

XIV. *The Reflexion of the X-rays.* By H. G. J. MOSELEY, M.A., John Harling Fellow, and C. G. DARWIN, M.A., Lecturer in Mathematical Physics in the University of Manchester\*.

THE discovery in July 1912 by Friedrich and Knipping † that the X-rays can show interference has opened up a new region to research. It had been suggested to them by Laue that the ordered arrangement of the atoms in a crystal would do the same for X-rays that a diffraction-grating does for light. They therefore sent a fine beam of X-rays through a thin crystal of zinc-blende on to a photographic plate. After prolonged exposure to the radiation the plate showed an elaborate system of spots surrounding the central image. These spots were caused by fine beams of X-rays, which came from the crystal. Explanations of this phenomenon have been offered by Laue † and by W. L. Bragg ‡. Laue assumes the radiation to consist of a finite number of monochromatic waves, while Bragg believes it to be similiar in constitution to white light. The explanation of the latter accounts perfectly for the positions of the spots in question. The present paper shows that a large proportion of the radiation from an X-ray tube has no definite wave-length, but that five types of monochromatic waves are also present in the radiation from a tube with a platinum target. The following is a brief statement of W. L. Bragg's theory.

It is known that every atom in a crystal scatters a small fraction of the incident radiation. Since the atoms are regularly arranged these scattered disturbances have definite time relations. In certain directions a number of scattered disturbances reinforce one another, and so give rise to a new disturbance of considerable strength. The co-operating disturbances come from atoms which lie in a plane. This plane, the incident beam, and the direction of the new beam have the same geometrical relation as have a mirror, an incident ray, and the reflected ray. Wherever a plane containing many atoms can be drawn in the crystal a ray will be found in the direction which is the reflexion in this plane of the incident beam. The position of every spot in Friedrich and Knipping's photographs can be found from this principle. A cleavage-plane is exceptionally rich in atoms, and Bragg §

\* Communicated by Prof. E. Rutherford, F.R.S.

† Laue, Friedrich and Knipping, *Kgl. Bay. Akad.* p. 303 (1912).

‡ W. L. Bragg, *Proc. Camb. Phil. Soc.* xvii. p. 43.

§ W. L. Bragg, 'Nature,' vol. xc. p. 410, Dec. 12 (1912).

confirmed his theory by throwing a beam of X-rays on to a cleavage surface of mica, and observing a strong image in the position which an optically reflected beam would occupy.

So far, the subject had been developed entirely by photography. The first object of our investigation was to test whether the "reflected" radiation, which was so clearly shown to have an extended wave-front, possessed those properties which have led Prof. W. H. Bragg to suggest that X-rays are corpuscular. It was still possible that the X-rays were really a mixture of two different constituents.

At first we followed the experimental arrangement of Friedrich and Knipping. When, however, W. L. Bragg discovered that a greater effect was obtained by "reflexion" from mica, his method was adopted, and was found to be much more convenient. Later W. H. Bragg\* announced in 'Nature' that the reflected radiation ionized air, and we† were able to add that it had all the principal properties which characterize ordinary X-rays. We then tried to determine how the intensity of reflexion depends on the angle of incidence on the surface of a crystal. We first measured the reflexion from selenite, and found that as the angle of incidence was varied from  $88^\circ$  to  $55^\circ$  the reflexion fell to a thousandth of its initial value. Our results showed certain singularities, which we hoped to explain by studying other crystals. At this stage, however, Prof. Bragg very kindly communicated to us results obtained with other crystals, which he has since published in conjunction with Mr. W. L. Bragg‡. They found the intensity of reflexion to be abnormally large at certain angles. They attributed this selective reflexion to the presence in the radiation of monochromatic waves. These waves were thought to be characteristic radiation from the platinum target in the X-ray tube. When the phase difference between the waves "reflected" from successive layers of atoms is  $2\pi$ , the waves reinforce one another in the reflected beam. In the light of this communication we re-examined the reflexion from selenite. We found that this was the cause of the singularities which we had noticed. By using very fine slits we had missed the position at which the principal selective reflexion occurs. Our later work, in which selenite, rocksalt, and potassium ferrocyanide have been used as reflectors, fully confirms the conclusions of Prof. and W. L. Bragg. At the

\* W. H. Bragg, 'Nature,' Jan. 23 (1913).

† Moseley and Darwin, 'Nature,' Jan. 30 (1913).

‡ Proc. Roy. Soc. A, vol. lxxxviii, p. 428.

same time a careful study of the general reflexion from these crystals has thrown some light on the nature of the ordinary X-rays.

### *The Apparatus.*

In the course of the present work the experimental arrangements have frequently been refined, and the methods which finally proved satisfactory will alone be described. The general plan was to allow a beam of X-rays to fall on to a crystal and examine the radiation from its surface by means of a movable ionization chamber. This part of the apparatus will in future be called the detector.

*The X-rays.*—The source of the X-rays was a Müller tube with platinum target. The beam of radiation was limited by two parallel lead slits 3 cm. long, 0.5 mm. broad, and 20 cm. apart. They are marked P and Q in fig. 1, which

Fig. 1 B.

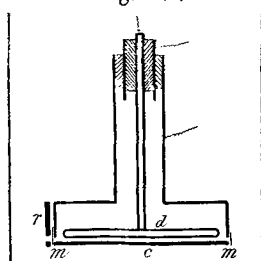
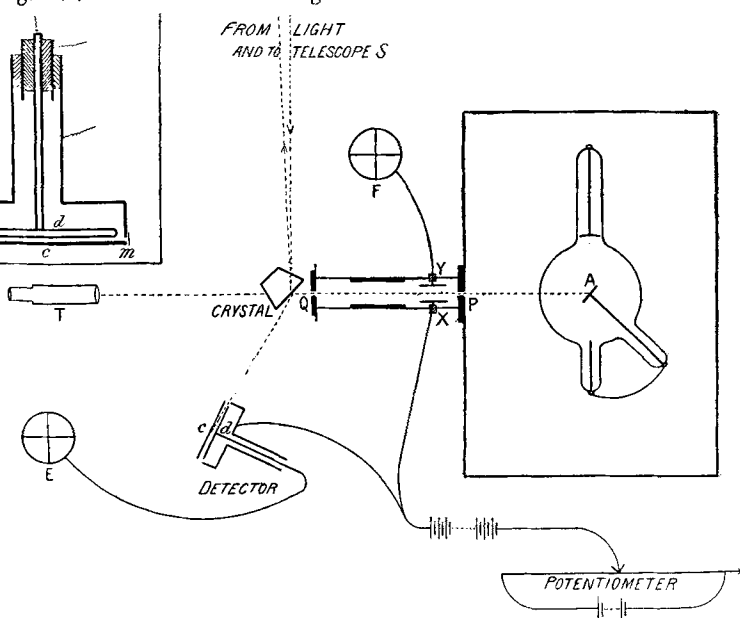


Fig. 1.



shows the general arrangement of the apparatus. The slits stopped the radiation from all but a small strip of the target, and it was therefore important that the cathode stream falling on it should be focussed to a point. The tube was excited by a coil with a Sanax mercury break. A current of some 6 amps. was sent through the primary circuit, and

this gave a current of about 0.3 milliamp. through the tube. The tube was enclosed in a thick lead box, and the coil surrounded by an earthed tin case.

