The Transmission of Slow Neutrons through Microcrystalline Materials*

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The transmission of monochromatic slow neutrons through microcrystalline Be and BeO has been determined. The source of neutrons was the Argonne heavy water pile. These neutrons were monocromatized by means of a mechanical velocity selector for low energies and a neutron crystal spectrometer for higher energies. The results are in excellent agreement with the theory of elastic scattering from crystals. It is found by comparison of the results on BeO with the theory that the scattering amplitudes of Be and O have the same sign. This method may be used to determine the relative scattering phases of other pairs of nuclei which can be combined to form a crystalline material. The sample must consist of crystals smaller than a micron in linear dimensions. Other possible sources of disagreement between theory and experiment are discussed in Section 5.

1. INTRODUCTION

THE measurement of the effective scattering cross section of crystalline materials for slow neutrons is of considerable interest in connection with problems of neutron diffusion. If the material consists of nuclei having a small absorption cross section, the scattering cross section can be determined by measuring the transmission coefficient for slow neutrons. It is known that the scattering cross section determined in this way is a very sensitive function of neutron energy which shows violent fluctuations for small changes in the energy. These fluctuations are associated with the appearance of Bragg reflections from the appropriate planes in those of the microcrystals which are properly oriented. The shape of the transmission curve has been determined theoretically by Halpern, Hammermesh, and Johnson[1] and by Weinstock[2] for materials containing one atomic species. The generalization to multi-atomic materials is simple and will be given in Section 4.

With the intense monochromatic neutron sources which are now available, it is possible to obtain a rather precise measurement of transmission curves as a function of energy. Measurements of this kind have been carried out for microcrystalline Be and BeO. It is the purpose of this paper to present the results of these measurements and to compare them with theory. In particular, it will be shown in Section 4 how the results can be used to determine the relative phases of the Be and O scattering.

In Section 2, the experimental methods for obtaining monochromatic neutron beams are discussed and in Section 3 the experimental results obtained with these beams are given. Section 4 contains a discussion of the theory with particular reference to its application to materials containing more than one atomic species. The limits of applicability of the theory are considered in Section 5.

2. EXPERIMENTAL METHOD

The monoenergetic neutron beams required for the measurements were obtained by means of two velocity selectors in conjunction with the heavy water pile at the Argonne Laboratory. The first was the mechanical velocity selector first used by Fermi, Marshall, and Marshall as modified by Brill and Lichtenberger,[3] which by use of a motor driven cadmium shutter and proper timing devices can be applied to the measurement of cross sections for the range of neutron energies between 0.004 and 0.20 electron volt. The second, which extended the measurements in one case to about 1.0 electron volt,

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[1] O. Halpern, M. Hammermesh, and M. H. Johnson, Phys. Rev. 59, 981 (1941).

[2] R. Weinstock, Phys. Rev. 65, 1 (1944).

[3] This velocity selector will be described in forthcoming papers by E. Fermi, L. W. Marshall, and J. Marshall and by T. Brill and H. D. Lichtenberger. This description of a use of the instrument before publication of their papers is presented with the permission of these authors.

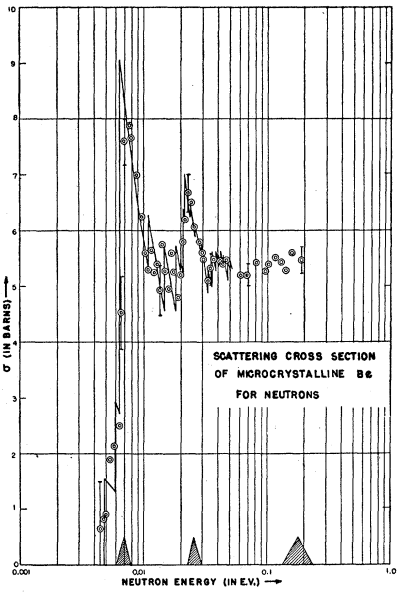

FIG. 1. The microcrystalline beryllium cross section. The solid line represents the cross-section function as calculated in Section 4. Triangles at the base of the curve indicate the resolution of the instrument in the various ranges of measurement. The cross sections, σ, are given in units of 10^{-24} cm^2/atom (barns).

