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Transmission of X-rays through Calcite near the Bragg Angle*†

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Transmitted and diffracted intensities of monochromatic x-ray radiation through single crystals of calcite have been recorded. Wavelengths between 0.631A and 2.29A and crystal thicknesses from 0.1 mm to 3.06 mm were used. A double-crystal spectrometer served as monochromator. Intensities were measured with a Geiger counter. The measurements permit an angular correlation between the transmitted and diffracted curves to within one second of arc. The results show an anomalous absorption near the Bragg angle. Characteristically different results are obtained depending on the crystal thickness and the wavelength. For "thin" crystals the transmitted intensity decreases at the Bragg angle. For "thick" crystals the transmitted intensity increases at the Bragg angle. At each particular wavelength there exists a region of "intermediate" crystal thicknesses for which the transmitted intensity passes through a maximum and minimum as the crystal is rotated through the Bragg angle.

I. INTRODUCTION

THE diffraction of x-rays by large single crystals has received much attention in regard to both theory and experiment. There still exist, however, problems in this field with relatively little experimental data and until very recently incomplete theoretical treatment. Such a problem is that of the transmission of x-rays through a crystal in the direction of the incident beam at a position where a Laue diffraction occurs. Investigations of this transmitted and reflected radiation have been made in only a few cases and without the precision of x-ray spectroscopic techniques available today.

Bragg and others[1] have observed that when a rock-salt crystal is oriented so that Mo $K\alpha$ radiation undergoes a Laue reflection, the transmitted intensity is less at this Bragg angle than that which is transmitted when the crystal is off the Bragg angle. This decrease in intensity is due to primary extinction.[2] Campbell[3] has recently shown that the transmitted intensity may undergo large increases when the crystal is set at a Bragg angle. The work of Campbell was done with single-crystal monochromatization using Cu $K\alpha$ and Cu $K\beta$ radiation and Geiger counter intensity recording. Borrmann,[4] using a divergent beam technique and photographic recording, has investigated this effect with quartz and calcite crystals.

We have investigated the transmission of monochromatic x-rays between 0.63A and 2.29A through single crystals of calcite of thicknesses 0.1 to 3 mm and have recorded the changes in transmitted and reflected intensities near the Bragg angle with a much higher precision than previously reported. New results have been obtained showing that at the Bragg angle, the transmitted intensity may increase, decrease, or fluctuate with both an increase and decrease, depending on the wavelength of the radiation and the thickness of the crystal.

II. EXPERIMENTAL

The experimental arrangement is shown in Fig. 1. A standard full-wave rectified high voltage supply was used in this work. The input voltage was stabilized with a Sorenson model 2000-S electronic voltage regulator. The high voltage applied to the x-ray tube was electronically regulated with a modified Pepinsky type circuit.[5] The x-ray tube current was stabilized using a circuit similar to the one described by Le Mieux and Beeman.[6] Commercial Machlett A-2 x-ray tubes with Cr, Cu, Mo, and W targets were used. Tests showed that after an initial warm-up period, the intensity of x-ray radiation was constant over long periods.

A double-crystal spectrometer was used to monochromatize the x-rays. The first crystal was mounted on a slide perpendicular to a line connecting the focal spot and the axis of rotation for the second crystal and

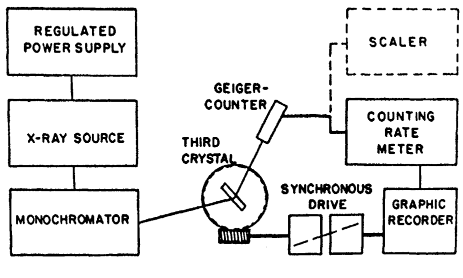

FIG. 1. Block diagram of apparatus used.

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* A preliminary report of this work was presented at the spring meeting of the American Physical Society in Washington, D. C., 1952. See also Guenter Schwarz and George L. Rogosa, Phys. Rev. **86**, 421 (1952).

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[1] Bragg, Bosanquet, and James, Phil. Mag. **42**, 1 (1921).

[2] See, e.g., A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand and Company, Inc., New York, 1935).

