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## LXXVIII. The theory of X-ray reflexion. Part II

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- LXXVIII. The Theory of X-Ray Reflexion. Part II. By C. G. DARWIN, M.A., Lecturer in Mathematical Physics in the University of Manchester\*.
- 1. TN the First Part of this paper † formulæ were obtained giving the intensity of reflexion of X-rays by a crystal, and by a discussion of the results of experiment; it was concluded that a factor had been neglected which in fact must be of some importance, and that to represent the case at all accurately an improved theory was necessary. It was indicated that the factor to be included is the influence of the vibration of each atom on that of the others. If this is not done, cases will present themselves in which the conservation of energy is apparently violated. The experiment which was discussed is one of these cases. In the present paper this mutual influence is allowed for and a revised formula is found for the reflexion from a crystal. Comparison with experiment shows that the new formula is no better able than the old to account for the observed strength of reflexion. It appears, however, that this may be attributed to the fact that in all crystals there is a considerable amount of distortion, so that there are a great many separate small regions in which reflexion takes place. As a consequence of this fact it will be deduced that, constant factors apart, the old reflexion formulæ may be allowed to stand. We shall first of all deal with the reflexion from a perfect crystal.

#### 2. Reflexion from one Plane.

In the earlier work the procedure was first to calculate the reflexion from a single plane of atoms and then to combine the effects of the different planes. The amplitude of the reflexion from one plane was represented by a coefficient -iq, where q is made up in the following way. A wave of unit amplitude and length  $2\pi/k$  falls on an atom. Let  $f(\psi, k) e^{-ikr}/r$  be the amplitude of the wave it scatters, measured at a distance r in a direction inclined at angle  $\psi$  to the direction of the incident wave. In addition to  $\psi$  and k, f will depend on the direction of polarization of the incident wave. Let N be the number of atoms per c.c. (for the present we shall suppose them all identical) and a the distance between successive planes; then Na is the areal density

<sup>\*</sup> Communicated by Prof. Sir Ernest Rutherford, F.R.S.

<sup>†</sup> Darwin, Phil. Mag. vol. xxvii. p. 315 (1914). † Loc. cit. p. 331.

of atoms in a plane. Let  $\theta$  be the glancing angle between the direction of incidence and the plane. Then\*

$$q = \frac{2\pi \operatorname{N} a f(2\theta, k)}{k \sin \theta}. \quad . \quad . \quad . \quad (1)$$

As long as the atoms are supposed not to influence one another's motion, every plane however deep in the crystal scatters the same amount of radiation, and if no allowance were made for the absorption of the transmitted wave the reflexion would become indefinitely large. Now an atom in rocksalt may be supposed to have about 10 electrons, and so

f is probably about 10  $\frac{e^2}{mc^2}$ †. Taking N=4.50×10<sup>22</sup>,

 $a=2\cdot81\times10^{-8}$  and reflexion in the first order so that  $k\sin\theta=\pi/a$ , we find that q is about  $2\times10^{-4}$ . According to the assumptions of the earlier paper, the amplitude of the wave is reduced by absorption in passing through one plane by an amount  $\frac{1}{2}\mu a \csc\theta$ , and for soft X-rays this is  $4\times10^{-6}$ . Thus we should expect reflexion to be much more efficient than absorption in extinguishing the transmitted wave. Indeed, we shall find that over a certain small range of angles of incidence the reflexion is practically complete and does not depend on the absorption coefficient.

In the earlier paper the coefficient of reflexion q was calculated by considering a spherical wave coming from a point source. In view of the greater complexity of the present problem it is more convenient to deal with plane waves, and we must therefore first observe that the evaluation of q could have been done equally well with these. It is only necessary to find the amplitude of reflexion at a point so distant from the crystal, that in the principal part of the field the phases of waves from adjacent atoms are sensibly the samethis permits the summation to be replaced by an integration -while yet the point is not so distant that the crystal has to be regarded as finite—this introduces the Fresnel factors in the integral and makes it converge. There can be little doubt that the procedure gives the right value, though it is not of course mathematically rigorous. It has the great advantage that the formulæ do not involve the exact number of atoms in the whole plane, which is obviously quite irrelevant to the final results.

The formula for q cannot be quite general, if f is supposed to depend only on the atom itself. For we might then make

<sup>\*</sup> Loc. cit. pp. 319-320. † Loc. cit. pp. 326-329.

