

The Reflection of X-rays by Crystals

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Proc. R. Soc. Lond. A 1913 **88**, doi: 10.1098/rspa.1913.0040, published 1 July 1913

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investigation given above has been confined to that portion of the spectrum on the red side of the blue. If we wish to extend our investigation into the blue and violet then we should have to take account of the effect of the blue sensation. Further, although it seems quite clear that the major part of the luminosity perceived in that part of the spectrum including the red, yellow and green, at any rate with fairly bright spectra, is due to the red and green sensations, there is much evidence to indicate that, at any rate in the green and blue, there is a fourth sensation which may be called the fundamental white sensation. This probably corresponds to the rod-effect of von Kries, which would have to be taken into account if extremely minute differences of luminosity were under consideration, or if we were dealing with spectra of small intensity.

The Reflection of X-rays by Crystals.

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(Received April 7,—Read April 17, 1913.)

In a discussion of the Laue photographs it has been shown* that they may conveniently be interpreted as due to the reflection of X-rays in such planes within the crystal as are rich in atoms. This leads at once to the attempt to use cleavage planes as mirrors, and it has been found that mica gives a reflected pencil from its cleavage plane strong enough to make a visible impression on a photographic plate in a few minutes' exposure. It has also been observed that the reflected pencil can be detected by the ionisation method.†

For the purpose of examining more closely the reflection of X-rays in this manner we have used an apparatus resembling a spectrometer in form, an ionisation chamber taking the place of the telescope. The collimator is replaced by a lead block pierced by a hole which can be stopped down to slits of various widths. The revolving table in the centre carries the crystal. The ionisation chamber is tubular, 15 cm. long and 5 cm. in diameter. It can be rotated about the axis of the instrument, to which its own axis is perpendicular. It is filled with sulphur dioxide in order to increase the ionisation current: both air and methyl iodide have also been used occasionally to make sure that no special characteristics of the gas in

* W. L. Bragg, 'Proc. Camb. Phil. Soc.,' vol. 17, Part I, p. 43.

† W. H. Bragg, 'Nature,' Jan. 23, 1913.

the chamber affect the interpretation of the results. The ionisation current is measured directly. A balance method has not been used as we have not found it possible to deflect a suitable portion of the primary rays into a balance chamber.

The face of the box containing the X-ray bulb is covered with a special shield of lead, 5.5 mm. thick; the general lead covering of the box is 1 mm. thick, which is not always enough to screen the chamber from penetrating X-rays that produce an effect comparable with the effect of the reflected rays. The circular end of the ionisation chamber is also protected by lead. The slit through which the primary pencil of X-rays emerges from the box is 3.3 mm. long; its width has been 2 mm. for the rougher measurements and 0.75 mm. for the finer. Since the slit is 12 cm. from the anticathode the emerging pencil has an angular width of about a third of a degree in the latter case. In the same way a slit 2 mm. wide and 5 mm. long admits the reflected pencil to the ionisation chamber when preliminary measurements are being made, or when the whole effect is feeble; and this width can be cut down to 0.75 mm. when desired. The distance from either slit to the axis of the apparatus is 8 cm.

We have found it best to keep the bulb very "soft." The cathode stream has often been visible over its whole length.

As will be seen later it is desirable to determine angles of incidence and reflection with great accuracy. This was not anticipated, and the circular scale was only divided into degrees, and was made too small. Nevertheless, it is possible to read tenths of a degree; a better and more open scale is now being put in.

Let us suppose that a crystal is placed on the revolving table so that the cleavage face passes through the axis of the instrument. Let the incident pencil fall on the face and make an angle θ with it; and let the crystal be kept fixed while the ionisation chamber is revolved step by step through a series of angles including the double of θ , the ionisation current being measured at each step. The results of such a set of measurements are shown in fig. 1. In this case the crystal is rock-salt; and it has been placed so that the incident pencil makes an angle of 8.3° —as given by the apparatus—with the incident beam. The points marked in the figure show the result of setting the ionisation chamber at various angles and measuring the current in each case.

