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W. Lawrence Bragg M.A. , R.W. James M.A. & C.H. Bosanquet

To cite this article: W. Lawrence Bragg M.A. , R.W. James M.A. & C.H. Bosanquet (1921) XXIX. The intensity of reflexion of X-rays by rock-salt , Philosophical Magazine Series 6, 41:243, 309-337, DOI: [10.1080/14786442108636225](https://doi.org/10.1080/14786442108636225)

To link to this article: <http://dx.doi.org/10.1080/14786442108636225>



Published online: 08 Apr 2009.



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THE
LONDON, EDINBURGH, AND DUBLIN
PHILOSOPHICAL MAGAZINE
AND
JOURNAL OF SCIENCE.

[SIXTH SERIES.]

MARCH 1921.

XXIX. *The Intensity of Reflexion of X-Rays by Rock-Salt.*
By W. LAWRENCE BRAGG, M.A., *Langworthy Professor of Physics, The University of Manchester*; R. W. JAMES, M.A., *Senior Lecturer in Physics, The University of Manchester*; and C. H. BOSANQUET, *Balliol College, Oxford* *.

Introduction.

1. ACCURATE comparisons of the intensity of reflexion of X-rays by crystal-faces were first made by W. H. Bragg †, who measured by the ionization method the energy of the X-rays reflected by various faces of rock-salt. He showed that if X-rays of definite wave-length are reflected at a glancing-angle θ by a face with one or more even crystallographic indices (*e. g.* (100) (110)), the intensity of the reflected beam can be expressed approximately by the formula

$$I_{\theta} = \frac{A}{\sin^2 \theta} e^{-B \sin^2 \theta},$$

where A and B are constants and I_{θ} measures the intensity of the reflected beam of rays. In the case of even orders of reflexion from faces with odd indices, the intensities of reflexion may be expressed by the same formula. When the observed intensities for various faces and orders are plotted

* Communicated by the Authors.

† W. H. Bragg, *Phil. Mag.* vol. xxvii, p. 881 (May 1914).

against $\sin \theta$, the points lie on a smooth curve, showing that the intensity is a function of θ and does not otherwise depend on the indices of the face or the order of the reflexion.

Theoretical expressions for the intensity of the reflected beam have been deduced by Darwin* and Compton†. Any theoretical formula for the intensity of reflexion or diffraction by a crystal must contain a factor whose value depends upon the number and arrangement of the electrons in the atoms. In the case of reflexion, the intensity falls off more quickly with increase of glancing-angle than the theoretical formula would indicate, and this falling off must be accounted for by the factor referred to above. In his paper, Compton makes use of the relative measurements of intensity made by W. H. Bragg, and assuming the theoretical formula for the intensity of reflexion to be correct, he obtains the relative value of the factor at different angles, and thence deduces certain possible arrangements for the electrons in the atom.

A very interesting paper by Debye and Scherrer‡ deals with the same question from a slightly different standpoint. The relative intensities of the diffraction haloes obtained by Debye's powder method are measured photometrically. The authors come to the same conclusion as had been drawn from the reflexion measurements§, that the falling-off in the intensity with increasing angle of scattering must be partly ascribed to the variation in the factor we are considering.

The experiments described in the present paper have been made with the object of extending the measurements of intensity over a larger range of glancing-angles. Further, a direct comparison has been made between the energy of an incident homogeneous beam, and its reflexion by the crystal. The results so obtained have been compared with those given by the theoretical formula; and it will be shown that there is strong evidence that the formula is accurate. From the observations it is possible to calculate not only the relative values of the factor, which depends on the arrangement of the electrons in the atom, but also its absolute value over a range of angles, so that a direct comparison may be made between the observed value and the value calculated for various models of the atomic structure.

* C. G. Darwin, *Phil. Mag.* vol. xxvii. pp. 315 & 675 (Feb. and April 1914).

† A. H. Compton, *Phys. Rev.* ix. p. 1 (Jan. 1917).

‡ *Phys. Zeitschr.* xix. pp. 474-483 (1918).

§ *Cp.* W. H. Bragg, *Trans. Roy. Soc. A*, cxxv. pp. 253-274 (1915).

Comparison of the Intensity of Reflexion by different Faces.

2. The method employed is fundamentally the same as that described by W. H. Bragg. Homogeneous rays are emitted from the focal spot on the target, and are limited to a narrow beam by a slit termed the bulb-slit. The beam falls on the crystal, and the reflected beam is received by an ionization-chamber through a second slit. If the chamber-slit and the bulb-slit are equidistant from the axis of the instrument with which the crystal face coincides, the chamber-slit can be set so that it receives all rays of any particular wave-length reflected by the crystal face, although the reflexion may take place at various points on the face owing to irregularity of the crystal structure. This focussing effect has been described by W. H. Bragg and one of the authors*.

The chamber may therefore be placed so as to receive all reflected homogeneous rays of any required wave-length—for instance, those corresponding to the K_{α} -line of Rhodium. If this is done, and the crystal is slowly rotated about the axis of the spectrometer, very little effect is observed until the crystal planes come into the position where the equation for reflexion is satisfied. The ionization then rapidly rises to a maximum, and falls away again as the crystal passes beyond this position. The more perfect the crystal, the narrower the range over which reflexion takes place. No crystals are perfect; in all cases the rays are reflected by a number of facets making a small angle with each other. As the crystal turns, these come, one after the other, into the correct position for reflexion, the reflected beam from each falling on the chamber-slit in virtue of the focussing effect described above.

The intensity of the reflexion cannot be measured by the effect observed when the crystal is set at the position which gives the most intense reflected beam, for the strength of this beam is dependent on the degree of perfection of the crystal face. It is measured by sweeping the crystal with uniform angular velocity through the entire range over which it reflects, and by observing the total ionization produced in the chamber during this process. In this way, every part of the pencil of homogeneous rays will fall at some time on a portion of crystal which reflects it, and will contribute its share to the whole effect. Experiments made with different crystals show that the intensity, measured in this way, is not dependent on the degree of perfection of the crystal,

* 'X-Rays and Crystal Structure,' p. 31.

that it is the same for any one face and order, however the crystal may be distorted, and is, in fact, a definite physical quantity on which theoretical calculations may be based.

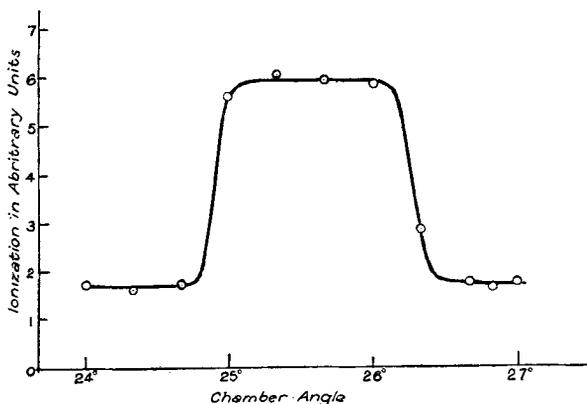
3. The X-ray spectrometer is of the type devised by W. H. Bragg, which has been described in former papers. The ionization-chamber is filled with methyl bromide, and a potential of 320 volts is applied between the outer walls of the chamber, and the inner electrode which is connected to the electrometer. This potential is sufficient to prevent appreciable recombination of ions with the strongest ionization produced. A Lutz-Edelmann string electrometer is used to observe the charge communicated to the electrode, its sensitivity being adjusted to about 100 divisions to a volt. A null method is employed to measure the charge. The inner rod of a small cylindrical condenser is connected to the electrometer, the outer cylinder being raised to any desired potential by a potential divider and battery. When the crystal is swept through the reflecting angle, a charge is communicated to the electrometer. This charge is neutralized by adjusting the potential of the outer cylinder of the condenser until the string in the electrometer returns to its zero. The potential applied to the condenser is then proportional to the total charge which has passed through the ionization-chamber.