made use of the monoenergetic beams of neutrons diffracted from LiF(100) in the neutron crystal spectrometer which has been previously described.[4]

The bursts of neutrons periodically released by the rotating shutter of the mechanical velocity selector passed through a neutron detector after traversing a measured path between the shutter and detector. The output of the detector, recorded during short measured intervals after the release of the initial burst and during the time the burst was passing through the counter, made it possible to record only neutrons in a very small velocity range. By introducing a sample of a microcrystalline substance into the periodic beam released by the shutter, it was possible to measure the transmission of the substance as a function of energy by measuring the fraction of neutrons scattered by the sample in each small energy interval. The value of the effective scattering cross section was calculated for this interval from the measured transmission data.

[4] W. J. Sturm and S. H. Turkel, Phys. Rev. **70**, 103 (1946); W. H. Zinn, Phys. Rev. **70**, 102 (1946).

The neutron source was a beam from the graphite thermal column of the chain reactor collimated by a series of three absorbing slits to a $\frac{3}{4}''\times 3''$ area. The spectrum of neutrons emitted from this source had an approximate Maxwellian distribution of velocities with its peak at about 0.04 electron volt. The intensities in the range between 0.004 to 0.20 electron volt were sufficient for these measurements; outside this range intensities were generally too low to be used. Passing through the shutter the interrupted beam fell upon the absorber and reached the detector, 150 cm from the rotating cylinder. The detector, mounted with its axis normal to the beam, was a BF_3 proportional counter, 3.8 cm in diameter and 11 cm long, having a central wire 0.002 inch in diameter. The filling gas was enriched in the isotope B^{10} to increase its counting efficiency. In order to minimize the effect of background neutron radiation, the whole counter except for a port to admit the beam was surrounded by a 2.5-cm shield layer of boron carbide.

Measurement of transmission was made by comparing the beam intensity at the various energies with the scattering sample interposed in the beam to the intensity of the unfiltered beam. A second series of similar measurements was taken at each energy to evaluate the intensity of neutrons leaking through the shield plus those fast neutrons which passed through the closed shutter. This measurement was made by stopping the shutter and setting it at the position which it assumed at the time the signal from the detector was sent to the recorders. In this position only fast neutrons could pass through the shutter to reach the detector. In all cases this background intensity was less than 5 percent of the total flux in each energy interval, reaching this fraction only in the extremes of the energy range. The transmission was then calculated from the relation

$$T=R(a)/R(0),$$

where $R(a)$ is the counting rate in the filtered beam corrected for background, and $R(0)$ is the counting rate of the open beam similarly corrected.

3. RESULTS

A. Beryllium

The sample for the measurements was prepared from a block of very pure microcrystalline beryllium metal reduced to chips by turning on a lathe. These chips, subsequently further pulverized by trituration in a mortar and pestle made of beryllium metal, were reduced to their final size in a beryllium ball mill. A 3.25″ diameter cylindrical disk of the material having a surface density of 2.45 grams/square centimeter was prepared by pressing in a die. The cross section per atom shown plotted in Fig. 1 was calculated from the relation

$$\sigma = -\ln T/N$$

in which N is the number of atoms of Be per cm^2 and T is the transmission measured in the manner discussed above. Solid lines represent theoretically predicted values. Uncertainties in the cross section were evaluated by assigning a statistical uncertainty equal to the square root of the number of counts to each of the four measured components of the transmission value. Because the intensities were lowest at the extremes of the energy range, cross-section measurements at these energies show the greatest statistical uncertainty.