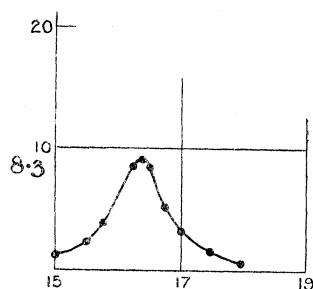


FIG. 1.—Regular reflection from cleavage face of rock-salt, glancing angle 8.3° .

The maximum effect is not quite at 16.6° , but at a point somewhat less than 16.4° . The defect from the double angle is due in part to want of symmetry and accuracy of the apparatus; but not much of it is caused in this way. It is rather due to the difficulty of setting the crystal face exactly; sometimes this is much accentuated by "steps" on the face of the crystal. The error can be eliminated by swinging over the ionisation chamber to the other side and taking corresponding observations, in a manner analogous to the method of finding the angle of a prism on the spectrometer.

The finer slits were used in obtaining this curve, and it may be inferred from the figure that the source of the X-rays is practically a point. For the width of the pencil from a point source by the time it reaches the slit of the ionisation chamber is $0.75 \times 28/12$ or 1.75 mm. The chamber slit being 0.75 mm. wide, the whole effect observed is comprised within a lateral movement of the chamber equal to $1.75 + 0.75$ or 2.50 mm. Since the chamber slit is 8 cm. from the axis of the apparatus this implies a rotation of the chamber through $(2.50 \times 180)/(\pi \times 80)$ or 1.78° . The figure shows that these limits are actually observed; the whole curve lies well within the range 15° to 18° . The source must therefore be nearly a point.

When the actual relation between the angles of the crystal mirror and the ionisation chamber has been determined, the mirror and chamber may be swept together through an extended range, keeping the relation between the angles such that the chamber always shows the maximum current for each setting of the crystal. It is convenient to use the wide slits for a preliminary examination of this kind. When the effect is small the wide slits can alone be used. But in a number of cases it is possible to use the narrow slits in order to make a closer survey, and where this is done much more information can be obtained.

The curve in fig. 2 shows the results of a sweeping movement of this kind, the crystal being iron pyrites. Curves for rock-salt are drawn in figs. 3, I, and 3, II. It will be observed that there are peculiar and considerable variations in the intensity of the reflection at different angles. The three peaks marked A, B, and C are common to the curves of all crystals so far investigated, *e.g.* zinc blende, potassium ferrocyanide, potassium bichromate, quartz, calcite, and sodium ammonium tartrate. They are readily distinguishable by their invariable form, relative magnitudes, and spacings. Moreover, the absorption coefficients of the rays reflected at these separate angles do not vary with the nature of the crystal or the state of the bulb. It happens that the actual angles of reflection of the three sets of rays are nearly the same for several crystals.

The use of the narrow slits permits a closer examination of these

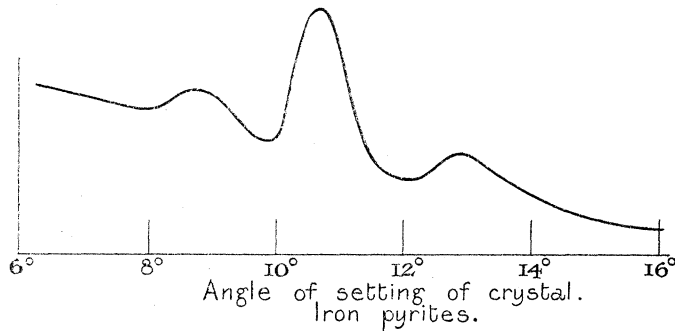


FIG. 2.—Reflection from face (100) of iron pyrites, at varying angles of incidence. Abscissa—Angle of incidence of rays on crystal face; Ordinate—Strength of reflected beam, arbitrary scale.

