

Anomalous Absorption of Slow Neutrons and X-rays in Nearly Perfect Single Crystals

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Two nearly perfect calcite crystals were set in the parallel arrangement of a two-crystal neutron spectrometer and the γ -rays from neutron capture by the calcium of the second crystal, which was 2 in. thick, were measured with a NaI scintillation detector. A maximum change in γ -ray intensity of $2.5 \pm 0.6\%$ occurred near the angle of Laue reflection. An application of the dynamical diffraction theory gave a 'dispersion curve' variation with angle for the γ -ray intensity. This theory agreed quantitatively with the experimental observation. In a second experiment concerning a search for anomalous absorption of neutrons incident at the Bragg angle, the transmission of neutrons by a 2 mm. thick cadmium sulphate crystal was measured and compared with the transmission of X-rays. The incident radiations (1.30 Å neutrons and 0.708 Å X-rays) were selected by reflection from calcite crystals. When the 2 mm. crystal was set near the position of Laue reflection the X-ray transmission increased from 1.5×10^{-4} to 4.5×10^{-4} but the neutron transmission decreased from 3.3×10^{-2} to 2.9×10^{-2} , this decrease being equal to the reflected intensity. Both observations may be explained by postulating a mosaic block size of 0.4 mm. in the cadmium sulphate crystal.

Introduction

Anomalies in the absorption of X-rays have been observed by several workers when the X-rays were incident on nearly perfect crystals set at, or almost at, a position of Laue reflection (Borrmann, 1941, 1950; Campbell, 1951; Schwarz & Rogosa, 1954; Brogren & Adell, 1954). The anomalies have been shown to be predicted quantitatively by the dynamical theory of X-rays (Laue, 1949; Hirsch, 1952; Zachariasen, 1945, 1952). According to this theory, when the Laue conditions are satisfied one of the Fourier components of the electromagnetic field inside the crystal is enhanced. The interaction with the spatially periodic electron density then results in absorption which differs from absorption at random settings of the crystal.

Similar effects are to be expected if neutrons are substituted for X-rays. The present paper reports the results of two experiments of this nature.

In the first experiment a calcite crystal was placed in a monoenergetic neutron beam, and the γ -ray intensity emitted from the crystal was measured as the crystal was turned through an angle of Laue reflection. The γ -rays are attributed mainly to neutron capture in the calcium, and the changes in intensity are attributable to changes in the neutron field at the calcium sites. The results confirm the periodicity of the fields inside the crystal.

In the second experiment a cadmium-containing crystal, $3 \text{ CdSO}_4 \cdot 8 \text{ H}_2\text{O}$, was placed in the neutron beam and the neutron transmission was studied in the neighbourhood of a Laue position. No anomalous absorption of the transmitted neutron beam was observed. The same crystal, using X-rays, showed

striking anomalous absorption. Calculation assuming an ideal absorbing crystal predicted a marked effect with both neutrons and X-rays. The difference is considered to have arisen because the crystal was not ideal, and can be accounted for quantitatively by assuming the crystal to have a mosaic structure.

Experiment with a calcite crystal

Plan diagrams of the experimental arrangement for the first experiment are shown in Fig. 1(a) and Fig. 2. A 2 in. diameter beam of slow neutrons having an angular divergence of about 1° was Laue reflected from the (211) plane of a cleaved calcite crystal A' in Fig. 1(a). Crystal A' is enclosed by 9 in. of boron carbide and paraffin and an outer $\frac{1}{2}$ in. sheath of lead, except for the inlet channel V and the outlet channel W. The extension of the outlet channel W is a 1.5 in. square aperture through an 8 in. thick lead wall. The beam I emerging from this 1.5 in. aperture, shown in Fig. 2, was incident on a second calcite crystal at A, set in the parallel position for Laue reflection. This second crystal, which has dimensions roughly 1 in. \times 1.75 in. \times 2 in., was placed so that the neutrons entered through the 1 in. \times 1.75 in. face, and were reflected from the (211) planes parallel to the 1 in. \times 2 in. face. The neutron beam, incident on the second calcite crystal A, was further defined by an aperture Y in a boron carbide shield. These neutrons, after reflection from crystal A, were detected by a BF_3 counter D, Fig. 1(a).

To detect the 2 MeV. γ -rays from the $\text{Ca}(n\gamma)$ reaction in calcite, a $1\frac{1}{2}$ in. cylindrical NaI crystal was used in conjunction with a 5819 photomultiplier. This

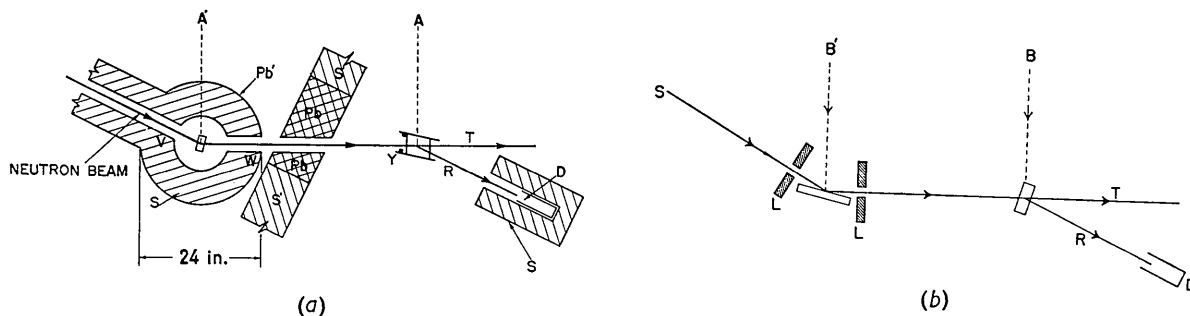


Fig. 1. (a) Experimental arrangement of neutron spectrometer. V, 2 in. diameter inlet channel for neutrons. A', calcite monochromatizing crystal. W, outlet channel for monochromatic neutrons $\lambda = 1.30 \text{ \AA}$. Pb, lead wall with 1.5 in. square aperture. Pb', $\frac{1}{2}$ in. lead shielding. Y, boron carbide defining aperture. A, specimen calcite crystal. D, BF₃ counter for measuring reflected beam R or transmitted beam T. S, boron and paraffin shielding. S', Masonite and steel shielding.

