

AN EXPERIMENTAL STUDY OF THE REFLECTION OF
X-RAYS FROM CALCITE.

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SYNOPSIS.

Per cent., of monochromatic X-rays reflected from calcite, for wave length ranging from .3Å. to .8Å. was studied by means of a *double X-ray spectrometer*. A narrow beam of general radiation fell at an angle upon a crystal designated as *A* and the reflected beam was allowed to fall upon a second crystal designated *B*. The intensity of the beam from *B* was then compared, by means of the ionization produced in an ionization chamber with the direct beam from *A*. *Per cent., reflection was found surprisingly large* when the surfaces of the two crystals were strictly parallel. The per cent., reflection will depend upon the character of the beam reflected and therefore upon crystal *A*.

Significant energy distribution curves were found when the crystal *B* was rocked through a small angle on either side of parallelism. *A very sensitive micrometer motion* was provided for rotating crystal *B* about both a horizontal and also about a vertical axis, necessitated by the extremely narrow energy distribution curves obtained.

Three pairs of crystals were investigated. Pair (A_1-B_1) where *A* was a rather imperfect specimen of *Montana calcite* and B_1 was a clear specimen of *Iceland spar*. The surface of both crystals were polished. Pair ($A_2 - B_2$). Here a clear specimen of *Iceland spar* was split. The two split surfaces were polished and the crystals mounted so that reflection was from the surfaces that had been contiguous before splitting. Pair ($A_3 - B_3$) was another pair obtained by splitting a clear specimen of *Iceland spar*, but leaving surfaces unpolished. Mounted same as ($A_2 - B_2$).

The rocking curves vary greatly in width, depending upon the nature of the reflecting surface; 16'' of arc for width of half maximum when crystal is a good sample and the surface is the natural split surface (not polished). When crystals are not perfect samples and surface polished as much as 57'' of arc was obtained. For any one pair of crystals the width of the half maximum of these rocking curves is a *linear function of the wave length* with a *definite finite intercept* depending upon the condition of the surface. The more perfect the crystal the less does this width depend upon wavelength.

Per cent., reflection is a function of wave length, but is less so the more perfect the crystal. For the pair ($A_3 - B_3$), it was nearly independent of wave length. It would seem that the *reflection from a really perfect crystal would be independent of wave-length and would probably be greater than fifty per cent.*

A KNOWLEDGE of the proportion of the incident X-rays of any frequency that are reflected from the cleavage surface of a crystal is of great importance in determining the distribution of energy in the continuous X-ray spectrum and also the partition of energy among the many lines of the characteristic spectra. It is also of prime importance in the development of an adequate explanation of crystal reflection itself.

Measurements of the relative reflection of X-rays in the several orders have been made by Professor W. H. Bragg and W. L. Bragg, as well as the relative reflectivity of various crystals for the same radiation. The percentage reflection or the coefficient has also been measured by Professor Bragg, using a single crystal. Such determinations must contain an error due to the presence of the general radiation in addition to the line radiation in the original beam before reflection. A. H. Compton¹ has made a direct determination of the coefficient of reflection of rock salt and calcite for one particular wave-length. Compton allowed the radiation reflected from a crystal placed on one spectrometer to fall on a second crystal placed on another spectrometer suitably placed near the first. The apparatus was arranged so as to give an integral area of the curves as the crystals were rocked through a small angle. The small value of the reflection obtained by Compton would

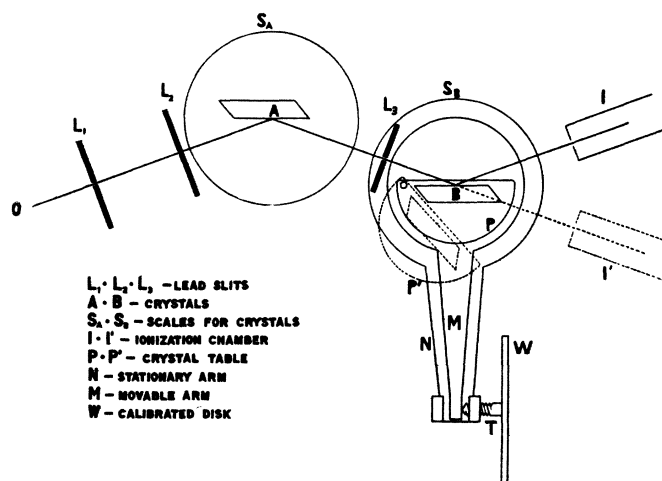


Fig. 1.

indicate that the two crystals were not placed with the crystal faces parallel, as we have done, but were placed nearly $(180^\circ - 2\theta)$ to this position so that only a small portion of the ray from first crystal could be reflected from the second crystal in any one position. (This point is not mentioned in his description of the experiment.)

For the purpose of this study of crystal reflection a special double X-ray spectrometer was constructed. A plan of this instrument is shown in Fig. 1. The arrangement of the parts is sufficiently shown in the figure as not to need a detailed description. The heterogeneous beam passing through the slits L_1L_2 falls on Crystal *A*. The position of this

¹ Proc. Am. Phys. Soc., PHYS. REV., July, 1917.

crystal is read on the vernier scale S_A in the usual manner. The pencil of rays reflected from A is nearly homogeneous, the range of wavelengths ($\Delta\lambda$) included depending on the degree of divergence of the original beam. The beam reflected from A falls on crystal B and suffers a second reflection. The intensity of this reflected beam is measured by the ionization current in the ionization chamber I as shown. The crystal B is mounted on a movable plate P which can be thrown back as indicated by the dotted lines in the figure. The beam from crystal A can then pass directly into the ionization chamber placed in the position I' . The intensities of the two beams before and after reflection are thus compared by the ionization produced in the chamber in the two positions I and I' . A stop is provided so that the crystal B will return exactly to the same position each time after displacement.

The source of X-rays was a standard Coolidge tube with a tungsten target. This tube was energized by a high tension transformer supplied by a 500-cycle generator. The alternating potential was rectified by a kenetron set and the electricity stored in a large plate-glass condenser immersed in oil. The capacity of this condenser was such that the voltage applied to the tube was practically constant, the momentary fluctuations being of the order of one half of one per cent. of the total voltage (50,000) when a current of 2 milliamperes was flowing through the tube. The speed of the 500-cycle generator was also controlled to a constancy of one half of one per cent. The voltage applied to the tube and the current flowing could be controlled and measured to this degree of accuracy. The voltage was measured by a specially designed repulsion electrometer that had a range from 5,000 to 100,000 volts.

