LONG WAVELENGTH NEUTRON TRANSMISSION AS AN ABSOLUTE METHOD FOR DETERMINING THE CONCENTRATION OF LATTICE DEFECTS IN CRYSTALS

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Recent work has shown that very long wavelength neutrons (>6Å) are scattered isotropically by point defects in a way that is accurately known from other measurements. Thus an absolute determination of the concentration of defects is available if this scattering can be measured. Although the scattered intensities available are very low, it is possible to make a transmission measurement and obtain the same data. Such experiments have been carried out on samples irradiated in the Brookhaven reactor at about 50°C. For an exposure of 1.1×10^{20} neutrons/cm² the fraction of displaced atoms in graphite was found to be 0.0263. This value is in excellent agreement with that derived from Seitz's theory; the theoretical estimate being of lesser accuracy because of uncertainties in exposure. The change in neutron transmission arising from reactor irradiation anneals out upon heating the sample at high temperatures, showing that a true irradiation effect was measured.

INTRODUCTION

Of obvious importance to those concerned with research on solids, is a knowledge of the kind and number of defects contained in the solid. We shall describe some work in which we have employed the scattering of very slow neutrons by radiation-induced point defects in graphite to give an absolute determination of the number of these defects. The point defects may be atoms at sites not on the crystal lattice (interstitials), or vacant lattice sites (vacancies).

THEORY

The scattering of slow, long wavelength neutrons (approx. 8 Å) by such defects is simply described. The incident neutrons are considered as a plane wave, and the defects scatter isotropically. Thus the neutrons scattered by m defects are described by the radially symmetric wave

$$\psi = \sum_{j=1}^{m} a_k e^{i(\mathbf{s}\cdot\mathbf{r}_j)}. \qquad (1)$$

a_k is the neutron "scattering length" for a carbon atom and is positive for interstitials and negative for vacancies. $\mathbf{r}_j$ is the vector distance to the jth atom from an arbitrary origin, and $|\mathbf{s}| = (4\pi \sin\theta)/\lambda$ (θ = scattering angle, λ = neutron wavelength).

The differential cross section $\sigma(\theta)$, which corresponds to intensity in a diffraction pattern, is given by the square of Eq. (1).

$$\sigma(\theta) = \psi\psi^* = \sum_{j,j'}^{m} a_k^2 e^{i\mathbf{s}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})}, \qquad (2)$$

where the primes distinguish between identical atoms. This expression is correct if it

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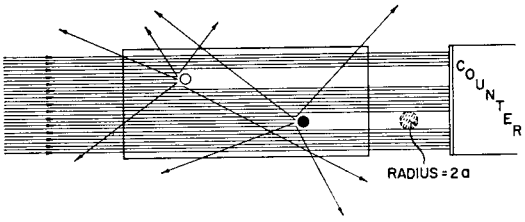

FIG. 1. Illustration of long wavelength neutron cross section for point defect scattering.

is assumed that the defects are randomly arranged and at a sufficient distance from one another that they do not interact while scattering. Upon performing the sum in Eq. (2), only when $j = j'$ is there a non-zero result, so that

$$\sigma(\theta) = \overset{\vee}{m}a^2$$

The cross section is obtained by integration over all space, thus

$$\sigma = 4\pi m a^2. \qquad (3)$$

Since a is squared, both vacancies $(-a)$ and interstitials $(+a)$ will scatter similarly. In optical scattering, this is known as Babinet's principle. The cross section per atom is

$$\sigma_d = \frac{4\pi m a^2}{N} \qquad (4)$$

N = number of atoms per unit volume. The fractional number of defects is $f = m/N$. The value $4\pi a^2$ is called the bound atom cross section σ_b and is readily obtained from other measurements. Substituting for m/N and $4\pi a^2$ in Eq. (4), the concentration of defects is

$$f = \sigma_d/\sigma_b .$$

Equation 3 suggests a geometrical description of the scattering as illustrated in Fig. 1. A beam of neutrons appears to be intercepted at each defect by an opaque circular area of radius $2a$. The cross section σ of Eq. 3 is merely the sum of these cross sectional areas, clearly a measure of the absolute number of defects present.