B. Beryllium Oxide

A sintered block of BeO, prepared from very fine powder crystals was used to measure the scattering cross section for this compound. Cross-section values were calculated from transmission measurements and, as can be seen from Fig. 2, agree very well with the theoretical treatment given below.

In the case of the beryllium oxide curve, mechanical velocity selector measurements were repeated on the neutron crystal spectrometer in the range between 0.04 and 0.20 electron volt and extended to 1.0 electron volts by this method.

4. THEORY

Although the theory of the coherent scattering of neutrons by microcrystalline materials has been given,[1,2] it seems worth while to indicate the physical factors which go into the results. For this purpose, we consider a beam of neutrons moving in a given direction incident upon a single microcrystal. The wave-lengths of the neutrons in the beam are assumed to be distributed uniformly between λ and $\lambda+\Delta\lambda$. The orientation of the microcrystal is such that one of the wave-lengths in the interval $\Delta\lambda$ undergoes Bragg reflection from some particular set of lattice planes. Then, since the scattered wave is undergoing constructive interference, the scattered intensity is proportional to the square of the number of scattering centers. Thus, if N_0 is the number of nuclei in the microcrystal and F the scattering cross section of a nucleus, the scattered intensity is proportional to N_0^2F.

Since the crystal is finite in size, the scattered beam is spread by diffraction over a solid angle of the order of $(\lambda/l)^2/\cos\theta$ where l is the average linear dimension of a microcrystal and θ is the angle between the scattered beam and the

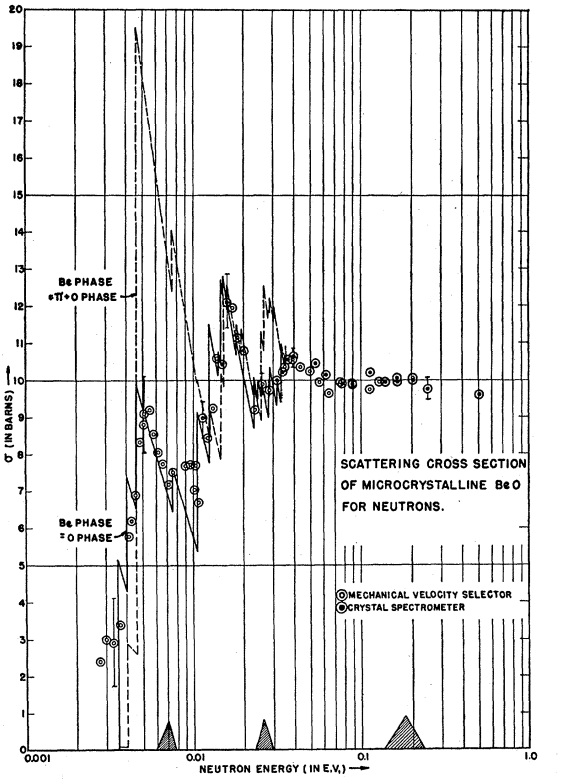

FIG. 2. Scattering cross section of BeO. Alternate theoretical curves for same and opposite phase of neutron scattering are shown as solid and dotted lines, respectively. Results indicate that the Be phase is the same as the oxygen phase.

normal to the crystal plane responsible for Bragg reflection. Since $\cos\theta = b\lambda/2$ where b is the product of the order of the reflection and the reciprocal of the spacing between lattice planes, the intensity of the scattered beam integrated over its spatial width is proportional to

$$\lambda N_0^2 F/l^2 b. \quad (1)$$

Only neutrons within a small range of wave-lengths in the interval $\Delta\lambda$ contribute to this scattering. The fraction of the neutrons with the proper wave-length can be obtained from the resolving power of the crystal which is $d\lambda/\lambda = 1/bl$ where bl is equal to the product of the order of the reflection and the number of crystal planes contributing to the reflection. The fraction of the incident neutrons which are scattered is therefore $d\lambda/\Delta\lambda = \lambda/bl\Delta\lambda$, so the ratio of the intensity scattered to the incident intensity is proportional to

