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X-rays as a branch of optics

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One of the most fascinating aspects of recent physics research has been the gradual extension of familiar laws of optics to the very high frequencies of X-rays, until at the present there is hardly a phenomenon in the realm of light whose parallel is not found in the realm of X-rays. Reflection, refraction, diffuse scattering, polarization, diffraction, emission and absorption spectra, photoelectric effect, all of the essential characteristics of light have been found also to be characteristic of X-rays. At the same time it has been found that some of these phenomena undergo a gradual change as we proceed to the extreme frequencies of X-rays, and as a result of these interesting changes in the laws of optics we have gained new information regarding the nature of light.

It has not always been recognized that X-rays is a branch of optics. As a result of the early studies of Röntgen and his followers it was concluded that X-rays could not be reflected or refracted, that they were not polarized on transversing crystals, and that they showed no signs of diffraction on passing through narrow slits. In fact, about the only property which they were found to possess in common with light was that of propagation in straight lines. Many will recall also the heated debate between Barkla and Bragg, as late as 1910, one defending the idea that X-rays are waves like light, the other that they consist of streams of little bullets called "neutrons"». It is a debate on which the last word has not yet been said!

The refraction and reflection of X-rays

We should consider the phenomena of refraction and reflection as one problem, since it is a well-known law of optics that reflection can occur only from a boundary surface between two media of different indices of refraction. If one is found, the other must be present.

In his original examination of the properties of X-rays, Röntgen¹ tried unsuccessfully to obtain refraction by means of prisms of a variety of mate-

rials such as ebonite, aluminum, and water. Perhaps the experiment of this type most favorable for detecting refraction was one by Barkla². In this work X-rays of a wavelength which excited strongly the characteristic K-radiation from bromine were passed through a crystal of potassium bromide. The precision of his experiment was such that he was able to conclude that the refractive index for a wavelength of 0.5 \AA probably differed from unity by less than five parts in a million.

Although these direct tests for refraction of X-rays were unsuccessful, Stenström observed³ that for X-rays whose wavelengths are greater than about 3 \AA , reflected from crystals of sugar and gypsum, Bragg's law, $n\lambda = 2D \sin \vartheta$, does not give accurately the angles of reflection. He interpreted the difference as due to an appreciable refraction of the X-rays as they enter the crystal. Measurements by Duane and Siegbahn and their collaborators⁴ showed that discrepancies of the same type occur, though they are very small indeed, when ordinary X-rays are reflected from calcite.

The direction of the deviations in Stenström's experiments indicated that the index of refraction of the crystals employed was less than unity. If this is the case also, for other substances, total reflection should occur when X-rays in air strike a polished surface at a sufficiently sharp glancing angle, just as light in a glass prism is totally reflected from a surface between the glass and air if the light strikes the surface at a sufficiently sharp angle. From a measurement of this critical angle for total reflection it should be possible to determine the index of refraction of the X-rays.

When the experiment was tried⁵ the results were strictly in accord with these predictions. The apparatus was set up as shown in Fig. 1, reflecting a

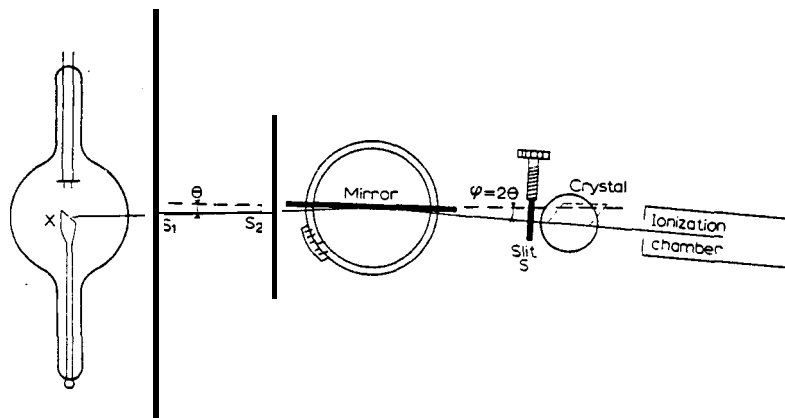


Fig. 1. Apparatus for studying the total reflection of X-rays.

narrow sheet of X-rays from a polished mirror on the crystal of a Bragg spectrometer. It was found that the beam could be reflected from the surfaces of a polished glass and silver through several minutes of arc. By studying the spectrum of the reflected beam, the critical glancing angle was found to be approximately proportional to the wavelength. For ordinary X-rays whose wavelength is one half an ångström, the critical glancing angle from crown glass was found to be about 4.5 minutes of arc, which means a reflective index differing from unity by less than one part in a million.