[3] H. N. Campbell, Acta Cryst. **4**, 180 (1951); J. Appl. Phys. **22**, 1139 (1951).

[4] G. Borrmann, Physik. Z. **43**, 157 (1941); G. Borrmann, Z. Physik **127**, 297 (1950).

[5] R. Pepinsky and Paul Jarmotz, Rev. Sci. Instr. **19**, 247 (1948); Guenter Schwarz and Earl H. Byerly, Rev. Sci. Instr. **19**, 273 (1948).

[6] A. F. Le Mieux and W. W. Beeman, Rev. Sci. Instr. **17**, 130 (1946).

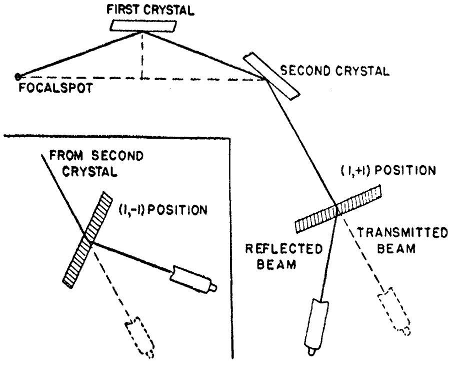

FIG. 2. Arrangement of crystals.

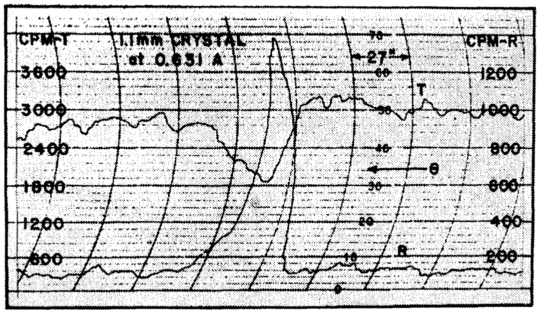

FIG. 4. Transmitted and reflected intensities through a 1.1-mm calcite crystal showing the transition to asymmetrical transmission.

halfway between them. The second crystal was mounted on a table which could be rotated in steps of a second of arc by means of an arrangement of levers and a precision micrometer head. This table was supported by a set of high precision Timken roller bearings.[7] The micrometer screw was calibrated by measuring the separation of the Cu $K\alpha$ lines, using different parts of the screw. The calibration was linear over the range of the screw which covered an angular region of 83′ of arc. The accuracy of resetting was checked by measuring $(1, -1)$ curves. It was possible to reset the crystal to within a second of arc.

The perfection of the cleaved calcite crystals on the spectrometer was checked by measuring the width of $(1, -1)$ curves and line widths in the $(1, +1)$ position. The half-width of the $(1, -1)$ curve at Cu $K\alpha$ was 10.3″ which is very close to the theoretical value of 9.8″. The half-width of the Cu $K\alpha_1$ line was 44″.

The monochromatic beam coming from the second crystal passed over the axis of a Gaertner spectrometer[8] on which the crystal to be investigated was mounted. The crystal table of this spectrometer could be rotated by a synchronous motor drive at angular speeds of 7.5 or 27 seconds of arc per minute of time. Manual setting of this table could be done to within 1″. The angular position of the table could be read accurately by means of two microscopes. The circle had been recently re-engraved by the Gaertner Company.

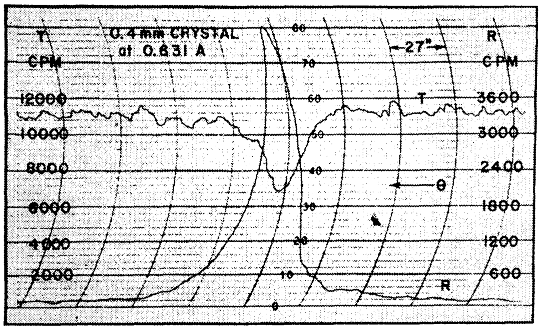

FIG. 3. Transmitted and reflected intensities through a 0.4-mm calcite crystal showing extinction at the Bragg angle.

