



I. The intensity of reflexion of X-rays by rock-salt.—Part II

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- I. *The Intensity of Reflexion of X-Rays by Rock-Salt.*—
Part II. By W. L. BRAGG, F.R.S., Langworthy Professor
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Reflexion Formulæ.

1. **I**N a previous paper in the Philosophical Magazine †, we described a series of measurements of the intensity of reflexion of X-rays by various planes of the rock-salt crystal structure. In analysing the results obtained, a formula for the intensity of reflexion was assumed which is essentially the same as that obtained by Darwin ‡ and Compton §.

Let the Intensity I_0 of a beam of homogeneous X-rays, at a given point, be defined as the total amount of radiation falling on an area of one square centimetre at right-angles to the direction of the beam. If a crystal-element of volume dV , supposed to be so small that absorption of the rays by the crystal is inappreciable, be placed so that it is bathed by the X-rays, and if it is turned with angular

* Communicated by the Authors.

† Bragg, James, and Bosanquet, *Phil. Mag.* vol. xli. p. 309 (March 1921).

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

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

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velocity ω through the angle at which some plane in it reflects the X-rays, the theoretical expression for the total quantity of radiation E reflected states that

$$\frac{E\omega}{I_0} = \frac{N^2\lambda^3}{\sin^2 2\theta} F^2 \frac{e^4}{m^2c^4} \frac{1 + \cos^2 2\theta}{2} e^{-B \sin^2 \theta} dV \quad \dots (1)$$

$$= Q dV. \quad \dots \dots \dots (2)$$

In this expression :

- N = No. of diffracting units per unit volume,
- λ = wave-length of X-rays,
- θ = glancing angle at which reflexion takes place,
- e = electronic charge,
- m = „ mass,
- c = velocity of light.

The constant B, which occurs in the Debye factor $e^{-B \sin^2 \theta}$, was assumed to be 4.12, on the basis of an experimental determination by W. H. Bragg*.

The contribution of a single electron is represented by the factor $\frac{e^4}{m^2c^4}$ in this formula.

The factor F depends on the number and arrangement of the electrons in the diffracting unit. At zero glancing-angle, it has a maximum value equal to the total number of electrons in the unit. As the glancing-angle increases, F falls off, owing to interference between the wave-trains diffracted by the separate electrons.

2. In the case of a large crystal, the linear absorption-coefficient, μ , of the rays in the crystal has to be taken into account in calculating the intensity of reflexion. Two special cases present themselves.

In the first place, a narrow beam of rays may be reflected from the face of a crystal cut parallel to the reflecting planes. In this case, if the intensity I of the incident beam is defined as the total amount of radiation falling on the crystal per second, calculation shows that

$$\frac{E\omega}{I} = \frac{Q}{2\mu}, \quad \dots \dots \dots (3)$$

where Q has the same significance as in equation (2). This is the case dealt with in our previous paper.

* W. H. Bragg, Phil. Mag. vol. xxvii. p. 897.

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The second case is illustrated by fig. 1. A beam of homogeneous X-rays is obtained by reflexion by the first crystal C. This beam is reflected by a second crystal

Fig. 1.

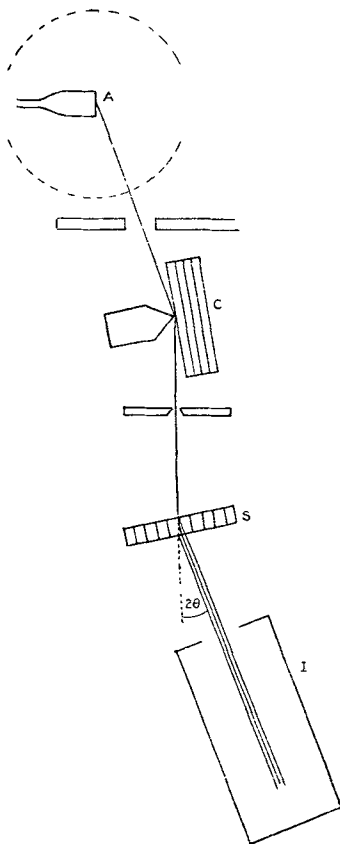


plate S, which is cut so that the reflecting planes are at right angles to its surface, and is turned with angular velocity ω .