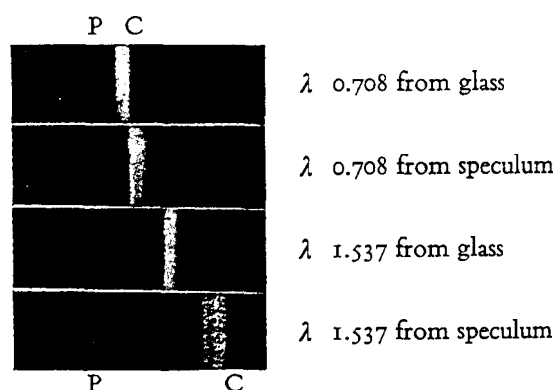


Fig. 2. Total reflection of X-rays from polished glass and speculum metal (Doan).
P = direct beam; C = critical angle of the totally reflected beam.

Fig. 2 shows some photographs of the totally reflected beam and the critical angle for total reflection taken recently from Dr. Doan⁶ working at Chicago. From the sharpness of the critical angle shown in this figure, it is evident that a precise determination of the refractive index can thus be made.

You will recall that when one measures the index of refraction of a beam of light in a glass prism it is customary to set the prism at the angle for minimum deviation. This is done primarily because it simplifies the calculation of the refractive index from measured angles. It is an interesting comment on the psychology of habit that most of the earlier investigators of the refraction X-rays by prisms also used their prisms set at the minimum deviation. Of course, since the effect to be measured was very small indeed, the adjustments should have been made to secure not the minimum deviation but the maximum possible. After almost thirty years of attempts to refract X-rays by prisms, experiments under the conditions to secure maximum re-

fraction were first performed by Larsson, Siegbahn, and Waller⁷, using the arrangement shown diagrammatically in Fig. 3. The X-rays struck the face of the prism at a fine glancing angle, just greater than the critical angle for the rays which are refracted. Thus the direct rays, the refracted rays, and the totally reflected rays of greater wavelength were all recorded on the same plate.

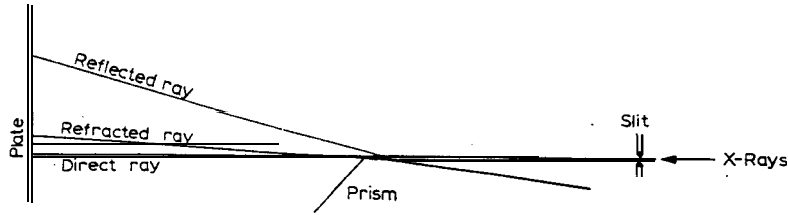


Fig. 3. Refraction of X-rays by a glass prism. (Arrangement of Larsson, Siegbahn, and Waller.)

Fig. 4 shows one of the resulting photographs. Here we see a complete dispersion spectrum of the refracted X-rays precisely similar to the spectrum obtained when the light is refracted by a prism of glass. The presence of the direct ray and the totally reflected ray on the same plate make possible all the angle measurements necessary for a precise determination of the refractive index of each spectrum line.

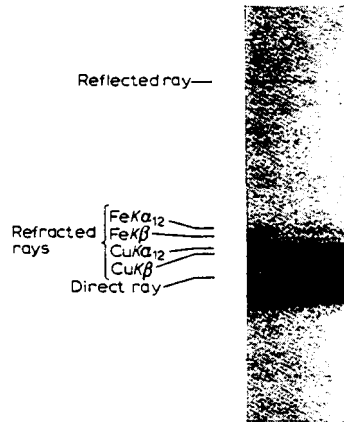


Fig. 4. Prism spectrum of X-rays obtained by Larsson, Siegbahn, and Waller.