The scale S_B , the mounting for the crystal and the ionization chamber I were all mounted on an arm (not shown in the figure) and could be rotated about center of scale S_A as axis and the angle read by a vernier on scale S_A in the usual manner. The angle of crystal B was read by a vernier on scale S_B . The position of the ionization chamber was read by another vernier on this scale. The arm M was attached to the table carrying crystal B . This arm extended out from the axis as shown and could be moved through a small angle when so desired by the finely threaded screw T . The disc W attached to the screw was graduated on its edge. The parts were so proportioned that one scale division on the periphery of disc W represented a rotation of crystal B through 2 seconds of arc. In this way it was possible to give crystal B an angular displacement as small as one fifth second of arc and read the same with considerable accuracy.

The slightly divergent beam passing through the slits L_1 , L_2 after

reflection from crystal *A* falls on the reflecting face of crystal *B*. A third slit L_3 was introduced in the path of this beam as shown in the figure. This narrow slit was necessary to keep the beam sufficiently narrow that it would not spread over an area broader than the face of crystal *B* at small glancing angles.

Maximum reflection was obtained at a certain setting of crystal *B*, presumably this is the position in which the reflecting planes of *B* are parallel to those of *A*. The position of parallelism referred to in this paper is the position at which maximum reflection is obtained. When the reflecting planes of the two crystals are parallel, the ray ee' from crystal *A* for instance will strike crystal *B* in such manner that angle b will equal angle a . Another ray striking crystal *A* at f will meet *B* at f' and the angle c will equal angle d . The same is true for every ray re-

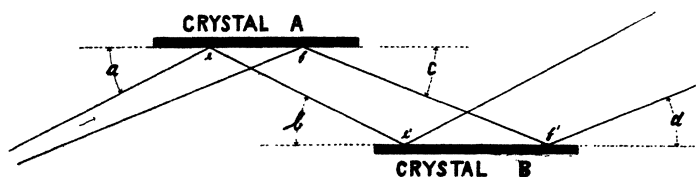


Fig. 2.

lected from *A* between the points e and f . *Every ray reflected from A will meet B at the proper angle for maximum reflection for the same setting of B.* The above statement is only strictly true if the planes of *A* and *B* are perfect and the reflection follows strictly the law $\lambda = 2d \sin \theta$. An imperfection of any kind in crystal *A* would make the reflected ray slightly non-homogeneous, that is along any path as ee' for instance the radiation would not only consist of a beam of wave-length given by $\lambda = 2d \sin \theta$, but would contain other radiation of slightly different wave-length.

Since every pencil of rays reflected from *A* to *B* falls on *B* at the proper angle for best reflection (when the planes are parallel) we shall consider the coefficient of reflection as determined in this investigation to be the ratio of the intensity of the beam after reflection from *B* to its intensity before such reflection. These two intensities are measured by means of the ionization chamber in the two positions I and I' .

It is at once evident from Fig. 2 that a ray ee' of given wave-length reflected from *A* and a ray ff' of another wave-length so reflected cannot both meet the crystal *B* at proper angle for reflection if the two crystals differ at all in structure or in crystal spacing. If the crystals *A* and *B* do differ in structure or spacing a small rotation of crystal *B* will bring

the crystal to the proper position for reflection of the rays falling on different portions of its surface.

In addition to the lever arm M and slow screw device T for rotating B through small angles about a vertical axis, a similar device (not shown in the figure) was constructed for rotating crystal B about a horizontal axis. In this way the reflecting planes of B could be brought accurately parallel to those of crystal A .

The first pair of crystals investigated was A_1 a Montana calcite, and B_1 a very old and clear specimen of Iceland spar. The reflection was from the (100) planes of each crystal. The surfaces were polished on a plane parallel plate in the usual manner of obtaining an optical polish. The crystals being carefully mounted and crystal A_1 set for

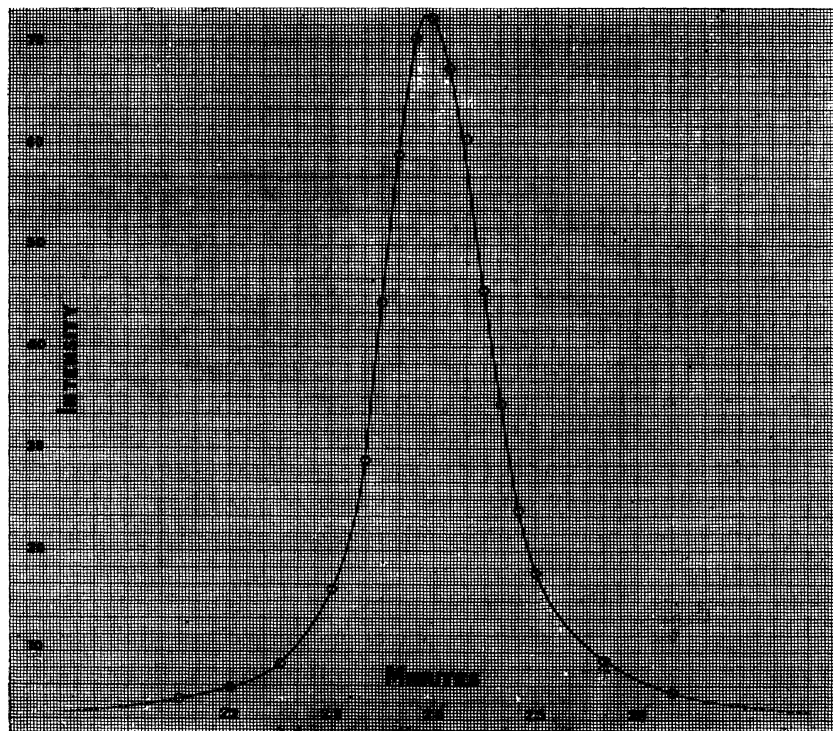


Fig. 3.

reflection of a portion ($\Delta\lambda$) of the general radiation of a desired wavelength, it was found on rotating crystal B_1 through small angles each side the maximum, that a very regular curve of intensity was obtained. A typical curve for these two crystals is shown in Fig. 3 and is given to indicate how accurately such curves can be obtained. The observed

points lie on a very smooth regular curve. It should be pointed out that the principal irregularities in X-ray spectrometer observations do not arise from irregularities in the radiation but are due to the fact that close settings and readings of small angular changes can not be made with verniers. The smoothness of the curves here presented is due to the accuracy of measuring small angular changes by the slow screw device.