In this case, all the rays have to traverse a depth $t_0 \sec \theta$ in the crystal, where t_0 is the thickness of the plate. Calculation shows that

$$\frac{E\omega}{I} = Q \cdot t e^{-\mu t}, \dots \dots \dots (4)$$

where $t = t_0 \sec \theta$, and Q and I have the same meaning as in equation (3).

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This expression has a maximum value $\frac{Q}{\mu e}$, when

$$t = \frac{1}{\mu} \dots \dots \dots (5)$$

The Determination of Q.

3. The object of the measurements described here and in the previous paper has been the determination of the absolute value of the quantity F over a range of glancing angles. F can be calculated if Q is determined experimentally, since the other quantities in the expression for Q are known.

In the former paper, Q was found by determining the reflecting power $\frac{E\omega}{I}$ for a crystal face, for which the formula $\frac{E\omega}{I} = \frac{Q}{2\mu}$ holds good. The linear absorption-coefficient μ was measured directly, by passing the homogeneous beam through plates of rock-salt of various thicknesses. It was pointed out, however, that in the case of reflexions at small glancing-angles, the value of μ determined in this way is certainly too low. When X-rays pass through a crystal in such a direction that the crystal reflects the rays, the absorption of the transmitted beam is greater than that for other directions. This effect was first noticed in the case of the diamond by W. H. Bragg*, and the effect also exists in the case of rock-salt †. Fig. 2 shows some determinations made with the apparatus arranged as in fig. 1, but with the ionization-chamber placed so as to receive the transmitted beam. The ordinates represent the strength of the transmitted beam, plotted for a number of crystal settings. It will be seen that the transmitted beam is reduced by 20 per cent. when the crystal is set at $4^\circ 50'$ ‡, the angle at which it reflects the X-rays most strongly.

The value of μ is therefore greater than its normal value ($10\cdot7$), for beams passing through the crystal in any direction in which they are reflected strongly, and the values of Q, calculated using the value $10\cdot7$ for μ , will

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

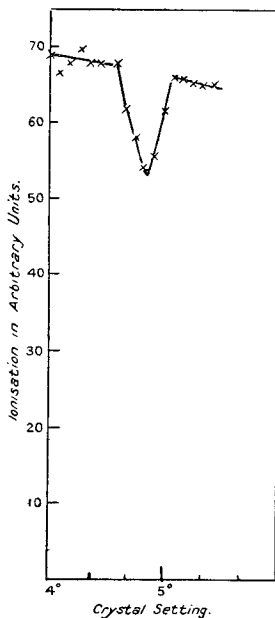
† *Cp.* the figures obtained by Rutherford and Andrade in their determination of the wave-length of γ -rays, *Phil. Mag.* ser. 6, vol. xxviii. no. 164, p. 263 (August 1914).

‡ Owing to a zero error in setting the crystal plate, this angle differs from the reflexion angle $6^\circ 17'$.

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be too small. This was pointed out in the former paper, but no allowance could be made for this effect, as the increase in μ had not been measured. This increase has

Fig. 2.



now been determined by a direct method, and in this paper the necessary corrections have been made to the values of Q , and so of F .

4. The increase in μ cannot be determined from a curve such as that of fig. 2. The beam falling on the crystal is not absolutely parallel; therefore it is not possible to set the crystal so that all the rays of the beam pass through at that angle for which absorption is a maximum. The apparent increase in μ indicated by the curve sets a lower limit to the effect, which may be considerably greater.