For a generation we have been trying to obtain a quantitative test of Drude and Lorentz' dispersion theory in the ordinary optical region. But our ignorance regarding the number and the natural frequency of the electron oscillators in the refractive medium has foiled all such attempts. For the extreme frequencies of X-rays, however, the problem has become greatly simplified. In the case of substances such as glass, the X-ray frequencies are much higher than the natural frequencies of the oscillators in the medium, and the only knowledge which the theory requires is that of the number of electrons per unit volume in the dispersive medium. If we assume the number of electrons per atom to be equal to the atomic number, we are thus able to calculate at once the refractive index of the medium for X-rays. In the case of glass this calculation gives agreement with experiment within the experimental error, which is in some cases less than one per cent. So we may say that the laws of optical dispersion given by the electron theory are first established on a quantitative basis by these experiments on the refraction of X-rays.

Another way of looking at the problem is to assume the validity of the dispersion equation developed from the electron theory, and to use these measurements of refraction of X-rays to calculate the number of electrons in each atom of the refracting material. This affords us what is probably our most direct as well as our most precise means of determining this number. The precision of the experiments is now such that we can say that the number of electrons per atom effective in refracting X-rays is within less than one half of one per cent equal to the atomic number of the atom.

Thus optical refraction and reflection are extended to the region of X-rays, and this extension has brought with it more exact knowledge not only of the laws of optics but also of the structure of the atom.

The diffraction of X-rays

Early in the history of X-rays it was recognized that most of the properties of these rays might be explained if, as suggested by Wiechert⁸, they consist of electromagnetic waves much shorter than those of light. Haga and Wind performed a careful series of experiments⁹ to detect any possible diffraction by a wedge-shaped slit a few thousandths of an inch broad at its widest part. The magnitude of the broadening was about that which would result¹⁰ from rays of 1.3 Å wavelength. The experiments were repeated by yet more

refined methods by Walter and Pohl¹¹ who came to the conclusion that if any diffraction effects were present, they were considerably smaller than Haga and Wind had estimated. But on the basis of the photometric measurements of Walter and Pohl's plates by Koch¹² using his new photoelectric microphotometer, Sommerfeld found¹³ that their photographs indicated an effective wavelength for hard X-rays of 4 Å, and for soft X-rays a wavelength measurably greater.

It may have been because of their difficulty that these experiments did not carry as far as their accuracy would seem to have warranted. Nevertheless it was this work perhaps more than any other that encouraged Laue to undertake his remarkable experiments on the diffraction of X-rays by crystals.

Within the last few years Walter has repeated these slit diffraction experiments, making use of the $K\alpha$ -line of copper, and has obtained perfectly convincing diffraction effects¹⁴. Because of the difficulty in determining the width of the slit where the diffraction occurs, it was possible to make from his photographs only a rough estimate of the wavelength of X-rays. But within this rather large probable error the wavelength agreed with that determined by crystal spectrometry.

While these slit diffraction experiments were being developed, and long before they were brought to a successful conclusion, Laue and his collaborators discovered the remarkable fact that crystals act as suitable gratings for diffracting X-rays. You are all acquainted with the history of this discovery. The identity in nature of X-rays and light could no longer be doubted. It gave a tool which enabled the Braggs to determine with a definiteness previously almost unthinkable, the manner in which crystals are constructed of their elementary components. By its help, Moseley and Siegbahn have studied the spectra of X-rays, we have learned to count one by one the electrons in the different atoms, and we have found out something regarding the arrangement of these electrons. The measurement of X-ray wavelengths thus made possible gave Duane the means of making his precise determination of Planck's radiation constant. By showing the change of wavelength when X-rays are scattered, it has helped us to find the quanta of momentum of radiation which had previously been only vaguely suspected. Thus in the two great fields of modern physical inquiry, the structure of matter and the nature of radiation, the discovery of the diffraction of X-rays by crystals has opened the gateway to many new and fruitful paths of investigation. As Duc de Broglie has remarked, "if the value of a discovery is to be measured by fruitfulness of its consequences, the work of Laue and his collaborators

should be considered as perhaps the most important in modern physics".

These are some of the consequences of extending the optical phenomenon of diffraction into the realm of X-rays.

There is, however, another aspect of the extension of optical diffraction into the X-ray region, which has also led to interesting results. It is the use of ruled diffraction gratings for studies of spectra. By a series of brilliant investigations, Schumann, Lyman, and Millikan, using vacuum spectrographs, have pushed the optical spectra by successive stages far into the ultraviolet. Using a concave reflection grating at nearly normal incidence, Millikan and his collaborators¹⁵ found a line probably belonging to the L-series of aluminum, of a wavelength as short as 136.6 \AA , only a twenty-fifth that of yellow light. Why his spectra stopped here, whether because of failure of his gratings to reflect shorter wavelengths, or because of lack of sensitiveness of the plates, or because his hot sparks gave no rays of shorter wavelength, was hard to say.