(b) Experimental arrangement of X-ray spectrometer. S, X-ray source, Mo $K\alpha$, $\lambda = 0.708 \text{ \AA}$. B', calcite monochromatizing crystal. B, specimen crystal. D, krypton, methane (Kr, 65 cm. Hg; CH₄, 6.5 cm. Hg) proportional counter for measuring reflected beam R or transmitted beam T. L, limiting apertures.

detector was mounted rigidly, directly above the calcite crystal A, Fig. 2. Background radiation from the pile room was reduced by surrounding the NaI crystal with 4 in. of lead. The counting rate from the background radiation was further reduced by making use

Fig. 2, and with the first crystal A', in Fig. 1(a), turned so that the Laue-reflected neutrons did not emerge from the aperture. The upper curve of Fig. 3 was obtained when the neutron beam was incident on crystal A. The absorption of thermal neutrons in

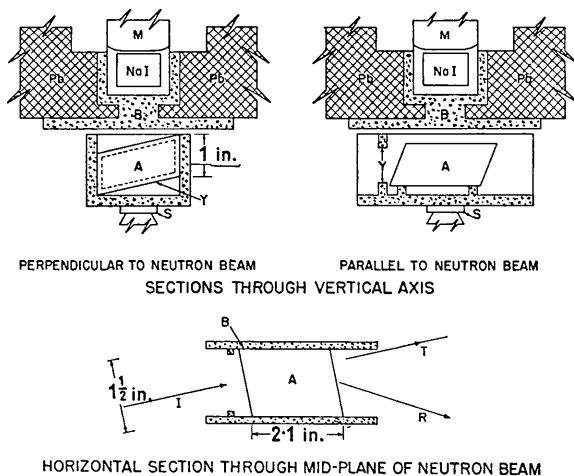


Fig. 2. Experimental arrangement of calcite crystal and sodium iodide crystal detector. S, spectrometer table. Pb, lead shielding. B, boron carbide shield. Y, aperture in boron carbide shield. M, photomultiplier. I, the 1.5 in. square incident neutron beam. R and T, the reflected and transmitted beams respectively.

of a single channel pulse-height analyser to select pulses of appropriate magnitude. It was important that γ -rays originating from crystal-scattered neutrons captured outside the crystal should not be counted. Shields of boron carbide $\frac{1}{4}$ in. thick were placed around the NaI crystal and about the calcite crystal in order to capture the scattered neutrons. The γ -rays produced by capture in boron are too low in energy to be detected with the channel settings used.

The pulse-height distribution due to background radiation is shown in the background curve of Fig. 3. The curve was obtained with the geometry shown in

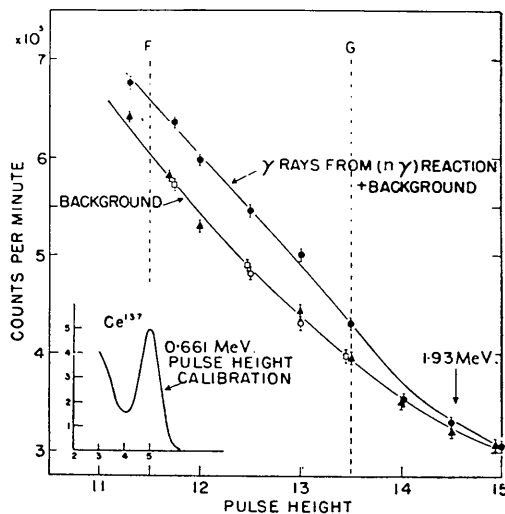


Fig. 3. Pulse-height distribution using a $1\frac{1}{4}$ in. cylindrical NaI scintillation detector. Solid circles: neutron beam incident on calcite at A; squares: neutron beam incident on graphite block at A; open circles: neutron beam present, but no scatterer present at A; triangles: neutron beam absent at A. The insert is the photo-peak of Ce¹³⁷ used to calibrate the pulse-height analyser. The vertical lines on each point measure the statistical standard deviation. F and G bracket the pulse-height channel region used in the crystal rotating experiment of Fig. 7.

natural calcium occurs predominantly in Ca⁴⁰ and Ca⁴². A γ -ray of 1.93 MeV. has been observed (Braid, 1953). It appears in the reference to have been assigned to the first excited state in the reaction Ca⁴⁰(n, γ)Ca⁴¹ and to have an emission probability of greater than 80% (Kinsey & Bartholomew, 1954). The interaction of these γ -rays with the NaI crystal results in a

broadly distributed pulse-height spectrum, with maximum pulse height corresponding to about 1.93 MeV. A knowledge of the neutron flux incident on the calcite crystal at A, of the $\text{Ca}^{40}(n\gamma)$ cross section and of the geometry and detector efficiency allowed an estimate of the counting rate due to the $\text{Ca}(n\gamma)$ reaction to be calculated. This estimate agreed with the area between the two curves of Fig. 3. The difference between the two curves cannot be attributed to secondary processes following neutron scattering, such as γ -rays following the capture of neutrons scattered into the neighbouring lead. This was demonstrated by showing that the background intensity, given by the lower curve of Fig. 3, was unaltered when the calcite crystal A was removed from its position, and also when the calcite crystal A was replaced with an equal volume of graphite, which is a good scatterer of neutrons; the neutron beam reflected from the first crystal A' in Fig. 1(a) was at full strength in both cases.