When reflexion takes place inside a crystal plate, as in fig. 1, the strength of the reflected beam is given by equation (4),

$$\frac{E\omega}{I} = Qt e^{-\mu t},$$

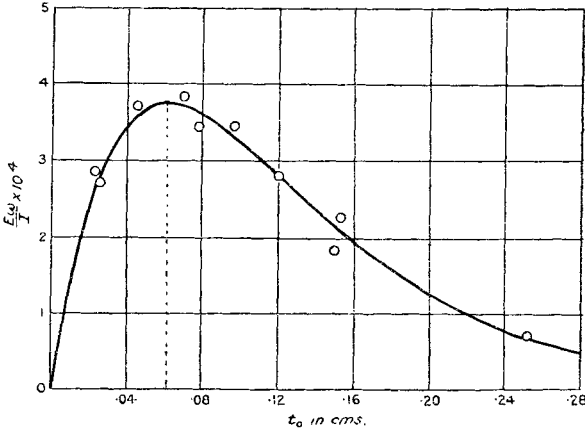
where $t = t_0 \sec \theta$.

In this equation, μ is the effective absorption-coefficient

appropriate to rays passing through the crystal at the reflecting-angle, since all rays entering the ionization-chamber have passed through at this angle.

If a number of plates of different thickness are taken, the intensity of reflexion measured in each case, and its value plotted against t_0 , the points should lie on a curve such as that in fig. 3. The circles in this figure are the

Fig. 3.



experimental determinations of the intensity of reflexion (I_{00}) for a number of plates varying in thickness from 0.2 to 2.5 mm., and a curve of the form $y = ate^{-\mu t}$ has been drawn so as to conform as closely as possible to the absolute experimental values.

The constants of this curve give the values of Q and μ . “ μ ” can be measured by noting the value of t at which the curve has its maximum, since at this point $t_0 \sec \theta = \frac{1}{\mu}$.

The maximum occurs at $t_0 = 0.610$ mm.; whence $\mu = 16.30$. The value measured directly is $\mu = 10.7$, so that the increase in μ at the reflecting angle is 52 per cent.

The value of Q is given by the equation

$$\left(\frac{E_{\omega}}{I}\right)_{\max.} = \frac{Q}{e\mu} \dots \dots \dots (6)$$

The mean of our experimental determinations gave

$$\left(\frac{E_{\omega}}{I}\right)_{\max.} = 3.65 \times 10^{-4}.$$

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Substituting $e = 2.718$, $\mu = 16.30$, we obtain

$$Q_{(100)} = 16.16 \times 10^{-3}.$$

From the value of $Q_{(100)}$ the absolute value of $(F_{Cl} + F_{Na})$ can be calculated, from equation (1).

This yields

$$F_{Cl} + F_{Na} = 20.10$$

at the glancing-angle $6^\circ 17'$, corresponding to reflexion from (100). In other words, when the rays are diffracted through an angle of $12^\circ 34'$, the total effect of the 28 electrons in a pair of sodium and chlorine atoms is reduced, by interference, to an effect 20.10 times that due to a single electron.

This value for $\frac{E\omega}{I}$ may be compared with that obtained by reflexion from the face (100) of rock-salt. Formulæ (3) and (4) state that

$$\frac{E\omega}{I} = \frac{Q}{2\mu} \text{ for reflexion at a face.}$$

$$\left(\frac{E\omega}{I}\right)_{\max.} = \frac{Q}{e\mu} \text{ for reflexion through a plate.}$$

The experimental value for the reflexion in the first case is

$$\frac{E\omega}{I} = 5.41 \times 10^{-4} *.$$

If the effective coefficient of absorption μ were the same for the crystal face and for the plate, the value of $\frac{E\omega}{I}$ for reflexion through the plate would be

$$5.41 \times 10^{-4} \times \frac{2}{e} = 3.99 \times 10^{-4}.$$

The experimental value, as stated above, is 3.65×10^{-4} . As will be seen, the value of μ for a crystal plate depends on the way in which the plate has been prepared, so that too much stress must not be laid on the numerical agreement; but this comparison serves to show that μ is approximately the same in both cases, and that the absolute values for the reflecting power of a small crystal element got by these different methods are in agreement with each other.

* This redetermined absolute value is lower than that given in the former paper (6.12×10^{-4}).

Effect of Imperfection in the Crystal.