4. It is necessary that the incident beam of rays should remain constant in intensity. A Coolidge bulb is used, in which the anticathode consists of a button of rhodium embedded in a tungsten block. A large induction-coil and Sanax break supply a current of 1.5 milliamperes at a potential of about 50,000 volts. It is possible to keep the intensity of the rays constant to within 2 or 3 per cent. and with the Coolidge tube it is also possible to repeat the conditions of the experiment on successive occasions in a satisfactory manner. Such variations in intensity as do occur are probably due to the irregular action of the break.

5. Superimposed on the homogeneous rays, there is a general radiation of all wave-lengths which is also reflected by the crystal. In making a measurement of intensity it is necessary to allow for this general radiation. When comparing the intensity for two faces or orders, a preliminary survey is made in each case to enable a measurement to be made of the effect of the general radiation. The chamber is set at a series of angles over a range including the angle at which it receives the homogeneous beam: At each position of the chamber, the crystal is swept through the corresponding reflecting angle and the total ionization measured.

A series of readings plotted in this way is shown in fig. 1. The readings are at first approximately constant, being due to the general radiation. As the position at which the chamber is set approaches that at which homogeneous rays

Fig. 1.



are received, the ionization rises rapidly, remains constant again as long as the whole pencil of homogeneous rays enters the chamber, and then falls to a value approximately equal to its former steady value when the homogeneous rays are no longer received.

6. When comparing two crystal faces, this survey is made in each case. One of the faces is then mounted in the spectrometer, the chamber set so that it receives the homogeneous beam, and a series of readings taken by sweeping the crystal backwards and forwards. The crystal faces are interchanged, the chamber reset, and a series of readings taken for the other face. This process is repeated several times, and the means of the intensities for the faces are compared. The preliminary survey indicates what fraction of the total intensity observed must be subtracted, for each face, in order to allow for the general radiation; and when this has been done, the ratio of the corrected readings gives the ratio of the intensity of reflexion by the two faces. A series of readings obtained in this way is given below. It is a comparison of the reflexion by the (311) face of NaCl, mounted so as to face left on the spectrometer, of the same face turned through 180° so that it faces right, and of the third-order reflexion from the face (100) mounted so as to face right. The difference between the values for (311) L and (311) R is due to inaccurate grinding of the crystal

surface, the effect of which will be discussed later. It can be shown that, although they differ greatly, their mean represents accurately the strength of reflexion if the face were cut true. In taking the readings, the crystal was turned 5 minutes of arc for every beat of a metronome, beating 100 to the minute.

Comparison of (311) L, (311) R, and (300) R.

Face.	Sweep of crystal.	Chamber angle.	Potentiometer scale.	Readings.	Mean of readings.
(311) L	8 50-11 20	20 50	2	(71, 73, 73, 72)	72.2
(311) R	10 05-12 35	21 00	2	(57, 55, 58, 57, 56, 56)	56.5
(311) L	8 50-11 20	20 50	2	(73, 74, 74, 75)	74.0
(300) R	17 30-20 00	38 25	3	(77, 78, 78, 78)	77.8
(311) L	8 50-11 20	20 50	2	(72, 71, 70, 72, 71, 70)	71.0
(300) R	17 30-20 00	38 25	3	(78, 78, 79, 80, 80)	79.0

A survey of the three reflexions showed that the homogeneous radiation was responsible for 76.9 per cent. of the total effect in the case of the (300) R reflexion, 33.0 per cent. for the (311) R, and 32.2 per cent. for the (311) L reflexion. Since the intensity is very much greater for the (300) face than for the (311) face, different scales on the potentiometer were used. A reading of 72.2 on the second scale represents 72.2 per cent. of a total voltage of 15.72, the corresponding voltage for the third scale being 22.79.

Taking this into account and allowing for the general radiation, one gets a ratio

$$\frac{\text{Mean intensity, face (311)}}{\text{Intensity, face (300) R}} = \frac{3.22}{13.45} = 0.2395.$$

In another experiment, (300) R and (300) L were compared, and in this way the relative mean intensities of (311) and (300) measured.

In order to have a uniform system of indicating both the order of reflexion and the face at which it is taking place, the convention of multiplying the indices of the face by the order has been adopted. Thus, by the reflexion from (622) is meant the second order of reflexion from the face (311).

The crystal is not turned continuously during each reading; its setting is altered five minutes of arc at each beat of a metronome by means of a series of spokes on the tangent screw. It would be preferable to turn the crystal with a uniform angular velocity, but it is unlikely that any appreciable error was caused by the method used. In order

to make certain that this was the case, the crystal was turned slightly between each reading, in order to ensure that the halting-places did not occur at exactly the same angles.

7. The faces used in this experiment were prepared by grinding, and were of sufficient area to intercept the whole of the incident beam of rays. In general, faces were prepared 3 or 4 centimetres in length and breadth. The perfection of the crystal structure may be judged from the range of angles at which reflexion takes place. In most cases the greater part of the effect was observed to take place within less than a degree of arc as the crystal was turned, the faces being prepared from large blocks of rock-salt which were very little distorted.

The face should be cut so that it is as nearly parallel to the planes of the crystal structure as possible. If this is not the case, there will be a difference in the intensities of reflexion when the crystal is mounted facing right and left on the spectrometer table. This effect is described and explained in the paper by W. H. Bragg referred to above (Phil. Mag. *loc. cit.* p. 888). When the crystal face is not parallel to the planes of the structure, the incident and reflected beams do not make equal angles with the face of the crystal. If the glancing-angle of incidence is less than that of reflexion, the rays suffer less absorption in the crystal than when the reverse is the case. The smaller the angle of incidence, the greater is this effect.

As an example of this effect, the following table gives a comparison of the 1st-, 2nd-, and 3rd-order reflexions from a natural face ($1\bar{1}0$) of a ruby crystal (Al_2O_3). The crystal had the form of a six-sided prism bounded by the faces ($1\bar{1}0$), but had so developed that this prism tapered towards one end. The angle between the face of the prism and the crystal planes could be measured by comparing the angles at which reflexion took place on the right-hand and left-hand sides. In this case it was $1^\circ 49'$. It will be observed what a large effect is produced by the small deviation from truth in the orientation of the face.

Comparison of ($1\bar{1}0$), ($2\bar{2}0$), ($3\bar{3}0$) Al_2O_3 .

Face	$1\bar{1}0$	$2\bar{2}0$	$3\bar{3}0$
Right-hand side	100	61.0	41.5
Left-hand side	52	38.9	39.5

The corresponding figures for the face (100) of NaCl are given below :—

	(100).	(200).	(300).
Right-hand side	100	21.3	5.08 *
Left-hand side	116.6	21.8	

The error in the orientation of the (100) face was in this case too small to be measured with accuracy; it was less than 30 minutes of arc.

Since the effect of inaccurate grinding of the face is so much less for the second order than for the first order, the intensity of reflexion from (200) NaCl was taken as standard, and all other intensities compared with it. On account of the difficulty of grinding the face accurately, there was generally a difference in the intensities on the two sides. It can be proved, however, that the mean of the intensities on the two sides can be taken as the right value without making an appreciable error so long as the difference in the value does not exceed about 25 per cent. The intensities were in all cases measured on both sides and the mean taken. In most cases the difference between them was small; for the higher orders it did not exceed 5 per cent.

8. It is necessary to use faces which have been ground. The strength of the reflexion is very different, especially for small glancing-angles, when a cleavage surface is compared with one which has been ground. As an example, a comparison is given below of the reflexion from a very perfect cleavage face of a rock-salt crystal (A) with a similar cleavage face on a crystal (D), which was afterwards ground until a layer 1 millimetre thick had been removed.

The intensity of reflexion from crystal (D) was measured with two orientations of the face. In the first, the crystal was set so that the edge, on which the knife was pressed in cleaving the crystal, was horizontal. In the second position, the edge was vertical and therefore at right angles to the horizontal beam of X-rays.