The observation on the variation of γ -ray intensity with the rotation of the crystal was made with the pulse-height channel between F and G in Fig. 3. The difference of the areas under the two curves between F and G divided by the area of the lower curve between F and G gave 0.126 as the fraction of the γ -ray intensity due to the $\text{Ca}(n\gamma)$ reaction.

A measure of the perfection of crystal A was made by analysis of the ratio of the neutron intensity $P_H(y)$ reflected from the H planes of crystal A, to the intensity P_0 incident on this crystal. The geometry of the double crystal spectrometer in Fig. 1(a), which has the two crystals set in the parallel position, is such that P_H/P_0 is given by (Zachariasen, 1945)

$$\frac{P_H(y)}{P_0} = \frac{1}{R'_H} \int_{-\infty}^{+\infty} \frac{I_H(y)}{I_0} \cdot \frac{I'_H(y-y')}{I_0} dy', \quad (1)$$

where

$I'_H(y)/I_0$ is the ratio of the intensity reflected from the H planes of the first crystal A' to the intensity I_0 incident on that crystal,

$I_H(y)/I_0$ is the same ratio for the H planes of the second crystal A,

$$R'_H = \int_{-\infty}^{+\infty} \frac{I'_H(y)}{I_0} dy \text{ is the integrated reflecting power}$$

from the H planes of the crystal A',

and

$$y = \frac{b(\theta_B - \theta) \sin 2\theta_B + \frac{1}{2}(1-b)\psi'_0}{K|\psi'_H||b|^{\frac{1}{2}}}, \quad (2)$$

where $b (= \gamma_0/\gamma_H)$ is the ratio of direction cosines of incident and reflected beams relative to the normal to the surface of the crystal. (In the above experiment $b \approx +1$, since both crystals A and A' are set for symmetrical Laue reflection with respect to the incident beam.)

K is the polarization factor in X-ray diffraction

($K = 1$, for neutron diffraction since the angular variation with polarization present in the diffraction of X-rays, is not present in the diffraction of neutrons).

For X-rays, $\psi_H = \psi'_H + i\psi''_H$ is the Fourier component of index H of 4π times the polarizability.

For neutrons,

$$\psi'_H = -\frac{1}{\pi} \frac{\lambda^2}{V} \sum_j a_j \exp [i\mathbf{B}_H \cdot \mathbf{r}_j]$$

and

$$\psi''_H = -\frac{\lambda}{2\pi} \mu_H,$$

where

a_j is the coherent scattering length for atom (j) at position \mathbf{r}_j ,

\mathbf{B}_H is the reciprocal-lattice vector, of magnitude $\mathbf{B}_H = 2\pi/d_H$ (d_H is the interplanar spacing of the planes H),

μ_H is the Fourier component of the linear absorption coefficient μ for the crystal (see equation (6)),

V is the volume of the crystal unit cell, and

λ is the wavelength of the incident radiation.

$(\theta_B - \theta)$ is the angular deviation of the diffracted neutron beam from the Laue reflecting angle θ_B .

For the neutron-diffraction experiments equation (2) becomes

$$y = \frac{\sin 2\theta_B}{|\psi'_H|} (\theta_B - \theta). \quad (3)$$

The double-crystal integrated reflecting power R is obtained by integrating equation (1) with respect to the parameter y

$$R = \int_{-\infty}^{+\infty} \frac{P_H(y)}{P_0} dy = R_H, \quad (4)$$

where R_H is the reflecting power from crystal A. Thus the double-crystal integrated reflecting power is dependent only on the integrated reflecting power of the second crystal. The real crystal integrated reflecting power approaches a minimum value the more perfect the crystal. For an ideal 'thick' crystal with small absorption, $R_H = \frac{1}{2}\pi \exp[-\mu_0 t/\gamma_0]$ (Zachariasen, 1945). For an ideal calcite crystal with 6% absorption, $R_H = 1.48$.

Fig. 4(a) shows the rocking curve for neutrons diffracted from the (211) planes in the calcite crystal at position A of the double crystal spectrometer. The integrated reflecting power R (the area under the curve), is 1.63, which is nearly equal to 1.48, showing that crystal A is nearly ideal. However, the width, ω_y , at half-maximum intensity of the curve is 9.6, as compared to the calculated value 2.8 for two ideal crystals A and A'. It is concluded that the broadening of the curve must be due to imperfections in the first crystal A'. A more detailed idea of the perfection of the second crystal A was obtained by observing the rocking curves produced by neutrons scattered from a number of internal positions in crystal A. The arrangement shown in Fig. 5 was used. The beam inci-

