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The Effect of Crystal Orientation on the Scattering of Slow Neutrons*

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A simple theory is developed for taking into account non-random orientations in microcrystalline material of the hexagonal system. The transmission of slow neutrons through extruded and through powdered graphite is measured experimentally and compared with the theory.

INTRODUCTION

THE transmission of slow neutrons through microcrystalline materials has been the subject of several investigations.[1-4] However, a theoretical treatment of the effect of non-random orientations of the microcrystals on neutron scattering has not been considered in detail. A simplified scheme is developed for taking account of the orientation effect when a single symmetry axis only needs to be considered. A comparison is made with the measurements of the transmission of slow neutrons through randomly oriented and through extruded graphite. It is possible to obtain an estimate of the amount of crystal orientation in this instance.

THEORETICAL

If ψ is the angle between the direction of extrusion and the symmetry axis of a given microcrystal, the probability of finding a crystal oriented in the solid angle $d(\cos\psi)d\beta$ is assumed to be

$$P(\cos\psi)d(\cos\psi)d\beta. \qquad (1)$$

If $\mathbf{b}$ denotes the product of the order of the Bragg reflection and the reciprocal lattice vector and the symmetry axis of the crystal is taken in the direction of one of the basic reciprocal lattice vectors $\mathbf{b}_3$, then

$$\mathbf{b} = l\mathbf{b}_1 + m\mathbf{b}_2 + n\mathbf{b}_3, \qquad (2)$$

where l, m, n are the products of the order of the

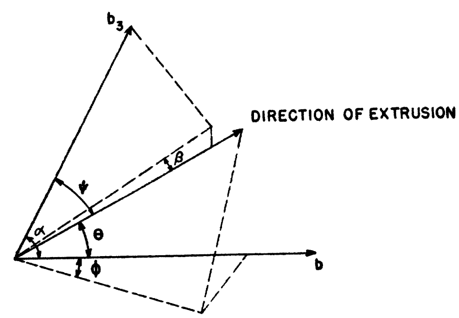

FIG. 1. Angles used in calculating the orientation effect. ϕ is generated perpendicular to a plane through $\mathbf{b}$ and $\mathbf{b}_3$, β is generated perpendicular to a plane through $\mathbf{b}_3$ and the extrusion direction.

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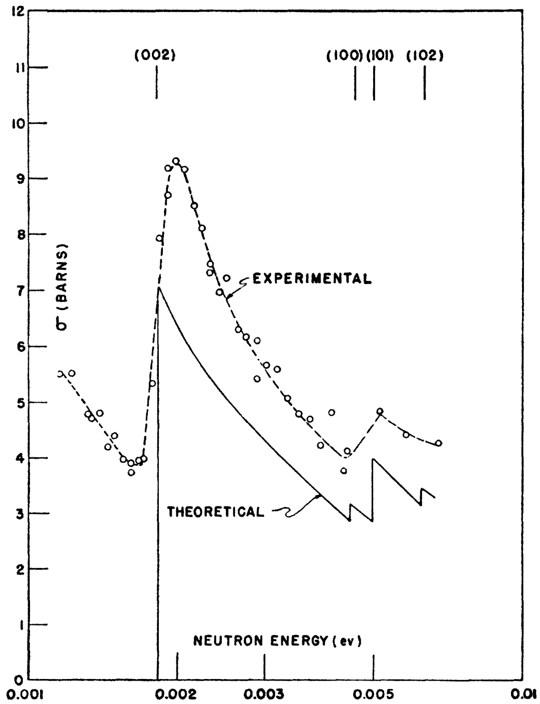

FIG. 2. Total cross section *vs.* neutron energy for randomly oriented (powdered) graphite. The positions of the various Bragg low energy limits are shown by the vertical lines with Miller indices above the curves.

reflection and the Miller indices for the particular Bragg reflection under consideration.

It is sufficient to consider the incident neutron beam either parallel or perpendicular to the extrusion direction since the experimental measurements have been made for these two cases for which it is possible to estimate separately the amount of microcrystalline orientation.