Na, the areal density of atoms, so large that the conservation of energy would be violated. In nature this is of course obviated by the fact that if the atoms are too closely crowded together, the wave from each will influence the others. We have seen that for rocksalt q is only of the order  $10^{-4}$ , so that the conservation of energy is in no danger, and we shall continue to use q as it stands. The direct calculation of the influence of all the atoms on one of their number leads to a double series of some complexity. I am informed by a friend\* to whom I referred the matter, that the series does in general converge (which was not at all obvious at first sight), but that the question is quite a difficult problem in pure mathematics. I do not give the form of the series as no use is to be made of it. It will be found that the forces from the other atoms exert an effect like an addition to the radiation term in the vibration of an electron. radiation term hardly influences the amplitude of vibration of an electron under the influence of X rays (except in the case of resonance), and so we may conclude that the mutual influence of the atoms in a plane may be neglected.

When a wave falls on a single plane of atoms, besides the directly reflected wave there are others scattered. Thus there will be a wave given off in any direction for which a line of atoms are in phase together, while the next parallel line is a phase  $2\pi$  behind. These diffracted waves are destroyed by the operation of the other planes of the crystal; but there remains a wave scattered in the same direction as the transmitted beam. The amplitude of this wave is given by  $-iq_0$ , where  $q_0$  is obtained from q by replacing  $f(2\theta, k)$  by f(0, k). As was shown in the earlier work, it is the wave  $-iq_0$  which gives rise to the refractive index  $\dagger$ .

### 3. Combination of Planes.

The difficulty of the problem of allowing for the mutual influence of the atoms in one plane is the complete absence of phase relations between the waves arriving at one atom from the rest, but this is also the reason why it is justifiable to neglect it. For the combination of all the planes the matter is quite otherwise. Here, when the radiation is at the angle of reflexion, all the waves reflected from the successive planes are in phase together, and they must be supposed to give rise to a secondary reflexion, which

<sup>\*</sup> Mr. G. H. Hardy, F.R.S., of Trinity College, Cambridge, to whom I must express my thanks for his kind interest in the question. † *Loc. cit.* p. 320.

contributes a component in the direction of the transmitted wave. It was this secondary reflexion that was neglected in the earlier work, on the assumption that the radiation scattered by one atom had no effect on the others. The recombination into a single wave of the wavelets from the atoms in one plane will not be very complete in the short distance between adjacent planes, but the error in assuming it complete will not be systematic. Moreover, the mutual influence of two planes alone is very small; it is only the cumulative effect that is important. We thus have a problem very similar to that of the Fabry-Perrot étalon, only with an infinite number of parallel equidistant plates.

We shall suppose that independently of the scattering of the atoms there is also a small absorption. Thus if a plane wave  $e^{ik(Ct-x\cos\theta+z\sin\theta)}$  falls on a single plane of atoms, the reflected wave is  $-iq \ e^{ik(Ct-x\cos\theta+z\sin\theta)}$  and the transmitted wave is  $(1-h-iq) \ e^{ik(Ct-x\cos\theta+z\sin\theta)}$ . The term h represents the absorption and may be taken as  $\frac{1}{2} \mu a \csc\theta$ .

We consider a crystal composed of atoms of a single substance arranged in planes at distance a. Let  $T_r$  represent in amplitude and phase the total transmitted wave just above the (r+1)th plane,  $S_r$  the total reflected wave in the same position. Then  $T_0$  is the incident wave, and  $S_0$  the reflected wave.  $S_r$  is derived from two components, the part of  $T_r$  reflected by the r+1th plane and the part of  $S_{r+1}$  transmitted through it. The latter must be multiplied by a phase factor  $e^{-ika\sin\theta}$  to give its value just below the r+1th plane instead of just above the r+2th. Thus

$$S_r = -iqT_r + (1-h-iq_0) e^{-ika\sin\theta} S_{r+1}$$
.

Again  $T_{r+1}$  is made up of the part of  $T_r$  transmitted through the r+1th plane and the part of  $S_{r+1}$  reflected by it. Putting in the proper phase factors we have

$$\mathbf{T}_{r+1} e^{ika\sin\theta} = (1 - h - iq_0) \mathbf{T}_r - iq e^{-ika\sin\theta} \mathbf{S}_{r+1}.$$