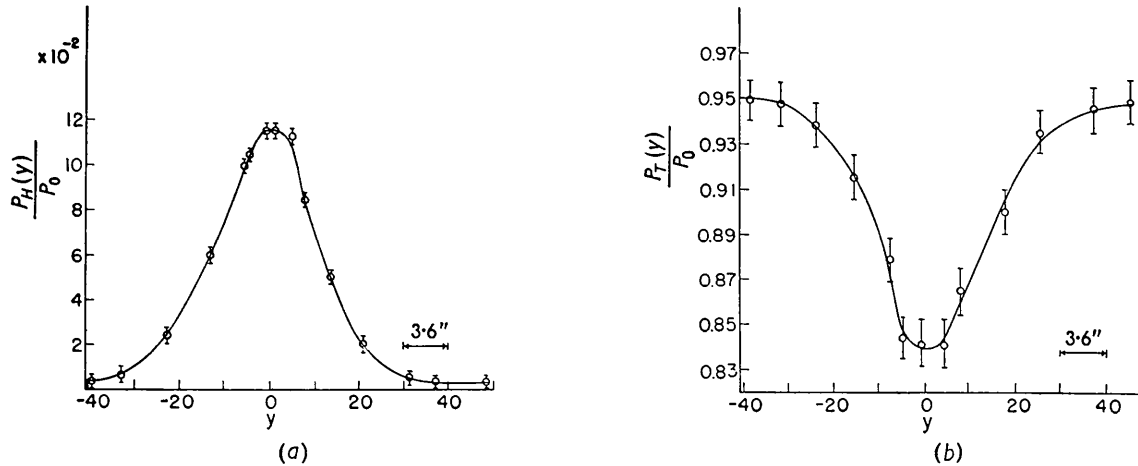


Fig. 4. (a) The reflection of 1.30 Å neutrons by a calcite crystal 5.3 cm. thick plotted as a function of the variable y in the region of Laue reflection from the (211) planes. R_H , the area of the peak in the reflection, and ω_y , (ω_θ in seconds), the width at half-maximum intensity of the peak, are tabulated below with theoretical values calculated for an ideal low-absorbing crystal. The vertical lines on each point measure the statistical standard deviation.

	ω_y	ω_θ	R_H
Experiment	13.0	9.4''	1.63
Theory	12.7	9.2''	1.48

(b) The transmission of 1.30 Å neutrons by a calcite crystal 5.3 cm. thick plotted as a function of the variable y in the region of Laue reflection from the (211) planes. R_T , the area of the dip in the transmission, and ω_y , (ω_θ in seconds), the width at half-maximum intensity of the dip, are tabulated below with theoretical values calculated for an ideal low-absorbing crystal. The vertical lines on each point measure the statistical standard deviation.

	ω_y	ω_θ	R_T
Experiment	13.0	9.0''	1.65
Theory	12.7	9.2''	1.48

dent on crystal A was 1 mm. wide. The 1 mm. slit E was set at a number of positions across the crystal face. The plot of peak height versus position of the slit is given in Fig. 6. This curve shows that the crystal reflecting power is uniform throughout its interior and is somewhat greater at its surfaces.

The direct observation of anomalous absorption by the calcium was made as follows. With the BF_3

counter set to detect the neutrons diffracted by A, the γ -ray and neutron counting rates were measured simultaneously for several angles of incidence near the angle of Laue reflection. The measurement at each

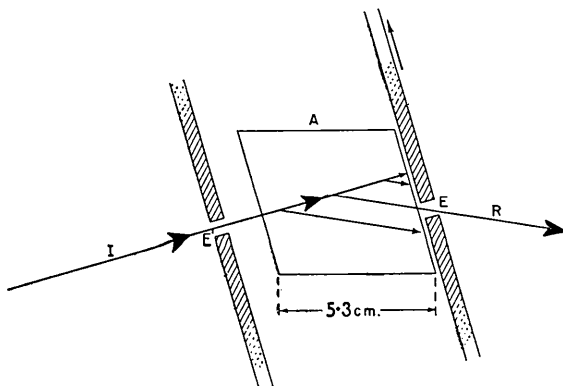


Fig. 5. Experimental arrangement for measuring the efficiency of reflection of slow neutrons, $\lambda = 1.30 \text{ \AA}$, from inside a calcite crystal A. I, the incident neutron beam. R, the reflected beam, which is detected in a wide BF_3 counter. E', a slit 1 mm. wide \times 10 mm. high in a cadmium screen. E, a 1 mm. slit in a movable cadmium screen.

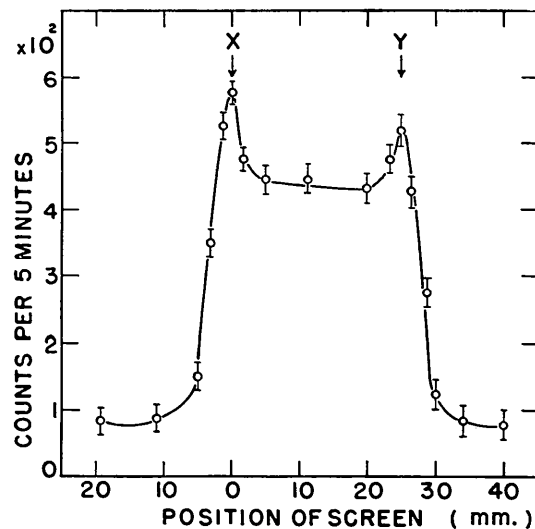


Fig. 6. Peak height versus position of slit E, Fig. 5. X and Y are the positions corresponding to the surfaces of the calcite crystal A. The vertical lines on each point measure the statistical standard deviation.

angle was followed immediately by a measurement at a chosen reference angle to permit normalizing the counting rates to a standard incident intensity. The points in Fig. 7 show the γ -ray intensity variation

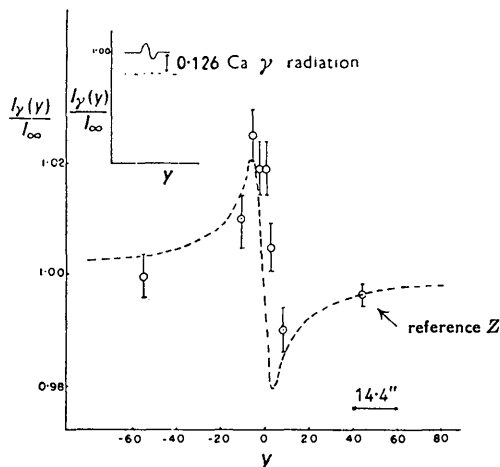


Fig. 7. The measured γ -ray intensity as a function of incident angle, when the 1.30 Å neutrons are incident on the calcite crystal in the region of Laue reflection from the (211) planes. The broken curve is the calculated γ -ray intensity based on the dynamical theory. The insert shows the proportion of the γ -rays attributed to calcium. The vertical lines on each point measure the statistical standard deviation.