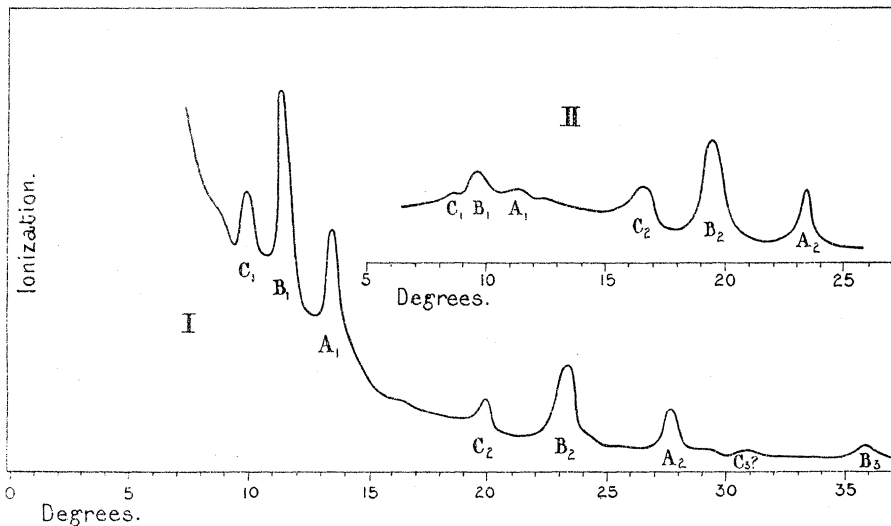


FIG. 3.—Reflection (I) from face (100) and (II) from face (111) of rock-salt. The curves show the variation of strength of reflected beam with angle of incidence.

effects; but, of course, it takes much longer time to make, and more space to exhibit. The results for iron pyrites are shown in the series of curves of fig. 4: a series in which each curve is obtained in the same way as the curve of fig. 1, the crystal being set at some definite angle which is altered in going from curve to curve. The curves are arranged so that the vertical distance between the horizontal lines of reference of any pair is proportional to the difference in the angles of setting of the crystal in the two cases.

In comparing the curves at the different angles two principles must be borne in mind. In the first place if there is a general reflection of rays throughout the whole range of the pencil which is emerging from the slit near the bulb, the curves show, as in fig. 1, a maximum with similar slopes