If we eliminate the S's from these difference equations we obtain

$$\begin{split} (1-h-iq_0)\,e^{-ika\sin\theta}\,(\mathbf{T}_{r-1}+\mathbf{T}_{r+1}) = & \left[1+q^2\,e^{-2ika\sin\theta}\right. \\ & \left. + (1-h-iq_0)^2\,e^{-2ika\sin\theta}\right]\mathbf{T}_{r}, \end{split}$$

and the solution is given by  $T_r = T_0 x^r$ , where x is the root of

$$(1-h-iq_0) e^{-ika\sin\theta} \left(x+\frac{1}{x}\right)$$

$$=1+q^2 e^{\frac{-2ika\sin\theta}{2}}+(1-h-iq_0)^2 e^{-2ika\sin\theta}. \qquad (2)$$

The product of the roots of this equation is unity and that one is to be taken which makes |x| < 1. Otherwise the intensity would increase with r. If we substitute back with this solution we find

$$S_r = T_0 \frac{-iqx^r}{1 - x e^{-ika\sin\theta} (1 - h - iq_0)},$$

and in particular

$$\frac{S_0}{T_0} = \frac{-iq}{1 - x \, e^{-ika\sin\theta} \, (1 - h - iq_0)}. \quad . \quad . \quad (3)$$

This expression holds for any angle of incidence.

We shall now approximate by allowing for the fact that q,  $q_0$ , h are small and by supposing that the incident wave is very nearly at the angle of best reflexion. Then  $\theta$  is very near  $\phi$ , where  $ka\sin\phi=n\pi-q_0$ . The presence here of  $q_0$  represents the shift in the angle of best reflexion due to the refractive index, as explained in the former paper\*. We have then

$$ka\sin\theta = n\pi - q_0 + v$$

where

$$v = ka \cos \phi (\theta - \phi)$$
. . . . . (4)

To the degree of approximation needful we have

$$e^{-ika\sin\theta} = (-)^n (1 + iq_0 - iv),$$

so that

$$\frac{S_0}{T_0} = \frac{-iq}{1 - (-)^n x (1 - h - iv)}$$

and x is that root of

$$(-)^n(1-h-iv)\left(x+\frac{1}{x}\right)=1+q^2+(1-h-iv)^2,$$

for which |x| < 1.

The roots of this equation are very nearly  $(-1)^n$ , so to solve it we put  $x=(-)^n(1-\epsilon)$ .

Substituting in the equation we have

$$(1-h-iv)(2+e^2) = 1+q^2+(1-h-iv)^2,$$

so that

$$\epsilon = \sqrt{\{q^2 + (h+iv)^2\}}.$$

The ambiguity is to be determined so that the real part is positive. Thus

$$\frac{S_0}{T_0} = \frac{-iq}{h + iv + \sqrt{\{q^2 + (h + iv)^2\}}}. \quad . \quad . \quad (5)$$

\* Loc. cit. p. 318.

The earlier paper was written under an assumption which may be seen to be equivalent to taking q much smaller than

h, so that 
$$\frac{S_0}{T_0} = \frac{-iq}{2(h+iv)}$$
\*. It is quite possible to evaluate

the expressions required with any values of h and q, but the formulæ involve elliptic functions, so that their numerical values are not easy to see. Now as we saw q is probably about  $2 \times 10^{-4}$  while h is only  $4 \times 10^{-6}$ , so that not much error is introduced by supposing h/q negligible. It would not, however, have been permissible to have supposed that h vanished at the beginning of the work, because if this were done it would be found that for some angles the reflexion tends to no definite value as the number of planes tends to infinity.

In discussing the ambiguity of  $\sqrt{\{q^2 + (h+iv)^2\}}$  when h vanishes it will be convenient to suppose q positive. We do not know whether this is true, but if q is really negative the modification is very simple. When -q < v < q we have simply  $\sqrt{(q^2 - v^2)}$ , the positive square root being taken. When v > q we must write the expression in the form  $\pm i\sqrt{(v^2 - q^2 - 2ivh)}$ , and if the radicle is expanded it will be seen that the proper value is  $+i\sqrt{(v^2 - q^2)}$ . Without the presence of h this could not have been determined. Similarly when v < -q we have to take  $-i\sqrt{(v^2 - q^2)}$ . Thus the amplitude of reflexion is

Transfer is
$$-\frac{q}{v-\sqrt{(v^2-q^2)}} \quad \text{for } v < -q$$

$$-\frac{q}{v-i\sqrt{(q^2-v^2)}} \quad \text{for } -q < v < q$$

$$-\frac{q}{v+\sqrt{(v^2-q^2)}} \quad \text{for } q < v$$

$$(6)$$

To express the intensity of reflexion we take the moduli of the squares of these quantities. In the middle region this is unity and reflexion is perfect. Now  $v=ka\cos\phi(\theta-\phi)$ , so this is the region

$$\theta = \phi \pm s$$
, where  $s = q/ka \cos \phi$ .