with incident angle. Fig. 4(a) shows the corresponding diffracted neutron intensity variation. In both of these measurements $y = 0$ corresponds to the angle of Laue reflection. This angle is determined by the position of the peak of the neutron-diffracted intensity in Fig. 4(a). The broken curve in Fig. 7 shows the calculated γ -ray intensity variation. The curve is fitted to the experimental points horizontally by having the theoretical $y = 0$ coincide with the measured $y = 0$ and vertically by having the theoretical curve pass through the reference intensity at angle Z . The maximum observed variation of intensity is about 2.5% of the total γ -ray intensity. This corresponds to a 15% variation of the calcium γ -ray intensity.

The transmission of the calcite crystal A is shown in Fig. 4(b). The area R_T of the dip in transmission is experimentally equal to the area R_H of the reflection peak, Fig. 4(a). This shows that a portion of the incident radiation has been deflected into the Laue reflected beam, and that anomalies in the absorption of the transmitted intensity, which would result in slight asymmetries in the curve shape, are too small to be observed in such a weakly absorbing crystal. The total absorption of the beam in the 5.3 cm. calcite crystal is 6%.

Theory and discussion

The basic equation of the dynamical theory is the equation for self-consistency of the wavefields inside the crystal (Zachariasen, 1945). It has been shown

that when the direction of the radiation incident on the crystal is in the neighbourhood of a Laue reflection the equation of self-consistency representing the internal field reduces to two sets of plane wave systems. Each set consists of an internal incident and diffracted wave. Starting from the equations representing these plane wave systems, and using the boundary conditions at the face of a semi-infinite crystal, the following expression for the internal field produced by a parallel monochromatic beam of radiation in the region of Laue reflection was developed. The ratio of intensity $I^i(\mathbf{r})_H$ at position \mathbf{r} inside the crystal to the incident intensity I_0 when the incident radiation is in the neighbourhood of the Laue reflection angle of the H planes of the crystal is

$$\frac{I^i(\mathbf{r})_H}{I_0} = \exp \left[-\frac{\mu_0 r}{\gamma_0} \right] \left\{ 1 + K \frac{(\chi'_H)}{|\psi'_H|} \cos(\mathbf{B}_H \cdot \mathbf{r}) \cdot \frac{y}{1+y^2} \right\}, \quad (5)$$

where ψ'_H is the real part of ψ_H defined previously, and μ_0 is the average linear absorption coefficient for the crystal.

The conditions under which formula (5) holds are: that the crystal slab is set for symmetrical Laue reflection, that the crystal is thick, that the total absorption is small and that the crystal unit cell has a centre of symmetry.

When the absorption processes occur in a single crystal which has physical properties periodic in position, it is convenient to expand $\mu(\mathbf{r})$, the linear absorption coefficient, as a Fourier series:

$$\mu(\mathbf{r}) = \sum_H \mu_H \exp [i\mathbf{B}_H \cdot \mathbf{r}]. \quad (6)$$

The energy $I''_H(y)$ absorbed per second throughout the volume V of the ideal crystal is

$$I''_H(y) = C \int_V \mu(\mathbf{r}) I^i(\mathbf{r})_H dV, \quad (7)$$

where C is a proportionality constant for the particular radiation incident on the crystal. Substituting equations (6) and (5) into (7) gives

$$\frac{I''_H(y)}{I_\infty} = \left[1 + K \frac{|\psi'_H|}{(\psi''_H)} \frac{(\psi''_H)}{(\psi''_0)} \frac{y}{1+y^2} \right], \quad (8)$$

where

$$\frac{\mu_H}{\mu_0} = \frac{(\psi''_H)}{(\psi''_0)},$$

and

$$I_\infty = C \mu_0 \int_V \exp \left[-\frac{\mu_0 r}{\gamma_0} \right] dV$$

is the energy absorbed in the crystal when the angle of the incident radiation is far removed from the Laue reflection; i.e. $y \rightarrow \infty$. The thermal motion of the atoms in a real crystal may be introduced into the theory for an ideal crystal by Debye-Waller terms which reduce the Fourier coefficients ψ'_H , ψ''_H of the scattering and absorption. In formulae (8) and (2),

Table 1. Comparative factors in the crystal diffraction of X-rays and neutrons

	Calcite		Cadmium sulphate	
	X-ray	Neutron	X-ray	Neutron
Wavelength, λ (Å)	0.708	1.30	0.708	1.30
b ($= \gamma_0/\gamma_H$)	1.07	1.07	1	1
Laue reflecting angle, ($^\circ$)	6.73	12.42	6.67	12.25
C^*	1.61	0.72	1.83	0.27
ψ_H ($= \psi'_H + i\psi''_H$) $\times 10^8$	$192 + i(2.35)$	$151 + i(0.0016)$	$-211 - i(4.72)$	$-52\ddagger - i(3.60)$
$\psi'_0 \times 10^8$	2.45	0.0016	4.98	3.60
k ($= \psi'_H/\psi''_H$)	1.22×10^{-2}	1.06×10^{-5}	2.23×10^{-2}	6.9×10^{-2}
t'_0 (cm.) \ddagger	0.223	5.50	0.210	0.210
A \S	193	208	188	24.4
kA	2.37	0.022	4.17	1.68
kA (mosaic)**	—	—	0.8	0.4

* $\Delta\theta = \theta - \theta_B = C\Delta y$ sec. of arc.