$$N_0^2 F\lambda^2/l^3 b^2 \Delta\lambda. \quad (2)$$

This scattered intensity is now to be averaged over all orientations of the microcrystal. Since the crystal gives rise to a Bragg reflection only if its orientation is such that the Bragg condition is satisfied for some wave-length within $\Delta\lambda$, a contribution to the scattering is obtained only in the solid angle $2\pi\Delta(\cos\theta) = 2\pi b\Delta\lambda/2$. The average over-all orientations, therefore, gives rise to the factor $\Delta(\cos\theta)/2 = b\Delta\lambda/4$. The final result for the scattering cross section per nucleus corresponding to scattering of a particular order from a particular set of lattice planes is, apart from numerical factors,

$$\sigma_b = NF\lambda^2/b, \quad (3)$$

where N is the number of nuclei per unit volume. This cross section is to be summed over all sets of crystal planes and all orders of reflection which are consistent with the Bragg condition; i.e., for which $b \leq 2/\lambda$.

From this result it can be seen that for very low neutron energies of wave-length large compared to twice the spacing between any adjacent lattice planes, no Bragg reflection can occur so there is no coherent scattering. When the energy is increased to the point at which the Bragg condition is just satisfied for the most widely spaced pair of lattice planes, there is a discontinuous jump in the scattering given by Eq. (3). Then the cross section decreases proportionally to the square of the neutron wave-length until the Bragg condition is satisfied for the next most widely spaced pair of lattice planes. At that point, another term of the type Eq. (3) is introduced so the cross section again increases discontinuously. This behavior repeats itself as the energy is increased so a jagged curve of the form shown in Fig. 1 is obtained.

The exact form of the elastic scattering cross section with the correct numerical factors including a factor which takes into account the zero point and thermal oscillations of the crystal is[2]

$$\sigma_c = \sum_{b \leq 2/\lambda} FN\lambda^2 \exp(-\omega b^2)/8\pi b, \quad (4)$$

where ω is a constant which depends on the Debye temperature and temperature of the lattice and is to be obtained from reference 2, Eq. (30). F is to be interpreted as the form factor of the unit cell in the crystal and N the number of unit cells per unit volume. Then σ_c is the cross section per unit cell.

Since the Be lattice is hexagonal close packed, the form factor is given by

$$F_{\mathrm{Be}} = 2\sigma_{\mathrm{Be}}[1+\cos\pi(2l+4m+3n)/3]/\mu_{\mathrm{Be}}^2, \quad (5)$$

where σ_{Be} is the cross section of the free Be atom, μ_{Be} is the reduced mass, in units of the neutron mass, of the neutron and a free Be atom, and l, m, n are the products of the order of the reflection and the Miller indices of the plane leading to the particular Bragg reflection under consideration. The appearance of the factor $1/\mu_{\mathrm{Be}}^2$ is owing to the very large effective mass of a Be atom which scatters neutrons with an energy small compared to the binding of the Be in the lattice.[5] The theoretical curve obtained from Eq. (4) for Be at room temperature ($T = 293°\mathrm{K}$) is given in Fig. 2. The cross section σ_{Be} has been taken to be 6.1×10^{-24} cm^2. This value is obtained from the mean experimental cross section for a neutron energy of the order of several volts (an energy at which the Be atom may be treated as free). The Debye temperature of Be has been taken to be[6] $\Theta = 1000°\mathrm{K}$.

[5] E. Fermi, Ricerca Scient. **7**, No. 2, 13 (1936). See also R. G. Sachs and E. Teller, Phys. Rev. **60**, 18 (1941).

[6] Mott and Jones, *Properties of Metals and Alloys* (Oxford, 1936).