The curve (Fig. 3) is seen to have considerable width. The width of curve referred to throughout this paper is the width at half-maximum indicated by the length of the lines aa' , bb' , cc' in Figs. 4, 5, 6 and 7.

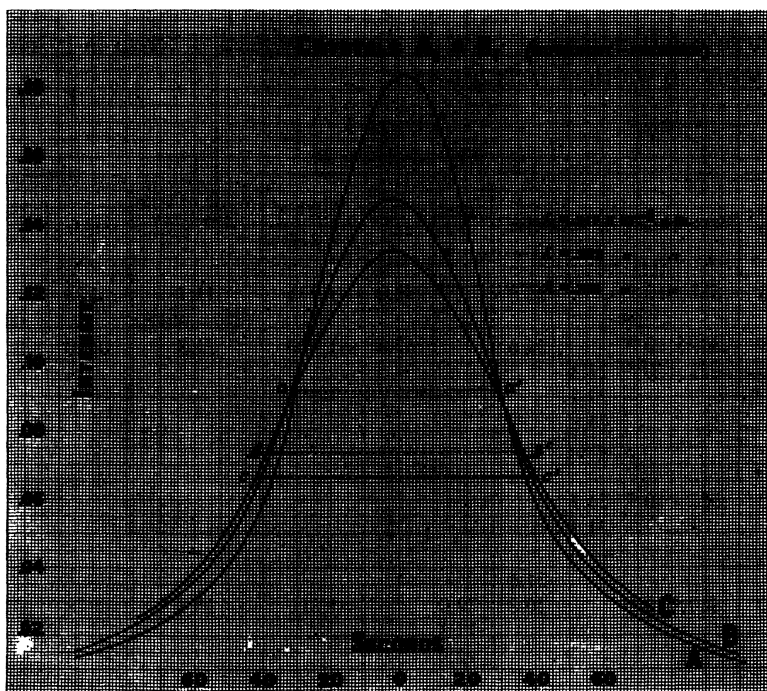


Fig. 4.

After observation of this effect due to *rocking the crystal* through small angles a systematic investigation was undertaken to ascertain its cause.

The width of curve at half-maximum was found to be independent of:

- (a) Width of the slits L_1L_2 .
- (b) Voltage applied to X-ray tube.
- (c) Current through X-ray tube.

It was found to depend on:

- (a) Angle of reflection (wave-length).
- (b) The nature of the crystals.
- (c) The state of polish of the surfaces and the similarity of crystals.

Three combinations of crystals were investigated designated as follows:

Crystals $A_1 - B_1$. Crystal A_1 was a Montana calcite. Slight flows were visible in the interior. Surface polished. Crystal B_1 . An old clear specimen of Iceland spar. Surface polished.

Crystals $A_2 - B_2$. A clear specimen of Iceland spar was split in two portions. The surfaces polished. The two reflecting surfaces as mounted (Fig. 2) were the surface planes that had been in contact before splitting.

Crystals $A_3 - B_3$. A clear specimen of Iceland spar was split into portions. The freshly split surfaces were *not polished*. The two reflecting surfaces as mounted were the contiguous planes before splitting.

These combinations of crystals showed marked differences in reflectivity and in width of *rocking curves*.

TABLE I.
Crystals A_1 and B_1 .

Grazing Angle.	Wave-length.	% Reflection.	Width of Half Maximum.	Width of Half Max. \times % Reflec.	Width of Half Maximum Minus Intercept = Q .	Sin θ .	$\frac{\text{Sin } \theta}{Q}$.
3° 00'	$.317 \times 10^{-8}$	25.8	58.0	1,495	11.7	.0523	
3 30	$.369 \times 10^{-8}$	24.6	67.8	1,670	(58.0 - 46.3)	.0610	.0045
4 00	$.422 \times 10^{-8}$	23.3 22.2 24.8			21.5		.0028
		23.4					
5 00	$.527 \times 10^{-8}$	21.7 21.3					
		21.5					
5] 30	$.580 \times 10^{-8}$	20.8	72.0 73.5 72.5	1,510	26.4	.0958	.0037
			72.7				
6 00	$.632 \times 10^{-8}$	19.5 19.1 20.7					
		19.8					
7 00	$.737 \times 10^{-8}$	18.3 19.0					
		18.7					
7 30	$.790 \times 10^{-8}$	18	78.2 79.6				
			78.8	1,475	32.5	.1305	.0040
8 00	$.842 \times 10^{-8}$	17.3	83.4	1,445	37.1	.1392	.0038

The curves obtained by slightly rocking crystal B_1 through small angles each side of the maximum are shown in Fig. 4. Observations were made at three different wave-lengths. The coefficients of reflection obtained at a number of glancing angles are given in Tables I and IV.

The fourth column of Table I. gives the widths at half-maxima (aa' , bb' and cc') expressed in seconds of arc. The fifth column gives the product of width at half-maximum and the per cent. reflection (proportional to maximum ordinate). This product is probably proportional to the areas of the curves. The table shows these areas to be nearly constant. The widths at half-maxima are plotted in Fig. 8. The

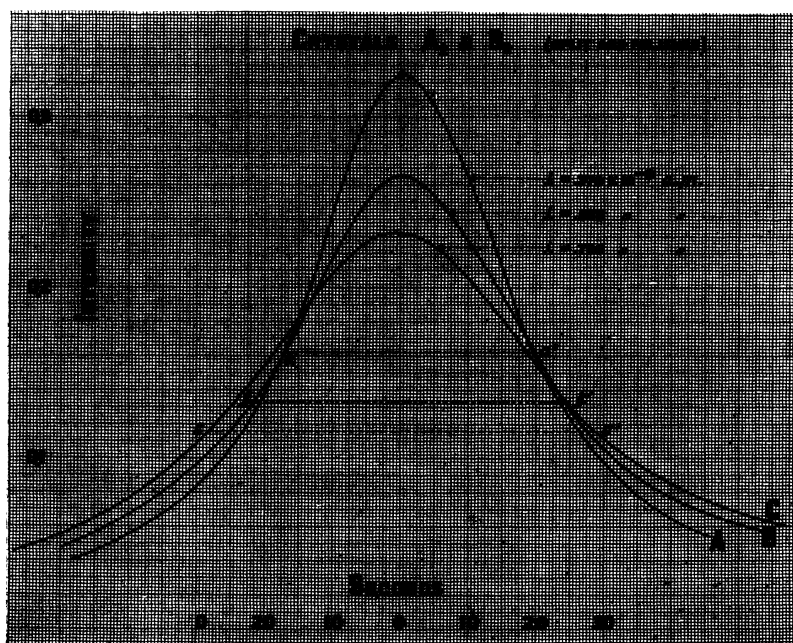


Fig. 5.

straight line drawn through the points intercepts the ordinate axis at points designated by g_1 . The width at half-maxima minus the intercept g_1 is given in the sixth column of the table (Q).