Röntgen had tried to get X-ray spectra by reflection from a ruled grating, but the task seemed hopeless, How could one get spectra from a reflection grating if the reflection grating would not reflect? But when it was found that X-rays could be totally reflected by fine glancing angles, hope for the success of such an experiment was revived. Carrara¹⁶, working at Pisa, tried one of Rowland's optical gratings, but without success. Fortunately we at

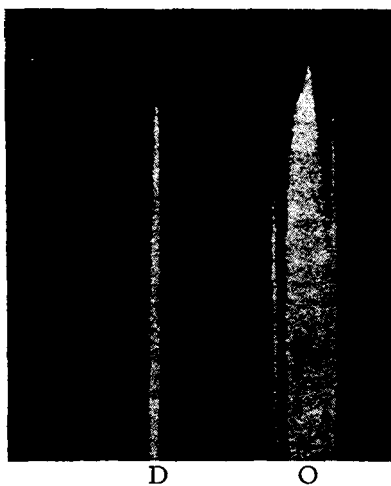


Fig. 5. Spectrum of the $K\alpha_1$ -line of molybdenum, $\lambda = 0.708 \text{ \AA}$, from a grating ruled on speculum metal (Compton and Doan). *D* marks the direct beam, and *O* the directly reflected beam.

Chicago did not know of this failure, and with one of Michelson's gratings ruled specially for this purpose, Doan found that he could get diffraction spectra of the K-series radiations both from copper and molybdenum¹⁷. Fig. 5 shows one of our diffraction spectra, giving several orders of the $K\alpha_1$ -line of molybdenum, obtained by reflection at a small glancing angle. This work was quickly followed by Thibaud¹⁸, who photographed a beautiful spectrum of the K-series lines of copper from a grating of only a few hundred lines ruled on glass. That X-ray spectra could be obtained from the same type of ruled reflection gratings as those used with light was now established.

The race to complete the spectrum between the extreme ultraviolet of Millikan and the soft X-ray spectra of Siegbahn began again with renewed enthusiasm. It had seemed that the work of Millikan and his co-workers had carried the ultraviolet spectra to as short wavelengths as it was possible to go. On the X-ray side, the long wavelength limit was placed, theoretically at least, by the spacing of the reflecting layers in the crystal used as a natural grating. De Broghe, W. H. Bragg, Siegbahn, and their collaborators were finding suitable crystals of greater and greater spacing until Thoraeus and Siegbahn¹⁹, using crystals of palmitic acid, measured the $L\alpha$ -line of chromium with a wavelength 21.69 Å. But there still remained a gap of almost three octaves between these X-rays and the shortest ultraviolet in which, though radiation had been detected by photoelectric methods, no spectral measurements has been made.

Thibaud, working in de Broglie's laboratory at Paris, made a determined effort to extend the limit of the ultraviolet spectrum, using his glass grating at glancing incidence²⁰. His spectra however stopped at 144 Å, a little greater than the shortest wavelength observed in Millikan's experiments.

But meanwhile, Dauvillier, also working with de Broglie, was making rapid strides working from the soft X-ray side of the gap. First²¹ using a grating of palmitic acid, he found the $K\alpha$ -line of carbon of wavelength 45 Å. Then²² using for a grating a crystal of the lead salt of melissic acid, with the remarkable grating space of 87.5 Å, he measured a spectrum line of thorium as long as 121 Å, leaving only a small fraction of an octave between his longest X-ray spectrum lines and Millikan's shortest ultraviolet lines. The credit for filling in the greater part of the remaining gap must thus be given to Dauvillier.

The final bridge between the X-ray and the ultraviolet spectra has however been laid by Osgood²³, a young Scotchman working with me at Chicago. He also used soft X-rays as did Dauvillier, but instead of a crystal

grating, he did his experiments with a concave glass grating in a Rowland mounting, but with the rays at glancing incidence. Fig. 6 shows a series of Osgood's spectra. The shortest wavelength here shown is the $K\alpha$ -line of carbon, 45 \AA , and we see a series of lines up to 211 \AA . An interesting feature of the spectra is an emission band in the aluminum spectrum at about 170 \AA , which is probably in some way associated with the L-series spectrum of aluminum. These spectra overlap, on the short wavelength side, Dauvillier's crystal measurements, and on the other side of the great wavelengths, Millikan's ultraviolet spectra.

