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Concerning the detection of X-ray interferences

Nobel Lecture, June 3, 1920

If other Nobel Prize winners express their thanks for this high honour at this juncture by recounting the history of their discovery they are able to report how, at the outset, they envisaged a high, but nevertheless attainable, objective and how they strove toward that objective by many paths, most of which proved initially to be false and how then, after many years of protracted endeavour, they finally achieved their objective. In my eyes the credit to which they are entitled increases in direct proportion to the magnitude of the difficulties which they finally surmounted. By comparison, what I have to say here differs somewhat from those examples. There is no doubt that I, also, had long been aware of the problem, i.e. producing X-ray interferences, before the inherent difficulties had finally been surmounted. But I never believed that it would be my personal good fortune to make a contribution in that direction, and it was for that reason that I did not concern myself unduly in that respect until suddenly I perceived the way which subsequently proved to be the shortest path to success. I am not, therefore, able to recount many details in connection with my personal preparatory work, so I shall confine my comments to explaining the combination of scientific and personal circumstances from which the idea first arose.

In the case of X-rays their discoverer had already made efforts to locate diffraction or interference phenomena in order to solve the question of whether or not they represent a wave phenomenon or the ejection of any small particles. But in this quest his research, which had otherwise been so successful, met with failure. Nevertheless, from the outset there were supporters for the former opinion; for Stokes and Wiechert could point out with complete justification that, according to Maxwell-Lorentz electrodynamics, electromagnetic waves must be produced whenever electricity carriers alter their velocity; one was also aware that cathode rays, which induce X-rays on striking an obstacle, are made up of electrons. W. Wien endeavoured to reconcile this consideration with what was, at that time, the very young quantum theory in order thus to arrive at an estimate of the relevant wavelengths. He arrived at a value lying between 10^{-10} and 10^{-9}cm, which was in accordance with the fact that, if they were at all made up of waves, the X-rays could only have a shorter wavelength than visible light. For in 1900 all electromagnetic radiation of longer wavelengths was already known at least to the extent that one could not seek in it the more striking characteristics of X-rays such as, for example, the strong penetrating power.

If diffraction or interference phenomena were to be sought it was therefore necessary, in accordance with the basic principles of wave theory, to select for the test arrangement far smaller decisive dimensions than those employed in corresponding tests with visible light. Thus Haga and Wind, as well as Walter and Pohl, selected a wedge-shaped pointed slit at the pointed end of which they were able to prove a broadening of the transmitted X-ray bundles which could be attributed to diffraction. The conclusive force of these tests has frequently been a subject of dispute; for example there was one school of thought which tried to explain the broadening as a subjective optical illusion. However, the photometric measurement carried out by P. P. Koch on the best photograms of this type and the estimated wavelength of 4×10^{-9} cm which Sommerfeld connected with those results would appear to prove that there is an objective basis for the broadening theory. For now that the relevant wavelengths have been confirmed we must regard this value as representing a very good determination for such tests.

Further evidence in support of the wave theory of X-rays was provided by Barkla's discovery of polarisation. Barkla permits rays, such as those originating from the anticathode of an X-ray tube, to fall upon a substance such as, for example, carbon, after which he then carries on working with those secondary X-rays as result from scattering, which emanate from the substance in question perpendicular to the original direction of radiation. If the secondary radiation is again permitted to fall upon such a substance this causes a production, as a result of repeated scattering, of a tertiary radiation of X-rays; but this has, at the plane perpendicular to the secondary radiation, an intensity maximum in one direction, and perpendicular to that a minimum at which it may even disappear completely. This indicates most clearly that, for the secondary radiation, the direction of propagation is not a symmetry axis and that it is thus polarised. This fact can be explained only with extreme difficulty by a corpuscular theory - of this we are only too well aware from the history of optics. Conversely it is quite possible to explain this by wave theory based upon the fact that all electromagnetic waves oscillate transversally and are therefore generally polarised. This theory also readily explains the further fact that the direction of the vanishing intensity is parallel to the direction of the primary rays.

Notwithstanding these major arguments the wave theory initially did not meet with complete acceptance. For experts like W. H. Bragg maintained until 1912 the corpuscular view and, in support of that view, they were able to quote a series of phenomena which, in actual fact, have remained inaccessible to explanation by the wave theory. If X-rays do, in fact, fall upon a body, they not only excite the aforementioned secondary X-rays, they also free electrons. The velocities of these electrons do not now depend upon the temperature and other conditions obtaining with the body in question and one might therefore assume that their kinetic energy is derived from the incident X-ray radiation. But this is where the difficulty is encountered. For the velocities which have been observed are not dependent upon the intensity of radiation but, in fact, upon their degree of hardness, and specifically in such a manner that these velocities increase with increasing hardness. If the intensity is weakened this will result in a decrease in the number of electrons released, but it will have no effect upon their velocities. The greatest velocity which occurs is always that which was possessed by the cathode radiation by which the X-rays have been excited; it is almost as if X-ray radiation retains a "memory" of the process of its creation and expresses this fact when, for its part, it releases electrons.