The fact that the points in Fig. 8 lie approximately on a straight line suggests that the curve width is made up of two parts, one part a constant g_1 and another part proportional to the sine of the glancing angle. This is indicated by the constancy of the ratio $\sin\theta/Q$ shown in the last column of the Table, where Q represents the widths at half-maxima minus the intercept g_1 .

Crystals A₂ and B₂. A clear specimen of Iceland spar was split into two parts along a cleavage plane. The two split surfaces were polished on a plate using fine rouge and water as the abrasive. The crystals were mounted on the spectrometer in such position that the reflection took place at the two surfaces that had been contiguous before cleavage. The coefficient of reflection was considerably larger than that obtained with the two dissimilar crystals A₁ - B₁. The width of rocking curves

TABLE II.
Crystals A₂ and B₂.

Grazing Angle.	Wave-length.	% Reflection.	Width of Half Maximum.	Width of Half Max. × % Reflec.	Width of Half Maximum Minus Intercept = Q .	Sin θ .	$\frac{\text{Sin } \theta}{Q}$.
3° 00'	$.317 \times 10^{-8}$	37.3					
3 30	$.369 \times 10^{-8}$	(curve) 35.4	34.3 36.3 35.0	1,245	18.7 (35.2-16.5)	.0610	.00326
			35.2				
4 00	$.422 \times 10^{-8}$	34.3 32.9					
		33.6					
4 30	$.474 \times 10^{-8}$	(curve) 32.9	41.5 41.9	1,371	25.2	.0785	.00312
			41.7				
5 00	$.527 \times 10^{-8}$	30.5					
5 30	$.580 \times 10^{-8}$	(curve) 29.2	45.6 46.5	1,343	29.5	.0958	.00325
			46.0				
6 00	$.632 \times 10^{-8}$	27.9					
6 30	$.684 \times 10^{-8}$	(curve) 26.9	54.6 50.9	1,417	36.2	.1132	.00313
			52.7				
7 00	$.737 \times 10^{-8}$	26.0					
7 30	$.790 \times 10^{-8}$	(curve) 25.3	57.0 57.8	1,452	40.9	.1305	.00319
			57.4				
8 00	$.842 \times 10^{-8}$	24.6					

on the other hand was much less. The widths at half-maxima, the products of widths at half-maxima and per cent. reflection are given in Table II. The product of width at half-maxima and per cent. reflection is not quite constant but progressively increases with glancing angle as

shown in the fifth column of the table. The widths at half-maxima plotted against glancing angle lie sensibly on a straight line (Fig. 8). The widths at half-maxima minus the intercept g_2 is given in the sixth column of the table (Q). The ratio $\sin \theta/Q$ is a constant. The width of rocking curve is composed of two parts. One part is a constant g_2 , and the other part varies as the sine of the glancing angle.

Crystals $A_3 - B_3$. A clear crystal of Iceland spar was split into two parts along a cleavage plane. The crystals were so mounted that the molecular planes that were contiguous before splitting became the two reflecting surfaces of the crystals. The surfaces were not polished, and

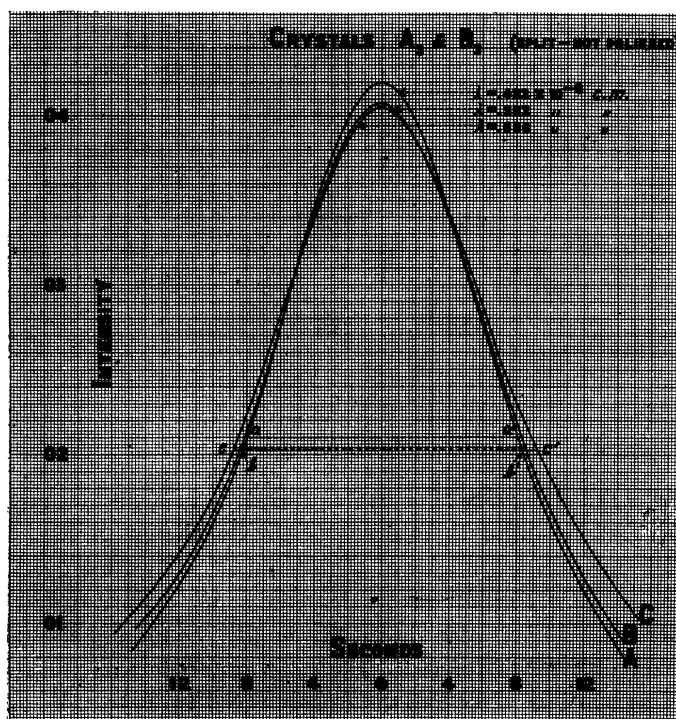


Fig. 6.

care was taken not to touch them in any way so that the surface molecular layers should remain in as perfect a state as possible.

The reflection was very much greater than that obtained with the other crystals described. The results are shown in Table III., where the several columns have the significance previously described. The curves obtained by *rocking* crystal B_3 are shown in Fig. 6. The product of widths at half-maxima and per cent. reflection is nearly constant, in-

creasing slightly with increasing wave-length. The intercept on the width at half-maxima ordinate at zero angle is shown at g_3 in Fig. 8. The ratio $\sin \theta/Q$ is a constant. The curve width may be regarded as composed of two parts, one part is a constant g_3 , the other part varies as the sine of the glancing angle.

TABLE III.

Crystals A_3 and B_3 .

