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THE SCATTERING OF X-RAYS AND BRAGG'S LAW

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In 1914 Darwin¹ showed that when the classical electromagnetic theory of light is applied to the regular reflection of X-rays by a crystal a small departure from Bragg's law is demanded if there is a refractive index for X-rays in the crystal. The theory further demands that the refractive index shall be $1-\mu$, where μ is a small positive quantity. Stenström² was the first to show a departure from Bragg's law experimentally. He found that the wave-length of a spectrum line as determined by applying Bragg's law to the second order reflection was slightly less than that as determined by the first order reflection, the same crystal being used in each case. This is as it should be according to Darwin's theory. Stenström's result has been confirmed by Siegbahn,³ Hjalmar,⁴ and by Duane and Patterson.⁵ Confirmation by another method of an index of refraction for X-rays in solids of a value less than unity has also been obtained by A. H. Compton⁶ in his experiments on total reflection.

A few months ago A. H. Compton published a quantum theory of the scattering of X-rays⁷ in which he showed that when X-rays are scattered by free electrons a change of wave-length in the scattered rays should be expected. This change of wave-length has been observed experimentally by Compton⁸ for Mo K α rays scattered by carbon. It occurred to Mr. C. H. Eckart and the present writer that, if crystalline reflection of X-rays is a special case of scattering, then the reflected ray should be of longer wave-length than the incident ray. Making this assumption the effect on Bragg's law was discussed in a letter to Nature.⁹ It was there shown that the experimental departures from Bragg's law could be explained by this assumption of a change of wave-length on reflection. The same explanation was made later by Wolfers¹⁰ and still later by Hulburt.¹¹ Hulburt, however, remarks that the theoretical deviation from Bragg's law is of the same order of magnitude as the experimental deviation but is of the wrong sign, while Jauncey and Eckart⁹ found the theory to give

the same sign as the experiments. There is a reason for this disagreement between Hulbert and Jauncey and Eckart, and we shall now proceed to consider the theory in detail.

Let the incident and reflected wave-lengths be λ_1 and λ_2 , respectively, the grazing angles of incidence and reflection being θ_1 and θ_2 , respectively. Then for reflection from a single plane of atoms we have

$$\cos \, \Theta_1 / \lambda_1 \,=\, \cos \, \Theta_2 / \lambda_2 \tag{1}$$

while for reflection from two consecutive planes of atoms

$$d \sin \theta_1 / \lambda_1 + d \sin \theta_2 / \lambda_2 = n$$
 (2)

where $2n\pi$ is the difference of phase between the rays reflected from the first and second planes, respectively. Further, the total angle of deviation between the incident and reflected rays is $(\theta_1 + \theta_2)$ and applying Compton's change of wave-length formula⁷ we have

$$\lambda_2 = \lambda_1 + 2\gamma \sin^2 (\theta_1 + \theta_2)/2 \tag{3}$$

where $\gamma = h/m \ c = 0.024 \ A$. These three equations may be solved for λ_1 in terms of d, n and Θ_1 or in terms of d, n and $(\Theta_1 + \Theta_2)/2$. To facilitate the first solution let us put $x = \lambda_2/\lambda_1$ and from eqs. (1) and (3) we obtain an equation for x in terms of Θ_1 and α where $\alpha = \gamma/\lambda_1$. This equation is a quadratic with the solutions x = 1 and

$$x = (1 + 2\alpha + \alpha^2 \cos^2 \theta_1) / (1 + 2\alpha \cos^2 \theta_1 + \alpha^2 \cos^2 \theta_1).$$
(4)

The meaning of the solution x = 1 is that there is a Laue spot in the forward direction of the primary beam. However, instead of expressing λ_1 in terms of d, n, and Θ_1 , we can obtain a relation between λ_1 and L', where L' is the apparent wave-length given by Bragg's law nL' = 2d sin Θ_1 , when the grazing angle of incidence Θ_1 is measured. This relation is

$$L' = \lambda_1 + \gamma - \gamma^2 \sin^2 \Theta_1 / (\lambda_1 + \gamma). \tag{5}$$

From eq. (5) we see that L' becomes less for greater values of θ_1 , that is, for higher orders of reflection and thus gives a deviation from Bragg's law of the right sign. The deviation is also of the right order of magnitude as shown in table I. The experimental values are taken from the paper of Duane and Patterson,⁵ who measured θ_1 using a crystal of calcite.