The generalization of these considerations to polyatomic crystals leads again to Eq. (4). In this case, however, the form factor depends on the positions of the different atomic species in the unit cell and on the relative signs of the amplitudes of the waves scattered from the different nuclei. If our considerations are limited to biatomic crystals, the form factor is given by

$$F=4\pi|A_1\sum_{abc}\exp[2\pi i(la+mb+nc) +A_2\sum_{\alpha\beta\gamma}\exp[2\pi i(l\alpha+m\beta+n\gamma)]]|^2, \quad (6)$$

where abc are the positions in terms of the primitive translations of the lattice of the nuclei which scatter with amplitude A_1, and $\alpha\beta\gamma$ are the corresponding positions of the nuclei which scatter with amplitude A_2. For the BeO crystal this reduces to

$$F_{\mathrm{BeO}}=4\pi|A_{\mathrm{Be}}+A_{\mathrm{O}}\exp 3\pi in/4|^2F_{\mathrm{Be}}$$

where F_{Be} is to be obtained from Eq. (3). In particular, for $n=\pm1$, the amplitude dependent factor is

$$A_{\mathrm{Be}}^2+A_{\mathrm{O}}^2-\sqrt{2}A_{\mathrm{Be}}A_{\mathrm{O}}.$$

Since A_{Be} and A_{O} are of the same order of magnitude, this quantity depends rather sensitively on the relative signs of the amplitudes A_{Be} and A_{O}. It is larger if the signs are opposite. Thus, for the reflections from the twelve sets of planes $(l, m)=(\pm1, 0)$, $(0, \pm1)$, $(1, 1)$, $(-1, -1)$ with $n=\pm1$, there will be a considerable difference in the strength of the Bragg peak between the two cases of like sign and opposite sign. This makes it possible to determine experimentally by a transmission measurement whether the signs A_{Be} and A_{O} are equal or opposite.

The theoretical curves for the cross section as a function of energy are given in Fig. 2 for both possible choices of sign. In obtaining these curves, the magnitudes of the scattering amplitudes were obtained from

$$4\pi|A_{\mathrm{Be}}|^2=\sigma_{\mathrm{Be}}/\mu_{\mathrm{Be}}^2, \quad 4\pi|A_{\mathrm{O}}|^2=\sigma_{\mathrm{O}}/\mu_{\mathrm{O}}^2, \quad (7)$$

where σ_{O} is the scattering cross section of a free O atom (found as before from the scattering of one volt neutrons to be 4.1×10^{-24} cm^2) and μ_{O} is the reduced mass of the neutron plus a free O atom. The Debye temperature of BeO was estimated from the velocity of sound and found to be $\Theta=1200$°K. Comparison of the experimental points and the theoretical curves shows clearly that the signs of the amplitudes of the Be and O scattering are the same.

5. CONCLUSION

The good agreement between theory and experiment indicates that the application of Eq. (4) to other crystalline materials should give an adequate representation of the scattering cross section as a function of energy. There are, however, a number of conditions that may lead to deviations from this result.

Probably the most important of these would be the occurrence of such large microcrystals in the sample that the peaks in the scattering are wiped out by extinction in the crystals. An estimate of the maximum size of the microcrystals to be used can be obtained by determining what fraction of those neutrons of a given energy which hit a particular crystal at the Bragg angle are scattered. This fraction can be obtained by dividing the expression (1) by the projected cross-sectional area of the crystal which is given by $l^2\cos\theta=l^2b\lambda/2$. If we introduce the correct numerical factor which can be obtained by comparing Eqs. (1)–(4), the fraction of the neutrons scattered by a particular set of crystal planes turns out to be

$$N^2l^2F/\pi b^2. \quad (8)$$

Since it is desirable to have not more than 1 percent of the beam scattered by one microcrystal, the upper limit on the crystal dimensions may be taken to be