This would be quite easy to comprehend if the energy of the X-ray radiation were to accumulate and remain accumulated at certain points, as would, for example, be the case with the motion of individual corpuscles. But in agreement with the wave theory it expands progressively over increasing areas. This poses us with a complete puzzle, which is merely rendered the more complex by the further observation that the electrons which have been released would appear to prefer the direction taken by the incident X-rays. And it is a source of only qualified comfort if we make clear to ourselves that with the short-wave visible and ultraviolet light rays the same phenomena occur in the photoelectric effect. In this case also, where we most certainly have before us a wave process, they are simply incomprehensible.

We can only understand the reasons why our understanding fails at this juncture. Einstein has provided us with a rule on the dependency of the greatest existing electron velocity upon the frequency of light waves; in this the decisive role is played by the universal constant *h,* designated by Planck, its discoverer, as the elementary quantum of action. Conversely, more recent measurements - among which particular accuracy was achieved in those made by Ernst Wagner in Munich - have shown that with the reverse process, specifically the excitation of X-rays by cathode rays, the same constant appears. This same Einstein law, namely, also links the greatest frequency in the X-ray spectrum with the kinetic energy of cathode rays. We are confronted here with just one special case of the great quantum mystery, the solution of which one may justly term the most important objective currently confronting the entire field of physics. However, it was quite understandable that before this relationship with the quantum theory had been ascertained researchers had based their arguments against the wave theory on the facts of electron emission.

But let us turn once again to the situation which obtained before 1912. For as far back as I can recall having had an interest in physics - and that goes back to the days in which I was privileged to receive first-class instruction at the famous old Protestant Gymnasium in Strassburg, Alsatia - my particular attention was drawn to the field of optics, and within that field the wave theory of light. The lectures which I heard during my student days from Voigt, Planck and Lummer provided me at that time, and especially in the afore-mentioned field, with thorough experimental and theoretical knowledge; and as I had finally been able to cultivate what one could almost term a special feeling or intuition for wave processes, it was quite natural that my first independent work should be concerned with the propagation of natural radiation in dispersing substances and the thermodynamics of interference phenomena. It turned out to be a matter of great good fortune that Sommerfeld passed to me the article "Wellenoptik" (Wave optics) at that time to work upon for the *Encyclopedia of Mathematical Sciences.* For it was during that project that I was obliged to seek a mathematical presentation of the lattice theory which - while it was not exactly what one might term an innovation - could be applied with considerable simplicity to lattice grids. Already at that juncture I was devoting considerable thought to the further transfer to space-lattices; but I did not pursue the matter further owing to the fact that they do not, after all, play any actual part in the field of optics.

On my arrival in Munich in 1909 my attention was drawn constantly - first owing to the influence of Röntgen's work at this University and subsequently by Sommerfeld's active interest in X-rays and γ-rays, which he had also testified in several works-back to the question of their actual nature. To this was added a further important circumstance. Since the times of Hauy and Bravais the basic crystallographic law of rational indices had been explained simply and visually by the mineralogists through space-lattice arrangement of the atoms. Sohncke, Federow and Schoenflies had brought the mathematical theory of possible space-lattices to the greatest possible degree of perfection. But no more far-reaching physical conclusion had evolved from this line of thought and thus, in the form of a questionable hypothesis, it remained a somewhat unknown quantity to physicists. But in Munich, where models of the Sohncke space-lattices were to be found in more than one university institute, it was P. Groth who expressed his defence of it, both orally and in writing, and I, also, thus learned from him. My interest in the theories which he was expounding increased progressively as, already at an early time, I had made sure - and contrary to the doubts which, in my day, were maintained by only a few philosophers concerning the reality of atoms - that there existed no watertight epistemological arguments which might contradict this fact, while practical experience was constantly providing fresh confirmation in support of it.