*The Reflector.*—Three different crystals were used for reflecting the radiation: rocksalt ( $\text{NaCl}$ ), selenite ( $\text{CaSO}_4, 2\text{H}_2\text{O}$ ), and potassium ferrocyanide ( $\text{K}_4\text{FeCy}_6, 3\text{H}_2\text{O}$ ). The last of these was a magnificent specimen, with a perfect face 6 cm. square. A perfect face of selenite was easily obtained by cleavage. All the available specimens of rocksalt were contorted, but one was eventually found with a sufficient area of flat surface. The faces used were a cube face in rocksalt, and the principal cleavage-planes in selenite and potassium ferrocyanide.

*The Mounting.*—A dismantled spectrometer was used to carry the principal parts of the apparatus. The crystal was set up with its surface on the axis of the spectrometer. The slits P and Q were fixed in the place of the collimator, and the detector on the arm which usually carries the telescope, while each crystal was permanently mounted on a table, the legs of which could rest in the V grooves of the prism table. The slits were parallel to this axis and pointed at it. The beam therefore struck the crystal surface in a fine line, and however the crystal was turned, the line remained stationary on the axis of the spectrometer. This line was perpendicular to the direction of the incident beam and so the angles of incidence of the rays at the ends and at the centre of the slit were almost exactly equal.

*The Adjustment.*—A telescope T was kept permanently pointing at the axis of the spectrometer. The focus spot on the target A, the slits P and Q, and the surface of the crystal were set accurately in the plane defined by the axes of the telescope and spectrometer. The first of these adjustments was made by covering with lead all but the centre of P, and moving the X-ray tube until the bright spot on a fluorescent screen placed at Q was brought on to the axis of the telescope.

*Measuring the angles.*—The position of the arm which carried the detector could be read on the divided circle of the spectrometer to within  $30''$ . The prism-table, however, carried no vernier and a second telescope S was used to define the position of the crystal. A piece of plate-glass was fixed on to the crystal at such an angle that when the crystal surface was parallel to the X-ray beam a fixed light was reflected by the glass on to the cross-wires of S, which was 3 metres distant. The crystal was first set to this standard position and then turned together with the detector through

the required angle. In practice the glass was never fixed on the crystal at exactly the correct angle, and the error had to be determined for each crystal by optical methods. At a later stage, after the fact that the X-rays obey the ordinary laws of reflexion had repeatedly been verified, it was found easier to calculate the error by setting crystal and detector at the estimated position of reflexion and then turning both together until the reflected beam entered the detector.

*The Path of the Rays.*—The detector turned on the telescope arm of the spectrometer, and was always 20 cm. from the crystal. The distance to the crystal from the target was 40 cm., and so the rays always passed through 60 cm. of air. A large fraction of the softest constituents of the beam was therefore absorbed before reaching the detector. Much would be gained by removing the air from the path of the rays, but this refinement would have been inconvenient, because the arrangement of slits and crystal had constantly to be altered. There were two reasons for putting the detector so far from the crystal. In the first place the effect of the general scattered radiation, which comes from the crystal in all directions, was thereby diminished. In the second place the direction of the reflected beam could be determined with increased accuracy. The distance between P and Q had to be made large to ensure the rays being sufficiently parallel. Distance, combined with the use of two fine slits, is the only possible way of collimating a beam of X-rays.

*The Detection of the Rays.*—The fine slits, the distance from the target, and the small reflexion coefficient of the radiation together reduced the intensity of the beam which reached the detector to a minute fraction of the total radiation. It was therefore necessary to measure this intensity by a method which should be both sensitive and fairly accurate over a very wide range. A method was also needed which would discriminate against the natural ionization, which was very troublesome. The following plan proved to be quite satisfactory. The reflected beam struck a metal plate at almost glancing incidence. Laub\* has shown that under these conditions the emission of corpuscular radiation by X-rays is very greatly increased. This effect was found to occur equally for the reflected radiation. The metal plate was surrounded by helium at nearly atmospheric pressure. The ionization of the helium by the corpuscular radiation was greatly increased by using the principle of ionization by collision. An electric field was

\* *Ann. d. Physik*, xxvi. p. 712 (1908).

maintained in the gas which was just not sufficient to cause the passage of a spark. Townsend has shown that both positive and negative ions then acquire sufficient speed to produce fresh ionization, and that the current through the gas is in this way multiplied by a large but constant factor. This method was first employed by Griffith \*, who used it in quantitative measurements of the photoelectric effect. Rutherford and Geiger † in their electrical method of counting  $\alpha$  particles were the first to use helium.

*The Detector.*—The detector was a gas-tight brass box 10 cm. long, 2 cm. broad, and  $4\frac{1}{2}$  cm. deep, fixed to the telescope-arm by an ebonite block. It is shown in section in fig. 1 B. The rays entered at one end by a thin mica window *m* surrounded by a lead screen *r*. They fell at a very oblique angle on the plate *c*. This plate was one side of the box, which was kept charged to a potential of about 800 volts. A plate *d* accurately parallel to *c* and 2.8 mm. from it was connected to an electrometer. The connecting wire, which was insulated from the box by ebonite plugs and a guard-ring, passed to the axis tube of the spectrometer, and down it to the electrometer. It was therefore not disturbed by moving the detector.

*The Helium.*—Ionization by collision may be obtained by the use of helium at a very much higher pressure for a given potential difference than is possible with air. The great ease with which helium may be purified is also an advantage. It was purified by passing it over charcoal cooled in liquid air, and it was then pumped into the detector, which was temporarily connected to the purifying apparatus by a glass tube fitted with ground joints. It was found advisable to clean the helium every few weeks, since the natural ionization in the detector, at first very small, increased markedly after some time. This curious effect suggests that, as in the stagnant air of cellars, radium emanation gradually diffused from the walls of the box. The properties of pure helium are affected to an extraordinarily marked degree by the presence of traces of impurity. It was therefore not surprising that when the apparatus was first used, the factor by which a given difference of potential increased the ionization current was found to fall off very rapidly, owing to the contamination of the helium by traces of other gases coming from the walls of the box. This trouble was overcome by using helium mixed with a trace of air. The sparking potential was in this way raised by about a hundred volts,

\* Griffith, *Phil. Mag.* xiv. p. 297 (1907).