X-ray intensities were measured with a high pressure argon filled counter in conjunction with a General Radio counting rate meter. The output was plotted by means of an Esterline Angus Recording Milliammeter. In some cases intensities were measured by counting for a fixed time, using a Berkeley model 2000 scaler and rotating the crystal manually in small steps.

Figure 2 shows a diagram of the arrangement of the calcite crystals. The crystal under study was mounted with a face parallel to and on the axis of the Gaertner spectrometer. In order to align the internal planes the Bragg reflection from the planes parallel to this face was first detected by the Geiger counter. A rotation of 105° 3.5′ or 74° 56.5′ depending on how the crystal was positioned would then bring a similar set of internal planes into position to give a Laue reflection. This technique enabled one to quickly find the diffraction positions of the crystal. The internal planes were carefully adjusted to give the narrowest rocking curve. We used two different diffracting positions for each crystal. These two positions can be conveniently designated as $(1, +1)$ or $(1, -1)$ using the standard double crystal spectrometer nomenclature and referring the crystal under investigation to the second crystal of the mono-

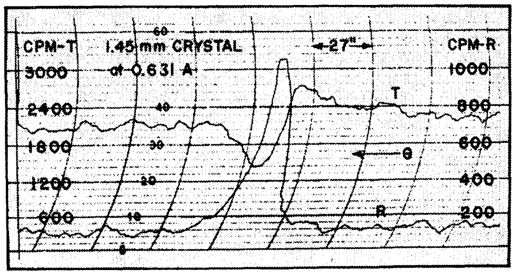

FIG. 5. Transmitted and reflected intensities through a 1.45-mm calcite crystal showing asymmetrical transmission.

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[7] We would like to express our appreciation to Mr. R. G. Harmon of the Timken Roller Bearing Company for providing us with a set of precision roller bearings.

[8] This spectrometer is on loan from the University of Chicago through the courtesy of Professor S. K. Allison.

chromator. The monochromator itself was used in the (1, +1) position for the work reported here except in the case of the Cr $K\alpha$ radiation.

The calcite crystals, through which the transmitted radiation was measured, were cleaved from large single crystals. With care it was possible to cleave suitable crystals as thin as 1 mm. The size of these crystals was approximately 10 mm by 20 mm. For thicknesses between 0.3 mm and 1 mm, cleaved crystals were ground with emery of mesh 275 and then etched in HCl.[9] In addition, thinner crystals were obtained by prolonged etching reaching 0.1 mm. Work was also done with crystals cut such that the reflecting planes were perpendicular to the surface. Crystal thicknesses were constant to less than 0.01 mm over the area used in the experiments. All calcite used came from optically clear pieces.

III. EXPERIMENTAL RESULTS

Experiments were performed at wavelengths corresponding to characteristic line radiation from the various targets. The lines used were Mo $K\beta_1$, Mo $K\alpha_1$, W $L\gamma_1$, Cu $K\alpha_1$, and Cr $K\alpha$ giving a spread of wavelength from 0.63A to 2.29A. The results indicated that there was no need to perform experiments at other intermediate wavelengths.

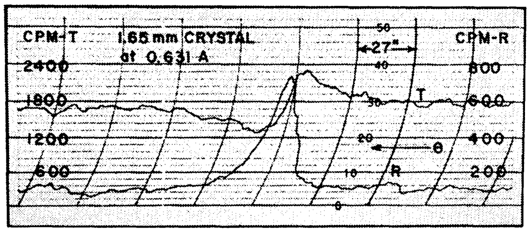

FIG. 6. Transmitted and reflected intensities through a 1.65-mm calcite crystal showing asymmetrical transmission.

Figures 3 to 8 show the results obtained at a wavelength of 0.631A for calcite of thicknesses 0.4 mm to 2.56 mm. These curves show the significant features observed in this work. All of them were recorded with an x-ray tube voltage of 28 kv and current of 20 ma. No second-order radiation was passed by the spectrometer. T represents the intensity of the transmitted beam, and R represents the Laue reflected intensity. These curves were all taken in the $(1, -1)$ position as illustrated in Fig. 2. θ represents the angle between the incident beam and the reflecting crystal planes and increases to the left in the curves. Some care has to be exercised in making quantitative calculations from these curves. With a counting rate meter, in regions where the intensity changes fast compared to the response time of the instrument, corrections have to be applied. In particular, peak intensities will be recorded too low. The transmitted and reflected curves were taken successively. Their abscissas are matched to within 1″.

