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Interference Phenomena with Compton Scattering

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For some years it has been known that some diamonds give excellent divergent-beam patterns even when the crystal is irradiated with a parallel beam of X-rays. By using a filter technique it is shown that the cause of the pattern is Compton radiation emitted from the atoms within the crystal. Only mosaic-type diamonds give such lines on the photographic film. It is suggested that the formation of the lines is mainly due to secondary extinction, the Compton radiation being weakened and reinforced through reflections from the lattice planes.

Introduction

Lonsdale (1947) has shown that mosaic-type diamonds of suitable thickness give excellent divergent-beam photographs. In investigating the graphitization of diamond and the nature of cliftonite, Grenville-Wells (1951, 1952) discovered that some natural cubes of Belgian Congo diamond, when irradiated with a parallel beam of Cu K+white radiation, gave a weak divergent-beam pattern superimposed on the Laue pattern of the diamond. In this case the source of the divergent radiation must lie within the crystal. Spectrographic analysis showed that the divergent-beam pattern could not be due to fluorescent radiation from metal impurities in the diamonds. Further, when the incident radiation was changed from copper to cobalt, the position of the continuous lines on the film shifted according to the increase in $K\alpha$ wavelengths. Grenville-Wells (1951) concluded that the origin of the divergent radiation either had to be Compton scattering or scattering due to imperfections in the crystal, or both.

The geometry of the interference pattern

Suppose that the atoms of the crystal lattice become independent sources of X-ray radiation. The waves spreading out from any atom must be diffracted by the other atoms of the lattice. Because the sources within the crystal are independent, the total intensity distribution outside the crystal will be the sum of those due to the individual atoms. We can make a plausible prediction of the type of disturbance to be expected by applying the elementary theory of reflection of plane waves at the lattice planes, although this approach requires further justification when the sources of radiation are close to the planes themselves. Neglecting this question for the moment, let us assume that the waves emanating from any point within the lattice are reflected at the lattice planes to an appreciable degree only at angles that correspond to Bragg's law $2d \sin\theta = n\lambda$. The directions of appreciable reflection from any set of planes will then lie on the surface of a set of cones whose common axis is normal to the planes. Since the X-ray sources are within the crystal, each set of planes will give complete double cones, corresponding to the reflection from both sides of the planes. The cones must be thought of as rigidly attached to the crystal. The intersection of these cones with the photographic film produces the interference pattern.

If the observed lines are due to Compton radiation, the value of θ for a single cone will vary, owing to the shift in wavelength of the Compton scattering when the angle φ between the beam incident on the crystal and the diffracted beam varies (see Fig. 1). The formula for this shift is

$$\delta\lambda = 0{\cdot}0242(1-\cos\varphi)\ \text{Å}\ .$$

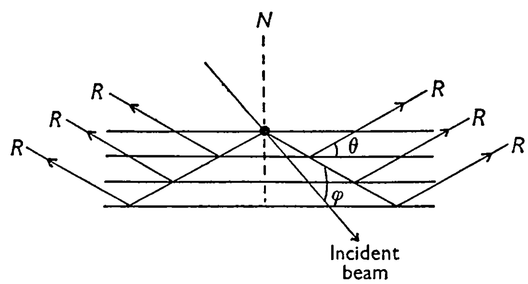

Fig. 1. Reflection from a set of lattice planes.

Only the Compton scattering gives a shift in wavelength of the above magnitude. From a careful examination of the position of the lines on the film it should in principle be possible to distinguish between the two cases of interference pattern due to Compton scattering, and interference pattern due to crystal imperfections.

Such a procedure was suggested by Grenville-Wells (1951). However, the interference pattern is mostly so weak that only lines from the strongest reflecting planes can be observed. For such low-index planes the

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shift in the position of the lines due to the change in wavelength of the Compton radiation is small, usually less than 1 mm. for a cylindrical film of diameter 60 mm. Since, moreover, the lines are weak and often broad, Grenville-Wells was not able to distinguish experimentally between the two possibilities.