L' A	м	N	L'M — L'N Experiment A	l'm — l'n Theory A
1.473	1	2	0.00015	0.00007
1.279	1	3	0.00025	0.00017
1.096	1	2	0.00006	0.00005

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In the above table *m* and *n* are the orders of reflection while L'm is the apparent wave-length when calculated by Bragg's law for the *m* th order. Now, however, let us examine the relation between L'' and λ_1 , where L'' is Vol. 10, 1924

or

the apparent wave-length given by Bragg's law when $(\theta_1 + \theta_3)$ is measured. Let $(\theta_1 + \theta_2)/2 = \beta$ so that $nL'' = 2d \sin\beta$ and we find

$$L'' = \lambda_1 (1 + 2\alpha \sin^2\beta) / \sqrt{(1 + 2\alpha \sin^2\beta + \alpha^2 \sin^2\beta)}, \qquad (6)$$
$$L'' = \lambda_1 + \gamma \sin^2\beta \qquad (7)$$

to the first power of γ . From this it appears that L'' becomes larger for higher orders of reflection, which is contrary to experiment. However, if the experimental values of Hjalmar,⁴ who used a crystal of gypsum and measured ($\Theta_1 + \Theta_2$), are used we obtain table II.

TABLE II

L" A	м	N	L"M — L"N Experiment A	L"M — L"N Theory A
1.389	1	2	0.00045	0.0006
2.279	1	2	0.0014	0.0016
2.279	1	3	0.0016	0.0043

The theoretical values are of the right order of magnitude. This is the result obtained by Wolfers,¹⁰ but he states that his theoretical values are the right sign. My values, however, are of the wrong sign, which result has also been reported by Hulburt.¹¹ Furthermore Θ_1 and β are approximately equal so we may apply eq. (5) to the experiments of Hielmar

imately equal so we may apply eq. (5) to the experiments of Hjalmar. We find, however, that although eq. (5) gives values of the right sign yet they differ from the experimental values of table II in order of magnitude, being some 0.02 times these values. It would seem therefore that the agreement between the experimental and the theoretical values of tables I and II is purely fortuitous.

Still further let us compare eqs. (5) and (7) and we find

$$L' - L'' = \gamma \cos^2 \Theta_1 \tag{8}$$

to the first power of γ . Or L'-L'' = 0.024 A approximately for the lower orders of reflection. Duane and Patterson⁵ have compared their results with those of Overn,¹² who measured ($\Theta_1 + \Theta_2$) for the same tungsten lines. Differences varying from -0.0023 A to +0.0011 A were found. These are much less than the 0.024 A predicted by the present theory. It is seen therefore that the experimental deviations from Bragg's law cannot be explained on the assumption of a change of wave-length on reflection. We are therefore forced to accept Darwin's theory of a refractive index as providing the only available explanation of the deviations from Bragg's law.

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² Stenström, Doctor's Dissertation, Lund, 1919.

⁸ M. Siegbahn, C. R., Paris Acad. Sci., 173, 1350.

⁴ E. Hjalmar, Zeit. Physik., 1, 439 (1920).

⁵ Duane and Patterson, Physic. Rev., 16, 526 (1920).

⁶ A. H. Compton, Phil. Mag., 45, 1121 (1923).

⁷ A. H. Compton, *Physic. Rev.*, **21**, 483 (1923).

⁸ A. H. Compton, *Ibid.*, **22**, 409 (1923).

⁹ Jauncey and Eckart, Nature, 112, 325 (1923).

¹⁰ M. F. Wolfers, C. R., Paris Acad. Sci., 177, 759 (1923).

¹¹ E. O. Hulburt, Phys. Soc. Meeting, Chicago, 1923.

¹² O. B. Overn, Physic. Rev., 14, 137 (1919).

REFRACTION OF X-RAYS IN PYRITES

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In all careful determinations of wave-lengths of X-rays by crystal reflection it has been found that the relation

$n\lambda = 2d \sin \Theta$

does not hold accurately for the several orders. The departure from this law has been rightly ascribed by Stenstrom¹ to refraction in the crystal. This refraction has been observed by Hjalmar,² Davis and Terrill³ and others, and has been directly confirmed by experiments of Compton on total reflection. If the angles are measured with respect to the crystal surface, the index of refraction μ is expressed by

$$\mu = \frac{\cos \Theta_1}{\cos (\Theta_0 - \varphi)} \tag{1}$$

where Θ_1 is the angle of incident rays to the surface outside the crystal and φ is the angle of the surface to the molecular planes and Θ_0 is the angle of the X-ray beam to the molecular planes inside the crystal.

Placing $\mu = 1 - \delta$, Stenstrom has derived the following expression for δ , for the case when $\varphi = 0$.

$$\delta = \frac{\left(\frac{\sin \Theta_m}{m}\right)^2 - \left(\frac{\sin \Theta_n}{n}\right)^2}{2\left(\frac{\cos \Theta_m}{m}\right)^2 - 2\left(\frac{\cos \Theta_n}{n}\right)^2},$$
(2)

where Θ_{m} , Θ_{n} are the observed glancing angles at the orders *m* and *n*.

The bending of the rays by refraction as they pass through the surface in the case of the natural cleavage surface is small. The bending for Mo, K_{α_1} radiation in calcite for instance is found to be about 3" arc. Since it is difficult to measure such a small effect accurately, the following method was proposed to increase the bending due to refraction. As in the case