\ddagger The estimation of ψ_H for neutron diffraction in cadmium sulphate is uncertain to about 50% since the distribution of the hydrogen atoms in this crystal is unknown, and since the coherent scattering cross section of cadmium for slow neutrons is uncertain to 50%.

\ddagger t'_0 is the radiation path length in the crystal.

\S For X-rays, A is different for the normal- and parallel-incident radiation. The values listed in the table are the 'mean' values for unpolarized X-rays.

** kA (mosaic) are calculated values based on a mosaic block size of 0.4 mm.

ψ'_H should be replaced by ψ'_H ($\exp[-M'_H]$) and ψ''_H by ψ''_H ($\exp[-M''_H]$). M'_H may differ from M''_H if the distribution of scattering and absorbing power in a given unit cell of the crystal are different. In these experiments the variation of absorption of the primary radiation in the crystal is observed by measuring the variation of intensity of the secondary radiation. Formula (8) still applies and $I''_H(y)/I_\infty$ is the ratio of secondary radiation, detected for a given angle of incidence of the primary radiation, to the secondary radiation detected at an angle of incidence far removed from the Laue reflection angle of the H planes. For example, in the case of a parallel monochromatic beam of slow neutrons incident on a slab of calcite, assumed to be an ideal crystal, $I''_H(y)/I_\infty$ represents the fractional variation of the calcium γ -rays with angle of incidence.

In the experiment with slow neutrons, the angle of incidence was near the Laue reflection angle for the (211) plane of calcite. As shown in Table 1, ψ_{211} is negative so that $|\psi_{211}| = -(\psi_{211})$. Because the neutron waves scattering from the calcium atoms in a (211) plane of calcite are in phase, and because absorption of neutrons occurs in the nucleus of the atom, which has a sphere of influence of linear dimensions small relative to the neutron wavelength, ψ''_H is equal to ψ'_0 . At room temperature the Debye-Waller term decreases ψ''_H by no more than 3% and this decrease is negligible when fitting the theory to experiment. With these considerations, formula (8) takes the simple form

$$\frac{I''_H(y)}{I_\infty} = 1 - \frac{y}{1+y^2}. \quad (9)$$

This formula predicts a variation of γ -ray intensity with angle of incident neutrons which has the shape of a dispersion curve. It says that the γ -ray intensity is the same at $y = 0$ as for large values of y , and in

the neighbourhood of $y = 0$ the intensity should vary by nearly 50% above and below the value at $y = 0$.

The theoretical variation of γ -ray intensity given by (9) cannot be compared directly with the experimentally measured variation from calcite crystal A of Fig. 1(a): the angular divergence of the neutron beam incident on crystal A must be considered. The theoretical variation $I_\gamma(y)/I_\infty$ was obtained from

$$\frac{I_\gamma(y)}{I_\infty} = \frac{1}{R'_H} \int_{-\infty}^{+\infty} \frac{I'_H(y')}{I_0} \cdot \frac{I''_H(y-y')}{I_\infty} dy', \quad (10)$$

where $I''_H(y)/I_\infty$ is taken from equation (9) and $I'_H(y')/I_0$ and R'_H were estimated for crystal A', using the experimental rocking curve, Fig. 4(a). Equation (10) was fitted to the experimental measurements of Fig. 7.

The anomalous absorption of the wavefield inside the ideal crystal can be pictured qualitatively quite simply. For incident radiation near the angle of Laue reflection, the internal field is represented in the theory by two sets of coupled plane wave systems, each set having slightly different wavelengths and slightly different propagation directions relative to the crystal reflecting planes. Each wave system, considered separately, has an intensity distribution which is periodic throughout the crystal lattice. If the crystal is oriented so that the angle of the incident radiation is exactly that of Laue reflection, the two wave systems have about the same amplitude; but because they differ in wavelength, they may, with equal probability, be in or out of phase with each other at equivalent lattice sites in different unit cells. Thus, at precisely the Laue reflecting angle the resultant internal field does not have the standing-wave properties of the individual wave systems with respect to the crystal lattice. This theory predicts that the variation of γ -ray intensity is zero at $y = 0$. The experimental measurements of

Fig. 7 agree with this conclusion. When the incident radiation is at an angle slightly removed from the Laue diffraction angle, one of the two wave systems will have smaller amplitude than the other, so that the standing-wave characteristics of one or the other separate wave system will be predominant. The absorbing atoms coincide with the nodes of the one system of standing waves when the angle of incidence is on the one side of the angle of Laue reflection and with the loops of the other system of standing waves when the angle of incidence is on the other side. This is illustrated by the dispersion shape of the observed γ -ray intensity curve, Fig. 7.

For a highly absorbing ideal crystal, the theory predicts that the amplitude of one of the two wave systems will be negligible after passage through a sufficient thickness of crystal. Under these conditions, the diffraction and transmission will be determined by the characteristics of the remaining wave system. The decrease of absorption of the transmitted and diffracted beams at the angle of Laue reflection requires that the nodes of the remaining wave system approximately coincide with the atomic positions in the crystal. In the case of neutron diffraction, where the atoms are essentially points, the decrease of absorption would be more complete than for X-rays. For this reason the following experiment was performed.

Experiment with a cadmium sulphate crystal, $3 \text{ CdSO}_4 \cdot 8 \text{ H}_2\text{O}$

An attempt was made to observe anomalous transmission of a neutron beam incident at the Laue reflection angle in cadmium sulphate. The dynamical theory of diffraction predicts (Zachariasen, 1945), for an ideal crystal, that anomalous transmission will be observed only for $kA > 1$, where

$$k = \frac{\psi_H''}{\psi_H'} \quad \text{and} \quad A = \frac{\pi K |\psi_H'| t_0}{\lambda (\gamma_0 \gamma_H)^{\frac{1}{2}}}, \quad (11)$$

t_0 being the thickness of the crystal slab.