EXPERIMENTAL CONSIDERATIONS

Figure 1 suggests also the type of experiments which may be used to measure this cross section. It would be most instructive if the intensity scattered by the defects could be measured directly. Unfortunately the intensities available are so small that they would be completely lost in the background intensity. The only practical technique is to measure the attenuation of a transmitted beam with the beam-specimen-counter arrangement illustrated.

It is instructive to study the graphite cross section over a wide wavelength range, Fig. 2. At very short wavelengths, approximately .05 Å to .1 Å, the cross section is quite constant at a value closely equal to the bound atom cross section σ_b. Independent measurements have tabulated this value[1] for most elements. As the wavelength is increased the cross section fluctuates greatly, the neutron being diffracted coherently by crystalline structure according to Bragg's law $n\lambda = 2d \sin\theta$, with $\sin\theta$ always equal to 1. At the point where the wavelength

$$\lambda = 2d_{\max} , \qquad (5)$$

$d_{\max}$ = largest interplanar spacing in the crystal, called the last cutoff, the cross section drops to a low value beyond which it increases proportional to the wavelength. Here the major portion of the cross section is (capture cross section σ_a) and inelastic cross section σ_i, both proportional to the wavelength. Other cross sections, represented by σ_{dis}, which are wavelength independent, occur from various nuclear disorder scattering: isotopic content, spin, etc. It is possible to select solids in which σ_a, σ_i and σ_{dis} are quite small relative to the cross section for defect scattering. With such a situation sensitive measurements of σ_d can be made at wavelengths beyond the last cutoff.

[1] Neutron Cross Sectons, AECU-2040, Department of Commerce, Washington, D. C., May 15 1952.

Graphite has small inelastic and absorption cross sections, which makes it quite suitable for this experiment. Graphite was expected also to retain enough radiation-induced damage for a room temperature measurement to be possible. Graphite features low extraneous cross sections relative to the defect scattering cross section:

$$\left.\begin{aligned} \sigma_i + \sigma_a &= 0.9 \text{ barns at 8 A}, \\ \sigma_{\text{spin}} &= 0, \\ \sigma_{\text{isotope}} &\cong 0, \end{aligned}\right\}$$

whereas

$$\sigma_b = 4.7 \text{ barns}.$$

Graphite has two undesirable features for this experiment. The many minute pores in graphite scatter neutrons consecutively through small angles and make the transmitted beam diverge considerably during passage through the long, 9-in. specimen used. A grossly incorrect cross section is measured if some of the small angle scattered neutrons are not counted. From Figure 3, it is seen that it was necessary to keep the counter within 1″ of the specimen. Another feature of graphite reduced considerably the accuracy possible in the determination. Upon irradiation the lattice parameter of graphite increases and changes the spectrum of transmitted neutrons in a way which is not easily corrected for. This will be made clear from the following.

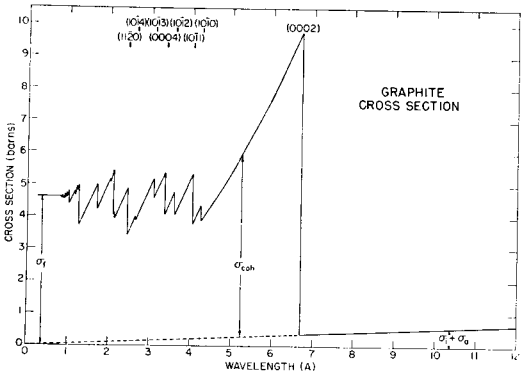

FIG. 2. Cross section plot for polycrystalline graphite. The bound atom cross section is obtained from $\sigma_b = \sigma_f(A+1)^2/A^2$; A = mass number.

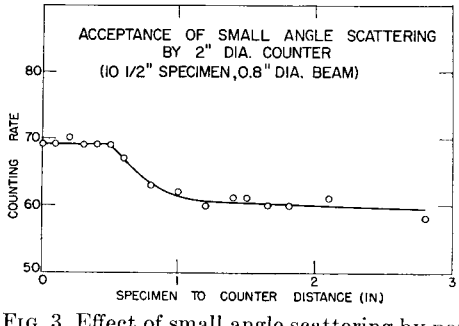

FIG. 3. Effect of small angle scattering by pores on the intensity transmitted by graphite. At a 1-in. separation the diverted neutron beam begins to miss the counter.