If we take the reflexion in the first order of rocksalt for the platinum radiation  $\beta$  ( $\lambda=1.11\times10^{-8}$  cm.) we find s=3''. For the second order it is about half this.

On account of the perfect reflexion the transmitted wave

\* This is equivalent to one of the equations on p. 322 of the earlier paper.

is rapidly extinguished. Since  $\frac{T_r}{T_0} = x^r = (-)^{nr} e^{-r\sqrt{(q^2-v^2)}}$  it follows that at a depth z in the crystal the intensity is only  $\exp{-2\frac{z}{a}\sqrt{q^2-v^2}}$  of its value at the surface, and so we may speak of an extinction coefficient

$$\frac{2}{a}\sqrt{q^2-v^2}. \qquad (7)$$

Averaged across the whole region of perfect reflexion this gives a coefficient

$$4q/\pi a$$
. . . . . . . . . (8)

For the value of q which we have been using, this gives about 8000, whereas the absorption coefficient, taken as  $\mu$  cosec  $\phi$ , is for the platinum rays only 300. Thus the extinction is complete long before the rays going in a slightly different direction are appreciably absorbed. This fact is important in explaining the reflexion from an ordinary imperfect crystal.

#### 4. Spherical Wave and Effect of a Slit.

We have so far only dealt with plane waves. A spherical wave can be made by compounding together in an integral a set of equal plane waves going in all different directions. If we put in the reflexion factor for each of these plane waves, we obtain an integral representing the diffraction pattern of the reflected beam. At the distances at which experiments are usually made this pattern would be of some complexity. Since it would never be observed in practice on account of the finite area of any actual source and the imperfection of the crystal, it is unnecessary to discuss it. To find the whole intensity of reflexion we may examine the effect at infinity. Here the waves are all plane, so that we can apply the formulæ (6) direct. If we take the intensity at a point at glancing angle  $\phi + \epsilon$ , we have  $v = ka \cos \phi \cdot \epsilon$ . So, making use of the abbreviation  $s=q/ka\cos\phi$ , we find as the intensity at a great distance  $\rho$ 

$$\frac{1}{\rho^{2} \cdot (\epsilon - \sqrt{\epsilon^{2} - s^{2}})^{2}} \quad \text{when } \epsilon < -s$$

$$\frac{1}{\rho^{2}} \cdot 1 \quad \text{when } -s < \epsilon < s$$

$$\frac{1}{\rho^{2} \cdot (\epsilon + \sqrt{\epsilon^{2} - s^{2}})^{2}} \quad \text{when } s < \epsilon$$
(9)

Suppose that the intensity of the monochromatic incident beam at a distance R is  $I/R^2$ , and that the whole effect is observed in an instrument having a slit of length l and sufficient breadth to include the whole beam. Then the instrument will measure

$$I_{\rho}^{l} \left\{ \int_{-\infty}^{-\epsilon} \frac{s^{2} d\epsilon}{(\epsilon - \sqrt{\epsilon^{2} - s^{2}})^{2}} + 2s + \int_{s}^{\infty} \frac{s^{2} d\epsilon}{(\epsilon + \sqrt{\epsilon^{2} - s^{2}})^{2}} \right\},\,$$

which reduces to

$$I\frac{l}{\rho}\frac{8}{3}s$$
.

If we put in the value of s this reduces to

$$I \frac{l}{\rho} \frac{8}{3\pi} N \left| f(2\phi, k) \right| \lambda^2 \csc 2\phi. \quad . \quad (10)$$

Of the two polarized components that for which the electric vibration is in the plane of incidence has in its f a factor  $\cos \phi^*$ . Introducing this and also the temperature factor  $\dagger$  we have

$$I\frac{l}{\rho} \frac{8}{3\pi} \frac{1 + |\cos 2\phi|}{2} N |f| e^{-\frac{1}{2} \frac{kT}{\sigma a^2} (2n\pi)^2} \lambda^2 \csc 2\phi. \quad (11)$$

As in the earlier paper, we next find the result of limiting the incident beam by a slit. To describe the diffractive effect of a slit it is usual to imagine that every point of the slit gives out a spherical wave, and that the separate waves are coherent. For our purpose it is better to resolve the waves from the slit into a set of plane waves. The amplitude of any of these waves is given by a Fresnel integral taken between the proper limits. The amplitude of reflexion in any direction will be determined by the product of this Fresnel integral and the reflexion factor for the corresponding direction. Let the slit be at a distance r from the source and of angular breadth  $\sigma$ . Then we saw in the earlier paper‡ that the intensity opposite the centre of the slit has practi-

cally its full value when  $\sigma^2 \frac{kr}{\pi} = 6$ , and so if the slit is placed symmetrically with regard to the reflexion, the intensity of

reflexion has its full value at the central point. If we take  $k=10^9$  and r=30 cm. this gives  $\sigma=5''$ . Now we know that

† Loc. cit. p. 323.