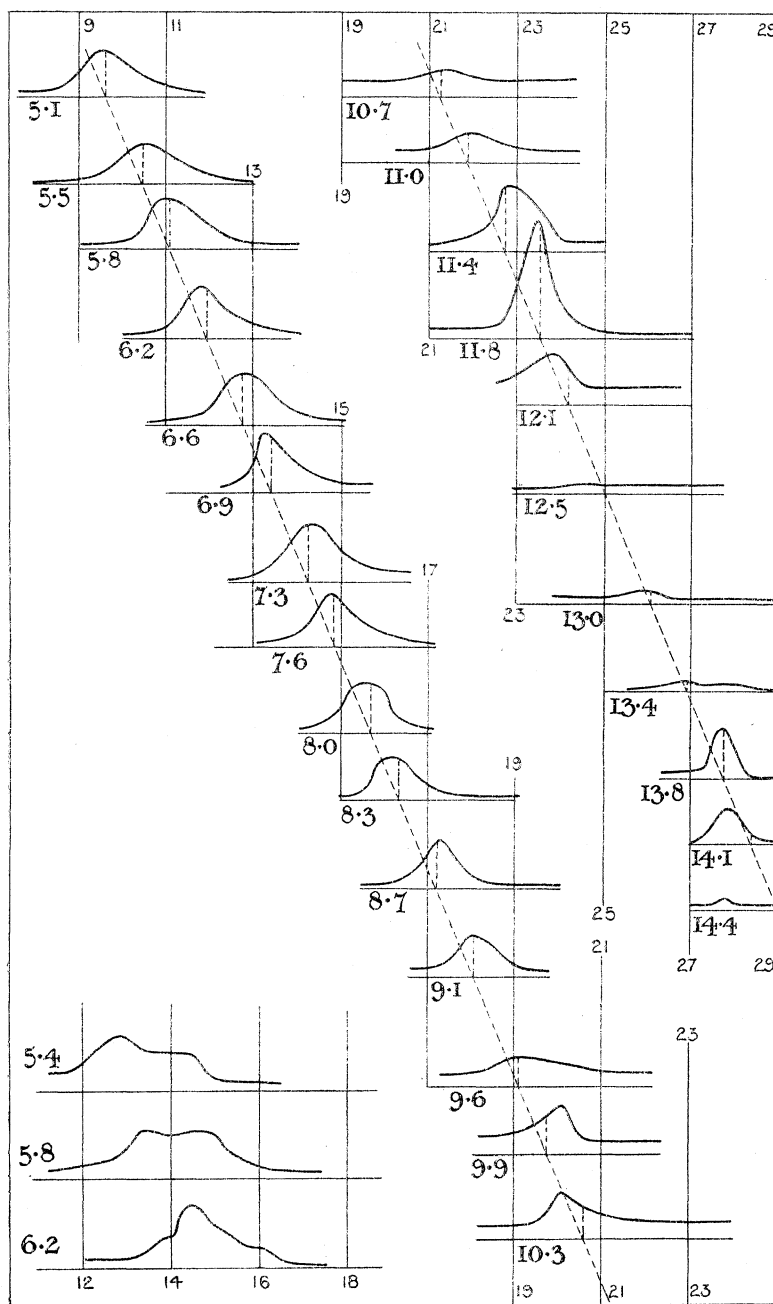


FIG. 4(a).

FIG. 4.

FIG. 4.—Detailed examination of reflection from iron pyrites (100).

on each side of it. The maximum occurs at that setting of the chamber which is twice the angle of setting of the crystal or differs from it only by that constant error of setting to which allusion has already been made. The maximum slowly marches across the page as we go down the series of curves, and its progress is marked by the dotted line.

In the second place there is a special reflection which manifests its presence in a curious and most convenient way. It often happens that the rays emerging from the bulb slit and falling on the crystal contain a large preponderance of rays of a given quality which can only be reflected at a certain angle. This angle is very sharply defined: even our present and somewhat rough apparatus shows that it is limited to a very few minutes of arc in either direction. In this case the radiation which is reflected is not distributed generally over the whole range bounded by the edges of the bulb slit, which it will be remembered is about a third of a degree, but is confined to a select small portion of that range. When this is the case the position of the maximum does not change at all as the crystal is moved from setting to setting, so long as any of this radiation is reflected. For example, the curves for 13.4° , 13.8° , 14.1° , 14.4° show the existence of a special reflection of this kind which is always at its maximum when the chamber is set at 27.7° . The reason for this may be understood from fig. 5.