Face.	Intensity.
A (100). (Cleavage face)	50.8
D (100) before grinding :—	
First position	25.4
Second position	12.9
D (100) after grinding	100

Hence D (100), after grinding, reflected eight times as well as in the second position before grinding.

* The difference between right-hand and left-hand sides was less than the error of determination.

Comparison of 1st, 2nd, and 3rd orders :—

	(100).	(200).	(300).
Crystal D (after grinding)	100	18.4	41
Crystal A (cleavage face)	50.8	18.1	47

These comparisons were made for reflexion on the right-hand side only, and must be regarded as approximate.

In this case, as in the case of the error due to inaccurate grinding, the effect is much greater for the first-order reflexion. On account of this effect, intensity measurements of the (100) reflexion are doubtful, and this provides an additional reason for using the reflexion (200) as standard.

The difference in intensity for the two positions of the D cleavage face indicates that the reason for the imperfect reflexion may be due to a distortion of a freshly-cleaved surface. A cleaved surface has a rippled appearance, the ripples being parallel to the line on which the knife-edge was pressed in cleaving the crystal. The range of angles, however, over which the crystal reflects is no greater for a cleavage surface than for a ground surface, so that it would not appear that the imperfect reflexion is due to small variations in orientation of the face. Measurements made at various stages in the grinding down of a crystal face indicate that the effect is deep seated, and a depth of a millimetre at least was removed from the face D (100) before it was used in obtaining the results given in this paper.

9. Certain precautions must be taken in order to ensure an accurate result.

The crystal must be swept through a range of angles sufficiently great to ensure that all the facets add their share to the total effect. A range of three degrees is generally used. To check whether this was sufficient, a larger range was used in certain cases. This increased the amount of general radiation, but when this was subtracted, the intensity due to the homogeneous rays was found to be the same as when the smaller range was used.

The crystal planes must be parallel to the axis of the instrument. The crystal is mounted on a table which can be rocked about a horizontal axis parallel to the faces, and the intensity of a high-order spectrum is measured for various tilts of the face. The crystal is fixed at that setting which gives a maximum effect. Unless this precaution is taken, the beam on reflexion may be thrown upwards or downwards, and not be completely received by the ionization-chamber.

A simple calculation shows what the width of the slit of the ionization-chamber must be in order that all the homogeneous rays may enter it. It is advisable to limit this as much as possible, as the amount of general radiation is directly proportional to its width. The breadth of the homogeneous beam may be found by a survey with a very fine chamber-slit set at a series of angles. For the higher orders, the chamber-slit must be wider in order to include the two components of the K_{α} doublet.

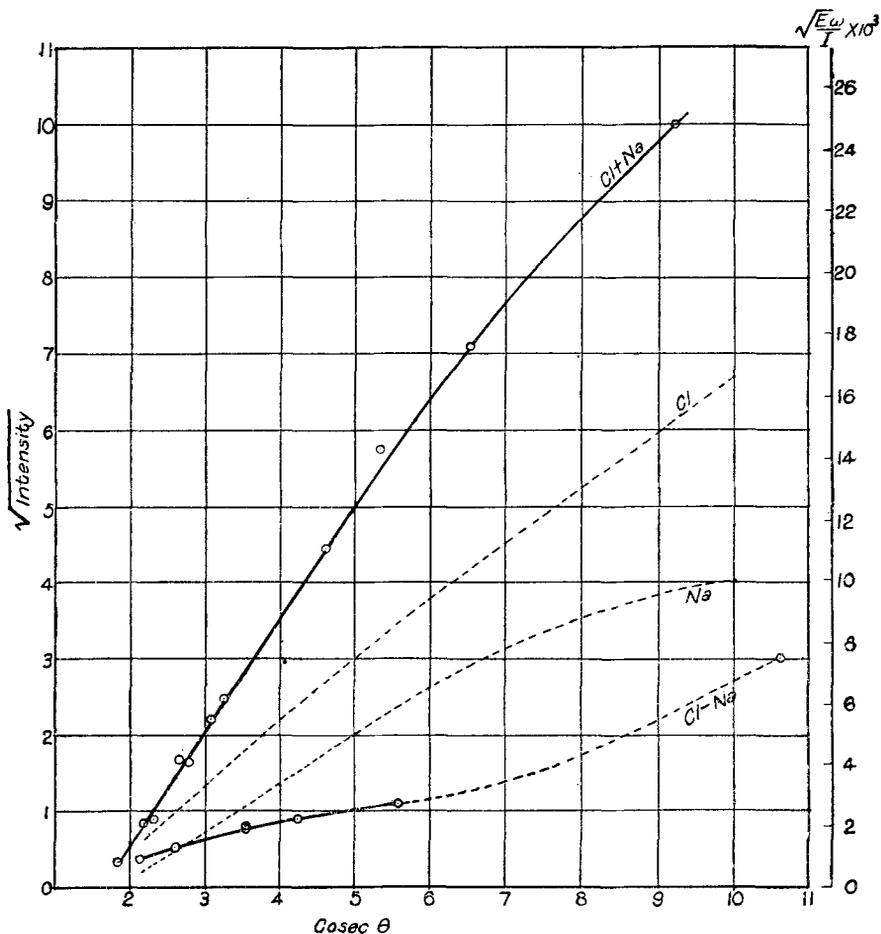
10. The comparisons which have been made are tabulated below, and the results are plotted in the form of a graph in fig. 2.

Plane.	Intensity.	$\sqrt{\text{Intensity.}}$	Cosec θ .	$\frac{E_{\omega}}{I} \times 10^6$.	$\sqrt{\frac{E_{\omega}}{I}} \times 10^3$
100	100	10	9.21	612	24.80
200	19.90	4.46	4.60	122	11.05
300	4.87	2.21	3.07	29.8	5.49
400	0.79	0.89	2.30	4.85	2.20
500	0.116	0.34	1.84	0.71	0.84
110	50.4	7.10	6.50	310	17.60
220	6.10	2.47	3.25	37.3	6.12
330	0.71	0.84	2.17	4.35	2.08
111	9.00	3.00	10.62	55.1	7.45
222	33.1	5.75	5.31	202	14.25
333	0.58	0.76	3.54	3.55	1.89
444	2.82	1.68	2.65	17.2	4.16
555	0.137	0.37	2.12	0.84	0.92
311	1.17	1.09	5.56	7.22	2.70
622	2.69	1.64	2.78	16.40	4.06
331	0.81	0.90	4.23	4.95	2.23
511	0.61	0.78	3.54	3.74	1.93
711	0.302	0.55.	2.58	1.87	1.37

The figures in the column headed "Intensity" were initially expressed in terms of the (100) reflexion, which was put equal to 100. Since it was discovered later that the (200) reflexion was a more reliable standard of intensity, all the other intensities have been determined relatively to it. In order to facilitate comparison with figures given by other authors, its value has been fixed at 19.90, since the ratio 100:19.90 was the most reliable value for the ratio

(100):(200). The values for (100) and (111) are, however, difficult to measure accurately, since the nature of the crystal face has so great an effect on the intensity, and the figures given here must be regarded as approximate.

Fig. 2.



For the other faces, it is believed that the average error does not exceed 2 per cent., except in the case of the very small intensities of high order. The smallest intensity measured is that denoted by (500), and is little more than one thousandth of the (100) reflexion.

The figures obtained by W. H. Bragg are given below for the sake of comparison :—

Plane.	Intensity.
100	100
200	18·7
300	6·25
110	41·0
220	7·05
222	24·4
444	4·20

11. In fig. 2 the square-root of the relative intensity has been plotted against the cosecant of the glancing-angle. By plotting the intensities in this way, the approximate relation found by W. H. Bragg—that the intensity varies inversely as $\sin^2 \theta$ —is made evident.