† Geiger and Rutherford, *Phil. Mag.* xxiv. p. 618 (1912).

and the gas was then no longer sensitive to impurities. After some months this trouble ceased, and the helium could be used almost pure.

*The Measurements.*—The factor by which the ionization current between *c* and *d* was increased by collision could be adjusted to any desired value by altering the potential of *c* by a few volts with the aid of a potentiometer. A factor of 50 was most frequently used, and it was usually not convenient to work with a factor much greater than 100. When the multiplication was too great the readings became unsteady and the natural ionization gave trouble. In order to detect variations in the primary X-rays a pair of parallel plates X and Y (fig. 1) were placed in front of the slit P. The beam of rays passed between them, and the resulting saturation ionization current was measured by a second electrometer F, with which was connected a large capacity.

#### *General Properties of the Reflected Radiation.*

Explanations of the “reflexion” of the X-rays are based on the assumption that the reflected beam is due to the co-operation of a number of the waves of scattered radiation which come from the atoms traversed by the primary beam. It is well known that the scattered radiation is in every way similar to the rays which excite it. A description will now be given of some simple experiments which show that this is equally true in the case of the “reflected” radiation.

(1) *Ionization of gases.*—A fine beam of radiation reflected from mica was sent straight down the detector and did not touch the plates *c* and *d*. Considerable ionization was produced in air, and slight ionization in helium.

(2) *Corpuscular radiation.*—The detector was moved a little to one side. The beam now touched *c* or *d*. The ionization in air was somewhat increased; the ionization in helium was increased some ten times. As the angle at which the beam hit the metal plate was altered, the ionization changed rapidly, being largest at almost glancing incidence.

(3) *Ionization at different pressures.*—The detector was set slightly aslant, so that the whole reflected beam fell on *c*, and the corpuscular radiation was a maximum. The ionization was measured in air at pressures between 10 cm. and an atmosphere. The crystal was then removed and the same process carried out with the primary beam. The two beams happened to be equally penetrating and the two curves were indistinguishable. In both cases the effect of the corpuscular radiation and its absorption by the air at the higher pressures

was very marked. Potassium ferrocyanide was here used as reflector.

(4) *Absorption*.—The primary and reflected beams were usually not equally penetrating. The cause of this was thought to be the unequal reflexion of different constituents. It was conceivable, however, that a special type of radiation was manufactured by the crystal. This point was tested in an experiment in which mica was used as reflector, and the reflected beam happened to be considerably the more penetrating. An aluminium sheet was placed alternately in the path of the primary and reflected beams. The reflected radiation was the same in the two cases.

All these experiments show clearly that the primary and reflected radiations consist essentially of the same constituents, but that these constituents are present in different proportions in the two beams.

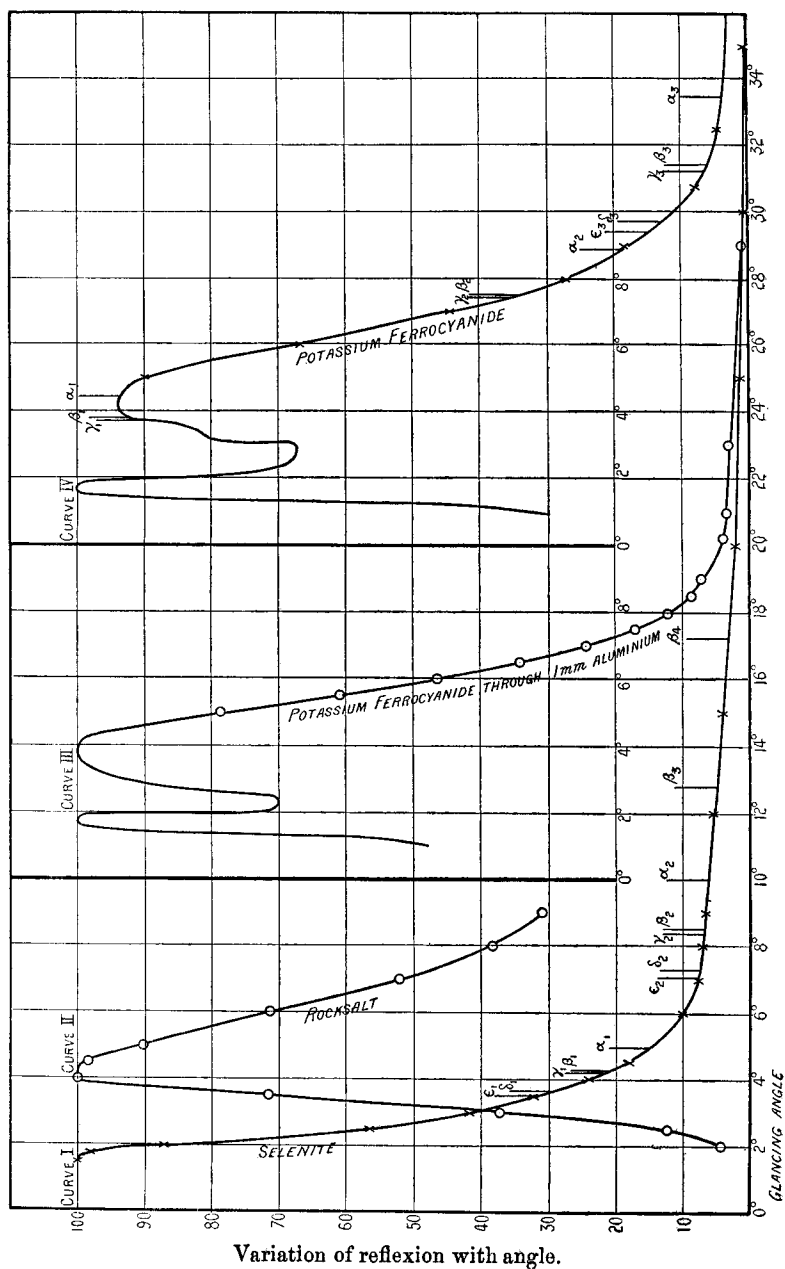
*The General Reflexion at different Angles of Incidence.*