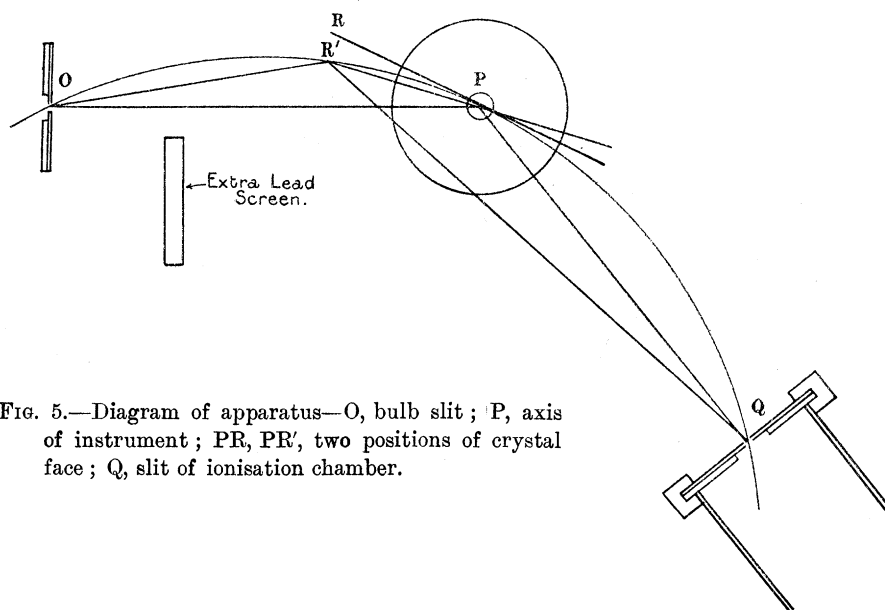


FIG. 5.—Diagram of apparatus—O, bulb slit; P, axis of instrument; PR, PR', two positions of crystal face; Q, slit of ionisation chamber.

Here O is the bulb slit, P the axis of the instrument, and Q the chamber slit. When the crystal face is in the position PR, let us say, the ray OP

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strikes at the right angle for reflection, and is reflected along PQ. But when the crystal is turned to OR', the ray OP of the radiation of this quality which we are considering is not reflected at all. It is now the ray OR', where R' lies on the circle OPQ; for the angles made by OR' and QR' with PR', and the angles made by OP and QP with PR, are all equal to each other. The ray OR' is reflected along R'Q, and still enters the ionisation chamber, though the latter has not been moved. When, therefore, we see a maximum persisting in the same angular position of the chamber for several successive positions of the crystal, we know that we have a case of this special reflection. There is a relatively large quantity of very homogeneous radiation of a certain kind present in the radiation from the bulb. The narrower we make the slits the more does it stand out, but the more difficult it is to find, if we do not know where to look for it.

It will be noticed how small the general reflection appears, in comparison with the special reflection between the angles (crystal settings) 12° and 14° . It is still small when the angle is reduced to 10.7° . At 10.3° there is enough of it to throw a hump on to one side of a peak of special reflection, and at 9.9° it has passed through, and thrown the hump upon the other side. Consideration of the whole series of curves shows that there are three strongly marked homogeneous pencils of sharply defined quality; they occur at (uncorrected chamber angles) 27.7° , 23.4° , and 20.0° . What we have called the general reflection may comprise many other definite pencils, but they are scarcely resolved at all in this series of curves. Their presence is, however, fairly obvious. A series of potassium ferrocyanide curves shows them much more clearly. Three of this series are shown in fig. 4 (*a*), and their peculiar forms indicate to what extent interpretation has yet to be carried.

When these homogeneous beams are isolated by the use of narrow slits, it is possible to determine their absorption coefficients in various substances. In the end, there is no doubt, this will be done with great accuracy; for the present, our results must only be looked on as provisional. They are, perhaps, right to 5 per cent.; for many purposes this is quite sufficient. In the case of rock-salt we find the mass absorption coefficients in aluminium of A, B, and C to be 25.5, 18.8, and 10.6 respectively, the last being the most doubtful and probably too low. The absorption coefficient of the B-rays in Ag is 74, in Cu 140, in Ni 138; these values are approximate. We have made no exhaustive determination of the coefficients in the case of various crystals, but in a number of cases, all those tried, we have found them to be the same. There can be little doubt the three peaks are, in all cases, due to the same three sets of homogeneous rays, rays which do not change with the state of the bulb, but may well do so with the nature of the anticathode. It

will be observed that the absorption coefficient of the least penetrating set is very nearly that found by Chapman for the characteristic radiation of platinum.

The angles at which the special reflections of these rays take place are not the same for all crystals, nor for all faces of the same crystal, as the following table shows. The angles can be determined with great accuracy; even with our rough apparatus they are probably within 1 per cent. of the truth.