<sup>\*</sup> Loc. cit. p. 323.

<sup>†</sup> Loc. cit. p. 325. The expression used is not applicable to low-temperatures.

the breadth of the good reflexion is 6" and if we calculate the intensity corresponding to a slit of the supposed breadth at 3" from the centre, we find it to be a little less than that at the centre. So we conclude that a slit of 5" will hardly give full reflexion. As the breadth is only required for the purpose of a rough estimate, we shall take it at 8". This is the narrowest slit which can be used if the intensity of reflexion of monochromatic rays is not to suffer.

#### 5. White Radiation.

We next find the intensity of reflexion of white radiation. Let the intensity of the incident radiation at distance R from the source be  $\frac{1}{R^2} \int_0^\infty u dk$ . Reflexion only occurs for values of k near those which satisfy the equation  $ka \sin \theta = n\pi - q_0$ . These values will be denoted by  $k_n$ . A value of k near  $k_n$  can be expressed in the form  $k = k_n \left(1 + \frac{x}{n\pi}\right)$ . The centre of best reflexion for k is at an angle  $\theta - \eta$  where

$$ka \sin (\theta - \eta) = n\pi - q_0$$

and it follows from this that  $x=ka\cos\theta \cdot \eta$ , so that x is the same as v in (6). The intensity of reflexion thus is

$$\frac{1}{\rho^{2}} \sum_{n} u_{n} \frac{k_{n}}{n\pi} \left\{ \int_{-\infty}^{-q} \frac{q^{2}}{(x - \sqrt{x^{2} - q^{2}})^{2}} dx + 2q + \int_{q}^{\infty} \frac{q^{2}}{(x + \sqrt{x^{2} - q^{2}})^{2}} dx \right\},$$
or
$$\frac{1}{\rho^{2}} \sum_{n} u_{n} \frac{k_{n}}{n\pi} \frac{8}{3} q.$$

If we introduce the value of q and the factors for polarization and temperature, and if we express the result in terms of the quantity  $E_{\lambda}$  where  $E_{\lambda} d\lambda = udk$ , we have

$$\frac{1}{\rho^2} \frac{16}{3\pi} \frac{1 + |\cos 2\theta|}{2} |\operatorname{N}a^2 \sum_{n} \frac{1}{n^2} e^{-\frac{1}{2} \frac{k_{\mathrm{T}}}{\sigma a^2} (2n\pi)^2} (|f| \operatorname{E}_{\lambda} \lambda)_n. \quad (12)$$

#### 6. Composite Crystal.

When the crystal is composite the complete discussion of any special case is rather more complicated. If, for example, the alternate planes are different in character, we obtain a set of four difference equations involving two different types of T's and S's. If three of these are eliminated we obtain a single difference equation for the fourth, the solution of

which depends on a quadratic equation rather more complicated than (2). The subsequent procedure follows the same course as in § 3. The general problem, though straightforward, might be rather complicated, but by the following argument is made unnecessary. The influence of a single atom on another is always very minute, and the effect only becomes important by its repeated recurrence. Consequently no error will be introduced by regarding as the unit of scattering, not the atom, but the group of atoms in a single unit of the crystal lattice. Let  $f_r$  be the scattering of an atom of type r, of which there are  $N_r$  per c.c., and let this atom occur at distance  $\alpha_r a$  from the first plane, a being the distance in which the structure of the crystal repeats itself. Then in the reflexion formulæ the expression

$$N \mid f \mid e^{-\frac{1}{2}\frac{k_{\mathrm{T}}}{\sigma a^2}(2n\pi)^2}$$

must be replaced by

$$\left|\sum_{r} \mathbf{N}_{r} f_{r} e^{-i2n\pi a_{r}} e^{-\frac{1}{2} \frac{k_{T}}{\sigma_{r} a^{2}} (2n\pi)^{2}}\right|^{*}.$$