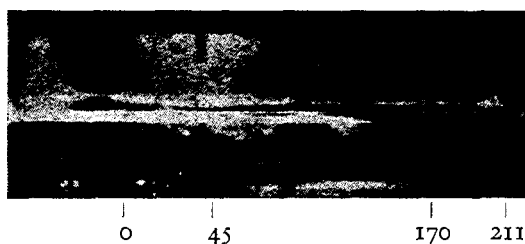


Fig. 6. Osgood's grating spectra of soft X-rays from Al, C, Mg, Fe, and Ni, showing lines from $\lambda = 45 \text{ \AA}$ to $\lambda = 211 \text{ \AA}$. These are the first spectra bridging the gap between soft X-rays and the ultraviolet.

In the September number of *The Physical Review*, Hunt²⁴ describes similar experiments, using however a plane ruled grating at glancing incidence, in which he has measured lines from 2 \AA down to the carbon line at 45 \AA , thus meeting the shortest of Osgood's measurements. On the other hand, Fig. 7 shows some beautiful spectra of the extreme ultraviolet obtained recently by Dr. Hoag, working with Professor Gale at Chicago, using a concave grating at grazing incidence. These spectra extend from 200 \AA to 1760 \AA , overlapping Osgood's X-ray spectra on the short wavelength side and reaching the ordinary ultraviolet region on the side of the great wavelengths. Thus from the extreme infrared to the region of the ordinary X-rays we now have a continuous series of spectra from ruled gratings.

Whatever we may find regarding the nature of X-rays, it would take a bold man indeed to suggest, in light of these experiments, that they differ in nature from ordinary light.

It is too early to predict what may be the consequences of these grating measurements of X-rays. It seems clear, however, that they must lead to a new and more precise knowledge of the absolute wavelength of crystals.

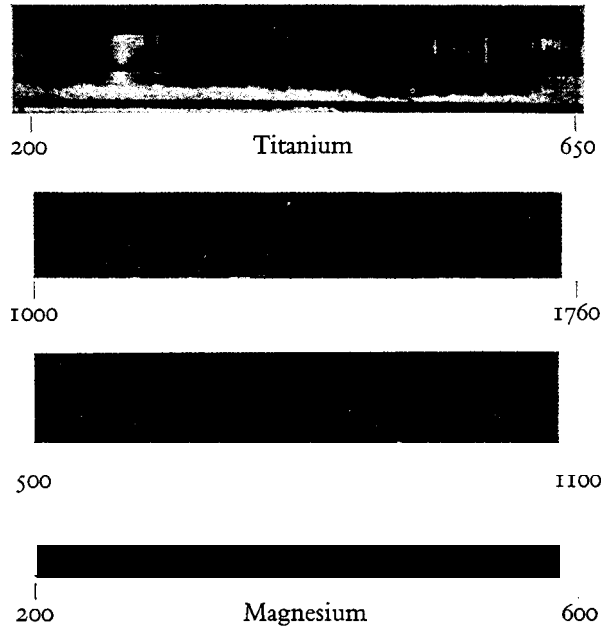


Fig. 7. Spectra of the extreme ultraviolet, from Mg and Ti, 200 Å to 1760 Å (Hoag).

This will in turn afford a new means of determining Avogadro's number and the electronic charge, which should be of precision comparable with that of Millikan's oil drops.

The scattering of X-rays and light

The phenomena that we have been considering are ones in which the laws which have been found to hold in the optical region apply equally well in the X-ray region. This is not the case, however, for all optical phenomena.

The theory of the diffuse scattering of light by turbid media has been examined by Drude, Lord Rayleigh, Raman, and others, and an essentially similar theory of the diffuse scattering of X-rays has been developed by Thomson, Debye, and others. Two important consequences of these theories are, (1) that the scattered radiation shall be of the same wavelength as the primary rays; and (2) that the rays scattered at 90 degrees with the primary rays shall be plane polarized. The experimental tests of these two predictions have led to interesting results.