5. The effect of irregularity in the crystal is discussed very fully by Darwin (Phil. Mag. April 1914, p. 685).

When X-rays are passing through a homogeneous element of the crystal at the correct angle for reflexion, the reflected beam is in its turn again reflected by the crystal. The beam which has undergone this double reflexion is parallel to the transmitted beam, but its phase differs from it by π , so that it tends to diminish it. The absorption of the transmitted beam is therefore greater than the normal absorption, for rays going through at any other angle. The greater the depth of the homogeneous element traversed by the X-rays, the greater is the increase in the effective absorption-coefficient. It is this which gives rise to the diminution of the transmitted beam when a crystal is set so as to reflect the rays. (See fig. 2.)

If the homogeneous elements in a composite crystal are small, so that the X-rays traverse a few planes only in each homogeneous fragment, the increase in the absorption-coefficient at the reflecting angle will be inappreciable. In this case the formulæ (3) and (4) will hold good for reflexion by a face and through a plate, μ being the normal coefficient of absorption. On the other hand, if the homogeneous fragments are so large that the increase in the absorption-coefficient is appreciable, the intensity of reflexion will be thereby diminished. The upper layers of each homogeneous element cut down the transmitted beam to an abnormal extent, so that the lower layers do not contribute so much to the total reflected beam. The more irregular the crystal, the stronger we should expect to find the reflected beam.

Darwin (*loc. cit.* p. 686) shows that, in a perfect crystal, all the radiation that can be reflected, is so, long before the depth is reached at which rays at a different angle are appreciably absorbed. Over a small range of angles reflexion is complete, the normal absorption-coefficient playing no part in diminishing the intensity. He takes the case of a crystal composed of homogeneous layers, each of sufficient depth to give the maximum possible amount of reflexion of the X-rays. The orientation of these successive layers is slightly different, so that the lower layers do contribute to the reflected beam when the X-rays fall on the crystal face at the angle which is appropriate to them, since rays reflected at them are able to pass through the upper layers.

To quote from Darwin's paper: "... We have strong experimental reason to believe that the crystals are even more imperfect than this. For when the reflexion is evaluated . . . it will be found that the second order of reflexion is as strong as the first—a result known to be untrue. This must be taken to indicate that the crystals are so badly twisted that their planes do not remain parallel even long enough to produce a single perfect reflexion."

We may therefore distinguish two degrees of irregularity in a crystal structure. On the one hand, the crystal may be so irregular that the absorption in each homogeneous fragment is entirely due to the conversion of the X-ray energy into cathode-ray energy. The absorption-coefficient will then be the same for rays passing through the crystal in any direction. This absorption-coefficient, which we will call μ_0 , can be found by direct measurement, and, in the case of these very irregular crystals, it is the correct coefficient to substitute in all the formulæ for reflexion. On the other hand, the homogeneous fragments, while still irregularly arranged, may each be sufficiently large for the extinction of the transmitted beam at the reflecting angle to be appreciable, in comparison with the reduction in intensity due to normal absorption. In this case, X-rays, passing through the composite crystal at such an angle that some of the fragments are reflecting, will be absorbed more strongly than rays passing through at other angles. There will be an increase in the effective absorption-coefficient. If the effective coefficient of absorption is now μ , we can state that

$$\mu = \mu_0 + \epsilon.$$

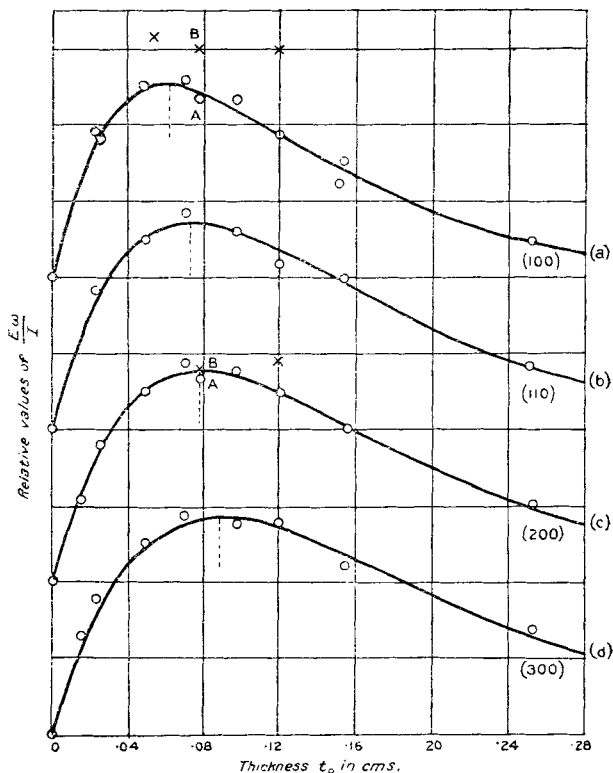
The coefficient ϵ will be called the "coefficient of extinction" of the X-rays in the crystal.