#### 7. Comparison with Experiment.

We now compare our result with experiment, and to do so shall take the same experiment as was discussed in the former paper. The elimination of the higher orders of reflexion follows the same course as before, but the numbers

resulting measure now not  $E_{\lambda} \frac{\lambda^2}{\mu}$  but  $E_{\lambda} \lambda$ . We thus obtain

revised values of  $E_{\lambda}$ . I do not give the details, because we shall see that a further modification is necessary in the reflexion formula. The quadrature of  $E_{\lambda}$  now gives that

$$E_0\lambda_0 = 1.3 \int E_{\lambda} d\lambda$$

where  $E_0$ ,  $\lambda_0$  refer to the wave-length  $3.92 \times 10^{-9}$  cm. which is reflected by rocksalt at  $4^{\circ}$ . We have seen that all the reflexion occurs in a region of about 8'' and is practically perfect in this region, so we estimate the efficiency for the

reflexion in rocksalt at 4° as  $E_0 \frac{d\lambda}{d\theta} \delta\theta / \int E_{\lambda} d\lambda$  where  $\delta\theta = 8''$ .

Using the quadrature this becomes 1.3 cot  $4^{\circ}$ .  $\delta\theta$  or 0.0004. The observed efficiency is 0.0035, and this calculated value is no better than the old one.

<sup>\*</sup> See the earlier paper, p. 325.

<sup>†</sup> Loc. cit. p. 330.

In spite of their failure to account for the amount of reflexion, the formulæ developed here are to be preferred to the earlier ones, since they include an effect which has been shown to have been unjustifiably disregarded in the former paper. The new formulæ make the reflexion independent of the absorption coefficient of the crystal. Now the work of W. H. Bragg\* has proved that absorption does play an important part in the reflexion, and this suggests a way out from the discrepancy. If a crystal is not perfect, so that the planes are not everywhere absolutely parallel, it may happen that some part of the beam which has not been extinguished by reflexion at the surface will find a piece of the inside of the crystal at the proper angle, and so will give rise to a second reflexion. Such an effect will obviously involve the absorption coefficient of the rays in the crystal.

#### 8. Imperfect Crystal.

The irregularity of a crystal is of necessity a rather indefinite matter, which it would be perhaps difficult to discuss with rigour. We shall only attempt to see the general type of change to be expected. We will first study the effect of supposing that the surface is irregular, without taking into account the possibility of interior reflexions. Suppose that the surface is divided up into a number of plates whose normals all point in slightly different directions. It is clear that in some cases there might be no reflexion or there might be several. Now the rays reflected from two different plates of the crystal will travel in slightly different directions, and if a photographic plate is put in their path they will strike it at different points. But if the distance of the photographic plate from the crystal is the same as that of the source, they will strike it at the same point; for on account of the constancy of the angle of reflexion, the locus of points which can reflect rays from a given source to a given point is a circle, and only when source and point are equidistant from it does this circle touch the crystal. In this case only is there any considerable area on the crystal which can all reflect to the same point. Moreover, as we shall see later, a very important fraction of the reflected radiation comes from reflexions inside the crystal, and these will be focussed to points only very slightly different from those coming from The accuracy with which Moseley t could the surface. determine his X-ray spectra is probably partly due to this

<sup>\*</sup> W. H. Bragg, Proc. Roy. Soc. A. vol. lxxxix. p. 430 (1914). † Moseley, Phil. Mag. vol. xxvi. p. 1024 (1913).

Phil. Mag. S. 6. Vol. 27. No. 160. April 1914.

focussing, since (though for a different reason) his photographic plate and source were at the same distance from the

crystal.

It is quite possible that a given crystal surface should systematically reflect more than its due share of radiation. For example, if it were of a wavy form, each separate wave would reflect a ray. But we can show that on the average there is no improvement in the reflexion when the surface is supposed divided into small plates, the normals of which deviate from their mean direction in a random manner. such an indefinite question as the present it is useless to proceed with any great rigour, and we shall be content with a rather general argument. In the first place, there is no need to allow for the fact that the normals of the plates deviate from the plane of incidence of the rays. The only effect of this is to shift the ray to a different part of the line of reflected rays. In considering the reflexion from a set of plates, the normals of which all lie in the plane of incidence. it will be sufficient to take it that a ray is reflected to the full extent given in (11), when a line can be drawn from the source to the plate, making exactly an angle  $\phi$ , the reflexion angle, with its plane. For a plate in any fixed position on the crystal there will be a certain small range of directions of the normal such that a line can be drawn from a given source to make angle  $\phi$  with the plate. This range is limited by the two positions when the line cuts the plate at either of its two edges, and therefore the range of inclinations of the plate which can give a reflexion is proportional to its breadth. The chance of a reflexion is thus not altered by cutting the plate in half, for if this is done either of the halves must be aimed in the proper direction with just twice the accuracy, that is to say each half is just half as likely to give a re-Thus there is on the average the same probable number of reflexions when the crystal is broken into many plates as when it is broken into few, or finally as when it is perfect. We conclude that there is no average improvement or deterioration of reflexion when the surface of the crystal is broken up.