A series of experiments performed during the last few years* has shown that secondary X-rays are of greater wavelength than the primary rays which produce them. This work is too well-known to require description. On the other hand, careful experiments to find a similar increase in wavelength in light diffusely scattered by a turbid medium have failed to show any effect^{2,5}. An examination of the spectrum of the secondary X-rays shows that the primary beam has been split into two parts, as shown in Fig. 8, one of the same wavelength and the other of increased wavelength. When

* For an account of this work, see e.g. the writer's *X-rays and Electrons*, Chap. 9, Van Nostrand, 1926.

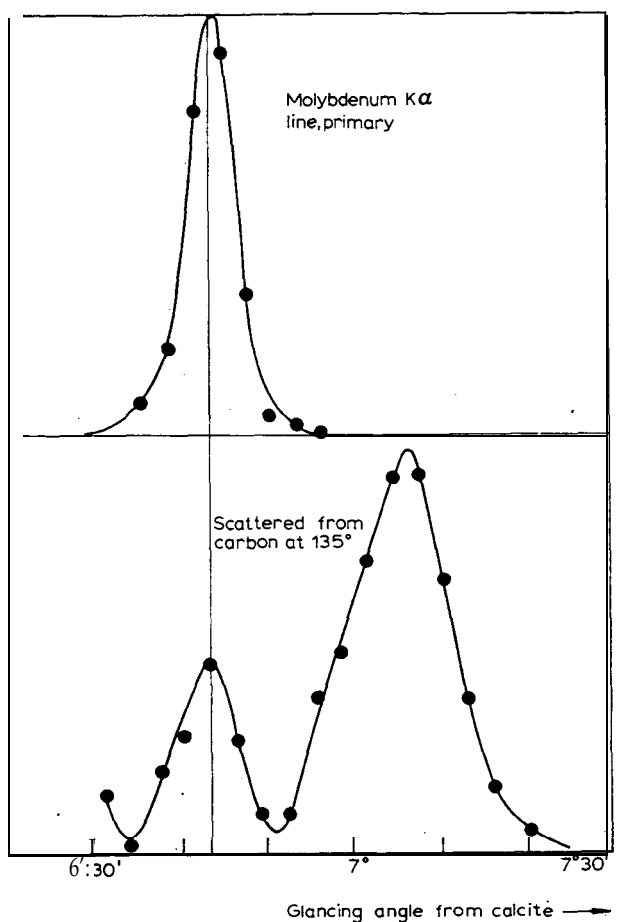


Fig. 8. A typical spectrum of scattered X-rays, showing the splitting of the primary ray into a modified and an unmodified ray.

different primary wavelengths are used, we find always the same difference in wavelength between these two components; but the relative intensity of the two components changes. For the longer wavelengths the unmodified ray has the greater energy, while for the shorter wavelengths the modified ray is predominant. In fact when hard γ rays are employed, it is not possible to find any radiation of the original wavelength.

Thus in the wavelength of secondary radiation we have a gradually increasing departure from the classical electron theory of scattering as we go from the optical region to the region of X-rays and γ -rays.

The question arises, are these secondary X-rays of increased wavelength to be classed as scattered X-rays or as fluorescent? An important fact bearing on this point is the intensity of the secondary rays. From the theories of Thomson, Debye, and others it is possible to calculate the absolute intensity of the scattered rays. It is found that this calculated intensity agrees very nearly with the total intensity of the modified and unmodified rays, but that in many cases the observed intensity of the unmodified ray taken alone is very small compared with the calculated intensity. If the electron theory of the intensity of scattering is even approximately correct, we must thus include the modified with the unmodified rays as scattered rays.

Information regarding the origin of these secondary rays is also given by their state of polarization. We have called attention to the fact that the electron theory demands that the X-rays scattered at 90 degrees should be completely plane polarized. If the rays of increased wavelength are fluorescent, however, we should not expect them to be strongly polarized. You will remember the experiments performed by Barkla²⁶ some twenty years ago in which he observed strong polarization in X-rays scattered at right angles. It was this experiment which gave us our first strong evidence of the similar character of X-rays and light. But in this work the polarization was far from complete. In fact the intensity of the secondary rays at 90 degrees dropped only to one third its maximum value, where as for complete polarization it should have fallen to zero.

The fact that no such unpolarized rays exist was established by repeating Barkla's experiment²⁷ with scattering blocks of different sizes. When very small blocks were used, we found that the polarization was nearly complete. The lack of complete polarization in Barkla's experiments was due chiefly to the multiple scattering of the X-rays in the large blocks that he used to scatter the X-rays. It would seem that the only explanation of the complete polarization of the secondary rays is that they consist wholly of scattered rays.