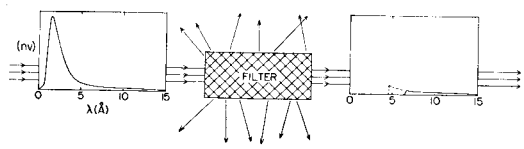

FIG. 4. Very slow neutron beam obtained by filtering. The spectrum at the left represents the intensity available from a reactor. After passage through a filter material, only neutrons of wavelengths longer than twice the largest lattice spacing in the filter remain in the beam. The solid curve is for graphite, the dashed curve for a material having a last cutoff, $\lambda = 2d_{\max}$, at a shorter wavelength.

The very slow neutrons were obtained from the Brookhaven Laboratory reactor by filtering—making use of the strong last cutoff (Eq. 5) of a suitable material. The high cross section at wavelengths lower than the cutoff remove the shorter wavelength neutrons by Bragg scattering, whereas longer wavelengths pass through the filter material almost unattenuated. Graphite, with its last cutoff at 6.70 Å is the best filter in this wavelength region. In these experiments the same piece of graphite was used both as specimen and filter and resulted in a most economic use of the low intensity available. Figure 4 illustrates the filtering process and indicates the relatively low intensities available for these scattering experiments.

It is clear now how a large lattice param-

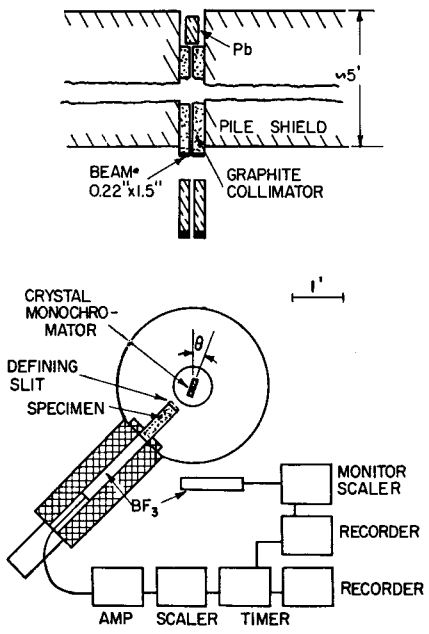

FIG. 5. Crystal spectrometer employed in these studies. The monochromating crystal is a laminate of mica sheets reflecting from the (001) cleavage planes, thus $\lambda = 19.908 \sin \theta$. The defining slit was 0.81 in x 0.81 in., the specimen approximately 2 inches x 2 inches x 10 inches. Six inches of Pb in the collimator provides γ-ray and thermal neutron attenuation. A portion of the beam transmitted by the Pb is intercepted by a BF_3 counter to provide monitoring of beam and background fluctuations.

eter change in graphite would change the transmitted neutron spectrum. Hence the transmitted intensity was plotted out on a neutron spectrometer, Fig. 5, and a correction made graphically.

EXPERIMENTAL DATA

We obtained a piece of AGHT-CS graphite which had been irradiated in the BNL reactor by a total fast neutron flux of

$$nvt = 1.1 \times 10^{20} \text{ neutrons/cm}^2,$$

during which time its temperature probably did not reach 80°C. As this sample was not available before irradiation, a standard piece of graphite had to be chosen which would represent the pre-irradiation state of the graphite. A piece of the same grade of graphite having closely the same density and length was selected from a number of samples cut from various longer extruded blocks. If the preferred orientation amongst samples varied considerably it would be impossible to decide upon a standard comparable in neutron filtering properties to the irradiated specimen. Therefore the neutron transmission of each sample as a function of orientation was checked. No measurable variation was found.

Figure 6, neutron transmission plots for the standard and irradiated samples, illustrates the pronounced effect of irradiation upon the transmitted neutron intensity. The increase in lattice parameter was allowed for by considering only intensities for wavelengths greater than 7.30 Å. These intensities were obtained by planimetering the areas under the curves. Upon annealing an irradiated sample at several temperatures, a series of curves between these extremes was obtained.