When we come to consider the inside of the crystal the matter is quite different. We saw in § 3 that if the crystal is perfect all the radiation that can be reflected, is so, long before the depth at which rays at a different angle are appreciably absorbed. Now if the crystal is twisted internally these unabsorbed rays may come on a part of it at the right angle, and so give rise to a second reflexion. We must estimate how this will affect the matter. Suppose d to be a

depth such that the crystal is twisted through an amount sufficient to allow of a new reflexion. Roughly speaking, then, at every successive d we shall get a reflexion, and the intensities of these reflexions will be 1,  $e^{-2\mu d \csc \phi}$ ,  $e^{-4\mu d \csc \phi}$ , The whole reflexion formula should then be multiplied

by  $\frac{1}{1-e^{-2\mu d \csc \phi}}$ , or  $\frac{1}{2\mu d \csc \phi}$  if the crystal is so badly

twisted that there are a number of reflexions. It appears that as it describes a property of the crystal, d ought to be taken constant. For a not very great distortion this might be justifiable, but we have strong experimental reason to believe that the crystals are even more imperfect than this. For when the reflexion is evaluated with this factor 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 crystals are so badly twisted that their planes do not remain parallel even long enough to produce a single perfect reflexion.

Suppose, therefore, that the crystal is composed of pieces each of depth d small compared with the amount necessary to produce a perfect reflexion. At the depth d the transmitted wave has on the average an intensity  $e^{-4qd/\pi a}$  (see (8)), and the wave reflected by the thickness d has intensity  $1-e^{-4qd/\pi a}$  or  $\frac{4qd}{\pi a}$ . Suppose one of the reflecting pieces is at depth z. Then the amount reflected from it is proportional to  $\frac{4qd}{\pi a} e^{-2\mu z \csc \phi}$ . The number of such pieces in a length dz is dz/d, so that the reflexion formula is to be multiplied by a factor

$$\frac{4qd}{\pi a} \int_0^\infty e^{-2\mu z \, \mathrm{cosec} \, \phi} \, \, dz/d \ \, \mathrm{or \, to} \, \, \frac{2}{\pi a} \, \frac{q \, \sin \, \phi}{\mu} \, .$$

If we multiply (11) and (12) by this we see that apart from a numerical factor they lead to the old expressions for the reflexion\*. That this should be so is not remarkable, since each reflecting piece of the crystal consists only of a few planes, so that the mutual influence of the atoms becomes unimportant. The chief difference is that the whole reflexion now no longer takes place in a band 5" broad, so that the argument t which pointed to the insufficiency of the earlier formulæ loses its validity. The displacement of the reflexion

due to refraction remains, since the distortion of the crystal does not influence the phase relations of the waves scattered in the direction of the transmitted beam. The pattern observed on a photographic plate will in general be very irregular, but when the plate and source are equidistant from the crystal it should have the general character of a band one side of which shades off exponentially. It is readily calculated that the intensity in the band falls to a fraction 1/e in a breadth  $\sin \phi \cos \phi/\mu$ .

For convenience I repeat the reflexion formulæ.

The effect of monochromatic radiation measured in an instrument at distance  $\rho$  from the image of the source with a slit of length l and sufficient breadth is

$$I\frac{l}{\rho} A \frac{1+\cos^2 2\phi}{2} N^2 \frac{f^2}{\mu} e^{-\frac{k\Gamma}{\sigma a^2}(2n\pi)^2} \frac{1}{2} \lambda^3 \csc 2\phi.$$
 (13)

The intensity of reflexion of white radiation is

$$\frac{A}{\rho^2} \frac{1 + \cos^2 2\theta}{2} N^2 a^2 \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-\frac{kT}{\sigma a^2}(2n\pi)^2} \left(\frac{f^2 E_{\lambda} \lambda^2}{\mu}\right)_n \cdot (14)$$

The factor A will probably varyl from crystal to crystal according to the amount of contortion, but we should not expect it to depart widely from unity, its value in the earlier paper. As we are now free from the argument about the limitation of reflexion to a breadth of  $5^{\prime\prime}$ , we may use the experimental value of the efficiency to determine  $f^2$ . When A was unity it was found to be  $26(e^2/mC^2)^2$ , and this is of the order to be expected from atoms with about 10 electrons, of which some, but not all, are concentrated close together.