All the points lie on two smooth curves, showing that they form two groups within each of which the intensity is a function of the glancing-angle alone. For instance, the reflexions from the faces (511) and (333) occur at the same angle, and the corresponding intensities 0·74 and 0·72 are identical within the error of observation.

The points which lie on the lower curve are those for which all the indices are odd—the faces (111), (311), (331), (333), (511), (711), (555). These reflexions are from planes which contain alternately sodium and chlorine atoms. The wave-train reflected from the planes containing sodium atoms is 180° out of phase with that reflected by the plane containing chlorine atoms. The other reflexions are either from planes which contain both sodium and chlorine atoms and are identical in their nature, or are reflexions of an even order from planes containing sodium and chlorine atoms alternately. In both cases the sodium and chlorine atoms reflect wave-trains which are in phase with each other.

Since the square-root of the intensity has been plotted, this may be taken as being proportional to the amplitude of the reflected wave-trains. The upper curve, therefore, represents the sum of the amplitudes due to sodium and chlorine atoms, the lower, the difference of these two amplitudes.

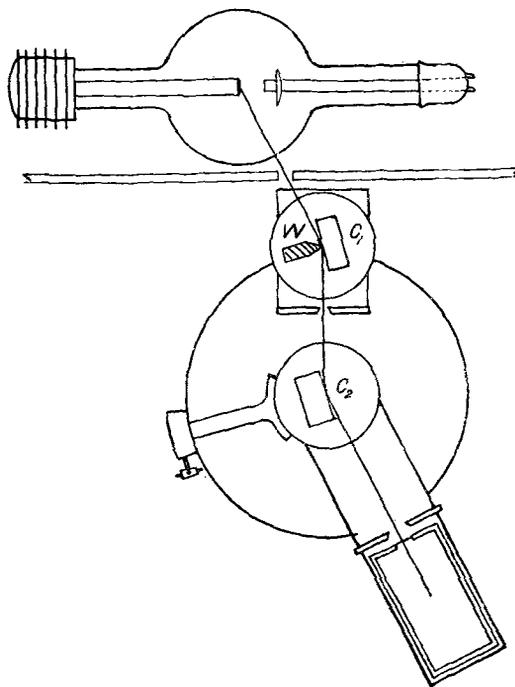
Comparison of Incident and Reflected Beams.

12. The rays from the bulb consist of heterogeneous radiation of all wave-lengths over a certain range, superimposed on the homogeneous radiation whose intensity of

reflexion has been measured. In order to compare directly the energy in the incident and reflected homogeneous beams, it is necessary to obtain a homogeneous beam by reflexion from a crystal face, and observe the total amount of radiation reflected by a second crystal, turning with constant angular velocity, on which this homogeneous beam is allowed to fall.

Fig. 3 shows the arrangement of the apparatus to effect this. The rays from the anticathode were reflected by the

Fig. 3.



crystal C_1 so as to pass after reflexion through the collimator slits of the spectrometer. The incident beam was not limited by slits, but by being forced to pass on reflexion around the lead wedge W^* , the edge of which was pressed against the crystal face. The position of the bulb and the orientation of the crystal face were adjusted until the reflected beam passed truly through the axis of the spectrometer. This beam fell on the second crystal C_2 , which was rotated with uniform velocity ω , and the total amount of radiation

* *Cp. Seeman, Phys. Zeit. xv. p. 795 (1914).*

reflected was measured in the ordinary way. In this case, since the rays are homogeneous, no allowance for general radiation need be made.

The amount of energy reflected is proportional to the intensity of the incident beam, and inversely proportional to the angular velocity of rotation. The quantity $\frac{E\omega}{I}$ is therefore a constant characteristic of any one face and order, where

E = Total amount of energy reflected when the crystal is rotated with angular velocity ω radians per second.

I = Total amount of energy passing into ionization-chamber when the incident beam enters it for one second.

This constant $\frac{E\omega}{I}$ will be defined as the "Reflecting Power" of the crystal face for the wave-length λ .

The chamber was placed so as to receive the whole of the incident beam (*i. e.* that reflected from C_1), and the effect measured when the rays entered the chamber for a known time. It was then turned so as to receive the beam reflected from C_2 when the second crystal was mounted on the spectrometer table and turned with a known angular velocity.

This was done for the face (100) of NaCl. The reflexions from other faces are so much weaker that it was not convenient to compare them directly with the incident beam. As a check, an absolute measurement was made of the reflecting power for (222), which was found to be in agreement with that calculated by a comparison with (100).

A series of measurements gave for the constant

$$\frac{E\omega}{I} = 0.000612 \text{ for NaCl (100)*.}$$

Since the reflecting powers of the other faces have been determined in terms of that from the face (100), their absolute reflecting powers may now be calculated. These values are given in the fifth column of Table I.

[The reflecting power of a face can only be defined satisfactorily in this way. It may be of interest, however, to give approximately the proportion of homogeneous radiation reflected when the crystal face is set at the most favourable

* More recent determinations of this value have shown that the figure 0.000612 is too high. The value varies somewhat with the nature of the crystal face, and a better mean value is .00055.

angle, although this depends on the state of perfection of the face as explained above. A direct comparison showed that when a narrow beam of X-rays falls on the face (100) set so as to reflect it, the intensity of the reflected beam is about one twenty-fifth of the incident beam.]

Theoretical Formula for the Intensity of Reflexion.

13. Formulæ for the intensity of reflexion have been deduced by Darwin and Compton (*loc. cit.*). The formula given by Compton is directly applicable, for he calculates the total amount of energy reflected when the crystal is turned at a uniform rate through the reflecting-angle. That given by Darwin may be extended to this case, and is in agreement with Compton's formula.

These formulæ are based on the amount of radiation scattered by a free electron when set in oscillation by rays of given intensity. It has been shown by J. J. Thomson * that the amount of energy S radiated per second by a single electron is given by

$$\frac{S}{P} = \frac{8\pi}{3} \frac{e^4}{m^2 c^4},$$

where P is the energy of the incident radiation falling on 1 sq. cm. per second, e and m are the charge and mass of the electron respectively, and c is the velocity of light. This expression is confirmed by Barkla's work † on the total amount of radiation scattered by elements of low atomic weight, from which he deduced that the number of electrons in the atoms of these elements is approximately equal to one-half the atomic weight. If the incident radiation is plane polarized, the relation between the amplitude of the electric vector of the incident radiation, and that of the radiation scattered in any direction perpendicular to the direction of the electric vector, is given by

$$\frac{A'}{A} = \frac{e^2}{mc^2} \cdot \frac{1}{R},$$

where R is the distance from the electron. To simplify matters, we will consider that the radiation reflected from the crystal face is plane polarized in such a manner that the electric vector is perpendicular to the plane of incidence, and allow for the "polarization factor" at a later stage of the calculation.

* J. J. Thomson, 'Conduction of Electricity through Gases,' p. 321.

† C. G. Barkla, *Phil. Mag.* vii. p. 543 (1904), and xxi. p. 648 (1911).

The following is a brief summary of the calculation, treated in a slightly different manner from that in Darwin's and Compton's papers, where it is worked out more completely.

If an atom contains Z electrons, and the waves scattered by these electrons are in phase, the amplitude of the scattered wave will be

$$A' = \frac{A}{R} \cdot Z \frac{e^2}{mc^2} \dots \dots \dots (1)$$

If the spatial distribution of the electrons is such that the scattered waves are not in phase, the factor, Z must be replaced by a function F , which depends on the angle of scattering and the positions of the electrons. F tends to its maximum value Z at small angles of scattering.