According to the classical theory, an electromagnetic wave is scattered when it sets the electrons which it traverses into forced oscillations, and these oscillating electrons reradiate the energy which they receive. In order to account for the change in wavelength of the scattered rays, however, we have had to adopt a wholly different picture of the scattering process, as shown in Fig. g. Here we do not think of the X-rays as waves but as light corpuscles, quanta, or, as we may call them, photons. Moreover, there is nothing here of the forced oscillation pictured on the classical view, but a sort of elastic collision, in which the energy and momentum are conserved.

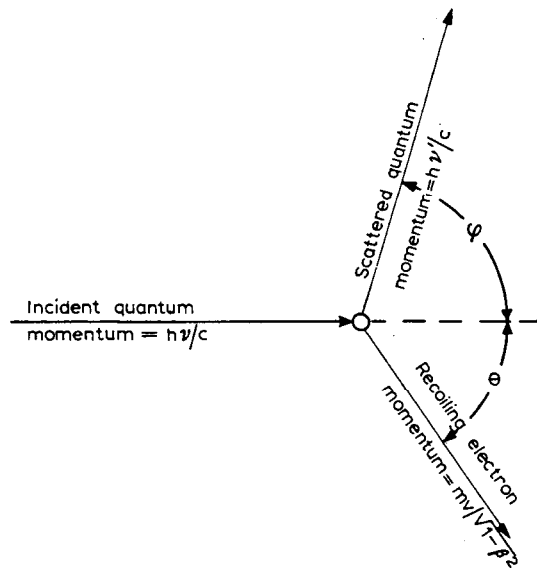


Fig. 9. An X-ray photon is deflected through an angle ϕ by an electron, which in turn recoils at an angle θ , taking a part of the energy of the photon.

This new picture of the scattering process leads at once to three consequences that can be tested by experiment. There is a change of wavelength

$$\delta\lambda = \frac{h}{mc} (1 - \cos \phi) \quad (1)$$

which accounts for the modified line in the spectra of scattered X-rays. Experiment has shown that this formula is correct within the precision of our

knowledge of h , m , and c . The electron which recoils from the scattered X-rays should have the kinetic energy

$$E_{kin} = h\nu \cdot \frac{h\nu}{mc^2} \cos^2\theta \tag{2}$$

approximately. When this theory was first proposed, no electrons of this type were known; but they were discovered by Wilson²⁸ and Bothe²⁹ within a few months after their prediction. Now we know that the number, energy, and spatial distribution of these recoil electrons are in accord with the predictions of the photon theory. Finally, whenever a photon is deflected at an angle ϕ , the electron should recoil at an angle φ given by the relation

$$\cot \frac{1}{2}\varphi = \tan \theta \tag{3}$$

approximately.

This relation we have tested³⁰, using the apparatus shown diagrammatically in Fig. 10. A narrow beam of X-rays enters a Wilson expansion

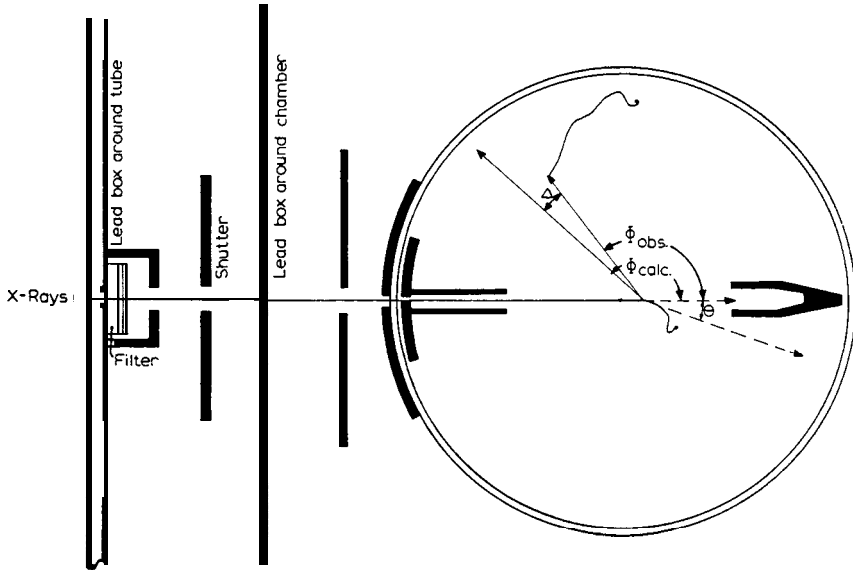


Fig. 10. An electron recoiling at an angle φ should be associated with a photon deflected through an angle θ .