Such was the state of affairs as, one evening in February 1912, P. P. Ewald came to visit me. On Sommerfeld's instigation he was working on a mathematical investigation into the behaviour of long electromagnetic waves in a space-lattice and subsequently he published a dissertation on the theory of crystal optics which was based upon that work. But he was faced at that time with certain difficulties and came to me with a request for advice. Now it was not, however, possible for me to assist him at that time. But during the conversation I was suddenly struck by the obvious question of the behaviour of waves which are short by comparison with the lattice-constants of the space-lattice. And it was at that point that my intuition for optics suddenly gave me the answer: lattice spectra would have to ensue. The fact that the lattice-constant in crystals is of an order of 10^{-8} cm was sufficiently known from the analogy with other interatomic distances in solid and liquid substances, and, in addition, this could easily be argued from the density, molecular weight and the mass of the hydrogen atom which, just at that time, had been particularly well determined. The order of X-ray wavelengths was estimated by Wien and Sommerfeld at 10^{-9} cm. Thus the ratio of wavelengths and lattice-constants was extremely favourable if X-rays were to be transmitted through a crystal. I immediately told Ewald that I anticipated the occurrence of interference phenomena with X-rays.

It was not long before W. Friedrich also heard of this. While he immediately expressed his willingness to carry out a relevant test, the acknowledged masters of our science, to whom I had the opportunity of submitting it, entertained certain doubts about this viewpoint. A certain amount of diplomacy was necessary before Friedrich and Knipping were finally permitted to carry out the experiment according to my plan, using very simple equipment at the outset. Copper sulphate served as the crystal, since large and regular pieces of it can easily be obtained. The irradiation direction was left to chance. Immediately from the outset the photographic plate located behind the crystal betrayed the presence of a considerable number of deflected rays, together with a trace of the primary ray coming directly from the anticathode. These were the lattice spectra which had been anticipated (Fig. 1).

In continuation of the work, Friedrich and Knipping, with the aid of the considerable amount of equipment then placed at their disposal by the Institute of Theoretical Physics of the Munich University, substituted random irradiation of triclinic copper sulphate with irradiation, in the crystallographically-indicated axial directions, of crystals of maximum possible symmetry, i.e. regular crystals. In principle the theory had already been completed by transfer from the ordinary and from the grid lattice, and, on June 8, 1912, Sommerfeld was able to submit to the Munich Academy the joint work of Friedrich, Knipping, and myself on X-ray interferences, which work, apart from the theory itself, also contained a series of very characteristic exposures (Figs. 2 and 3). Four weeks later followed the first application of the theory for interpreting the points of interference, produced by a regular crystal after irradiation along a four-fold axis (Fig. 2). Each point of interference corresponds with three integral numbers, originating from the three space-lattice periodicities, the ratios of which determine its position. I have attributed these numbers to the points of interference in the second publication which appeared simultaneously with the first. And said numbers, or their ratios, have proven thoroughly reliable, even after re-checking by the Braggs and other researchers. Thus the initial probe into the rightness of the interference theory proved favourable for this new phenomenon. This benefitted not only the wave conception of X-rays but also the lattice theory of crystals.

The wavelength can also be determined from the three specified numbers and the latter publication contains therefore also information in that connection. Nevertheless I made no secret of the fact that I could not attribute to these values the same degree of reliability as to the ratios of the numbers. For this there were two reasons. On the hand there may occur in each interference maximum - as has been adequately known in the science of optics - apart from a basic wavelength, in addition its half, third, etc. For as long as one has no further point of reference, apart from the position of the maximum, the wavelength thus remains uncertain by an integral factor. The wavelengths

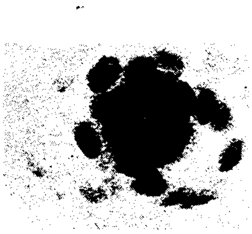

Fig. 1.

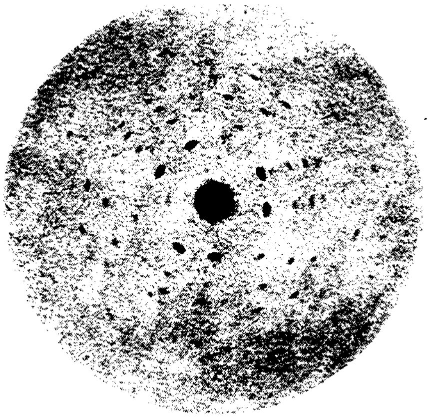

Fig. 2.

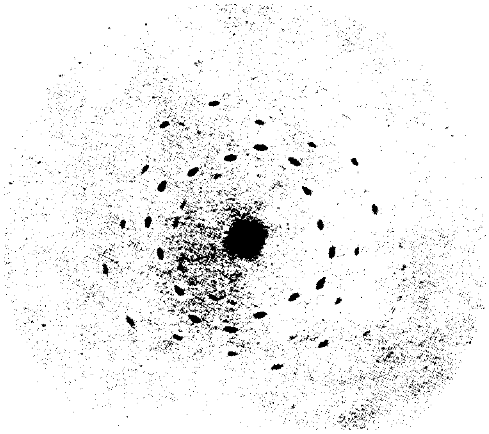

Fig. 3.

were then measured by the dimensions of the space-lattice. As has already been mentioned, these could certainly be estimated by the order of magnitude; but in order to compute them with some degree of accuracy it would be necessary to know the molecular structure of the crystals down to the last detail. Instead of that one was aware, solely from the research carried out by the mathematicians, that, apart from the simple space-lattices, many other types of lattice were possible, for example, face- and body-centred lattices. The type of structure which is found in certain crystals was first determined by the Braggs after they had made a daring guess in producing an accurate hypothesis as to the structure of sodium chloride. And it was thus that only they were also able to carry out the final measurements on the wavelengths.