The next step was to determine how the intensity of reflexion depends on the angle at which the radiation strikes the surface of the crystal. It was soon apparent that the strongest reflexion occurs at almost glancing incidence, and that at an angle less than  $55^\circ$  the reflexion becomes too small for measurement. For the future therefore the glancing angle, that is the angle made by the rays with the surface of the crystal, will be used instead of the angle of incidence. The crystals used were all so thick that at no angle would an increase in thickness have improved the reflexion. For the smaller angles the primary beam was limited by fine slits at P and Q. As the crystal was turned round the detector was turned through twice the angle, so that it always caught the whole reflected beam. At the larger angles it was necessary to use wider slits, and then the detector only intercepted the central part of the reflexion. When changing the slits careful comparative measurements were made at one or two angles, so that all the results could be reduced to a common standard. There were two reasons for using fine slits at small angles. In the first place it was essential that the whole beam should strike the crystal, and no radiation get past it. Secondly, the variation of reflexion with angle was sometimes so rapid that it was necessary to avoid any sensible divergence in the beam. This was particularly important near those angles at which the selective reflexion occurred. When these special angles had been accurately measured, they could always be avoided in finding the shape of the general curve. In giving the results obtained for the general reflexion we have as far as



possible eliminated the effect of selective reflexion, since the prominence of the latter in the general intensity curves depends entirely on the arrangement of the slits. In the curves given in fig. 2 the special angles are marked with letters, the meaning of which will be explained later. In these curves the intensity is plotted against the glancing angle. It was found that the shape of a curve was greatly influenced by the state of the tube. As an example of this, an experiment may be mentioned in which rocksalt was used as the reflector. The radiation from the tube was allowed to become gradually softer. The maximum of the curve was at first close to  $4^\circ$ . During the course of the experiment it shifted systematically towards the broader angles and finally approached  $6^\circ$ . Doubtless if a great part of the soft radiation had not been absorbed in its passage through the air, the maximum would have occurred at a still larger angle. It was therefore most important that the state of the tube should remain constant, and the results suffer somewhat from the impossibility of fulfilling this condition accurately. It was found that the tube usually remained much steadier if it was kept warm by burning an electric lamp inside the lead box. The tube was run for half a minute at regular intervals of two minutes. As soon as the rays were cut off, the electrometer E was disconnected from the detector, in order to avoid the natural ionization in the detector during the time taken by the needle to come to rest. The ratio of the readings of the two electrometers E and F was taken to be a measure of the intensity of reflexion, after allowing for the natural effect in the detector. In the more accurate work the current through the detector was measured by a balance method. It was found in practice that F did not give an entirely reliable measure of the intensity of the primary beam, probably because the focus point of the cathode stream was liable to shift, and this affected F very little, while it made a great difference in the amount of radiation which got through the second slit. It was therefore necessary to take the reflexion at one angle as a standard and return to it after every few readings. In fig. 2, curve I. refers to reflexion from selenite. The state of the tube remained very nearly constant during the course of the experiment, which took several days to complete. Curve II. was taken with rocksalt and curve IV. with potassium ferrocyanide. The state of the tube was roughly the same in the three cases and 1 mm. of aluminium cut down the primary beam by 50 or 60 per cent. Curve III. was taken with the ferrocyanide crystal after hardening the primary beam by passing it through 1 mm. of aluminium.

Fig. 2.



Much information on the way in which constituents of differing penetrating power are reflected at different angles can be obtained from a comparison of curves III. and IV. A direct determination was also made of the extent to which the rays, reflected at different angles from rocksalt, were absorbed by 1 mm. of aluminium. The state of the tube was the same as for curve II. The absorption was found to be 23 per cent. at  $3^\circ$ , 33 per cent. at  $4^\circ$ , 53 per cent. at  $6^\circ$ , and 63 per cent. at  $8^\circ$ , while for the primary beam it was 59 per cent. The measurements at very small angles were not easy to make, since, if the adjustment either of the slits or of the crystal was slightly in error, part of the primary beam failed to strike the surface of the crystal. This point was carefully tested optically, and the diminution of the efficiency of reflexion from ferrocyanide and rocksalt at very small angles is certainly a genuine effect. A similar result was obtained with selenite but was not accurately measured. The upper parts of curves III. and IV. have been drawn from the results of a number of experiments. All showed the general characters depicted, though the results obtained on different days were not in perfect agreement. These curves will be discussed later, after the simpler case of reflexion of monochromatic radiation has been considered.

Even at the most favourable angle a crystal reflects only a small fraction of the general incident radiation. The actual reflexion coefficient depends to a certain extent on the quality of the primary radiation. Under the usual working conditions it was found that the selenite crystal gave a maximum reflexion coefficient  $\cdot 0037$  at  $1\frac{1}{2}^\circ$ , while the ferrocyanide gave a maximum of  $\cdot 0035$  at  $1\frac{3}{4}^\circ$ . In the earliest experiments \* a reflexion coefficient of  $\cdot 01$  to  $\cdot 02$  was found for mica, but it is probable that in this case the surface was slightly curved and chanced to focus the very broad incident beam into the narrow opening of the detector. Since then mica has been little used, owing to the difficulty in obtaining a flat surface, but in the later work the reflexion coefficient found for this crystal has certainly not been greater than about  $\cdot 005$ .

### *The Selective Reflexion.*

It has already been mentioned that, while these experiments were in progress, Prof. W. H. and W. L. Bragg † discovered that certain crystals reflected exceptionally well at three

\* Moseley and Darwin, *loc. cit.*

† W. H. and W. L. Bragg, Proc. Roy. Soc. A. lxxxviii. p. 428 (1913).

special angles. They found that at each of these angles a particular type of radiation was selectively reflected. The special angles differed from crystal to crystal but the three types of radiation were always the same. They explained these results by assuming that the platinum target emitted three characteristic types of monochromatic waves. The beams reflected from the successive layers of atoms in the crystal generally destroyed each other by interference. When, however, the wave-length  $\lambda$ , the distance  $d$  between the layers, and the glancing angle  $\theta$  were connected by the relation

$$n\lambda = 2d \sin \theta . . . . . (1)$$

the separate beams reinforced one another, and the monochromatic radiation was selectively reflected. The three types of radiation represented three different values of  $\lambda$ , and in the reflexion from rocksalt special angles were found corresponding to  $n=1$  and to  $n=2$ . These angles may by analogy be said to refer to reflexion in the first and second order.

We have studied in detail the selective reflexion from the principal cleavage-planes of rocksalt, selenite, and potassium ferrocyanide. The most complete search was made in the case of ferrocyanide. The general radiation is reflected at all angles, while the selective reflexion may for practical purposes be said to occur only at precisely the correct angle of incidence. For this reason the ratio of the selective to the general reflexion is greatly increased by limiting the breadth of the slits and so increasing the parallelism of the primary beam. Unfortunately, a very small rotation of the crystal will then remove all traces of the selective effect. It therefore proved necessary to take readings with the crystal set at every  $5'$  of arc between  $1^\circ$  and  $14^\circ$ . In the case of selenite most of the special angles were already approximately known from the singularities in the original intensity curves, the meaning of which had not been appreciated at the time. Prof. Bragg most kindly told us the positions of his three special angles in rocksalt, and a search for others was made only in those places which theory suggested as possible. The positions of the special angles were first found roughly, and then the crystal was turned minute by minute until the angle which gave the maximum reflexion was definitely determined. The opening in the detector was wide enough to include the whole reflected beam, and it was made to follow this beam when the crystal was rotated. Several of the special angles were found with the crystal set on both sides of the

primary beam. As soon as the correct position was determined, the crystal was turned through an angle  $A$  into the position defined by the telescope  $S$ , the lamp, and the plate glass. The corresponding position was then found on the other side, and the crystal turned through the angle  $B$  into the standard position, using this time the other side of the glass. As the faces of the glass were strictly parallel, the special angle was given by  $\frac{1}{2}(A+B)$ .