The cross section, for calculation of the concentration of defects in the irradiated sample (Eq. (4)), is related to the intensities by the expressions

$$I_d = I_0 \exp -N\bar{x}(\check{\sigma}_i + \check{\sigma}_a + \check{\sigma}_{\text{dis}}) - mx\sigma_b$$

$$I_s = I_0 \exp -Nx(\sigma_i + \sigma_a + \sigma_{\text{dis}}).$$

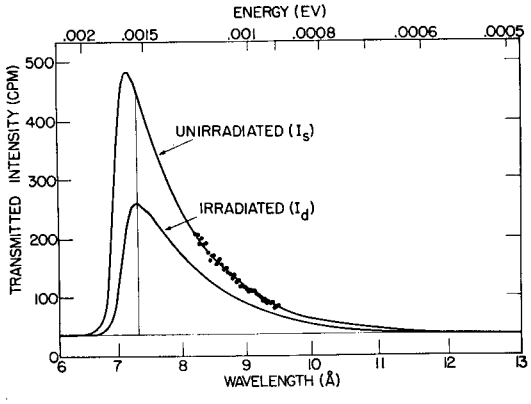

FIG. 6. Transmitted long wavelength neutron intensity for unirradiated and irradiated graphite. A representative portion of the data points is shown to indicate the number and spread. The intensities I_s and I_d were measured as the areas under the curves to the right of the vertical line at 7.30 Å.

TABLE I

Concentration of Defects Remaining after Annealing Treatment

Annealing Treatment	f
None	0.033
3 hrs at 250°C	0.021
1 hr at 500°C	0.011
1 hr at 900°C	0.00
1 hr at 2000°C	0.00

I_d = intensity transmitted by the irradiated specimen, I_s = intensity transmitted by the standard specimen, I_0 = incident intensity, and x = specimen thickness. The ratio of intensities is

$$\frac{I_d}{I_s} = e^{-mx\sigma_b}$$

or, since $f = m/N$,

$$\frac{I_d}{I_s} = e^{-Nxf\sigma_b},$$

from which,

$$f = \frac{-ln(I_d/I_s)}{Nx\sigma_b} \qquad (5)$$

Because the specimen acted also as a filter I_0 could not be measured, but as Equation 5 shows, comparison with a standard sufficed.

The data of Fig. 6 gives

$$f = 0.0526.$$

Therefore,

Concentration of displaced atoms $= f/2$
$= 0.0263$, or 2.63 %.

Another sample of irradiated graphite was annealed at various temperatures giving the results in Table I. An anneal at 2000°C was expected to return the graphite to its unirradiated state. Thus the zero value for f after this anneal confirmed the choice as a truly representative standard.

DISCUSSION

It is possible to make a calculation according to Seitz's theory[2] to predict the number of atomic displacements. For the *nvt* of the samples, one obtains

$$f/2 = 0.021 \text{ or } 2.1\%$$

in excellent agreement with the experimental value. A lack of precise knowledge of (*nvt*) places an accuracy of about 50 % on this figure.

Such agreement does not insure the success of all future experiments, however. Because irradiation of a material generally causes crystalline disorder, one might expect the neutron scattering to be affected by two other factors. Serious distortion of the lattice by interstitials would change the inelastic cross section σ_i and the apparent value of σ_b. Aggregation of single defects into pairs or larger groups would produce interference effects in the neutron scattering. Each of these factors however, would manifest themselves as a cross section which was wavelength dependent. Within the accuracy of the data obtained, over the wavelength range 7.3 Å–9.5 Å no wavelength dependence was found. Further, the generally uniform increase in transmitted intensity upon annealing clearly indicates the disappearance rather than the aggregation of defects. Experiments with increased accuracy and better temperature control during irradiation are planned to investigate such effects further. Thus we have found little reason not to believe that the results actually represent the number of vacancies and/or interstitials produced by the irradiation of these samples within an accuracy of, say, 20 %. The actual experimental error is ±9 %.

ACKNOWLEDGEMENT

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[2] F. Seitz, Disc. Faraday Society, No. 5, 271 (1949).