Grazing Angle.	Wave-length.	% Reflection.	Width of Half Maximum.	Width of Half Max. \times % Reflec.	Width of Half Maximum Minus Intercept = Q .	Sin θ .	$\frac{\sin \theta}{Q}$.
4 00'	$.422 \times 10^{-8}$	44.4	16.1	717	3.6	.0698	.0199
		44.4	16.2				
5 30	$.580 \times 10^{-8}$	42.9	17.3	752	4.9	.0958	.0195
		43.3	17.6				
6 30	$.686 \times 10^{-8}$	43.1	17.5	787	5.8	.1132	.0195
		42.4	18.1				
7 30	$.790 \times 10^{-8}$	43.2	18.6	787	5.8	.1132	.0195
		42.8	18.4				
		42.8					

The effect of similarity of crystal and treatment of surface on the width of rocking curves and their areas is clearly shown in Fig. 7. These curves are for glancing angles of 3° - $30'$ and 4° . They are given to scale. The areas of curves progressively decrease as the crystal surfaces become more perfect. This is also shown by the fifth column of the tables.

For convenience of reference the per cent. reflection for the several crystals at various wave-lengths is assembled in Table IV. and shown graphically in Fig. 9.

The last two columns of the table give the calculated variation of reflection with wave-length by an equation derived by C. G. Darwin¹ for imperfect crystals. All of the constants in this equation including the Debye heat term are collected into a single constant A , and the equation written in the form

$$R = A \frac{\lambda^3(1 + \cos^2 2\theta)}{\mu \sin \theta \cos \theta}.$$

¹ Phil. Mag., Feb. and April, 1914.

For purposes of comparison the constant A is taken to be such as to give the observed reflection at a particular angle (6°). The reflection at other angles is then calculated for the absorption $\mu \sim \lambda^{2.4}$. For this

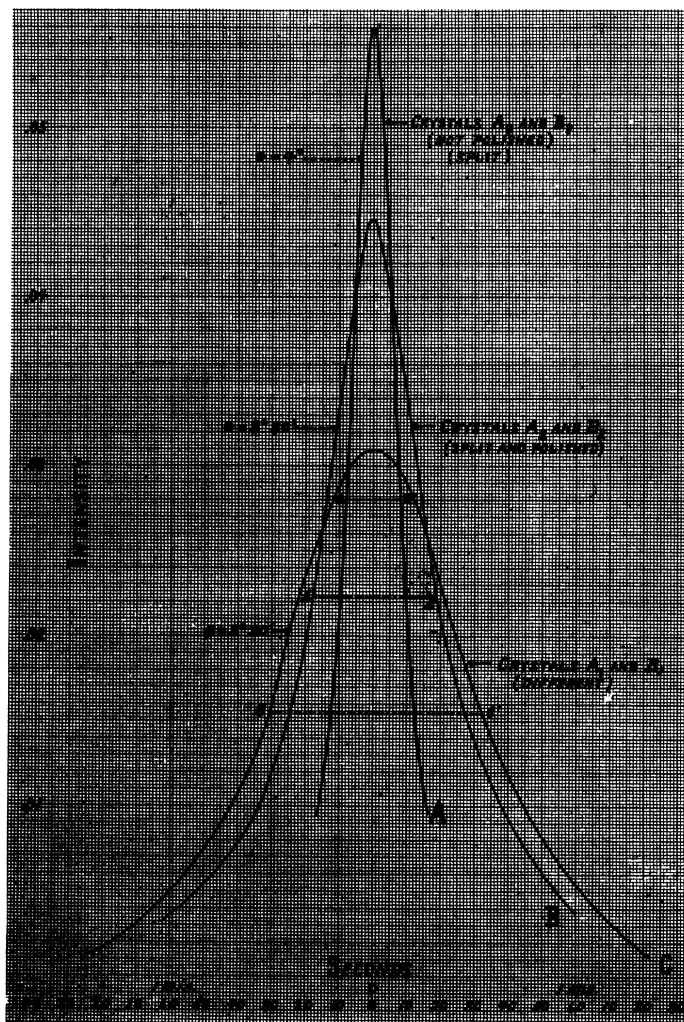


Fig. 7.

value of μ the equation agrees very closely with the observed results for the two pairs of polished crystals. This is particularly true for crystals $A_2 - B_2$.

It should be pointed out that the per cent. reflection or coefficient of reflection here determined is considered to be that of crystal B in every

TABLE IV.

Angle.	λ .	Per Cent. Reflection.				
		Observed.			Computed (Darwin).	
		A_1-B_1 .	A_2-B_2 .	A_3-B_3 .	A_1-B_1 .	A_2-B_2 .
3	.317	25.8	37.3		26.5	37.2
3-30	.369	24.6				
4	.422	23.4	33.6	44.4	23.5	33.1
5	.527	21.5	30.5		21.6	30.1
5-30	.580	20.8		43.1		
6	.632	19.8	27.9		19.8	27.9
6-30	.686			42.8		
7	.737	18.7	26		18.6	25.9
7-30	.790			42.8		
8	.842	17.3	24.6		17.5	24.6

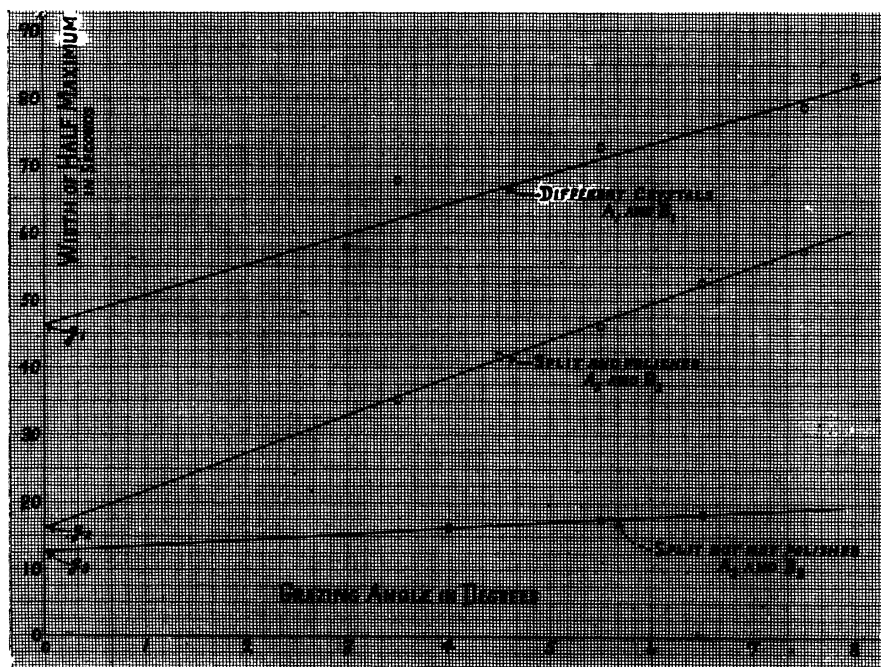


Fig. 8.

case. The reflection obtained from B however depends not only on its perfection but also on the perfection and surface of crystal A . This is indicated by the performance of the combination $A_1 - B_1$ which differed in character from that of the other set of crystals. Crystal B_1 was a perfect specimen of Iceland spar, as perfect in appearance as crystal B_2 .