Let rays from a source S fall at a glancing-angle θ on a plane containing n atoms per unit area, and be reflected. The amplitude at any point P is equal to one-half the total effect due to the scattering by the atoms lying in the first Fresnel zone around the corresponding point of incidence I . The area of the zone is equal to

$$\frac{\pi\lambda}{\sin \theta} \cdot \frac{r_1 r_2}{r_1 + r_2},$$

where $r_1 = SI$, $r_2 = IP$. The number of atoms it contains is therefore

$$\frac{n\pi\lambda}{\sin \theta} \cdot \frac{r_1 r_2}{r_1 + r_2},$$

and the amplitude at P is equal to

$$\frac{1}{2} \cdot \frac{2}{\pi} \frac{n\pi\lambda}{\sin \theta} \cdot \frac{r_1 r_2}{r_1 + r_2} \cdot A' = \frac{n\lambda}{\sin \theta} \cdot \frac{r_1 r_2}{r_1 + r_2} \cdot \frac{A}{r_2} \cdot F \cdot \frac{e^2}{mc^2}.$$

If r_1 is great compared with r_2 , so that the incident rays may be considered as a parallel beam, we get the relation

$$\frac{\text{Amplitude of reflected beam}}{\text{Amplitude of incident beam}} = \frac{D'}{D} = \frac{n\lambda}{\sin \theta} \cdot F \cdot \frac{e^2}{mc^2}. \quad (2)$$

Considering now a thin slip of crystal consisting of p planes at a distance d apart, the reflexion will be most intense when

$$m\lambda = 2d \sin \theta.$$

At a glancing-angle $(\theta + \epsilon)$ the amplitude of the reflected beam will be

$$D'p \frac{\sin\left(p \cdot \pi \cdot \epsilon \cdot \frac{2d \cos \theta}{\lambda}\right)}{p \cdot \pi \cdot \epsilon \cdot \frac{2d \cos \theta}{\lambda}}.$$

If we put $\phi = p \cdot \pi \cdot \epsilon \cdot \frac{2d \cos \theta}{\lambda},$

Amplitude $= D'p \cdot \frac{\sin \phi}{\phi},$

and the energy of the reflected beam is proportional to

$$(D')^2 p^2 \frac{\sin^2 \phi}{\phi^2}.$$

If, now, the crystal is rotated with constant angular velocity ω , the total amount of radiation reflected is proportional to .

$$\begin{aligned} & (D')^2 \cdot p^2 \cdot \int_{-\infty}^{+\infty} \frac{\sin^2 \phi}{\phi^2} dt \\ &= (D')^2 p^2 \int_{-\infty}^{+\infty} \frac{\sin^2 \phi}{\phi^2} \frac{d\epsilon}{\omega} \\ &= (D')^2 p^2 \int_{-\infty}^{+\infty} \frac{\sin^2 \phi}{\phi^2} \frac{\lambda \cdot d\phi}{2p\pi d \cos \theta \cdot \omega}. \end{aligned}$$

Since $\int_{-\infty}^{+\infty} \frac{\sin^2 \phi}{\phi^2} d\phi = \pi,$

this becomes

$$\frac{(D')^2 p \cdot \lambda}{2d \cos \theta \cdot \omega} = D^2 \cdot \frac{n^2 \lambda^2}{\sin^2 \theta} \cdot F^2 \frac{e^4}{m^2 c^4} \cdot \frac{p \lambda}{2\omega d \cos \theta}.$$

If $N =$ number of atoms per unit volume,
 $t =$ thickness of crystal slip,

we have $n = N \cdot d, \quad t = pd.$

The energy in both the reflected and incident beams is proportional to the square of the amplitude of the electric vector. If the energy of the incident beam falling on the

crystal per second is I , and it is all intercepted by the slip of crystal, the total energy reflected will be given by

$$\frac{E}{I} = \frac{n^2 \lambda^2}{\sin^2 \theta} \cdot F^2 \frac{e^4}{m^2 c^4} \cdot \frac{\rho \lambda}{2 \omega d \cos \theta},$$

or

$$\frac{E \omega}{I} = \frac{N^2 \lambda^3 t}{2 \sin^2 \theta \cos \theta} \cdot F^2 \frac{e^4}{m^2 c^4} \dots \dots \dots (3)$$

In this calculation it has been assumed that the absorption of the radiation is inappreciable.

As a corollary, we can calculate the reflecting power of a homogeneous fragment of crystal of volume V . The volume of the slip irradiated by a narrow pencil of rays is equal to $\frac{S}{\sin \theta} \cdot t$, where S is the area of cross-section of the pencil.

From the above formula

$$\frac{E \omega}{I} = \frac{N^2 \lambda^3}{2 \sin \theta \cos \theta} \cdot F^2 \cdot \frac{e^4}{m^2 c^4} \cdot \frac{t}{\sin \theta}.$$

Now, $I = S I_0$ when I_0 is equal to the intensity of the beam irradiating the crystal, defined as the amount of energy falling on one square centimetre per second, whence

$$\frac{E \omega}{I_0} = \frac{N^2 \lambda^3}{\sin 2 \theta} \cdot F^2 \frac{e^4}{m^2 c^4} \cdot V. \dots \dots \dots (4)$$

This result shows that the "Reflecting power" of a homogeneous fragment of the crystal is proportional to its volume, if the fragment be so small that absorption in it is inappreciable.

We will now assume that the crystal consists of a number of such homogeneous crystalline particles, set approximately parallel to each other, but not exactly so. When the rays are reflected from the face of a crystal, the reflexion by particles below the surface is diminished by absorption. It will be assumed that the linear coefficient of absorption μ is a constant. (This assumption will be discussed more fully below.)

Rays reflected by a particle at a depth z beneath the crystal surface suffer absorption by passing through a distance $\frac{2z}{\sin \theta}$ of the crystal. They are therefore reduced in intensity in the ratio

$$1 : e^{-\frac{2\mu z}{\sin \theta}}.$$

By equation (3) the reflecting power of a thickness t of the crystal is given by the formula

$$\frac{E\omega}{I} = \frac{N^2\lambda^3}{2 \sin^2 \theta \cos \theta} \cdot F^2 \frac{e^4}{m^2c^4} \cdot t.$$

The total reflecting power of the crystal face is therefore equal to

$$\begin{aligned} & \frac{N^2\lambda^3}{2 \sin^2 \theta \cos \theta} \cdot F^2 \frac{e^4}{m^2c^4} \int_0^\infty e^{-\frac{2\mu z}{\sin \theta}} dz \\ &= \frac{N^2\lambda^3}{2 \sin^2 \theta \cos \theta} \cdot F^2 \frac{e^4}{m^2c^4} \cdot \frac{\sin \theta}{2\mu} \\ &= \frac{N^2\lambda^3}{2\mu \sin 2\theta} \cdot F^2 \frac{e^4}{m^2c^4} \cdot \dots \dots \dots (5) \end{aligned}$$

Compton gives his formula in the form

$$R = \frac{E_r}{E_i} = \frac{N^2\lambda^3\phi^2\psi^2}{2\mu \sin 2\theta} \cdot \frac{1}{\Delta\theta},$$

which agrees with this, since his factors $N^2\phi^2\psi^2$ have the same meaning as $N^2F^2 \frac{e^4}{m^2c^4}$ defined as above. Compton

derives the equation by a more complete mathematical treatment, and has discussed very fully the effect of imperfection of the crystal and of the length of the wave-train. He arrives at the same formula, whatever assumptions are made.

This expression must now be multiplied by a "polarization factor" $\frac{1 + \cos^2 2\theta}{2}$ and a "Debye factor" $e^{-B \sin^2 \theta}$.

The complete expression for the reflecting power R is therefore

$$\frac{E\omega}{I} = R = \frac{N^2\lambda^3}{2\mu \sin 2\theta} \cdot F^2 \frac{e^4}{m^2c^4} \cdot \frac{1 + \cos^2 2\theta}{2} \cdot e^{-B \sin^2 \theta} \dots (6)$$

The Debye Factor for Rock-salt.

14. Debye * gives the formula for the factor which expresses the diminution of the intensity of reflexion with rise of temperature in the form

$$e^{-B \sin^2 \theta},$$

* P. Debye, *Ann. der Phys.* (4) xliii. p. 49 (1914).

where the constant B is a function of the temperature, the wave-length λ , the atomic weight, and the characteristic temperature θ of the crystal.