The values found for the special angles are tabulated below. Those in Tables I. and II. are probably correct to within about  $1'$ , but in Table III. the error may be twice as great.

*Angles of Special Reflexion.*

TABLE I. Rocksalt, NaCl. Cube face.	First Order...	$\alpha_1$ .	$\beta_1$ .	$\gamma_1$ .	$\delta_1$ .	$\epsilon_1$ .
	$\theta$ .....	$13^\circ 31'$	$11^\circ 28'$	$11^\circ 17'$	$9^\circ 47'$	$9^\circ 29'$
	$\sin \theta$ .....	$\cdot 2337$	$\cdot 1988$	$\cdot 1956$	$\cdot 1699$	$\cdot 1647$
	$\sin \theta \times 5\cdot030$ ...	$1\cdot176$	<u><math>1\cdot000</math></u>	$\cdot 984$	$\cdot 855$	$\cdot 829$
TABLE II. Potassium Ferrocyanide, $K_4FeCy_6 \cdot 3H_2O$ . Principal cleavage face.	First Order...	$\alpha_1$ .	$\beta_1$ .	$\gamma_1$ .	$\delta_1$ .	$\epsilon_1$ .
	$\theta$ .....	$4^\circ 26'$	$3^\circ 46\frac{1}{2}'$	$3^\circ 43\frac{1}{2}'$	not	found.
	$\sin \theta$ .....	$\cdot 0773$	$\cdot 0657$	$\cdot 0650$	—	—
	$\sin \theta \times 15\cdot11$ ...	$1\cdot168$	$\cdot 995$	$\cdot 982$	—	—
	Second Order...	$\alpha_2$ .	$\beta_2$ .	$\gamma_2$ .	$\delta_2$ .	$\epsilon_2$ .
	$\theta$ .....	$8^\circ 55\frac{1}{2}'$	$7^\circ 33\frac{1}{2}'$	$7^\circ 28\frac{1}{2}'$	not	found.
	$\sin \theta$ .....	$\cdot 1551$	$\cdot 1315$	$\cdot 1301$	—	—
	$\frac{1}{2} \sin \theta \times 15\cdot11$ ...	$1\cdot172$	$\cdot 994$	$\cdot 983$	—	—
	Third Order...	$\alpha_3$ .	$\beta_3$ .	$\gamma_3$ .	$\delta_3$ .	$\epsilon_3$ .
	$\theta$ .....	$13^\circ 28\frac{1}{2}'$	$11^\circ 27'$	$11^\circ 16'$	$9^\circ 45\frac{1}{2}'$	$9^\circ 27\frac{1}{2}'$
	$\sin \theta$ .....	$\cdot 2330$	$\cdot 1985$	$\cdot 1954$	$\cdot 1695$	$\cdot 1643$
	$\frac{1}{2} \sin \theta \times 15\cdot11$ ...	$1\cdot174$	<u><math>1\cdot000</math></u>	$\cdot 984$	$\cdot 854$	$\cdot 828$
TABLE III. Selenite, $CaSO_4 \cdot 2H_2O$ . Principal cleavage face.	First Order...	$\alpha_1$ .	$\beta_1$ .	$\gamma_1$ .	$\delta_1$ .	$\epsilon_1$ .
	$\theta$ .....	$4^\circ 59'$	$4^\circ 16\frac{1}{2}'$	$4^\circ 13\frac{1}{2}'$	$3^\circ 39'$	$3^\circ 32'$
	$\sin \theta$ .....	$\cdot 0869$	$\cdot 0745$	$\cdot 0736$	$\cdot 0637$	$\cdot 0616$
	$\sin \theta \times 13\cdot53$ ...	$1\cdot175$	$1\cdot009$	$\cdot 997$	$\cdot 861$	$\cdot 834$
	Second Order...	$\alpha_2$ .	$\beta_2$ .	$\gamma_2$ .	$\delta_2$ .	$\epsilon_2$ .
	$\theta$ .....	$9^\circ 58\frac{1}{2}'$	$8^\circ 30'$	$8^\circ 22\frac{1}{2}'$	$7^\circ 17'$	$7^\circ 3'$
	$\sin \theta$ .....	$\cdot 1731$	$\cdot 1478$	$\cdot 1457$	$\cdot 1268$	$\cdot 1228$
	$\frac{1}{2} \sin \theta \times 13\cdot53$ ...	$1\cdot172$	<u><math>1\cdot000</math></u>	$\cdot 985$	$\cdot 858$	$\cdot 830$

The positions of the special angles are indicated on the curves in fig. 2. The five angles found for rocksalt agree in position with the three found by Bragg. His slits were too broad to separate  $\beta$  from  $\gamma$  or  $\delta$  from  $\epsilon$ . If the way in which the atoms are packed in the simple cubical crystal rocksalt is known, the wave-lengths of these five characteristic radiations can be determined.

If  $N$  is the number of molecules in 1 gram mol.,  $M$  the molecular weight, and  $\sigma$  the density of the rocksalt, the number of molecules per c.c. is  $\frac{N\sigma}{M}$ . This number is, however,  $\frac{K}{d^3}$ , where  $d$  is the distance between successive layers of atoms and  $K$  a numerical constant, the value of which depends on the arrangement of the atoms in the crystal. Hence

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

$$\text{and} \quad \frac{K}{d^3} = \frac{N\sigma}{M}.$$

Now  $N = 6.22 \times 10^{23}$ ,  $M = 58.46$ , and  $\sigma$  was found to be 2.167. This gives  $d = K^{\frac{1}{3}} \times 3.513 \times 10^{-8}$  cm.

It follows that the wave-lengths of the five radiations  $\alpha, \beta, \gamma, \delta, \epsilon$  are 1.642, 1.397, 1.375, 1.194,  $1.157 \times K^{\frac{1}{3}} \times 10^{-8}$  cm. It is probably an accident that the first, second, and fourth, also the first, third, and fifth of these numbers are in geometrical progression. These values for the wave-lengths are no doubt slightly too large, as crystals are apt to contain minute inclusions of gas or liquid, which would make the experimental value of  $\sigma$  slightly too low. The absorption coefficient of the characteristic radiation  $\beta$  in aluminium was found to correspond to that of the K radiation from an element of atomic weight about 7.55. In order to excite this radiation a cathode particle should have a velocity\* about  $7.55 \times 10^9$  cm. per sec., which corresponds to an energy content  $2.5 \times 10^{-8}$  erg. If Planck's formula holds good the energy of the radiation  $\beta$  should be given by  $h\nu$ , where  $h$  is  $6.455 \times 10^{-27}$  erg. sec.† and  $\nu$  is the frequency of  $\beta$ . Therefore

$$h\nu = h \cdot \frac{c}{\lambda} = 1.37 \times K^{-\frac{1}{3}} \times 10^{-8} \text{ erg.}$$

If all the molecules are arranged in a simple cubical pattern

\* Whiddington, Proc. Roy. Soc. A. lxxxv. p. 323 (1911).