The Measurement of the Extinction-Coefficient.

6. Our experimental results show that the regularity in structure of rock-salt is such that ϵ is appreciable for strong reflexions, but is very small for the reflexions of high order. We have measured the normal coefficient of absorption μ_0 for rays passing through the crystal at any angle, and found it to be equal to 10.7. We have then determined the effective coefficient of absorption μ at the reflecting angle by the method described in paragraph 4. This has been done for the reflexions (100), (110), (200), and (300). The results are shown in figs. 4, 5, and 6.

In fig. 4 (a), for example, the intensities of the (100) reflexion through slips of various thicknesses are plotted against the thickness of the slip. Since the object of the experiment is to determine the position of the maximum of

Fig. 4.



the curve, the intensities are relative, that given by one of the slips whose thickness was 0.49 mm. being taken as standard in each case. The X-rays were not rendered homogeneous by a previous reflexion; but this does not affect the curves, since it was found that the allowance for "white" radiation of neighbouring wave-lengths was the same for all the slips. Figs. 4 (b), 4 (c), and 4 (d) are the curves for the reflexions (110), (200), and (300).

It will be seen that the maximum of the curve occurs at a progressively greater thickness in passing from one curve to that below it. Since the curves are placed in order of the

intensity of reflexion by the corresponding faces, this means that μ diminishes in passing to less intense reflexions (cf. equation (5)).

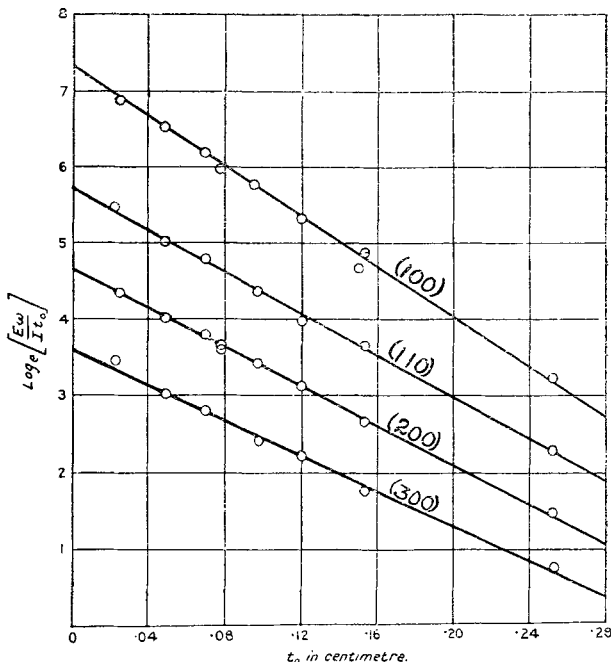
The effective coefficient μ can be calculated more readily by plotting the results as in fig. 5.

Since

$$\frac{E\omega}{I} = Q \cdot t_0 \sec \theta e^{-\mu t_0 \sec \theta},$$

$$\log_e \left(\frac{E\omega}{I t_0 \sec \theta} \right) + \text{cons} = -\mu t_0 \sec \theta.$$

Fig. 5.



