraints were present on the irradiated crystal.

[7] J. D. Eshelby, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 3, p. 79.

[8] G. W. Gobeli, Phys. Rev. **112**, 732 (1958).