But in reference to the manner in which the mathematical theory is to be generalised for such more complex types of structure, this I set forth almost one year later, when the necessity of that step had been shown with all urgency. The changes are minimal. The position of the points of interference remains the same as with a simple lattice of identical lattice-constants; it is only in the expression of their intensity that a new factor appears which is now designated as the structural factor. However, this factor can occasionally reduce to zero and then the relevant point is suppressed completely. It is due to this factor when an irradiation exposure plate gives some indication of the lower symmetry of the actual crystal instead of the full symmetry of the simple space-lattice.

The considerable interest which had been caused by the initial investigations was soon reflected in a large number of subsequent works. At first certain doubts were expressed as to whether, in this instance, an interference phenomenon really did exist. But these were soon overcome. In my opinion the most complete rebuff was embodied in the proof produced by Wagner and Glocker that the X-ray radiation at the points of interference contained only one wavelength, even if many of them should exist in the original ray. The experimental research diverged essentially into two branches. The crystals were either investigated with the aid of the interferences, which examined the crystals' space-lattices, or they were employed in spectroscopy in the X-ray range. Since 1912 much has been done in both fields, and in both sectors W. L. Bragg and W. H. Bragg have taken the first important step beyond the investigations carried out at Munich. It would range far too wide if I were, at this juncture, to compile a fairly comprehensive list of all the researchers who have gained distinction in that research work. I can mention here only the transformation of the process, which proved to be of great importance for the further conduct of the experiments and in which Debye transferred the examination of beautiful, well-formed crystal fragments - which sometimes are obtainable only with considerable difficulty - to research into the finest possible crystal powder. Mention should also be made here of the extremely painstaking and accurate determination of the characteristic spectra which, commenced by Moseley, have been carried on - especially here in Sweden by Manne Siegbahn and Stenström - on the great majority of chemical elements. This research, as is known, has become of fundamental importance for the further knowledge of the structure of the atom. Neither should I neglect to mention the fact that also the theory has been successfully extended. The important question as to why the thermal motion of the atoms does not result in any great amount of disturbance to the interference phenomena, has been settled by Debye, while H. A. Lorentz showed the manner in which the interference points which originate from the continuous spectrum obtain their intensity from the energy of many neighbouring wavelengths. For both reasons, owing to the thermal motion and to the working together of various wavelengths, factors arise which, in a similar manner to the structural factor, exert some influence upon the brightness of the interference points but not upon their location.

I would finally like to deal with a question which is still pending and to which, I trust, X-rays will shortly bring the answer. There are two contradictory opinions in respect of the mixed crystals. Opinions are united in respect of the fact that atoms form space-lattices, as is the case with chemically uniform crystals. There is also agreement in respect of the fact that in a mixed crystal of, e.g., potassium chloride and rubidium chloride the chlorine atoms lie in precisely the same position in relation to each other as in pure potassium chloride or pure rubidium chloride. (From the work carried out by the Braggs we are already aware of the fact that these two salts have the identical space-lattice.) But how are the potassium and rubidium atoms distributed over the positions reserved for the metal atoms? According to the one viewpoint, which probably originated from Van 't Hoff, this occurs entirely by chance, while the other viewpoint maintains that it occurs regularly, thus permitting the space-lattices to be composed of equal-sized parallelepipeds of molecular dimensions which are identical in every respect, including the atomic occupation. For purely theoretical reasons the former opinion would appear to me to be the more probable. If, as in the example given, every mixing ratio is possible, it is indeed difficult to conceive, in every case, of atomic distributions which observe crystallographic symmetry, and, in addition, leave sufficiently small dimensions for the specified elementary parallelepipeds. Also the irradiation tests conducted by Vegard and Skjeldrup on the above-mentioned mixtures would appear to have resulted in an even stronger contradiction of the second opinion than these researchers themselves had originally thought. Nevertheless during recent years Tammann has conducted chemical tests on mixed crystals of various metals such as, for example, copper and gold, which appear at first sight to support the second view. This should provide a further field of endeavour for a new application of X-ray interferences.