W. H. Bragg* made a series of measurements of the effect of temperature in reducing the intensity of reflexion by rock-salt. Intensities were compared at 288°K and 643°K . The results were, within the errors of experiment, consistent with the ratios given by Debye's formula. The latter gives different values for B according to the assumption or otherwise of the existence of the "Nullpunktsenergie." Compton (*loc. cit.* p. 47) gives, as the two values for B in NaCl , 4.6 and 3.6 respectively.

The mean value for B at 288°C ., calculated from W. H. Bragg's results, is equal to 4.12, and this will be assumed in the calculations which follow. To assume that the effect of temperature on both curves of fig. 2 is the same, is equivalent to supposing that the average amplitude of vibration of sodium and chlorine atoms is the same. This is very probably not the case, and the authors intend to measure the effect of cooling down the crystal in order to obtain an empirical law over a wider range expressing the temperature effect. However, the factor $e^{-B \sin^2 \theta}$ does not affect very greatly any but the smallest intensities measured, and will therefore not make much difference to the conclusions to be drawn from the curves.

The Linear Coefficient of Absorption " μ ."

15. The coefficient of absorption by rock-salt of the homogeneous radiation was measured in the usual way by interposing plates of rock-salt of various thickness in the path of the direct beam (fig. 3) and measuring the diminution in energy of the beam. Experiments were made with plates from 0.05 cm. to 0.15 cm. thick. The linear coefficient of absorption μ was found to be 10.7.

In the theoretical formula it has been assumed that μ is constant. Now, W. H. Bragg has shown that in the case of the diamond, when the crystal is set so as to reflect the radiation, the absorption-coefficient is abnormally large. It was therefore interesting to try whether such an effect is observable in the case of rock-salt. A slip of crystal 0.92 mm. in thickness with faces parallel to (100) was set on the spectrometer table at right angles to the homogeneous beam from C_1 in fig. 3 and the absorption measured. It was then turned through an angle of about 66° until the (100)

* W. H. Bragg, *Phil. Mag. loc. cit.* p. 897.

planes at right angles to the crystal face reflected the radiation, the reflexion being observed in the usual way. On redetermining the absorption-coefficient it was found to have increased by about 15 per cent. This effect is discussed in Darwin's paper referred to above, and will reduce the intensity of the reflected beam. Its effect will be smaller for higher orders of reflexion, since the increase in absorption is due to multiple reflexion within the crystal interfering with the primary beam, and reflexion is so much weaker in the higher orders.

The effect will not be taken into account in the calculations, since it is not obvious what allowance should be made for it. It is to be remembered, however, that the reflexions from (100) and (110) must be diminished by the increase in the absorption-coefficient.

It may possibly be the case that the effect of grinding a cleavage face, which increases so greatly the intensity of reflexion, is due to the fact that grinding breaks the crystal up into a number of small homogeneous crystals oriented in slightly different directions, so that absorption at the reflecting angle plays a less important part in diminishing the intensity of reflexion.

The Comparison of the Theoretical and Observed Results.

16. The formula for the reflecting-power of a face states that

$$\frac{E\omega}{I} = \frac{N^2\lambda^3}{2\mu \sin 2\theta} \cdot F^2 \frac{e^4}{m^2c^4} \cdot \frac{1 + \cos^2 2\theta}{2} \cdot e^{-B \sin^2 \theta}.$$

Since all the quantities have been measured except F, we can calculate the absolute value of F for a range of values of θ .

If the effect of the electrons in the chlorine atoms be represented by F_{Cl} , and of those in the sodium atom by F_{Na} , then for reflecting-powers corresponding to points on the upper curve of fig. 2 we have

$$F = F_{Cl} + F_{Na};$$

for those corresponding to points in the lower curve

$$F = F_{Cl} - F_{Na}.$$

From the formula

$$F = \sqrt{\frac{E\omega}{I} \frac{mc^2}{e^2} \cdot \frac{2\mu^{\frac{1}{2}}}{N\lambda^{\frac{3}{2}}} \sqrt{\frac{\sin 2\theta}{1 + \cos^2 2\theta}} \cdot e^{+\frac{B}{2} \sin^2 \theta}},$$

where N is the number of molecules of NaCl in unit volume of the crystal.

Taking

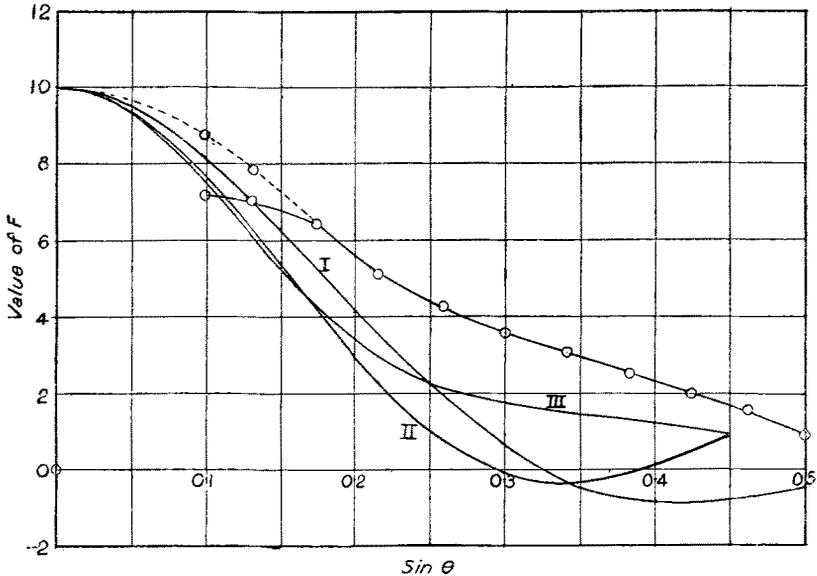
$$\begin{aligned} \frac{e}{m} &= 5.30 \times 10^{17}, & \mu &= 10.7, \\ e &= 4.77 \times 10^{-10}, & N &= \frac{1}{2} \left(\frac{1}{2.81 \times 10^{-8}} \right)^3, \\ c &= 3 \times 10^{10}, & \lambda &= 0.615 \times 10^{-8}, \\ & & B &= 4.12, \end{aligned}$$

this reduces to

$$F = 2143 \sqrt{\frac{E\omega}{I}} \cdot \sqrt{\frac{\sin 2\theta}{1 + \cos^2 2\theta}} \cdot e^{+2.06 \sin^2 \theta}.$$

The dotted curves in fig. 2 represent one-half the sum of the ordinates, and one-half the difference between the ordinates, of the upper and lower curves. From these

Fig. 4.