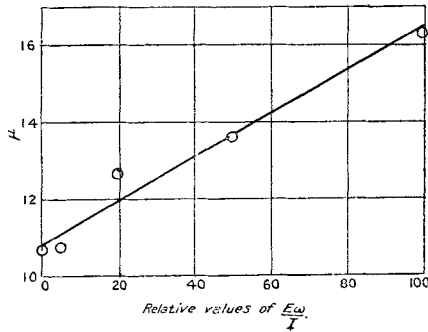
In fig. 5, $\log_e \left(\frac{E\omega}{I t_0} \right)$ is plotted against t_0 . The slope of the curve then gives μ directly, allowing for the factor $\sec \theta$. The results are tabulated below.

Reflexion.	Effective coefficient, μ .	Extinction coefficient, $\epsilon = \mu - \mu_0$.	Intensity of reflexion in arbitrary units.
100	16.30	5.60	100
110	13.60	2.90	50.5
200	12.66	1.96	19.90
300	10.72	0.02	4.87

Normal coefficient of absorption, 10.70.

The intensity of reflexion is that from a rock-salt face, determined as described in a previous paper. It would be perhaps more accurate to compare ϵ with Q , but the comparison has been made in this way in order to facilitate the application for the correction for ϵ to our results.

Fig. 6.



In fig. 6, μ is plotted against the intensity of reflexion. It will be seen that the points, with the exception of (200), lie on a straight line which cuts the y axis at 10.80, a value very close to the normal coefficient of absorption 10.70. We may therefore assume, without introducing any large error, that we can calculate for any reflexion from the formula

$$\mu = \mu_0 + kI, \quad \dots \dots \dots (6)$$

the value of k being determined from the slope of the curve.

The Relation between Regularity of Structure and Intensity of Reflexion.

7. An interesting point was observed in measuring the intensities of reflexion through the crystal plates. In the case of certain plates, the intensity was found to be abnormally high. In fig. 4, the points corresponding to these plates which gave the high values are denoted by crosses instead of circles. This abnormality is much more marked in the case of the (100) reflexions than for those which are less intense.

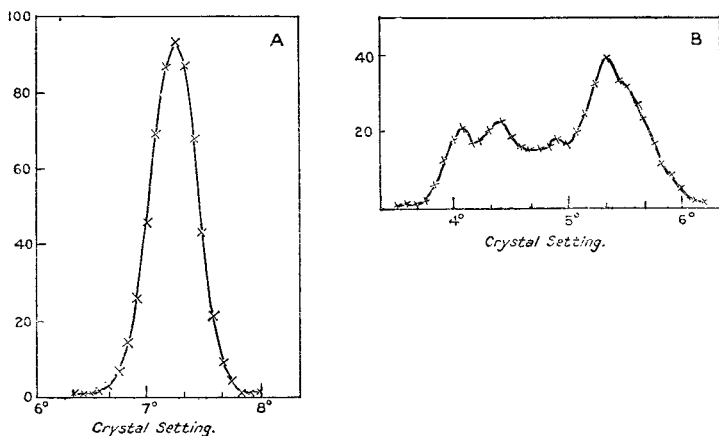
This effect was traced to a greater irregularity in structure of the crystal plates. In every case in which the value came to be abnormally high, it was found that the crystal gave reflexion over a wide range of angles. It was also found that this irregularity could be produced artificially by

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grinding the crystal plate on coarse emery-paper, plates prepared in this way giving a reflexion over a long range, and abnormally high values for the intensity.

Fig. 7 will illustrate this effect. A single crystal plate was cut across in two equal portions, and from these two plates of the same thickness (0.78 mm.) were prepared.

Fig. 7.



A. Intensity of reflexion 0.93.
 B. " " " 1.20.

The first plate was rubbed down on ground glass with water so as to avoid all mechanical strain. The second was ground down on coarse emery-paper. The curves of fig. 7 show the amount of radiation reflected by the plates over a range of a few degrees. The first gives a narrow maximum, showing that the structure is regular; the second, an irregular curve over a much larger angle. The intensities of reflexion in terms of that of the standard plate were 0.93 and 1.20 respectively for the (100) reflexion, showing how much greater the intensity is for the irregular than for the regular crystal. In fig. 4, the points corresponding to these two crystals are denoted by "A" and "B." The irregularity of the points in fig. 4 is therefore to be ascribed to slight differences in the perfection of structure of the crystal plates, and the fact that they can be made to fit with a fair approximation to a $te^{-\mu t}$ curve shows that all the plates have much the same degree of irregularity of structure. As stated above, the few abnormal points were all found to correspond to crystals which for some reason had an exceedingly irregular structure.