$$l\leq(\pi)^{\frac{1}{2}}b/10N(F)^{\frac{1}{2}}. \quad (9)$$

If $b=3\times10^7$ cm^{-1}, $F=10^{-23}$ cm^2 and $N=4\times10^{22}$, Eq. (9) becomes $l\leq5\times10^{-5}$ cm. Thus, for crystals of linear dimensions smaller than a micron, one would not expect an appreciable reduction in the peaks because of extinction. These dimensions refer to the prefect microcrystals of which the material is composed. Considerably larger crystallites would be acceptable if the crystallites consist of many such microcrystals oriented at small angles with respect to each other. In order to be sure that this condition was satisfied for the Be crystals on which were made the measurements discussed in Section 3, it was necessary to break

the material up mechanically; for BeO, there was no such difficulty.

Other sources of deviations from the theory may be the following:

(1) The microcrystals not randomly oriented. This may occur when the sample is prepared by extrusion.

(2) The temperature of the sample not small compared to its Debye temperature. Then an appreciable amount of inelastic scattering would be expected. This would render the peaks less sharp and would introduce appreciable scattering below the first Bragg limit.

(3) The crystal contains two or more isotopic constituents which have appreciably different scattering properties. The incoherent scattering background would then be appreciable. However, it is necessary that there be a very strong dependence of the scattering amplitude on the isotopic identity of the nucleus for this to be a large effect.

(4) The nuclei of the sample have a spin different from zero and the scattering is strongly spin dependent. This would again lead to an incoherent background which would be large only if the spin dependence of the scattering were very strong.

(5) If the sample used is made up of a very fine powder, the surface area will be so large that there is a chance that adsorbed surface films may contribute appreciably to the cross section. This may lead to an erroneous interpretation of the results, particularly if one is trying to distinguish between two possibilities corresponding to two different combinations of the signs of the scattering amplitudes. If a large amount of water, for example, is adsorbed on the crystals, there will be a smooth background cross section rising rapidly at low energies which could easily transform the lower curve in Fig. 2 into a curve that could hardly be distinguished from the upper curve.

If care is taken to avoid the difficulties mentioned above, it is believed that the transmission method offers a good procedure for determining the relative phases of the scattering of pairs of nuclei which can be compounded to form a microcrystalline material. This would then make it possible to determine the absolute sign of the scattering phases of a series of nuclei if the phase were determined for one particular nucleus.

The numerical work required for determining the theoretical curves in Figs. 1 and 2 was carried out by M. G. Goldberger.

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Peak Contour and Half-Life of Metastable Ions Appearing in Mass Spectra

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A method is described for determining the half-lives of metastable ions by varying the voltage which sweeps the ions from the ionization chamber of a mass spectrometer. The half-life of the state corresponding to the transition $C_4H_{10}^+ \rightarrow C_3H_7^+ + [CH_3]$ has been found to be 2.0×10^{-6} second and the value found for the transition $C_4H_{10}^+ \rightarrow C_3H_6^+ + [CH_4]$ was 1.7×10^{-6} second. In answer to the objection that the diffuse peaks should not be prominent in the 180° instrument if they are to be attributed to spontaneous dissociation during transit, it is shown that this result is to be expected for metastable transitions. The relative number of ions in the metastable states initially is estimated.

INTRODUCTION

THE diffuse peaks appearing in the mass spectra of hydrocarbons have recently been attributed to the spontaneous dissociation of metastable ions during transit.[1,2] Several workers in the field have objected to this interpretation on the basis that the appearance of diffuse peaks in the 180° instrument is unexplained—the peaks having roughly the same appearance as in the sectored-field instrument such as that employing a deflection of 90°. In the latter instrument there is a considerable distance between the ion source and the deflecting magnetic field where the dissociation could occur without an appreciable deflection of the ions, and this situation does not obtain in the former case. This paper will attempt to show that a diffuse peak observed

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[1] J. A. Hipple and E. U. Condon, Phys. Rev. 68, 54 (1945).

[2] J. A. Hipple, R. E. Fox, and E. U. Condon, Phys. Rev. 69, 347 (1946).