A. Incident Neutron Beam Parallel to Extrusion Direction

The details of geometry for the analysis are shown by Fig. 1. The coherent elastic scattering intensity for a particular Bragg reflection into the solid angle $d(\cos\psi)d\beta$ is given by[3]

$$\sigma_b d(\cos\psi)d\beta = (FN\lambda^2/2\pi b^2\Delta\lambda) \times\exp(-\omega b^2)P(\cos\psi)d(\cos\psi)d\beta, \quad (3)$$

where σ_b is the coherent elastic scattering cross section per unit cell for the Bragg reflection b, N is the number of unit cells per unit volume, $\Delta\lambda$ gives the spread in the wave-length of the incident neutron beam, $\exp(-\omega b^2)$ is the Debye-Waller temperature function,[2] and F, the *form factor* per unit cell of the graphite crystal, is calculated by

$$F = (4\sigma_{\text{free}}/\mu^2\{1+(-1)^n \cos^2 2\pi(l/3+2m/3) + [1+(-1)^n]\cos 2\pi(l/3+2m/3)\}, \quad (4)$$

in which*** σ_{free} is the free atom elastic scattering cross section and μ is the reduced mass of the carbon nucleus in units of the neutron mass.

Transforming to the variables θ, ϕ it follows from elementary analysis that

$$\cos\psi = \cos\theta\cos\alpha + \sin\theta\sin\alpha\cos\phi = (nb_3/b)\cos\theta + [1-(n^2b_3^2/b^2)]^{\frac{1}{2}}\sin\theta\cos\phi, \quad (5)$$

and $d(\cos\psi)d\beta = d(\cos\theta)d\phi$. Whence from the Bragg relation $\cos\theta = b\lambda/2$ and $\Delta\lambda$ small, Eq. (3) becomes

$$\int \sigma_b d(\cos\psi)d\beta = (FN\lambda^2/4\pi b)\exp(-\omega b^2) \times \int_0^{2\pi} P(\theta,\phi)d\phi. \quad (6)$$

In order to proceed further, it is necessary to make some specific assumptions concerning the function $P(\cos\psi)$. Consider a distribution function with two parameters, namely,

$$P(\cos\psi) = (1/4\pi) + x(\cos^2\psi - \tfrac{1}{3}) + y(\cos^4\psi - \tfrac{1}{5}). \quad (7)$$

The quantities x and y give a measure of the degree of correlation in orientation among the microcrystals and are subject to the limitation that $P(\cos\psi)$ shall remain positive for all values of ψ. The factors $\frac{1}{3}$ and $\frac{1}{5}$ in Eq. (7) are necessary in order that P shall be normalized to unity. Substituting the above expression for P in Eq. (6) yields

$$\begin{aligned}\int \sigma_b d(\cos\psi)d\beta = {} & (FN\lambda^2/4\pi b)\exp(-\omega b^2) \\ & \times\{[\tfrac{1}{2} - (2\pi x/3) - (2\pi y/5)] \\ & + 2\pi x[(b^2\lambda^2/4)\cdot(n^2b_3^2/b^2) \\ & + \tfrac{1}{2}(1-(b^2\lambda^2/4))(1-(n^2b_3^2/b^2))] \\ & + 2\pi y[(b^4\lambda^4/16)\cdot(n^4b_3^4/b^4) \\ & + 2(b^2\lambda^2/4)(1-(b^2\lambda^2/4))(n^2b_3^2/b^2) \\ & \times(1-(n^2b_3^2/b^2)) + \tfrac{3}{8}(1-(b^2\lambda^2/4))^2 \\ & \times(1-n^2b_3^2/b^2)^2]\}. \quad (8)\end{aligned}$$