chamber. Here it produces a recoil electron. If the photon theory is correct, associated with this recoil electron, a photon is scattered in the direction θ . If it should happen to eject a β ray, the origin of this β ray tells the direction in which the photon was scattered. Fig. 11 shows a typical photograph of the process. A measurement of the angle θ at which the recoil electron on this plate is ejected and the angle ϕ of the origin of the secondary P-particle, shows close agreement with the photon formula. This experiment is of especial significance, since it shows that for each recoil electron there is a scattered photon, and that the energy and momentum of the system photon plus electron are conserved in the scattering process.

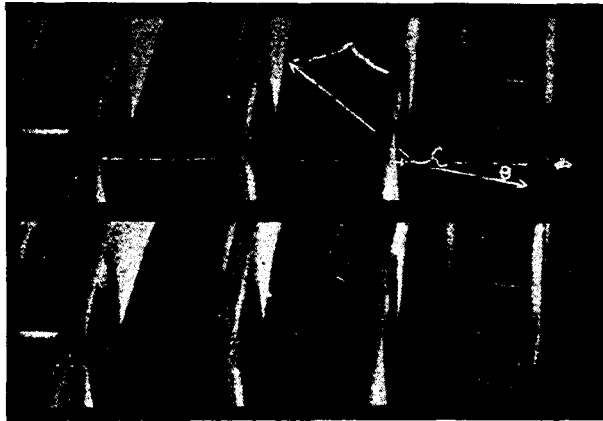


Fig. 11. Photograph showing recoil electron and associated secondary β ray. (The upper photograph is retouched.)

The evidence for the existence of directed quanta of radiation afforded by this experiment is very direct. The experiment shows that associated with each recoil electron there is scattered X-ray energy enough to produce a secondary β ray, and that this energy proceeds in a direction determined at the moment of ejection of the recoil electron. Unless the experiment is subject to improbably large experimental errors, therefore, the scattered X-rays proceed in the form of photons.

Thus we see that as a study of the scattering of radiation is extended into the very high frequencies of X-rays, the manner of scattering changes. For the lower frequencies the phenomena could be accounted for in terms of waves. For these higher frequencies we can find no interpretation of the scattering except in terms of the deflection of corpuscles or photons of radia-

tion. Yet it is certain that the two types of radiation, light and X-rays, are essentially the same kind of thing. We are thus confronted with the dilemma of having before us a convincing evidence that radiation consists of waves, and at the same time that it consists of corpuscles.

It would seem that this dilemma is being solved by the new wave mechanics. De Broglie³¹ has assumed that associated with every particle of matter in motion there is a wave whose wavelength is given by the relation

$$mv = h/\lambda$$

where mv is the momentum of the particle. A very similar assumption was made at about the same time by Duane³², to account for the diffraction of X-ray photons. As applied to the motion of electrons, Schrödinger has shown the great power of this conception in studying atomic structure³³. It now seems, through the efforts of Heisenberg, Bohr, and others, that this conception of the relation between corpuscles and waves is capable of giving us a unified view of the diffraction and interference of light, and at the same time of its diffuse scattering and the photoelectric effect. It would however take too long to describe these new developments in detail.

We have thus seen how the essentially optical properties of radiation have been recognized and studied in the realm of X-rays. A study of the refraction and specular reflection of X-rays has given an important confirmation of the electron theory of dispersion, and has enabled us to count with high precision the number of electrons in the atom. The diffraction of X-rays by crystals has given wonderfully exact information regarding the structure of crystals, and has greatly extended our knowledge of spectra. When X-rays were diffracted by ruled gratings, it made possible the study of the complete spectrum from the longest to the shortest waves. In the diffuse scattering of radiation, we have found a gradual change from the scattering of waves to the scattering of corpuscles.

Thus by a study of X-rays as a branch of optics we have found in X-rays all of the well-known wave characteristics of light, but we have found also that we must consider these rays as moving in directed quanta. It is these changes in the laws of optics when extended to the realm of X-rays that have been in large measure responsible for the recent revision of our ideas regarding the nature of the atom and of radiation.

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