Nature of Defects Arising from Fast Neutron Irradiation of Silicon Single Crystals*

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A method is described for determining the range of sizes that damaged regions created by fast neutrons may have in a silicon single crystal. In particular the use of precision ultrasonic velocity and attenuation measurements for compressional waves together with certain new results from scattering theory permit the determination of an upper and lower limit on the size of a damaged region. The lower limit is determined from the fractional velocity change arising from the irradiation and the upper limit is found from the corresponding attenuation change—or in this case the lack of it.

Under the assumption that such regions of damage are spherical the limits obtained lie in a range from 0.01 μ to 0.27 μ for the radius of the region in question.

IT has been shown[1] that defects introduced into silicon by collimated fission neutrons directed along an equivalent [100] direction produce an anisotropy in the elastic character of the silicon among the three equivalent [100] directions. This elastic anisotropy caused by directional neutron bombardment was examined by means of an ultrasonic double refraction effect arising when transverse waves were propagated through the silicon. The double refraction and the anisotropy in shear modulus arise from the anisotropic nature of the regions of displaced atoms produced by the recoiling atoms which were created by neutron collisions with the silicon atoms. On the average these regions of displaced atoms should be so oriented that an axis of symmetry of the region is parallel to the direction of bombardment.

It is the purpose of this article to point out that when the compressional wave velocity and attenuation measurements before and after irradiation are combined with certain results from the theory of multiple scattering of waves, the results yield estimates of the sizes of the damaged regions. In order to develop these ideas it is necessary first to state some of the results from scattering theory.

The results[2] of recent work in scattering theory show a detailed relationship, for an isotropic solid, between the fractional velocity change $\Delta v/v$ and the fractional volume $\Delta\tau/\tau$ of the material causing the change.[3]

The relation for the case of elastic scatterers in an elastic medium is (for compressional waves)

$$\frac{v-v'}{v'}=\left\{\frac{1}{2}\frac{\Delta\tau}{\tau}\left[\frac{\rho_2}{\rho_1}-\frac{\lambda_1+2\mu_1}{\lambda_2+2\mu_2-\frac{4}{3}(\mu_2-\mu_1)}+\frac{10(\mu_2-\mu_1)}{2(\mu_2-\mu_1)+3[(\mu_2/\mu_1)+\frac{3}{2}](\lambda_1+2\mu_1)}\right]\right\}, \quad (1)$$

where v' is the perturbed compressional wave velocity and v is the compressional wave velocity in the absence of scatterers. $\Delta\tau/\tau$ is the volume fraction of scatterers present and it may be written $(4/3)\pi a^3 n_0$, where a is the radius of the scatterer (assumed spherical) and n_0 is the density or number of such scatterers per unit volume of matrix. λ_1, λ_2, μ_1, μ_2 are the Lame elastic constants and ρ_1, ρ_2 are the densities of matrix and scatterer respectively. The use of the Lame constants requires an isotropic medium; in this case it is assumed that the material can be described approximately in this way. A simplification in the $\Delta v/v\sim\Delta\tau/\tau$ relationship is possible if one considers the scatterer to be a cavity in the matrix material. This simplification obviously involves the assumption that a damaged region can be represented as a cavity in the matrix. Since a spherical cavity in an elastic medium would scatter *more strongly* than an elastic sphere of the same size as the cavity within the elastic matrix, it seems clear that a spherical cavity of given size and scattering power would be *smaller* in size than an elastic sphere having the same scattering power. Consequently it is argued that if a damaged region be represented by a cavity in an elastic medium, then for elastic wave scattering purposes the cavity would be smaller than the actual damaged region because the damaged region is almost certainly not a simple cavity but a gradual transition in elastic properties from the inner core of the region out to the elastic matrix. For the purposes here the size of such a damaged region is considered to be determined by that distance over which the strain in the lattice falls from a maximum value to some factor $1/h$ of its maximum value. When h is specified, this determines a radius a of the region in which this change in strain occurs. The region of damage may be considered as composed of a core or central region of displaced atoms surrounded by a larger region of atoms which are "displaced elastically" from their normal lattice positions. As one proceeds outward from this inner core the "elastic displacement" of the atoms outside the core becomes smaller in amount until the normal lattice is reached. The resulting strain in

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[1] Truell, Teutonico, and Levy, Phys. Rev. **105**, 1723–1729 (1957).
[2] P. C. Waterman and R. Truell, Technical Report, Metals Research Laboratory, Brown University, October, 1957 (unpublished); C. F. Ying and R. Truell, J. Appl. Phys. **27**, 1086–1097 (1956).
[3] That there should be such a relationship between $\Delta v/v$ and $\Delta\tau/\tau$ is not surprising, but obtaining the factor for general use involves quite a lot of calculation.

the region surrounding the damaged region is going to be "seen" by an elastic wave propagating through that region. Obviously the sensitivity of the detecting device is important; in this case the strained region is detected by its scattering of elastic waves of megacycle frequency and by the associated changes in velocity and attenuation.