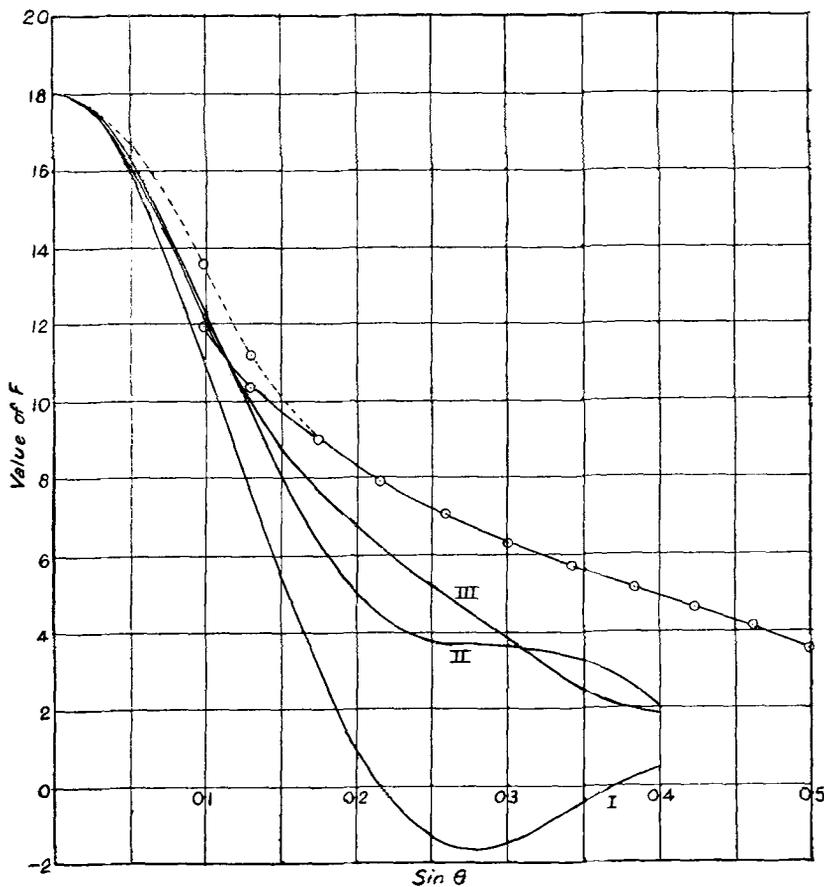


Values of F for Sodium.

The small circles indicate the observed values.

dotted curves the absolute value of F_{Cl} for chlorine, and F_{Na} for sodium, can be calculated directly. They are tabulated below, and the values are plotted against $\sin \theta$ in figs. 4 and 5.

Fig. 5.



Values of F for Chlorine.

The small circles indicate the observed values.

The values of F for Chlorine and Sodium are:—

Glancing-angle θ .	$\sin \theta$.	F_{Cl} .	F_{Na} .
$5^{\circ} 44'$	0.100	(11.67)	(6.90)
7 30	0.1305	10.11	6.88
10 0	0.1736	8.78	6.26
12 30	0.2164	7.72	4.98
15 0	0.2588	6.88	4.18
17 30	0.3007	6.14	3.47
20 0	0.3420	5.56	2.95
22 30	0.3827	5.00	2.41
25 0	0.4226	4.50	1.91
27 30	0.4617	4.01	1.49
30 0	0.5000	3.43	0.83

The angle between the scattered and incident beams is twice the glancing-angle θ .

It will be seen at once that the values of F are of the right order of magnitude. F should tend to a value 18 for chlorine, and 10 for sodium, as $\sin \theta$ approaches zero, assuming the atoms in the crystal to be ionized. The greatest value of F_{Cl} is 11.67, and of F_{Na} is 6.90, when $\sin \theta = 0.10$.

17. It now remains to take various models of the atom and see how the form of the function F calculated for these models agrees with that actually observed.

It is not intended here to lay much stress on the agreement between the calculated and observed forms of F for all values of θ . The object of the comparison is to demonstrate that any probable arrangement of electrons gives a close agreement between theory and experiment at small glancing-angles, and therefore to prove that the formula for the intensity of reflexion is very probably the true one.

The first atom model is one in which the electrons are supposed to be distributed uniformly throughout a sphere whose radius is 1.02×10^{-8} cm. in the case of chlorine, 0.67×10^{-8} cm. in the case of sodium.

In the second model the electrons are supposed to be arranged in a series of spherical shells. It is also assumed that, in considering the average effect of the atom, we may take the effect of the electrons in each shell to be equivalent to a uniform distribution of diffracting particles over the whole surface of the shell. The radii of these shells and the number of electrons in each are as follows :—

Chlorine.	No. of electrons.	Radius.	Sodium.	No. of electrons.	Radius.
1st shell ...	2	0.12	1st shell ...	2	0.40
2nd shell ...	8	0.41	2nd shell ...	8	0.67
3rd shell ...	8	1.02			

The diameters of the outer shells are those calculated by one of the authors* from crystal data.

In the third model the electrons are supposed to be arranged on shells of the same diameters as in the second model, but to be in oscillation about their mean positions along a line joining them to the centre of the atom with a total amplitude equal to their distance from the centre. This extreme case has been chosen to illustrate the effect of such an oscillation of the electrons on the form of the curve.

* W. L. Bragg, *Phil. Mag.* xl. p. 169, August 1920.

The values of F for chlorine and sodium calculated for these three types of atom model are plotted against $\sin \theta$ in figs. 4 and 5, the curves corresponding to the first, second, and third models being numbered I., II., and III. The measured values of F are shown for comparison as a series of small circles.

Of the three models chosen, the third type is the only one which gives diffraction curves of the same general shape as those actually observed. Both of the other models yield curves which have maxima and minima. The actual values of F fall off more slowly than do any of the theoretical curves, indicating that the distances of the electrons from the centre of the atom have been taken to be greater than their true value, both for sodium and chlorine.

The curve for sodium becomes nearly horizontal at some distance from the vertical axis, and the curve for chlorine shows a similar tendency. This must be ascribed to the fact that the reflecting power for small glancing angles is diminished by the increase of the absorption-coefficient, and that our values for (100) and (110) are too small. We know that this effect must exist, since measurement has shown that the absorption-coefficient increases by 15 per cent. in the neighbourhood of the angle corresponding to the (100) reflexion, and the increase at the exact angle of reflexion may be far greater than this. It is difficult to allow for this effect. In fig. 4 the circles on the dotted curve have been plotted so as to give that curve a more probable form passing through a maximum at 10. The actual values for $\sin \theta = 0.1$ and $\sin \theta = 0.13$ lie well below this dotted curve. In fig. 5 the two greatest values of F have been increased by the same amounts as for the other curve. This increase is of the order to be expected from the variation which was found to exist in the coefficient of absorption μ , but its exact value is, of course, merely conjectural.

There is another striking feature of the curves for the observed values of F . The curve for chlorine approaches the axis more slowly than that of the sodium. This is just the reverse of what would be expected from the relative dimensions of the two atoms, since we would expect the electrons to be on the whole at a greater distance from the centre in chlorine than in sodium. Here, again, the third type of atom chosen as a model gives results which agree most closely with those actually observed.

Although it is necessary to check the form of the curves for F by measurements on other crystals before drawing any

definite conclusions as to the arrangement of the electrons in the atom, the results so far obtained indicate that :—

(a) The formula for the amplitude of the wave scattered by each electron holds good, at any rate for small glancing-angles. The values for F when $\sin \theta = 0.1$ are of the order to be expected if the curves for chlorine and sodium have maxima at about 18 and 10 respectively.

(b) A uniform distribution of the diffracting points throughout a sphere cannot explain the form of the curve, for the theoretical form of the F function for such a model, which would coincide with the actual curve at small glancing-angles, meets and crosses the horizontal axis, whereas the observed curve falls away far more gradually with increasing θ .

(c) The general form of the curve makes it probable that as θ increases, the outer electrons for some reason become less and less effective in diffracting the X-rays. The result of assuming them to be in vibration is that the corresponding theoretical curve for F falls rapidly at first, and then very slowly, with increasing θ . It would seem necessary to make some such assumption, in order to obtain a theoretical curve approximating in form to that actually observed.

The electrons have been taken to be in radial vibration in the model, but this has only been done in order to obtain a theoretical expression in which the effect of the outer electrons falls away with increasing θ . It may be due to the electrons having a form such as that of the ring-electron assumed by Coster* in discussing diffraction by rings of connecting electrons in diamond.

Unless we suppose that the outer electrons become less effective with increasing θ , we must conclude, in order to explain the very gradual falling away of both curves, that the electrons are within a sphere of diameter smaller than is probable. By analogous reasoning, Debye (*Phys. Zeitschr. loc. cit.* p. 10) comes to the conclusion that in diamond the electrons are within a sphere of 0.43×10^{-8} cm. radius. This must not be excluded as impossible, or even very improbable; but if this is assumed, the difficulty remains of explaining why the curve for sodium approaches the horizontal axis more rapidly than that for chlorine, as if the former atom were the larger.