	A.	B.	C.
Rock-salt, cube face {100}	27·3	23·1	19·9
„ face {111}	48·5	40·2	34·0
Potassium ferrocyanide	27·2	23·1	19·8
Calcite, cleavage plane {100}	25·4	21·6	—
Iron pyrites, cube face {100}	28·5	24·2	20·8
Zinc blende, cleavage plane {110} ...	40·0	33·0	27·5
Potassium bichromate	(approx.) 22·4	19·8	(approx.) —

The readings for zinc blende and calcite are not corrected for errors of setting.

The difference in the case of the two faces of rock-salt suggested an attempt to find a repetition of the characteristic three peaks at multiples or sub-multiples of those at which they were first observed. For the sines of 11·55 and 20·1 (half the angles of the chamber settings of the B peak in the two cases) are 0·200 and 0·344 respectively. These are very nearly in the ratio $1 : \sqrt{3}$. If the effects are true diffraction effects such a relation might be expected. The {111} planes are further apart than the {100} planes in the ratio $2 : \sqrt{3}$; the sines of angles of special reflection should be in the inverse ratio, viz., $\sqrt{3} : 2$. True, the sines of the angles have been increased in the ratio $1 : \sqrt{3}$, instead of diminished in the ratio $2 : \sqrt{3}$, but it is not at all unlikely that a spectrum in one case is being compared with a spectrum of higher or lower order in the other. We, therefore, made a search for other spectra and found them at once. In the case of rock-salt we found traces of a third. The full rock-salt curves are shown in fig. 3 for the two kinds of face. The peaks first found are marked A_1 , B_1 , C_1 , and their repetitions A_2 , B_2 , C_2 ; there is a trace of B_3 also. The corrected angular positions of B_1 , B_2 , B_3 are $23·1^\circ$, $47·3^\circ$, and $73·3^\circ$. The sines of the halves of these angles are 0·200, 0·401, and 0·597, and are very nearly in the proportion $1 : 2 : 3$. The absorption coefficient of the rays at B_2 is the same as that of the rays at B_1 .

In the case of the rock-salt section {111} a spectrum occurs at half the angles first found. This is shown in fig. 3, II. It is not at all strongly

marked, and the question at once arises as to why the second spectrum should be so much stronger than the first in this case and so much weaker in the case of the face $\{100\}$. A large amount of the general falling away of intensity at small angles, so obvious in Curve II as compared with Curve I, is undoubtedly due to the fact that the $\{111\}$ face used was not extended enough to catch the whole pencil of rays from the bulb slit at so glancing an angle.

There can be little doubt as to the interpretation of these results. The three peaks A, B, and C represent three sets of homogeneous rays. Rays of a definite quality are reflected from a crystal when, and only when, the crystal is set at the right angle. This is really an alternative way of stating the original deduction of Laue. The three sets of rays are not manufactured in the crystal, because all their properties are independent of the nature of the crystal. An absorbing screen may be interposed with the same effect before or after the rays have struck the crystal. This was found by Moseley and Darwin,* and we have verified it in the case of aluminium.

Since the reflection angle of each set of rays is so sharply defined, the waves must occur in trains of great length. A succession of irregularly spaced pulses could not give the observed effect. In the application of electromagnetic theory to monochromatic light on the one hand, and to homogeneous X-rays on the other, there is no difference to be considered beyond that of wave-length.

These results do not really affect the use of the corpuscular theory of X-rays. The theory represents the facts of the transfer of energy from electron to X-ray and *vice versa*, and all the phenomena in which this transfer is the principal event. It can predict discoveries and interpret them. It is useful in its own field. The problem remains to discover how two hypotheses so different in appearance can be so closely linked together.

It is of great interest to attempt to find the exact wave-length of the rays to which these peaks correspond. On considering Curve I, fig. 3, it seems evident that the peaks $A_1 B_1 C_1$, $A_2 B_2 C_2$ are analogous to spectra of the first and second orders, because of the absence of intervening sets of peaks. The value of n in the equation

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

seems clear. The difficulty of assigning a definite wave-length to the rays arises when we attempt to determine the value of d , the distance of plane from plane.