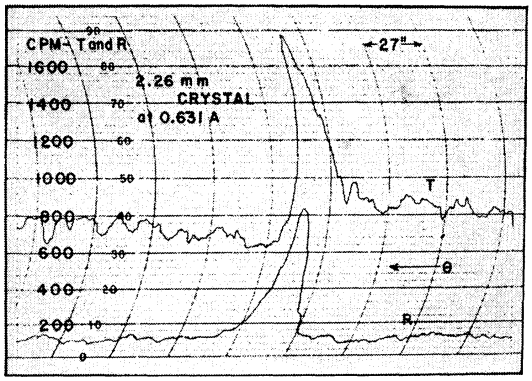

FIG. 7. Transmitted and reflected intensities through a 2.26-mm calcite crystal showing anomalous transparency.

The crystal thicknesses given in the figures represent the perpendicular distance between crystal faces. The crystal planes make an angle close to 15° with this perpendicular. In calculating absorption coefficients, the actual path of the radiation through the crystal has to be known.

Figure 3 shows an example of the "thin" crystal case. In this and all following curves the Bragg angle θ_B is defined by the peak of the reflection curve. There is a decrease in the transmitted intensity at the Bragg angle. In Fig. 4 an asymmetry appears showing a decreased absorption near θ_B on the side of $\theta<\theta_B$. The absorption on both sides of the Bragg angle is still different for angular separations of several minutes. This asymmetry becomes much more pronounced with increasing crystal thickness as seen in Figs. 5 and 6. The transmission curve in both cases shows the decreased absorption near θ_B for $\theta<\theta_B$ followed by an increased absorption for $\theta>\theta_B$. Even at an angular distance of several minutes off the Bragg angle the absorption itself is not the same on both sides. The transmission curve is characterized by an inflection point near the Bragg angle, and because of the increase and decrease we shall refer to this as "asymmetrical transmission." The crystals used in the curves of Figs. 7 and 8 approach the "thick" crystal case, and the curves show the

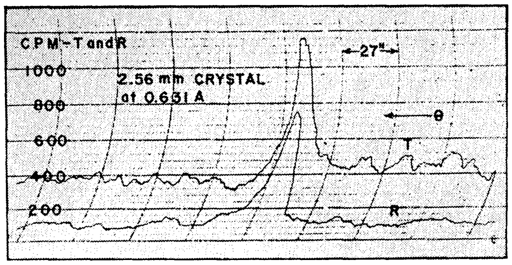

FIG. 8. Transmitted and reflected intensities through a 2.56-mm calcite crystal showing anomalous transparency.

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[9] K. V. Manning, Rev. Sci. Instr. 5, 316 (1934).

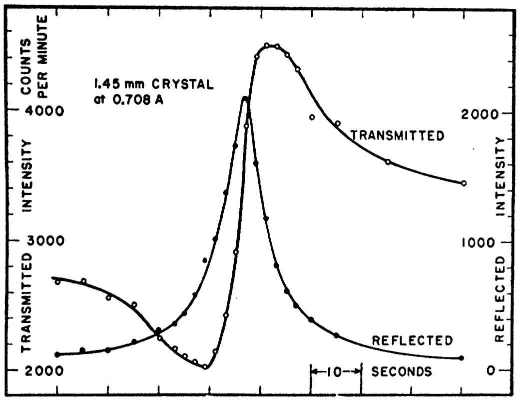

FIG. 9. Detailed plot of asymmetrical transmission case.

anomalous transparency at the Bragg angle. The absorption off the Bragg angle still remains different on both sides.