#### 9. Scattering from a Single Atom.

Since the first part of this paper was written two experimental results have been published by W. L. Bragg†, which have an important bearing on our subject. In the first place he has shown that we must suppose that each atom scatters a wave whose amplitude is proportional to its atomic weight. Thus in fluorspar the two fluorine atoms give waves which can destroy by interference the wave coming from one calcium atom. Since all experiments have shown that the atomic weight is proportional to the number of electrons  $\nu$  in the atom, we conclude that f is closely proportional to  $\nu$ . This result would hold if we could suppose that the majority

<sup>\*</sup> Loc. cit. p. 331. † W. L. Bragg, Proc. Roy. Soc. A. vol. lxxxix. p. 468 (1914).

of the electrons were crowded together in a region of the order of  $10^{-9}$  cm., and this they certainly will be for the heavier elements. But if this is so there is a certain amount of difficulty with regard to Bragg's second experimental result. From measurements of crystals of a good many substances, he concludes that on the average the relative strengths of reflexion of the several orders for monochromatic radiation are as the numbers 100, 20, 7, 3, 1. After allowing for the temperature corrections these numbers are

fairly well expressed by the formula  $\frac{1}{n^2}$ ; but since the radiation is not appreciably dispersed they are to be compared not with (14) but with (13), and in this formula the reflexion is proportional to  $\frac{1}{n}$ . Thus, we must attribute a factor  $\frac{1}{n}$  to  $f^2$ , the coefficient for the scattering of a single atom. Now we saw in the earlier paper\* that  $f^2$  will certainly decrease with the order of reflexion, and the expression there found seems capable of accounting for the excess scattering from amorphous substances, as in this case experiments have only been concerned with light atoms where there is no great concentration of electrons; but when we are dealing with heavier atoms we have seen that Bragg's first result points to a considerable crowding of electrons in a small space, and in this case it would hardly be expected that the excess effect should be so great as to give a factor

 $\frac{1}{n}$ . Involving as it does a knowledge of the arrangement of the electrons in the atom, it does not seem possible at present to make any better progress in discussing this question.

### Summary.

The paper attempts a more accurate solution than was given in the first part of the problem of X-ray reflexion, on the basis of allowing for the mutual influences of the scattering atoms.

(i.) It is shown that the mutual influences of the atoms in a plane together are unimportant.

(ii.) The interactions of the separate planes are allowed for, and revised reflexion formulæ are deduced. The reflexion is found to be practically perfect for a certain range of angles. The transmitted beam is extinguished much

<sup>\*</sup> Loc. cit. p. 329.

more rapidly than corresponds to the true absorption of the crystal.

(iii.) Comparison with experiment suggests that the new formulæ can account for the observed intensity as little as the old.

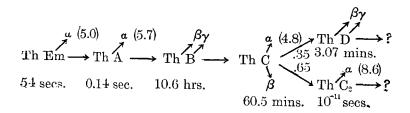
(iv.) This observed strength of reflexion is to be attributed to crystalline imperfection, and allowing for this the old formulæ of the first part are retained with a new numerical factor.

My thanks are due to Prof. Sir Ernest Rutherford for the kind interest he has taken in this research.

March 4, 1914.

LXXIX. The Transformations in the Active Deposit of Actinium. By E. Marsden, M.Sc., Lecturer in Physics, and P. B. Perkins, Ph.D., University of Manchester\*.

IT is well known that the "C" products of the active deposits of radium and thorium are anomalous in that in both cases the atom has two distinct modes of transformation, i. e. it breaks up with emission of either an  $\alpha$  or a  $\beta$  particle. In the case of thorium C, owing to the fact that the chance of disintegration in either of the two ways is of the same order, it has been found possible to examine the process in considerable detail, and the following scheme of transformation has been arrived at †:—



By analogy the method of production of the branch product,  $C_2$ , has been deduced  $\ddagger$ .

† K. Fajans, Phys. Zeit. xii. p. 369 (1911); xiii. p. 699 (1912).

<sup>\*</sup> Communicated by Sir E. Rutherford, F.R.S.

<sup>†</sup> Marsden & Barratt, Proc. Phys. Soc. xxiv. 1, p. 50 (1911); Marsden & Darwin, Proc. Roy. Soc. A. Ixxxvii. p. 17 (1912); Marsden & Wilson, P hil. Mag. xxvi. p. 354 (1913).