† Westphal, *Deutsch. Phys. Gesel.* xxi. p. 987 (1912).

$K=1$ ; and if it is the atoms which are so arranged,  $K=\frac{1}{2}$ . If, however, the atoms are arranged at the corners and the centres of faces of cubes,  $K=\frac{1}{8}$ . As the arrangement of the atoms in a salt crystal is still in doubt, there remains this factor of uncertainty in the determination of the wave-lengths.

It will be seen by comparing the results obtained with the three different crystals that equation (1) is accurately satisfied. First, in Tables II. and III. the five radiations will be seen repeated in different orders, that is with different values of  $n$ . Higher orders were also observed but not measured accurately. Secondly, it is clear that  $d$  is a constant for the same crystal, since the sines of the angles have the same ratio in each case. The characteristic radiations  $\beta_1$  and  $\gamma_1$  in ferrocyanide and in selenite were too close together to be properly separated. It was, however, quite clear in both cases that two components were present, and that they were about  $3'$  apart. For the purpose of finding wave-lengths the values found in the higher orders are, of course, the more accurate. Measurements of the absorption in aluminium of  $\beta_3$  from ferrocyanide and  $\beta_1$  and  $\beta_2$  from selenite showed that these were all the same radiation. The absorption coefficients were found by taking the difference between the intensities of reflexion at the special angle, and at a neighbouring angle, first with the detector bare and then with aluminium in the way. The approximate values found for the mass-absorption in aluminium of the four radiations  $\alpha_3, \beta_3, \gamma_3, \delta_3$  from ferrocyanide gave 34, 22, 19, 16 (g. per sq. cm.)<sup>-1</sup> respectively. The absorption of  $\epsilon_3$  was not measured, as this type of radiation was much weaker than the others. All these radiations were greatly reduced by absorption in passing through 60 cm. of air on their way to the detector, and no attempt was made to obtain at all accurate values for the absorption coefficients with this apparatus. As one would anticipate, the absorption increases rapidly with the wave-length. Thus the wave-lengths of  $\alpha$  and  $\delta$  are in the ratio of 1.37:1, while the absorption coefficients are roughly as 2.12:1. The so-called L characteristic radiation from platinum, which Chapman found to have a mass-absorption coefficient of 22.2\* in aluminium, seems to correspond roughly with the monochromatic radiation  $\beta$ . The absorption in aluminium of  $\beta_1$  from ferrocyanide appeared to be abnormally low. Here, however, the characteristic radiation contributes only 25 per cent. to the whole reflexion, and the discrepancy may be explained by supposing that the general reflexion, which is

\* Proc. Roy. Soc. A. lxxxvi. p. 439 (1912).

much more penetrating, had a slight maximum in the neighbourhood of this angle.

The different orders and different types of characteristic were by no means equally conspicuous. The relative intensities after subtracting the effect of the general reflexion are given for the case of ferrocyanide in Table IV. The

TABLE IV.  
*Intensity of Selective Reflexions.*

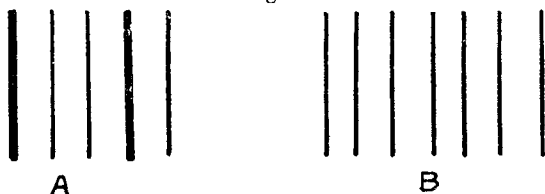
	$\alpha$ .	$\beta$ .	$\gamma$ .	$\delta$ .	$\epsilon$ .
First Order .....	38	62		—	—
Second Order.....	—	28	—	—	—
Third Order .....	58	100	74	44	14
Corrected for absorption by air }	99	140	100	56	17

relative intensities found for the different types of radiation have little definite meaning. In the first place they have been cut down by absorption to very different extents. If allowance were made for absorption in glass, air, and mica, probably the true intensities would be in the same order as the wave-lengths,  $\alpha$  being the strongest and  $\epsilon$  much the weakest. In the second place, definite evidence was obtained that the relative strengths depended on the state of the tube. The weaker types, indeed, almost disappeared sometimes as the tube became hard, and flashed up again when its condition was altered. The strength of the radiation  $\gamma$  relative to its neighbour  $\beta$  altered from 70 per cent. to 90 per cent. in the course of one short experiment. Comparison between the different orders is more profitable. It will be seen from Table IV. that in ferrocyanide the third order was enormously the most conspicuous, since the general reflexion was so much less intense at these large angles. The sixth order was quite easily found, the fourth order not at all. These facts can be very simply explained. If the crystal structure completely repeats itself only every three layers, the distance between the first and fourth layers gives the fundamental distance  $d$  in equation (1). It is possible either that the three layers are at nearly but not quite the same distance apart, or that every third layer is especially rich in the heavy atoms, iron for example, which most



efficiently scatter the primary radiation. The two cases are shown diagrammatically in fig. 3. In either case the different orders will all be present, but the third order and its multiples

Fig. 3.



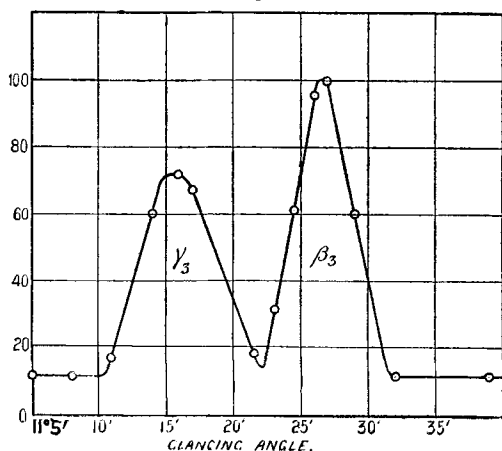
may well be much stronger than the others, because in the third order all the layers cooperate, while in the first order only every third layer does so. If all the layers become both similar and similarly situated, the previous third order becomes the fundamental and the others disappear. This is the case with rocksalt, for which no subsidiary orders could be found. In this case  $d$  is presumably the distance between successive layers, and this happens to be almost exactly the same as the average distance in ferrocyanide. The behaviour of selenite is similar. Here although the intensity of the first order was more than twice that of the second, the latter was the more conspicuous. The fourth order also was much more conspicuous than the third. In this case the crystal structure evidently repeats itself every second layer. This seems to offer a simple method of investigating the structure of crystals. Another way of looking at the matter, which is really equivalent, is to regard the most conspicuous order as the fundamental, and call all earlier orders "ghosts."