The scattering theory[2] mentioned previously yields as expression for the velocity change arising from spherical cavity scatterers

$$\frac{v-v'}{v'}=(9/2)\frac{\Delta\tau}{\tau}\times\left[\frac{(7/9)-(7/12)(\kappa/k)^2+(3/16)(\kappa/k)^4}{1-(9/4)(\kappa/k)^2}\right]+\cdots \quad (2)$$

for $ka\ll 1$, where κ is the transverse wave propagation factor and k is the compressional wave propagation factor. A corresponding expression for attenuation exists but is omitted from this discussion.

Using the above expression together with proper values of the ratio $\kappa/k=v/v_t=8.4/5.8=1.45$, where v is the longitudinal velocity and v_t the transverse velocity, we then have

$$(v'-v)/v'=\Delta v/v=0.47\Delta\tau/\tau. \quad (3)$$

This relation can be regarded as defining the size and number of scatterers for the case of spherical cavity scatterers.

Measurements made at 30 Mc/sec on an undamaged silicon cube show a compressional velocity of 8.368×10^5 cm/sec, and measurements made at the same frequency on a second similar cube, identical with the first cube before damage, show a compressional velocity of[4] 8.631×10^5 cm/sec after damage.[5] This is a velocity increase of approximately 3.1% and using this value in the expression

$$\Delta\tau/\tau\cong 2(\Delta v/v) \quad (4)$$

leads to $\Delta\tau/\tau=0.062$.

Taking 4.5×10^{22} atoms/cm³ for silicon, this means that 2.8×10^{21} atoms/cm³ are located in regions of damage. The irradiation was such that the fast neutrons produced about 2×10^{16} primary recoils/cm³ in the damaged silicon. The foregoing values lead to the conclusion that there should be about 1.4×10^5 atoms in a single region of damage, and if the region is assumed to be spherical, which it certainly is not, the radius of such a region would be about 32 atoms or about 9.0×10^{-7} cm.

This value of 9×10^{-7} cm is a measurement of the hypothetical cavity radius a_0:

$$a_0=9\times10^{-7}\text{ cm}. \quad (5)$$

It is not a measurement of a as defined by the lattice strain around a scattering center.

Since it has been assumed in the $\Delta v/v$ relation that the scattering region is a cavity of radius a_0, it is to be expected that a somewhat more realistic elastic region of radius a would necessarily be larger to produce a comparable velocity change. A cavity is a strong scatterer and any weaker scatterer, to produce comparable effects, would have to be larger in size. Consequently it seems that a damage region, under circumstances such as those given here, must be larger than the space occupied by 10^5 atoms, larger than 9×10^{-7} cm radius. If, however, one accepts these arguments regarding a cavity being the strongest type of scatterer, it follows that $9\times10^{-7}\text{ cm}=a_0<a$.

It is possible at the same time to obtain additional evidence about the size of such a damaged region from the results of attenuation measurements. With scattering regions of 10^{-6} cm to 10^{-5} cm in size (radius), one can calculate the scattering cross section γ as well as the attenuation α for ultrasonic waves. In this case $ka\ll1$ since at 100 Mc/sec

$$ka=\frac{2\pi10^8}{8.4\times10^5}\times10^{-5}=7.5\times10^{-3}\ll1. \quad (6)$$

With 2×10^{16} such regions considered as elastic scatterers the attenuation can under these circumstances be written[6]

$$\alpha=(\text{db}/\mu\text{sec})=\tfrac{1}{2}(8.68\times10^{-6})vn\gamma=3.6n\gamma, \quad (7)$$

where v = compressional wave velocity $=8.4\times10^5$ cm/sec (silicon), n is the density of scattering centers (i.e., $n=2\times10^{16}$ in this case), and γ is the scattering cross section for a cavity of radius a in the silicon. The frequency is taken to be 100 Mc/sec because the attenuation measurements were made in the range from 50 Mc/sec to about 250 Mc/sec. It turns out that $\gamma=3.1\times10^{12}a^6$ under these conditions; hence $\alpha=2.23\times10^{29}a^6$, so that

$$\begin{aligned}\alpha&=2.23\times10^{-7}\text{ db}/\mu\text{sec}\quad\text{for}\quad a=10^{-6}\text{ cm},\\ \alpha&=0.223\text{ db}/\mu\text{sec}\quad\text{for}\quad a=10^{-5}\text{ cm}.\end{aligned} \quad (8)$$

These attenuation values are for scattering only; they do not contain attenuation from any other causes.