* We learn that Messrs. Moseley and Darwin have lately been making experiments similar to some of those recorded here. Their results, which have not been published, agree with ours.

There is strong evidence for supposing that the atoms of a cubic crystal like rock-salt, containing two elements of equal valency, are arranged parallel to the planes $\{100\}$ in planes containing equal numbers of sodium and chlorine atoms. The atoms in any one plane are arranged in alternate rows of each element, diagonal to the cube axes, successive planes having these rows opposite ways. The question arises as to whether the value of d is to be taken as that between two successive planes, or two planes identical in all respects. The value of d in the one case is twice that in the other.

The centres of the atoms of sodium and chlorine, regarded for the time being as identical, are arranged in a point system, having as unit of its pattern a cube with a point at each corner and one at the centre of each cube face. The dimensions of this elementary cube can be found in the following way:—

If the side of the cube is of length a , the volume associated with each point in the point system will be $\frac{1}{4}a^3$.

The mass of a hydrogen atom being 1.64×10^{-24} gram. and the density of rock-salt 2.17, we have

$$\frac{1}{\frac{1}{4}a^3} \frac{1}{2}(35.5 + 23) \times 1.64 \times 10^{-24} = 2.17.$$

This gives

$$a = 4.45 \times 10^{-8}.$$

The distance between planes passing through atoms identical in all respects is this distance a . The wave-length, as calculated in this way, is

$$\lambda = 2a \sin \theta = 1.78 \times 10^{-8}$$

for the peak B.

But half-way between these planes which are identical in all respects are situated planes containing the same number of sodium and chlorine atoms, though the arrangement is not in all respects the same. Possibly this tends to make the odd spectra due to the first lot of planes disappear, and, if this is the case, we must halve the first estimate of the wave-length, and put

$$\lambda = 0.89 \times 10^{-8}.$$

The difference between these two values corresponds to taking as a unit of the point system—

- (1) The group 4NaCl , the smallest complete unit of the crystal pattern.
- (2) The individual atom of either nature, associated with only one-eighth of the volume of the complete unit.

We have also examined the reflection from the (110) face of the rock-salt, and have found the peaks situated at such angles as indicate that the ratio of

the distance between these parallel planes to the distance between planes parallel to the face (100) is as $1:\sqrt{2}$. Combined with the position of the peaks reflected from the (111) face, this indicates that the point system which the diffracting centres form has as element of its pattern that suggested above, a cube with a point at each corner and one at the centre of each face. Of the three elementary cubic space lattices, this is the only one in which the distance between the (111) planes is greater than that between any other of the planes of the system.

The wave-length as calculated from the reflection on the (110) face of zinc blende agrees within the errors of experiment with that calculated above.

The wave-lengths to be associated with the spots in the photographs taken by Laue of the diffraction of X-rays by crystals are much smaller than these values. They belong to the region in which we have found reflection to take place at all angles, a region in which the peaks do not obviously occur. This agrees with the distribution of intensity amongst the spots.

The experimental method can be applied to the analysis of the radiation from any source of X-rays. It may, however, be able to deal only with intense radiations. The three sets of rays issuing from the bulb we have been using have angles of reflection whose sines are 0.236, 0.200, 0.173. The reciprocals of these are 4.24, 5, and 5.78. The frequencies, and therefore, according to Planck, the corresponding quantum energies, are in arithmetical progression. In this there is some hint of analogy with Rutherford's recent work on the energies of the various types of β -ray from RaC.

Prof. Barkla has lately communicated to the Physical Society an account of certain experiments in which a diffuse pencil of X-rays, when reflected on the cleavage plane of a crystal, acted on a photographic plate, producing a series of bands. The effect which we have been describing is clearly identical in part with that which Prof. Barkla has described. It is impossible, of course, to criticise a communication of which we have seen an abstract only. But it seems probable that the ionisation method can follow the details of the effect more closely than the photographic method has so far been able to do: and that in this way it is possible to distinguish between those bands which represent distinct sets of rays, and those which are repetitions of one and the same set.