Crystal A_1 on the other hand, a specimen of Montana calcite showed imperfections that were visible in its interior. The irregular results and low value of the reflection obtained with these two crystals can be fairly attributed to the imperfection of A_1 . It will be specially noticed by Table I and Fig. 8 that the width at half-maximum in particular is too large at glancing angle $3^\circ + 30'$. The other points in Fig. 8 lie reasonably on a straight line. In addition to the visible evidence, the imperfections of crystal A_1 are indicated by curves of the continuous and line

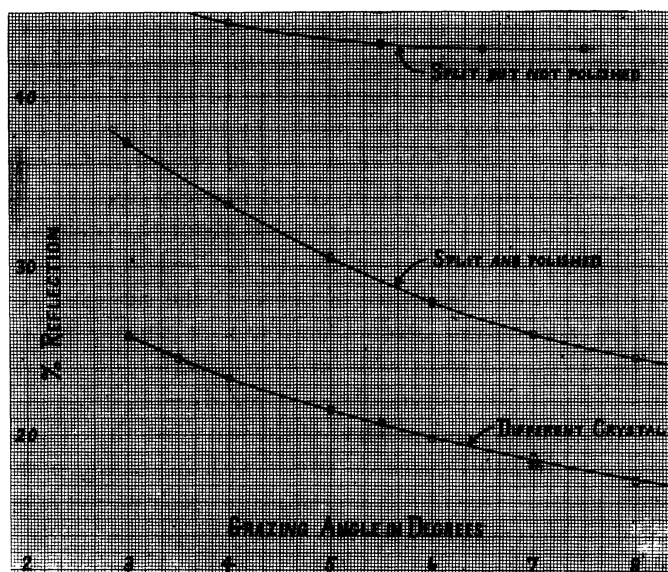


Fig. 9.

spectrum obtained by B. A. Wooten using this crystal (PHYS. REV., Jan., 1918—Fig. 1). The minute false lines there observed are undoubtedly due to imperfections in the crystal. Crystal B_1 on the other hand appears to be fairly perfect as indicated by smooth curves of the continuous and line spectrum obtained by C. T. Ulrey using this crystal (PHYS. REV., May, 1918).

What shall be defined as the coefficient of reflection is a matter for discussion. Using single crystal reflection Professor Bragg has taken the ratio of curve areas obtained by rocking the crystal and moving the slit of the ionization chamber across the beam. This of course can only be done in case of the line spectrum and even then is subject to the error of the general radiation which is present to a considerable extent.

A. H. Compton (*l.c.*) has applied the method of integration of curve

areas in case of reflection from two crystals. The coefficient of reflection is expressed in terms of angular measure (per degree). It is not clear how such a quantity which is really the ratio of intensities before and after reflection can contain angular measure.

We have taken as the coefficient the ratios of the intensities of the beam of X-rays before and after reflection from crystal B at position of maximum reflection. This is the coefficient for crystal B . The reflection from crystal B depends on the degree of homogeneity of the radiation falling on it. It is the function of crystal A to sort out from the general radiation a nearly homogeneous radiation for examination by crystal B . The degree of homogeneity depends on the perfection of crystal A . Since this crystal determines the character of the radiation falling on B it is necessary to describe the results in terms of pairs of crystals. The reflectivity measured however is that of crystal B for rays of the particular character furnished by crystal A in every case.

The curves obtained by rocking crystal B about the position of maximum reflection are found to depend to a marked extent on the perfection of the crystals and on the degree of polish of the surfaces. It will be observed that the curves become narrower at shorter wave-lengths. Also that the width decreases with increase of perfection of the crystals and their surfaces. The curves obtained for $A_3 - B_3$ are quite narrow and their width is nearly independent of the wave-length. The variation of reflectivity with wave-length decreases with increasing perfection of crystal surface also. *Our results suggest that a really perfect crystal (were such possible) would probably reflect more than fifty per cent. of the radiation falling on it at the proper angle, and that the reflectivity would be independent of wave-length.*

The attention of the reader is directed to a short paper on reflection of radiation from an infinite series of equally spaced planes appearing in this number of the PHYSICAL REVIEW. The results obtained are of considerable interest in view of the determination of reflectivity just described.

The width at half-maximum of $16''$ obtained for crystals $A_3 - B_3$ is many times too great to be attributed to the Fresnel line width that should be expected with radiation of such short wave-length and a grating of such a large number of elements. The width of $16''$ of arc for a reflectivity of about 45 per cent. would indicate that even in the case of a perfect crystal the curve width would still be of the order of a few seconds of arc. This width might be due to very minute differences in spacing in the case of the two crystals, but since contiguous planes were used for reflecting surfaces in case of $A_3 - B_3$ it is doubtful if the crystals

varied sufficiently in grating space in such a small distance as to produce so great an effect.

Another more reasonable explanation of curve width (for perfect crystals) is that it is due in some way to the width of each reflecting plane. That is, it may be due to the distribution of the electrons about the nucleus of the atoms of the crystal. One of us (Stempel) has recently developed a theory of crystal reflection involving the electron distribution that accounts with some success for these curves.

Since the width of the rocking curve would depend directly on a possible difference of crystal spacing in the two crystals one should obtain a great effect by heating one of the crystals slightly. Also by heating both crystals (*A* and *B*), keeping them at the same temperature it should be possible to measure the Debye heat effect with great accuracy by this arrangement of a double spectrometer.

PHOENIX PHYSICAL LABORATORY,
COLUMBIA UNIVERSITY,
January, 1921.