Table 1 gives comparative factors for the calcite and the cadmium sulphate crystals used, which are significant in the diffraction of X-rays ($\lambda = 0.708 \text{ \AA}$), and slow neutrons ($\lambda = 1.30 \text{ \AA}$). From the values of kA given in the second last row of Table 1, it is apparent that the anomalous absorption of the transmitted beam should be observable for calcite with X-rays, but should not be observable with slow neutrons. For an ideal cadmium sulphate crystal, 2 mm. thick, the anomalous absorption should be observed with both X-rays and neutrons.

Cadmium sulphate was selected because it contains cadmium which has a high absorption cross section for slow neutrons, it forms almost perfect crystals, it has a strong (202) reflection, and the (202) interplanar spacing is only 1% greater than the calcite (211) spacing. This latter property makes possible a

double-crystal spectrometer of low dispersion, having a calcite, a weakly absorbing crystal, in position A' and the cadmium sulphate crystal in position A, Fig. 1(a). To make the dispersion negligible, in the case of neutron diffraction it was necessary to limit the spread of energies in the neutron beam incident on the second crystal by interposing cadmium Soller slits between A and A', Fig. 1(a), which limited the beam to a 12' angular divergence.

A flat slab, 2 mm. thick, was cut from a cadmium sulphate crystal in such a way that the (202) plane was perpendicular to its surfaces. The crystal was etched in water to remove surface material disturbed by the cutting. Using a double-crystal X-ray spectrometer, Fig. 1(b), with a calcite crystal B' in the first position, the transmitted and diffracted intensities from this cadmium sulphate crystal B were measured. The proportional counter used for detection of the radiation was filled with a mixture of krypton and methane; Kr 65 cm. of Hg; CH₄ 6.5 cm. of Hg. The diffracted beam of Fig. 8(a) had a width at half-maximum intensity of 8.4'', and the transmitted beam of Fig. 8(b) a width of 8.9''. Both widths are greater than the theoretically expected value of 3.7''. Two freshly cleaved calcite crystals in the B and B' positions of the spectrometer of Fig. 1(b) gave a curve having a width of 3.7'', which is close to the theoretical value of 3.3''. The best cadmium sulphate crystal shows marked anomalous absorption of X-rays although its broad half-width indicates that it is not ideal.

The diffracted and transmitted neutron intensity in the 2 mm. thick cadmium sulphate crystal are given in Fig. 9(a) and (b). The transmitted beam is reduced in intensity at the diffraction position. This observation is contrary to the result predicted by the dynamical theory for an ideal crystal.

The X-ray measurements had been made before this neutron result was obtained and it now seems clear that a detailed analysis of the X-ray results provide an explanation. In fact, the neutron results can be explained by the imperfections of the cadmium sulphate crystal. If the cadmium sulphate crystal is assumed to be composed of a number of ideal crystallites, slightly tilted about a mean orientation, then the average size of the crystallites can be inferred from the relative areas under the peaks in Fig. 8(a) and (b). As was shown, equation (4), the total integrated reflecting power R of the double-crystal spectrometer depends only on the integrated reflecting power R_H of the second crystal. A similar argument shows that the area R_T under the peak or dip of the transmitted beam of the double-crystal spectrometer depends only on the properties of the second crystal. Provided that the angular dispersion of the mosaic blocks is sufficiently broad, then the ratio R_H/R_T is independent of the angular dispersion of the blocks and depends only on their size.

The X-ray measurements give $R_H/R_T = 1.57$. From Fig. 8 of Hirsch (1952), R_H/R_T corresponds to $kA =$

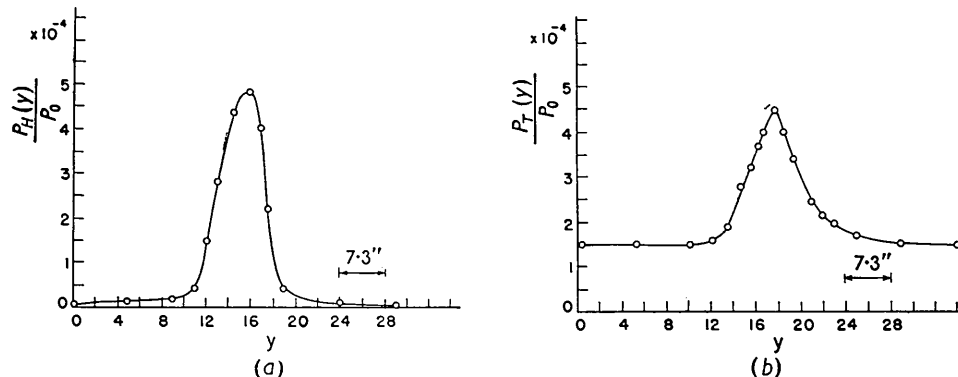


Fig. 8. (a) The reflection of 0.708 Å X-rays by a cadmium sulphate crystal 2 mm. thick plotted as a function of the variable y in the region of Laue reflection from the (202) planes. R_H , the area of the peak in the reflection is given below; also ω_y , (ω_θ in seconds), the width at half-maximum intensity of the peak, together with the theoretical value calculated for an ideal 2 mm. crystal. The statistical standard deviation of each point is approximately equal to the circle diameter.