*** The quantity enclosed by the braces of Eq. (4) is the x-ray *crystal structure factor* squared.

Equation (8) now must be summed over all possible Bragg reflections. Transforming from neutron wave-length to energy via the relationship $E_b/E=\lambda^2b^2/4$ yields

$$\sigma_c=\sum_{E_b\leq E}(FNE_b/2\pi b^3E)\exp(-\omega b^2)$$
$$\times\{[1-(4\pi x/3)-(4\pi y/5)]$$
$$+4\pi x[(E_b/E)\cdot(n^2b_3^2/b^2)$$
$$+\tfrac{1}{2}(1-(E_b/E)(1-(n^2b_3^2/b^2))]$$
$$+4\pi y[(E_b^2/E^2)\cdot(n^4b_3^4/b^4)$$
$$+2(E_b/E)(1-(E_b/E))(n^2b_3^2/b^2)$$
$$\times(1-(n^2b_3^2/b^2))+\tfrac{3}{8}(1-(E_b/E))^2$$
$$\times(1-(n^2b_3^2/b^2))^2]\},\quad(9)$$

where σ_c is the total coherent elastic scattering cross section at the neutron energy E and E_b is the neutron energy at a particular Bragg long wave-length limit.

B. Incident Neutron Beam Perpendicular to Extrusion Direction

If the incident neutron beam is perpendicular to the direction of extrusion, the previous analysis must be modified slightly. It is necessary to rotate the direction of the incident beam through 90°. Therefore, Eq. (5) has the form

$$\cos\psi=(nb_3/b)\cos\theta'$$
$$+[1-(n^2b_3^2/b^2)]^{\frac{1}{2}}\sin\theta'\cos\phi,\quad(10)$$

where $\theta'=\theta-90°$. Hence, corresponding to Eq. (9),

$$\sigma_c=\sum_{E_b\leq E}(FNE_b/2\pi b^3E)\exp(-\omega b^2)$$
$$\times\{[1-(4\pi x/3)-(4\pi y/5)]$$
$$+4\pi x[(1-(E_b/E))(n^2b_3^2/b^2)$$
$$+\tfrac{1}{2}(E_b/E)(1-(n^2b_3^2/b^2))]$$
$$+4\pi y[(1-(E_b/E))^2(n^4b_3^4/b^4)$$
$$+2(E_b/E)(1-(E_b/E))(n^2b_3^2/b^2)$$
$$\times(1-(n^2b_3^2/b^2))+\tfrac{3}{8}(E_b^2/E^2)$$
$$\times(1-(n^2b_3^2/b^2))^2]\}.\quad(11)$$

EXPERIMENTAL

A mechanical velocity selector[5] in conjunction with the heavy water pile at the Argonne Laboratory was used to measure the cross sections for randomly oriented (powdered) and extruded graphite. The results are shown in Figs. 2–4. All of the experimental curves exhibit a fairly large amount of scattering for energies below the region of Bragg effects (the (002) plane is the lower limit). This effect probably is due mainly to inelastic scattering of the "cool" neutrons by the crystal lattice. Graphite has an extremely low absorption cross section, 0.005 barn at thermal energy,[6] so that neutron capture should play an unimportant part. Scattering and absorption by adsorbed gases is a probable contributing factor also in the observed low energy scattering.

In the computation of the theoretical curves, a free atom cross section of 4.5 barns was used and the Debye temperature of graphite was estimated to be 1500°K from specific heat data.[7]

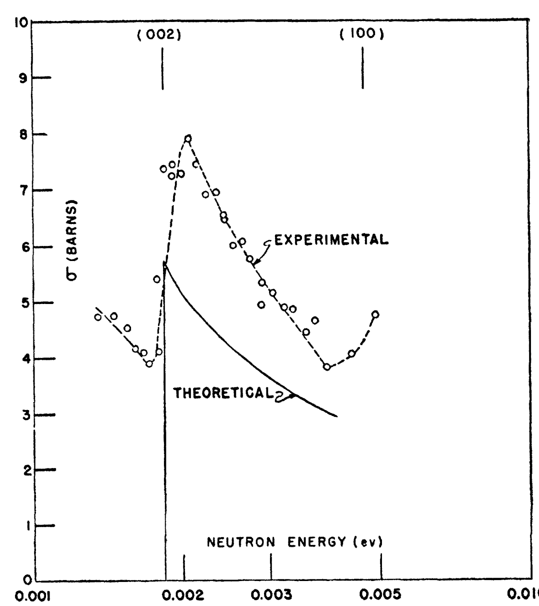

FIG. 3. Total cross section *vs.* neutron energy for extruded graphite with neutron beam incident *parallel* to the extrusion direction.