A uniform distribution of electrons throughout the volume of the atom will not account for the observed curves. A

* D. Coster, Proc. Roy. Acad. Sci. Amsterdam, xxi. No. 6, Oct. 1919.

model must be taken which has a greater concentration of electrons near the centre than that in the case of a uniform distribution.

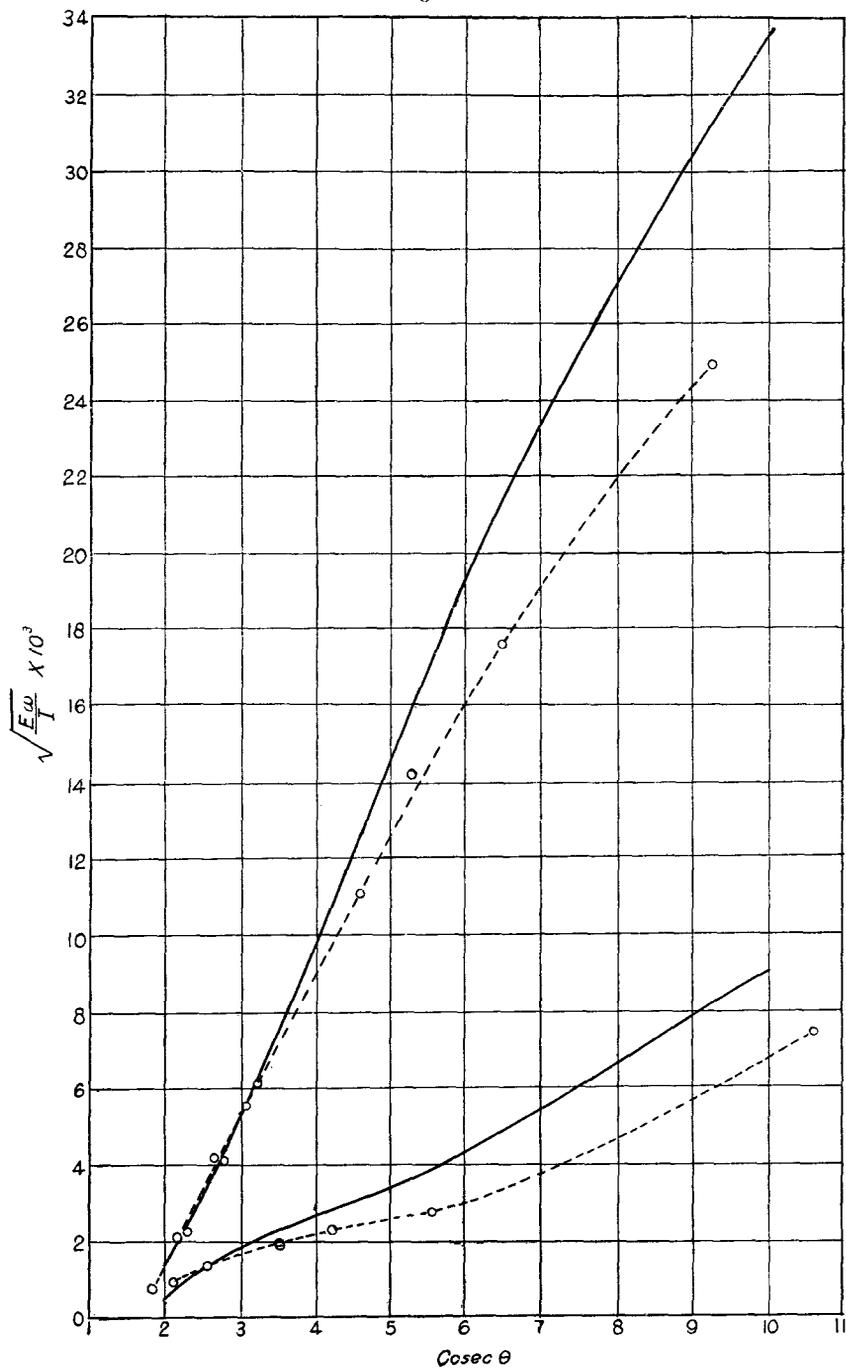
An important distinction must be made between the diffraction of X-rays by a crystal and the scattering of X-rays by an amorphous mass of material. In the formula

for the intensity of reflexion, the quantity $F \frac{e^2}{mc^2}$ represents

the amplitude of a polarized wave diffracted by a single atom in various directions. This amplitude must not be supposed to be necessarily the same as that which determines the amount of radiation scattered in various directions by an amorphous mass of the same atoms in a random arrangement. It is justifiable to consider the scattering by an amorphous substance as the summation of the intensities due to the separate atoms. If the electrons are in vibration, as has been supposed in the third atom model, their movements will be slow as compared with the frequency of the X-radiation. In the case of a single atom which is scattering the radiation, the arrangement of electrons in the atom at any one moment may be a random one, and the displacements may be so large and arbitrary that we may simply consider the scattering as due to a random arrangement of Z electrons, Z being the atomic number. If the amplitude of the scattered wave is $F' \frac{e^2}{mc^2}$, F' may be nearly equal to \sqrt{Z} for all angles of scattering, except for very small angles where all the electrons are in phase and "excess scattering" comes into play.

The factor F' will not be the same as the factor F in the case of the atoms of a crystalline substance. When examining the reflexion from a crystal, we have a large number of atoms diffracting waves which are exactly in phase with each other. $F \frac{e^2}{mc^2}$ is now the amplitude scattered by what we may term the "statistical" atom. In this case the movements of the electrons are allowed for by supposing that diffraction takes place, not at single electron points displaced from their mean positions, but from all over a certain region for each electron in which all its possible positions lie, due weight being given to each element of the region. This has been done in calculating F for the third atom model. This region may be so large that the effect of the outer electrons is practically zero for the higher orders, and this illustrates the essential difference between the two

Fig. 6.



Circles indicate observed values.

cases. F may become zero for certain values of θ . F' , depending on a random arrangement of electrons in each individual atom, will have no zero values.

18. The model of the atom which agrees most closely with the observed values of F for the large values of θ would be one of type 3, in which, however, all the figures given above for the distances of the electrons from the centre of the atom must be reduced in the ratio 3:2. Assuming this type of atom, it is possible to calculate from formula (6) the reflecting powers for any plane of the rock-salt crystal. The figures so calculated are shown by the continuous curves in fig. 6, and can be compared with those actually obtained (represented by points on the dotted curve). It is to be emphasized again that the comparison is not relative, but an absolute comparison of the reflecting powers actually observed and those calculated from the formula.

Summary.

The absolute values of the reflecting power for different faces of rock-salt have been measured. The reflecting power has been determined for eighteen glancing-angles over a range between $5^{\circ} 30'$ and $30^{\circ} 0'$.

The values obtained have been compared with those calculated from the theoretical formulæ for reflexion deduced by Darwin and Compton, and it has been shown that they afford strong confirmation of the accuracy of these formulæ.

The greatest care has been taken to make the measurements as accurate as possible, in order that they may serve as a basis for an analysis of the arrangement of the electrons in the atom. Possible arrangements are discussed.

In order to confirm the results, the effect of temperature on the intensity of reflexion must be more fully determined. The authors intend to make a series of determinations at liquid-air temperature, in order to be able to extrapolate to the values at absolute zero. It is hoped to extend the measurements over a wider range of angles at low temperatures.

It is further intended to repeat the experiments with KCl in order to check the formula in this case. Sylvine affords a simpler case for investigation than rock-salt, since the ions of potassium and chlorine will, in all probability, have a very similar structure.

The authors wish to acknowledge very gratefully the kind assistance given them by Dr. W. D. Coolidge, of the General Electric Company, Schenectady, to whom they are indebted for the gift of the Coolidge tube with which the investigations were carried out.