In the simple case of rocksalt the intensity falls off for the higher orders much more rapidly than theoretical considerations would suggest. This is doubtless due to the effect of the temperature oscillations of the atoms in the crystal. These oscillations will not affect the resolving power of the crystal, regarded for the moment as a grating, but will greatly reduce the intensity of the reflected beam. For suppose an atom displaced a distance  $b$  out of its plane. The wave scattered by this atom will have a phase difference from those of the other atoms in the plane of  $2b \sin \theta \cdot \frac{2\pi}{\lambda}$  or  $\frac{b}{d} 2n\pi$ , where  $d$  is the distance between successive planes; so the phase deviations are more marked in the higher orders and the relative intensity greatly reduced.

The next point investigated was the length of the train of

waves which makes up a characteristic radiation. An attempt was made to find over what range of angles reflexion of monochromatic radiation occurs. The beam was restricted by 0.5 mm. slits at P and Q, and in order to reduce the general reflexion still further a 0.5 mm. slit was placed in front of the detector. The variation of the intensity of reflexion with the angle at which the radiation struck the crystal of ferrocyanide was then measured in the neighbourhood of the special angles  $\beta_3$  and  $\gamma_3$ . Care was taken that the reflected beam always entered the detector. The curve is shown in fig. 4. The two maxima are caused by the

Fig. 4.



selective reflexions at  $\beta_3$  and  $\gamma_3$ , while the flat ends of the curve represent the general reflexion, which under these conditions was small. The shape of this curve is that which one would expect with a beam coming from a focus spot on the target not more than 0.5 mm. wide and limited by the second slit, if only those rays were reflected which struck the crystal at precisely the special angles  $\beta_3$  and  $\gamma_3$ . Now the characteristic rays are known to form only a fraction of the primary X-rays, and so the large intensity of the reflected beam, when the crystal is set at the special angle, shows that over a finite range of angles of incidence the characteristic rays can be reflected. This range cannot be estimated from the curve, as the exact size of the focus spot is not known, but the shape of the curve and also other experiments show that it is extremely narrow. The whole range over which reflexion can occur is certainly not more than 5', and from

this it may be concluded that these characteristic radiations are very nearly monochromatic. A mathematical treatment of this question shows that if these radiations are damped waves, the damping cannot reduce the amplitude to half value in less than 50 vibrations.

Probably the number of vibrations is really much greater than this. If it is assumed that the system which emits the characteristic radiation is a freely vibrating electron, the vibrator will, owing to its very high frequency, be appreciably damped by radiation. The equation of motion of such an electron is according to the ordinary electromagnetic theory

$$m \frac{d^2 x}{dt^2} + m \left( \frac{2\pi e}{\lambda} \right)^2 x - \frac{2}{3} \frac{e^2}{c^3} \frac{d^3 x}{dt^3} = 0.$$

The displacement is therefore  $e^{-\sigma \frac{ct}{\lambda}} \cos 2\pi \cdot \frac{ct}{\lambda}$  where  $\sigma = \frac{4\pi^2}{3} \cdot \frac{e^2}{mc^2 \lambda}$  ( $e$  is the charge,  $m$  the mass of the electron, and  $c$  the velocity of light). This gives rise in the case of a radiator with the frequency of the characteristic  $\beta$  to a wave, of which the amplitude is damped to half value in  $2680 K^{\frac{1}{3}}$  wave-lengths. A wave damped to this extent will be reflected over a range of angles of incidence large enough to be capable of being detected experimentally. An attempt will be made to measure this range, as the matter is of great theoretical interest.

The same experimental arrangements were used to test how precisely the laws of reflexion are obeyed. The slits were far too broad to introduce diffraction in the case of radiation of such short wave-length. Attempts are, however, sometimes made to explain the corpuscular properties of X-rays by endowing them with a very restricted wave-front. If such a beam struck a crystal the scattering from the atoms would give a system of waves, of which the interference would show the same arrangement as the diffraction pattern round a small hole. The crystal was set permanently at the special angle  $\beta_3$ , so that unless diffraction occurred, the reflected radiation would consist almost entirely of characteristic rays proceeding in a parallel beam 0.5 mm. wide. The opening of the detector, 0.5 mm. wide, was moved across this beam. The result showed no trace of diffraction. It was calculated that in each reflecting plane an area containing at least a million atoms must have been concerned in producing so perfect a reflexion.

*Discussion of the General Reflexion.*

We have seen that monochromatic radiation is reflected only at certain special angles, but that reflexion of the general radiation from an X-ray tube occurs for all angles of incidence. This shows that the general X-rays resemble white light. The primary radiation may therefore, for convenience, be imagined to contain constituents with wave-lengths varying continuously over a finite range, though, as a matter of fact, these constituents are produced by the analysing property of the crystal. These rays are sorted out into a continuous spectrum of monochromatic waves, and only those constituents are reflected at a glancing angle  $\theta$  of which the wave-lengths satisfy the condition  $n\lambda = 2d \sin \theta$ . With the help of this condition the variation of the reflexion with angle can be predicted in general terms.

Suppose that the energy in the spectrum is largely concentrated into a limited range of wave-lengths. For very small values of  $\theta$  the intensity of reflexion will be small, as only the constituents with the shortest wave-lengths will be present even in the first order. The radiation will here be exceptionally penetrating, since the absorption coefficient in aluminium increases very rapidly with the wave-length. As  $\theta$  is increased the intensity reaches a maximum, which roughly corresponds with that component which has the greatest intensity in the spectrum. At greater angles the intensity falls off, and the radiation becomes continuously softer. Here the effect of the short wave-lengths reflected in the second and higher orders begins to become noticeable. Under the conditions of these experiments but few of the softer types of radiation could reach the detector. At large angles therefore the effect of the constituents reflected in the first order will become small, and the penetrating power of the radiation will no longer even approximately correspond to it. Eventually the reflected beam will consist of such a mixture of constituents reflected in various orders that its quality will become approximately constant. As the X-ray tube is softened the angle for which the reflexion is a maximum will of course become greater.

The reflexion found for rocksalt is in perfect agreement with this theory. In fig. 2, curve II., the maximum close to  $4^\circ$  and the very rapid decrease in intensity for smaller angles are conspicuous. For larger angles the curve falls off less steeply, and at  $6^\circ$  the second-order spectrum is no doubt just making its appearance. We know, however, from the experiments with monochromatic rays that the second order in rocksalt has very little intensity compared with the first.