Corrected Values for F_{Cl} and F_{Na} .

8. From the curve in fig. 3, we can calculate the absolute value of $Q_{(100)}$ and therefore the value of $F_{Cl} + F_{Na}$ for rays diffracted through an angle of $12^\circ 34'$ (twice the glancing angle for reflexion from this face). This, as stated above, comes to be 20·10. Since allowance has been made for the extinction-coefficient, this is now an accurate determination. In the previous paper the value of $F_{Cl} + F_{Na}$ was obtained by measurements on the intensity of reflexion from a face, taking μ to be 10·7. These measurements give $F_{Cl} + F_{Na} = 16\ 68$. The difference between the two values shows the effect of allowing for the extinction-coefficient.

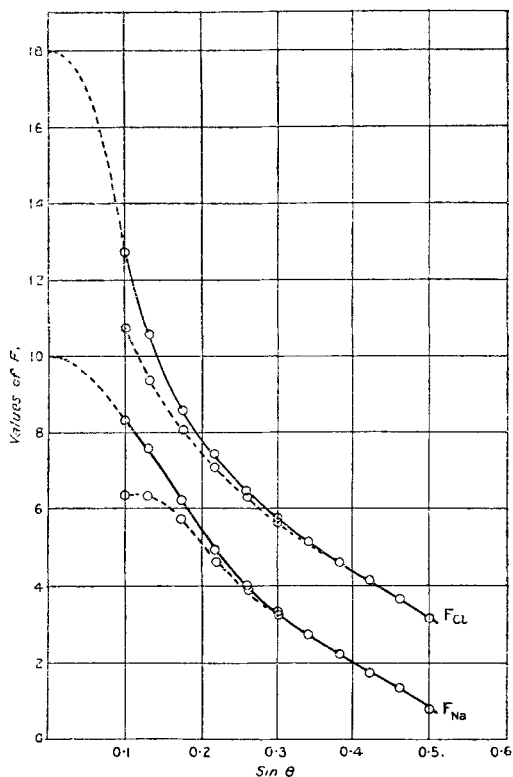
We now have sufficient data to calculate the correct values for F_{Cl} and F_{Na} over the whole range of angles which has been examined. When the glancing angle is large, the reflexion is so weak that the extinction-coefficient may be neglected. The values for $F_{Cl} - F_{Na}$, given in the previous paper, therefore hold good, allowance being made for the differences due to the re-determination of the absolute intensity of reflexion mentioned above. On the other hand, the measurements described in paragraph 4 make possible the absolute determination of $F_{Cl} + F_{Na}$ for the angle of most intense reflexion, by a method which is independent of any uncertainty as to the value of μ . We can therefore estimate to what extent the (100) reflexion is diminished by the extinction-coefficient in the case of the measurements with a crystal face, and using the formula $\mu = \mu_0 + kI$ we can raise the values of the (110), (222), (200), and other strong reflexions, so as to bring them into line with the new data. The values for F_{Cl} and F_{Na} so obtained are tabulated below, and plotted in fig. 8.

Values of F_{Cl} and F_{Na} .

$\sin \theta$.	$F_{Cl} + F_{Na}$.	$F_{Cl} + F_{Na}$. (Corr. for ϵ).	$F_{Cl} - F_{Na}$.	F_{Cl} .	F_{Na} .	2θ .
·1000	17·12	21·05	4·40	12·72	8·32	$11^\circ 24'$
·1305	15·66	18·21	2·98	10·59	7·61	15°
·1736	13·86	14·89	2·32	8·60	6·23	20°
·2164	11·71	12·35	2·53	7·44	4·91	25°
·2588	10·19	10·51	2·49	6·50	4·01	30°
·3007	8·86	9·11	2·46	5·78	3·37	35°
·3420	7·84	7·90	2·40	5·15	2·75	40°
·3827	6·83	6·83	2·39	4·61	2·22	45°
·4226	5·91	5·91	2·39	4·15	1·76	50°
·4617	5·07	5·07	2·33	3·70	1·37	55°
·5000	3·92	3·92	2·40	3·16	0·76	60°