Measured values of ultrasonic attenuation at 100 Mc/sec show an attenuation value of 0.2 db/μsec under normal conditions. Consequently an increase of another 0.2 db/μsec from the introduction of scattering

[4] This value is the average of three values for compressional velocities measured in the three equivalent [100] directions 8.778×10^5, 8.600×10^5, and 8.514×10^5 cm/sec. The differences arise because this crystal was bombarded by fast neutrons primarily in one direction, and it exhibits anisotropy after irradiation both for the shear[1] and compressional wave velocities. The three directions are equivalent [100] directions. The above values are all larger than the pre-irradiation value of 8.368×10^5 cm/sec.

[5] It is interesting to note that this increase in the compressional velocity was accompanied by a decrease in the transverse velocity so that while c_{11} increased, c_{44} decreased as a consequence of the fast neutron irradiation (see reference 1).

[6] See Ying and Truell, reference 2.

centers would represent a one hundred percent increase in attenuation; such an increase would certainly be noticed and easily measurable. No such increase was observed; in fact no detectable change was seen at 100 Mc/sec. It may be concluded that if the damaged regions are spherical cavities one can say, from attenuation considerations, that the value of a_0 is less than 10^{-5} cm. A determination of a_0 from attenuation values, for the cavity scatterer, should yield the same value of a_0 as the velocity measurements did, namely $a_0=9\times10^{-7}$ cm. On the other hand the attenuation values as they are used in this case can by themselves only set an upper limit on a_0—that is $a_0<0.1\ \mu$.

Thus far, from the velocity and attenuation evidence, it has been shown that the hypothetical spherical cavity equivalent of an average damage region should have a radius

$$9\times10^{-7}\ \text{cm}=a_0<10^{-5}\ \text{cm}.$$

where the left hand equality came from the velocity data, and the right-hand inequality came from limits on attenuation values.

In addition it is shown above that it is reasonable to conclude that

$$a_0<a,$$

where a is the radius of an arbitrarily chosen strain region around the region of damage, and where some reasonable fraction $1/h$ of the maximum strain is used as the boundary of the damage region.

One cannot thus far say anything about an upper limit on the value of a because the spherical cavity model can only set a lower limit on a, namely a_0. It is possible, however, to go further by considering the consequences of using the scattering cross section γ for an elastic body embedded in the matrix rather than the cavity case.

While the details of a damaged region are not known at present there is, nevertheless, a method of deciding on a reasonable set of elastic values to be used here. Normally $v=8.4\times10^5$ cm/sec and $v_t=5.8\times10^5$ cm/sec in silicon, hence

$$(\kappa_1/k_1)_{\text{matrix}}=v/v_t=8.4/5.8=1.45. \tag{9}$$

Now under irradiation conditions v increases to as much as 8.8×10^5 cm/sec and v_t decreases to 5.7×10^5 cm/sec, hence $(\kappa_2/k_2)=8.8/5.7=1.55$ but these are measured values in the entire medium so that (κ_2/k_2) for the actual region of damage must be considerably larger than this value. Consequently if one uses $(\kappa_2/k_2)=1.55$ for the scatterers in a matrix for which $(\kappa_1/k_1)=1.45$, this is certainly a conservative arrangement. The actual situation must have contained stronger hence smaller scatterers. Using these values and the assumption that $\mu_1=\mu_2$ (the shear moduli differ by 1% or less) one can calculate γ for this conservative elastic case. Using this resulting value of γ together with the fact that the total attenuation was about 0.2 db/μsec, and that it could not have changed by this amount, one finds a radius for the elastic scatterer

$$a_e=2.7\times10^{-5}\ \text{cm}.$$

This radius may be considered as an upper limit on the size of an actual scatterer. In other words

$$9\times10^{-7}=a_0<a<a_e\leqq2.7\times10^{-5}\ \text{cm}. \tag{10}$$

The results obtained by the velocity measurement, namely those for a_0 are in good agreement with estimates made on sizes of damaged regions from thermal spike ideas. For example, independent estimates[7] of the size of a damaged region and the way in which stress falls off in the neighborhood of such a region indicate a value of about 150 angstroms in one case to be compared with our smallest value of 90 angstroms.

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[7] G. J. Dienes and G. H. Vineyard, *Radiation Effects in Solids* (Interscience Publishers, New York, 1957), pp. 40, 85.