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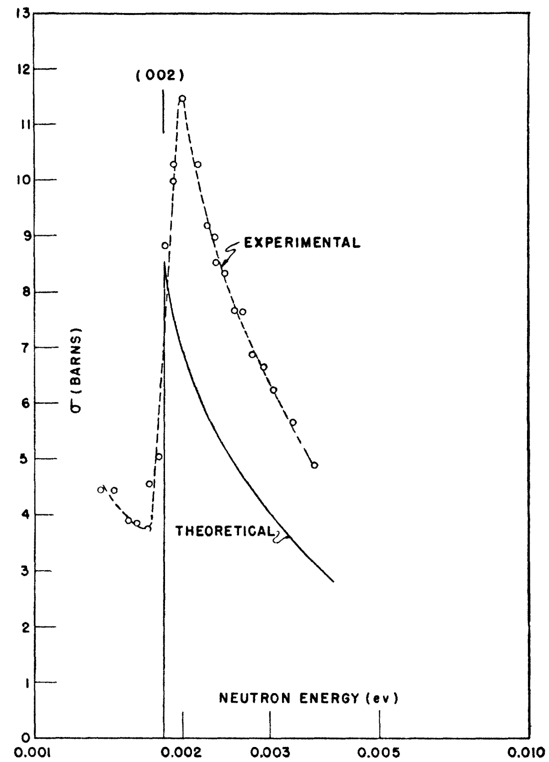

FIG. 4. Total cross section *vs.* neutron energy for extruded graphite with neutron beam incident *perpendicular* to the extrusion direction.

The theoretical curve for the powdered material (random orientation) is, of course, that corresponding to $x=y=0$. It is seen by Fig. 2 that the experimental curve lies appreciably above the theoretical curve in the region between the lower limits for the (002) and the (100) planes. This discrepancy also is probably due to the inelastic scattering of low energy neutrons. In the case of the extruded graphite, the theoretical curves are fairly sensitive to the choice of the parameters x and y. The values $x=-0.07$, $y=0.04$ yielded about the best agreement with the empirical data, and these are used in Figs. 3 and 4. Hence, the orientation function Eq. (7) becomes

$$\begin{aligned} P(\cos\psi) &= 1/4\pi - 0.07(\cos^2\psi - \tfrac{1}{3}) + 0.04(\cos^4\psi - \tfrac{1}{5}) \\ &= 1/4\pi(1.2 - 0.9\cos^2\psi + 0.5\cos^4\psi). \qquad (12) \end{aligned}$$

It is seen that $P(1) < P(0)$. This means that the sixfold symmetry axis of graphite tends to be perpendicular to the direction of extrusion so that the hexagonal basal planes show a preferential alignment along or nearly along the extrusion direction. This may be noted also in the experimental curves. The coherent cross section at the Bragg low energy limit for the (002) hexagonal plane is about twice greater for the neutron beam perpendicular (Fig. 4) than for parallel (Fig. 3) to the extrusion direction. Figure 2, for random orientation, has the (002) plane maximum properly between these two limits. These results have been confirmed by W. H. Zachariasen[8] who, by x-ray analyses of the same graphite samples, found the same approximate ratio 2:1, for coherent scattering of x-rays perpendicular and parallel to the extrusion direction.

In conclusion, it is seen that the effect of crystal orientations on the Bragg scattering of slow neutrons may be considerable and that measurement by neutron scattering affords a means of estimating the amount of microcrystalline orientation. The authors wish to thank Dr. R. G. Sachs for suggesting this problem and for his helpful comments.

[8] William H. Zachariasen, Argonne National Laboratory unpublished report.