	ω_y	ω_θ	R_H
Experiment	4.8	8.8''	25.8×10^{-4}
Theory	2.0	3.7''	—

(b) The transmission of 0.708 Å X-rays by a cadmium sulphate crystal 2 mm. thick plotted as a function of the variable y in the region of Laue reflection from the (202) planes. R_T , the area of the peak in the transmission is given below; also ω_y , (ω_θ in seconds), the width at half-maximum intensity of the peak, together with the theoretical value calculated for an ideal 2 mm. crystal. The statistical standard deviation of each point is approximately equal to the circle diameter.

	ω_y	ω_θ	R_T
Experiment	4.8	8.8''	16.5×10^{-4}
Theory	2.0	3.7''	—

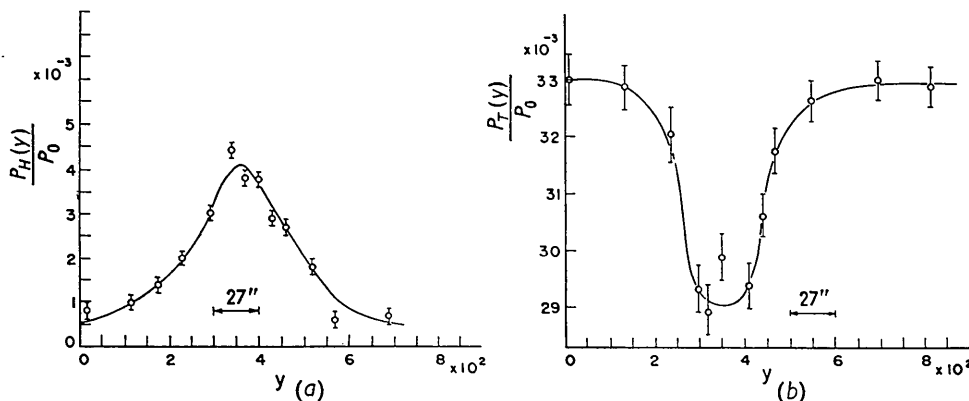


Fig. 9. (a) The reflection of 1.30 Å neutrons by a cadmium sulphate crystal 2 mm. thick plotted as a function of the variable y in the region of Laue reflection from the (202) planes. R_H , the area of the peak in the reflection is given below; also ω_y , (ω_θ in seconds), the width at half-maximum intensity of the peak together with the theoretical value calculated for an ideal 2 mm. crystal. The vertical lines on each point measure the statistical standard deviation.

	ω_y	ω_θ	R_H
Experiment	23×10	62''	86×10^{-2}
Theory	34×10	9.1''	—

(b) The transmission of 1.30 Å neutrons by a cadmium sulphate crystal 2 mm. thick plotted as a function of the variable y in the region of Laue reflection from the (202) planes. R_T , the area of the dip in the translation is given below; also ω_y , (ω_θ in seconds), the width at half value of the dip, together with the theoretical value calculated for an ideal 2 mm. crystal. The vertical lines on each point measure the statistical standard deviation.

	ω_y	ω_θ	R_T
Experiment	19×10	51.5''	92×10^{-2}
Theory	34×10	9.1''	—

0.8. Using this value of kA , the average crystallite size given by formula (11) is 0.4 mm.

For X-rays the 2.1 mm. thick ideal crystal of cadmium sulphate has $kA = 4.3$, for which Fig. 8 of Hirsch (1952), gives $R_H/R_T = 0.7$. For neutrons of wavelength 1.30 Å the 2.1 mm. crystal has $kA = 1.7$, while a 0.4 mm. crystal has $kA = 0.4$ (Table 1). Referring to Hirsch (1952), the former value of kA is in the region of anomalous absorption, the latter value in the region of extinction. Thus if it is assumed that the crystal used was composed of ideal 0.4 mm. crystallites, a decrease in the intensity of the transmitted neutron beam as observed (Fig. 9(b)) is to be expected. Again referring to Hirsch (1952), a value of $kA = 0.4$ gives $R_H/R_T = -1.4$. The experimental ratio, Fig. 9(a) and (b), is $R_H/R_T = -1$. The discrepancy between calculation and measurement is not considered significant since there are large statistical errors on R_H and R_T in Fig. 9(a) and (b).

A comparison of Fig. 9(a) and (b) with Fig. 8(a) and (b) shows that the neutron curves are 6.2 times as broad as the theoretical curves for ideal crystals, while the X-ray curves are only 2.4 times as broad. This difference is readily explainable because the crystallites are of a size that produces anomalous reduction in the absorption of X-rays but not of neutrons. The radiation in the peak of the observed X-ray curve has a large probability of passing through several crystallites with anomalously small absorption. Thus the central region of the X-ray curve is enhanced over the wings; and the result is a curve which is much narrower than the spread of crystallite orientations. On the other hand, the neutron absorption in the crystal is independent of the angle of incidence, so that there is no enhancement of the central region of the neutron curves. The neutron curves, Fig. 9, correctly represent the spread of crystallite orientation.

Conclusion

The observed variation of γ -ray intensity from the Ca ($n\gamma$) reaction in a nearly perfect calcite crystal gives direct demonstration of anomalous absorption of neutrons in thick crystals which have low absorption for neutrons. The absorption was too small for anomalous transmission to be observed. Both of these facts agree with the predictions of the dynamical diffraction theory for a semi-infinite ideal crystal. A study of the transmission of X-ray and neutrons in the neighbourhood of a Laue reflection position for a cadmium-containing crystal, $3\text{ CdSO}_4 \cdot 8\text{ H}_2\text{O}$, showed anomalous transmission of the incident beam with X-rays but showed no such effect with neutrons. The X-ray and neutron measurements are consistent with the theory for a crystal with a mosaic structure of 0.4 mm. average size, indicating that the cadmium sulphate crystal was imperfect.

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