Its effect will only become important when the constituent of which the wave-length corresponds with the first order is very weak. The steady increase in the absorption coefficient as the angle is increased from  $3^\circ$  to  $8^\circ$ , and the way in which the maximum shifts towards larger angles as the tube softens, have already been described. The effect of the different orders overlapping is very marked in the neighbourhood of the special angles  $\alpha_1$ , &c. Here the characteristic radiation, reflected in the first order, is stopped by a few tenths of 1 mm. of aluminium. A sheet of this thickness has, however, very little effect on the general reflexion at a neighbouring angle. In the reflexion from selenite the maximum intensity in the first order should occur at about  $1^\circ 5'$ . In this case, however, the second order is more important than the first, so that a strong second-order effect, with maximum at about  $3^\circ 0'$ , will be superimposed on it. The combined effect shows in curve I. a strong maximum at a rather smaller angle than would be anticipated, probably partly because a somewhat harder tube was used in this case. There is also, however, an important difference between the behaviour of monochromatic radiation and a continuous spectrum. In the latter case, owing to the increased dispersion, the second-order spectrum will be spread out over twice the range of angles occupied by the first order, and the intensity at any angle will consequently be halved. This is well shown in curves III. and IV. taken with the ferrocyanide crystal. Here the very narrow first order and the broader third order are conspicuous, while curve IV. shows also the effect of the weaker second order. At very fine angles curve IV. falls away much more than curve III., and at broader angles the reverse occurs, all in accordance with theory.

In the reflexion at small angles from rocksalt the first order predominates, and this gives an opportunity of finding the distribution of energy in the spectrum of the X-rays. The complete mathematical treatment of this question is much too long for this paper, and a simple argument which leads to the same conclusion must here suffice.

It is probable in the first place that, if no cooperation or interference took place, the amount of energy scattered from the surface of a thick sheet of any material, when struck by the X-rays, would be approximately independent of the quality of the radiation, and would be proportional to  $1 + \cos^2 \phi$  where  $\phi$  is the angle between the primary and scattered rays. The experiments of Crowther\* have shown that when  $\phi$  is small the scattering is much greater than

\* Crowther, Proc. Roy. Soc. A. lxxxvi. p. 478 (1912).

this ; but it has been suggested by Webster \* that this excess scattering is due to an irregular attempt at cooperation between the disturbances from the different atoms. If this is so no account need be taken here of this excess radiation. For glancing angles less than  $6^\circ$ , the factor  $(1 + \cos^2 \phi)$  is approximately constant and need not be considered. The intensity of reflexion will therefore depend on two factors : (1) the energy of the part of the incident radiation which is reflected, (2) the number of atoms of which the effects cooperate. If the components with wave-lengths between  $\lambda$  and  $\lambda + d\lambda$  are alone reflected, their energy may be expressed as  $E_\lambda d\lambda$ . Now consider the number of atoms that can cooperate. This is composed of two factors A and B. A represents the number of atoms in one plane which cooperate, B the number of cooperating planes. A is evidently proportional to the area of a Fresnel zone on the face of the crystal, that is to  $\lambda^2 \operatorname{cosec} \theta$ . To obtain B, consider a pulse with no definite wave-length coming from a point source and limited by a slit, so that it strikes the crystal only at angles between  $\theta$  and  $\theta + d\theta$ . Considering only first-order reflexion, components with wave-lengths between  $\lambda$  and  $\lambda + d\lambda$  will alone be reflected. Cooperation between the reflexions from successive planes will only continue so long as the extreme components  $\lambda$  and  $\lambda + d\lambda$  remain in step. They will be completely out of step in  $n$  layers where  $nd\lambda = \frac{1}{2}\lambda$  and so the factor B is proportional to  $\frac{\lambda}{d\lambda}$ . The intensity of the reflected beam is therefore proportional to  $E_\lambda d\lambda \cdot \lambda^2 \operatorname{cosec} \theta \cdot \frac{\lambda}{d\lambda}$ , that is to  $E_\lambda \cdot \lambda^2$ , since by equation (1)  $\lambda \operatorname{cosec} \theta$  is constant.

Now  $u_\nu$ , the energy between frequencies  $\nu$  and  $\nu + 1$ , is proportional to  $E_\lambda \cdot \lambda^2$ . If, therefore, this argument is based on correct assumptions with regard to the scattering, then curve II., fig. 2, gives the distribution in the spectrum of the energy of the X-rays. The maximum value of  $u_\nu$  occurs at a wave-length of about  $K^{\frac{1}{2}} \times 4.9 \times 10^{-9}$  cm. The potential measured by an alternative spark-gap was 50,000 volts. The curve does not show this distribution in its usual form, but  $u_\nu$  is plotted against  $\theta$ , which for these small angles is proportional to  $\lambda$ . The general form of the curve bears a striking resemblance to that of the radiation from a black body. The rapid fall of energy for high frequencies and the more gradual fall for low are very noticeable, but on both sides the rise to the maximum is much steeper than it is for the radiation from a black body.

\* D. L. Webster, *Phil. Mag.* xxv. p. 234 (1913).

*Summary.*

1. X-rays "reflected" from crystals have the general properties of ordinary X-rays.

2. The radiation from an X-ray tube with platinum target is of two kinds, (A) radiation of indefinite wave-length, analogous to white light, (B) five types of monochromatic radiation, probably characteristic of the platinum.

3. The radiation (A) is reflected at all angles of incidence. The reflexion has been studied at different angles, and the results have been interpreted.

4. Each of the radiations (B) is reflected only at special angles. These angles have been measured with three crystals, and the results are in good agreement with the simple theory deduced from the principle of interference.

5. The wave-lengths of these radiations have been calculated to be  $1.642, 1.397, 1.375, 1.194, 1.157 \times K^{\frac{1}{2}} \times 10^{-8}$  cm. where  $K$  is probably either 1,  $\frac{1}{2}$ , or  $\frac{1}{8}$ .

6. Their homogeneity has been examined.

7. An attempt has been made to estimate the distribution of energy in the wave-lengths for the general X-rays.

We wish to express our warmest thanks to Prof. Bragg for the information which he has given us from time to time about his work on this subject. This information has been of great value to us.

We also wish to thank Prof. Rutherford for the very kind interest which he has taken in our work.

*XV. Notices respecting New Books.*

*Results of Observations made at the Coast and Geodetic Survey Magnetic Observatory near Tucson, Arizona.* Washington. Government Printing Office, 1913.

TUCSON ( $32^{\circ} 14' 8$  N. lat.,  $110^{\circ} 50' 01$  E. long.) is a new magnetic station of the U.S. Coast and Geodetic Survey, taking the place of Baldwin, Kansas, which has been discontinued. The present volume describes the site, and the construction of the buildings, which were built in 1909, and gives particulars of the results obtained from the magnetographs and the principal earthquakes recorded by a Bosch-Omori seismograph down to the end of 1910. The volume resembles in its contents and the order of their presentation the biennial volumes issued for the older observatories of the Survey.