With Mo $K\alpha_1$ radiation of 0.708A, crystals from 0.4 mm to 3.06 mm were investigated. The 0.4-mm crystal showed a pronounced dip in the transmitted intensity, while the 3.06-mm crystal showed a pronounced increase. At 0.75 mm the effect was mainly a decrease, while at 2.56 mm it was still an increase. Between these two values the results gave asymmetrical transmission. Figure 9 shows the curves of the transmitted and reflected intensities for a crystal of 1.45 mm thickness. Here intensities were recorded by using the scaler and rotating the crystal manually in steps of 2″ near the Bragg angle. The transmitted and reflected intensity was measured by switching the Geiger counter to fixed positions as shown in Fig. 2 for each angular crystal setting.

At 1.096A, W $L\gamma_1$ radiation, a 0.9-mm crystal showed the "thick" crystal type increase in the transmitted intensity. The 0.1-mm crystal, which was the thinnest available in this work, showed a dip. Intermediate thicknesses produced asymmetrical transmission.

At Cu $K\alpha_1$ the thinnest crystal available at the time was 0.4 mm, and this gave a pronounced increase of transmission. All thicker crystals also gave anomalous transparency with no significantly new features. The sawcut crystals with the internal reflecting planes perpendicular to the faces were used at this wavelength and gave results similar to cleaved crystals.

Calcite is highly absorbing at 2.29A, Cr $K\alpha$, and crystal thicknesses were limited to less than 1 mm because of the x-ray intensity available. A 0.4-mm crystal gave a pronounced increase in the transmitted intensity when rotated through the Bragg angle.

In order to determine the effect of surface imperfections upon the results, a comparison was made between a cleaved crystal and a ground crystal of about the same thickness. No significant differences in the shapes of the transmitted curves were observed. Transmission and reflection curves were always taken in both $(1, -1)$ and $(1, +1)$ positions. In general, the effects were somewhat more pronounced in the $(1, -1)$ position.

IV. DISCUSSION

The theory of these phenomena has received the attention of Von Laue[10] and very recently Ramachandran,[11] Zachariasen,[12] Hirsch,[13] and Ewald.[14] Von Laue[10] has considered a possible explanation. Since his numerical calculations are for rocksalt and Cu $K\alpha$ radiation, they cannot be directly compared with our data. However, his predicted shapes of the transmitted curves are similar to those that we have recorded. Ramachandran[11] has used the theory of Von Laue to calculate the diffracted and transmitted intensities through "thick" crystals of calcite in order to compare with the results of Borrmann and Campbell. Ramachandran significantly points out that the peaks of the reflected and transmitted beams are not coincident but that a shift occurs which decreases for increasing crystal thickness. Our experimental results confirm this theoretical prediction.

Zachariasen[12] has derived an expression for the absorption coefficient at the Bragg angle for the "thick" crystal case, and he finds good agreement with data of Borrmann and Campbell. Zachariasen concludes that anomalous transparency only results when $\mu_0 \cdot t_0 > 1$, μ_0 being the ordinary absorption coefficient and t_0 being the thickness of the crystal plate. Zachariasen's results follow from equations in his dynamical theory of x-ray diffraction.[15]

Hirsch[13] has treated this problem starting with the dynamical theory as given by Zachariasen. He has calculated the intensity curves for both the transmitted and reflected beams for the "thin," "intermediate," and "thick" crystal cases. We find good agreement with the shapes of his calculated curves. Quantitative details are being further investigated.

We wish to express our appreciation to Professor P. P. Ewald for very helpful discussions. Thanks are due Mr. Lewis V. Eckhart who built the monochromator and made many valuable contributions to its design and to Mr. L. C. Brown for help in making the measurements.

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[10] M. Von Laue, Acta Cryst. **2**, 106 (1949).

[11] G. N. Ramachandran, J. Appl. Phys. **23**, 500 (1952); G. N. Ramachandran and Gopinath Kartha, Proc. Indian Acad. Sci. **35**, 145 (1952).

[12] W. H. Zachariasen, Proc. Natl. Acad. Sci. **38**, 378 (1952).

[13] P. B. Hirsch, Acta. Cryst. **5**, 176 (1952), and private communication.

[14] P. P. Ewald, private communication.

[15] A private communication from Zachariasen has pointed out that his theory predicts asymmetrical transmission for intermediate crystal thicknesses.