The figures for $(F_{Cl} + F_{Na})$ in the third column have been corrected for the extinction-coefficient. The intensity of reflexion is so weak in the case of the planes with odd indices that the extinction-coefficient is negligible, and the values for $F_{Cl} - F_{Na}$ need no correction. In the fifth and sixth columns are given the corrected values for F_{Cl}

Fig. 8.



The amplitude of the waves diffracted through an angle 2θ by the Chlorine and Sodium atoms are expressed in terms of that scattered by a single electron. The wave-length λ of the rays is equal to 0.616×10^{-8} cm.

and F_{Na} . The angle of 2θ through which the rays are diffracted is given in the last column.

In fig. 8, both the former values, and those corrected for the extinction-coefficient, are shown. The maximum values for F at $\theta=0$ would be expected to be 10 for sodium, 18 for chlorine, assuming both atoms to be ionized. The general

trend of the corrected curves is towards maxima in the neighbourhood of these values, whereas the uncorrected curves drop too quickly as they approach the y axis, this being particularly noticeable in the curve for sodium. The fact that the corrected curves tend towards these maxima confirms the accuracy, both of the experimental determinations and of the assumptions which have been made in the calculations.

SUMMARY.

The object of these experiments has been the determination of the amplitude of the wave diffracted in various directions by the chlorine and sodium atoms when homogeneous X-rays of unit amplitude fall on these atoms. This amplitude is expressed in terms of that diffracted by a single electron, the results for chlorine and sodium being shown in fig. 8.

The following formulæ for the intensity of reflexion have been used :—

$$\frac{E\omega}{I} = \frac{Q}{2\mu}$$

for reflexion at a face ;

$$\frac{E\omega}{I} = Qt_0 \sec \theta e^{-\mu t_0 \sec \theta}$$

for reflexion at glancing angle θ through a crystal plate of thickness t_0 cut perpendicular to the reflecting planes.

Q is given by the formula

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

Allowance has been made for the increase in the effective coefficient of absorption μ , in the case of strong reflexions, due to the existence of the "extinction-coefficient."

The amplitude of the wave diffracted by a pair of sodium and chlorine atoms is given by the factor $F \frac{e^2}{mc^2}$. The two atoms diffract waves which are in phase with each other for all planes except those with wholly odd indices, when the waves are opposed in phase. By measuring the intensity of reflexion for both types of plane, it has been possible to assign values to the amplitude diffracted by the chlorine and sodium atoms separately over a range of angles from 10° to 60° .

The results confirm the accuracy of the measurements and of the theoretical assumptions, since they indicate the correct number of electrons in the atoms of chlorine and sodium.

An estimate of the distribution of the electrons around the nuclei of these atoms may be obtained by analysis of the results, since the forms of the diffraction curves are dependent on the arrangement of the electrons. We hope to publish this analysis shortly.

Manchester University,
20th March, 1921.

II. *Triple Pendulums with Mutual Interaction and the Analogous Electrical Circuits.*—II. *By Prof. E. H. BARTON, F.R.S., and H. M. BROWNING, M.Sc., F.Inst.P.**

[Plate I.]

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I. INTRODUCTION.

THE present paper deals with the theory and confirmatory experiments carried out with the modified apparatus referred to in the previous paper on this subject †. All three pendulums in use still have bobs of equal masses as before, and their total lengths are still all equal. But a rather simpler system of suspension is here adopted which both facilitates the experiments and simplifies the theory. This mechanical form still secures a degree of generality equivalent to that which could be obtained with three equal electrical circuits in which each pair had a different mutual induction.

The equations of motion for these pendulums lead to a general solution, showing that each pendulum may execute

* Communicated by the Authors.

† See "Triple Pendulums," etc. I., *Phil. Mag.* Nov. 1920.