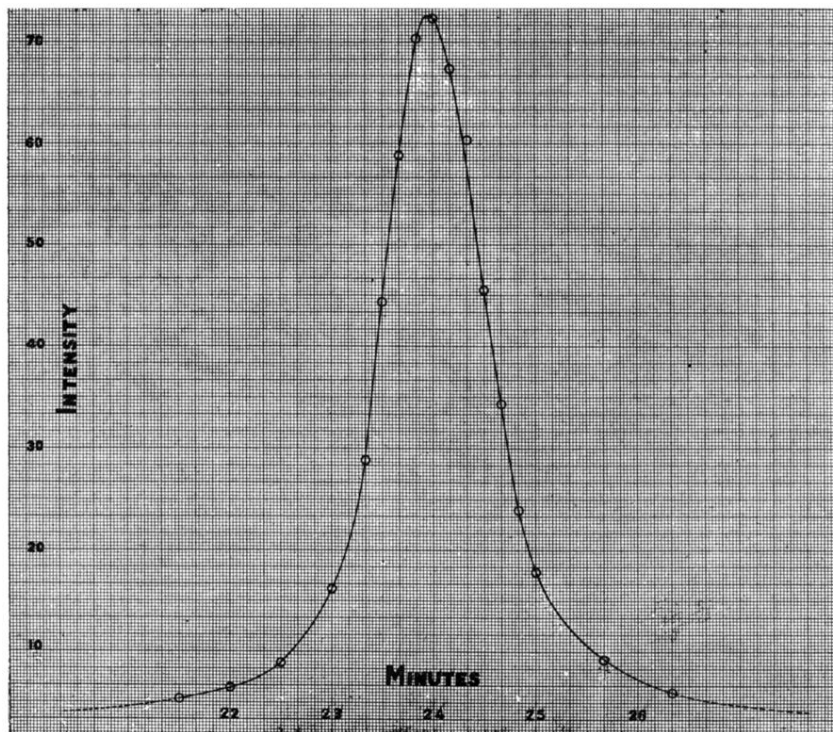


Fig. 3.

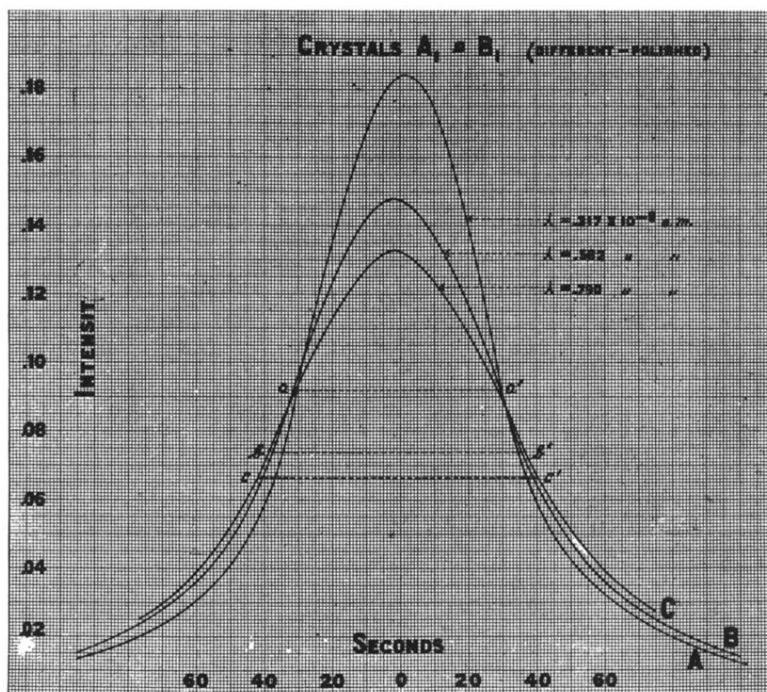


Fig. 4.

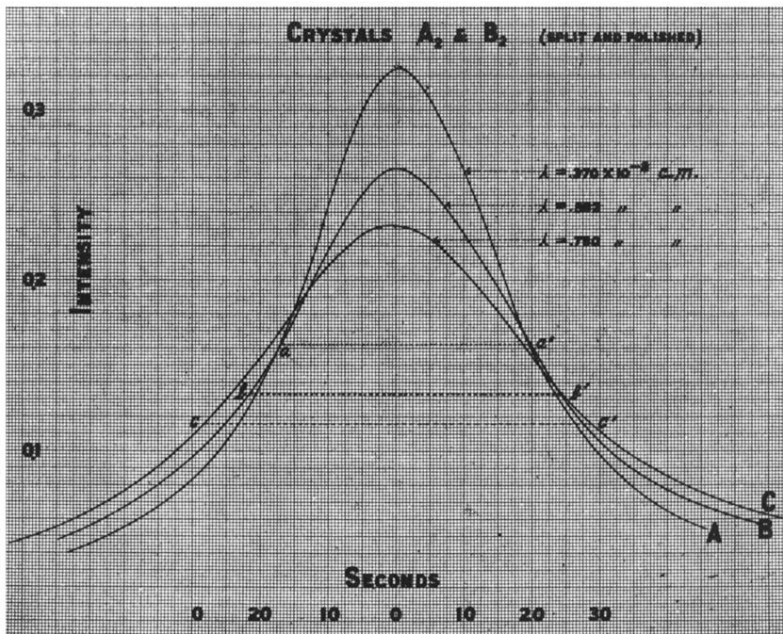


Fig. 5.

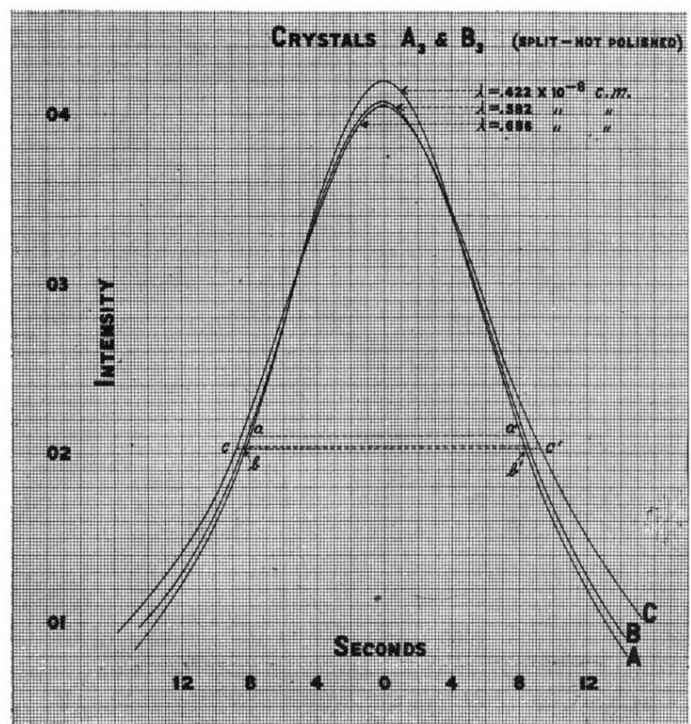


Fig. 6.

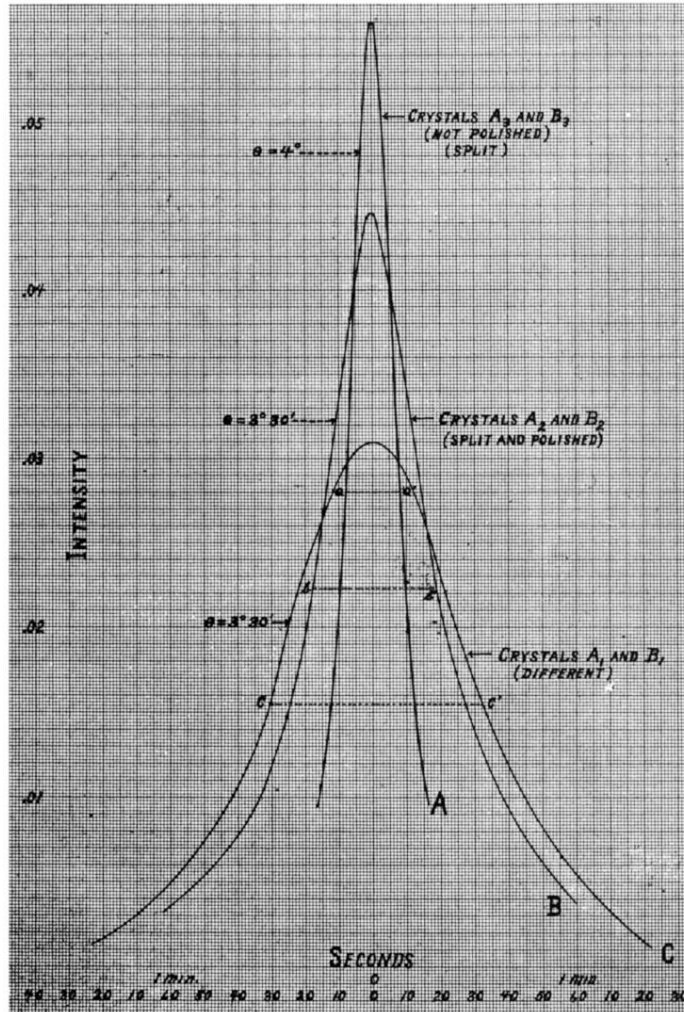


Fig. 7.

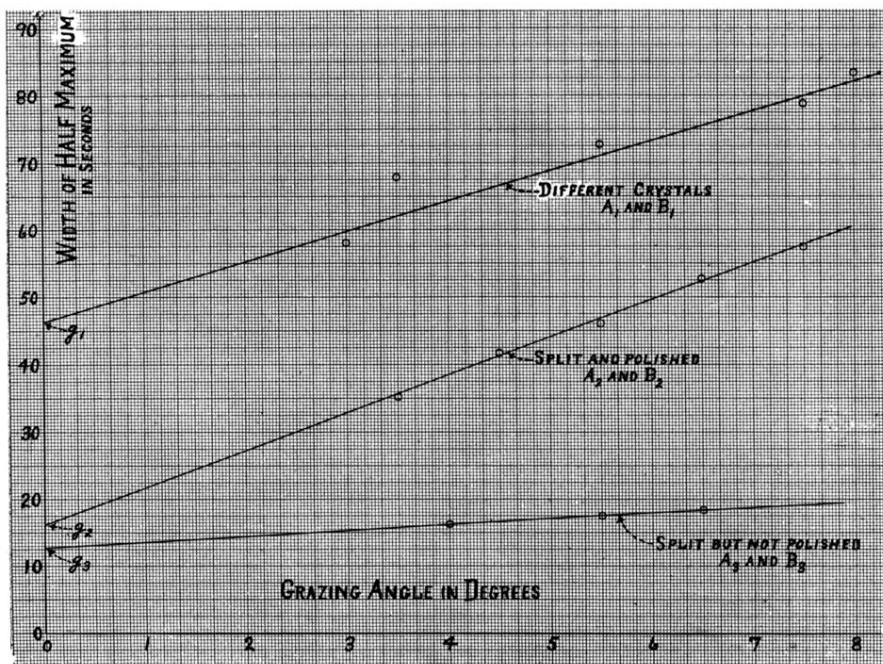


Fig. 8.

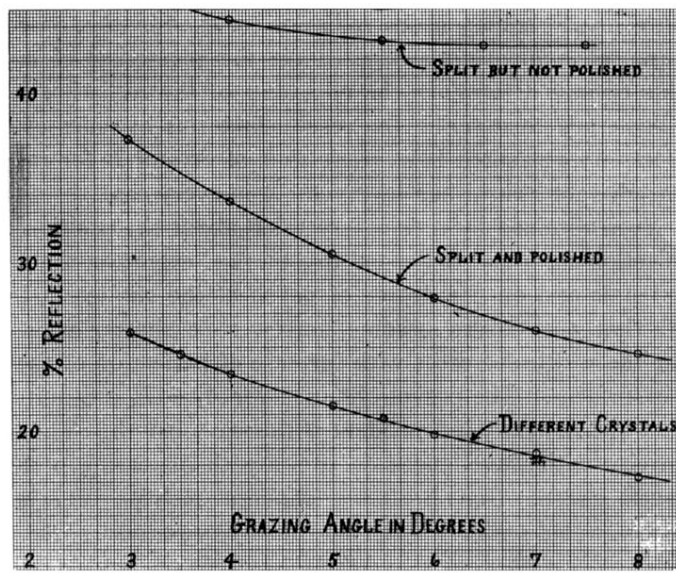


Fig. 9.