The filter technique

Several investigators (Wollan, 1934; Kappeler, 1936; Curien & Deroche, 1956) have used a system of three balanced filters to measure the amount of Compton radiation scattered at different angles. A simplified procedure can be used to show whether or not the observed interference pattern is due to Compton scattering. For an X-ray tube with molybdenum anode the wavelengths of the K radiation are

$$K\alpha_1\colon\ \lambda = 0{\cdot}7076\ \text{Å};\quad K\alpha_2\colon\ \lambda = 0{\cdot}7120\ \text{Å}\ .$$

For this radiation, the elements zirconium, yttrium and strontium, with absorption edges

$$\text{Zr}\colon \lambda_K=0{\cdot}6872\ \text{Å},\ \text{Y}\colon \lambda_K=0{\cdot}7255\ \text{Å},\ \text{Sr}\colon \lambda_K=0{\cdot}7693\ \text{Å},$$

form a natural filter triplet.

The absorption edge for yttrium lies in the region of the Compton wavelengths caused by molybdenum radiation. A filter of suitable thickness of this metal or of one of its compounds, placed between the irradiated crystal and the film, would only let through Compton scattering of wavelengths larger than the absorption edge for yttrium. If the observed interference pattern is due to Compton radiation, lines extending over a sufficiently large angular range in φ on an unfiltered film would on a filtered film start at a fixed minimum value of φ.

Experimental

The diamonds investigated were Belgian Congo cubes of the 'coated stone' type. They were yellowish-gray industrial diamonds of between 1 and 3 mm. edge. A parallel beam of Zr-filtered Mo radiation hit a diamond placed in the centre of a rotation camera, and diagrams were taken with and without an additional cylindrical filter of Y_2O_3 placed between the crystal and the film. Photographs for which the Y_2O_3 filter was replaced by a SrO filter were also taken. The main experimental difficulty consisted in preparing uniform filters of Y_2O_3 and SrO. Several procedures were tried. For the final filters the following method, suggested by Prof. Fankuchen, was used: Fine-ground powder of the oxide was suspended in a dilute solution of Duco cement in acetone. The suspension was sprayed on the lower side of a horizontally mounted clean glass plate, and the film was stripped after cutting the edges with a razor blade. Several films were superimposed to give a cylindrical filter of the desired thickness. When these filters were oscillated $\pm 10°$ during the exposure, variation in film density due to filter inhomogeneities was eliminated.

Experimental results

Figs. 2 and 3 show the result of the investigation. In both photographs the diamond is oriented with the incident beam in the [110] direction whereas $[1\bar{1}0]$ is parallel to the camera axis. The diagrams contain the region around the undeviated beam.

Fig. 2 represents an exposure without filter between crystal and film. In the photograph, a faint interference pattern can be seen, superimposed on the Laue spots. Four lines belonging to the $[1\bar{1}1]$ and $[\bar{1}11]$ cones should be easily recognizable. They run from φ almost zero to φ almost 180°. Two and two lines are parallel, parallel lines corresponding to reflections from opposite sides of the same set of planes. With Mo $K\alpha$ radiation the half angular opening of the [111] cones is 80·03°. For a cylindrical camera of diameter 60 mm., this means a shortest distance of 10 mm. between parallel lines on the film.

In Fig. 3 a filter of Y_2O_3 was placed between crystal and film. Also in this photograph faint lines can be seen. However, the lines now appear for large values of φ only. That is exactly what we would expect to find for lines caused by interference of Compton scattering.

The experiments thus prove that the interference pattern is due to Compton radiation emitted from the atoms in the crystal. This assumption is further confirmed by photographs where a filter of SrO was placed between crystal and film. Such photographs do not show any interference line even for exposure times twice that of Fig. 3, the SrO and Y_2O_3 filters being balanced to reduce the Mo $K\alpha$ radiation by the same amount. The absorption edge of Sr is larger than the maximum wavelength of the Compton scattering caused by Mo K radiation. A filter of SrO consequently absorbs all Compton radiation.

In Fig. 3 the interference lines do not start suddenly at a fixed value of the angle φ between incident beam and emitted Compton radiation. Instead, the intensity of the lines seems to increase gradually with φ. This is due to the existence of two incident wavelengths, Mo $K\alpha_1$ and Mo $K\alpha_2$, and to the comparatively large half-width of the Compton line, of the order of 15 X.U. (Du Mond & Kirkpatrick, 1931; Du Mond, 1933). For values of the angle φ that give a Compton wavelength close to the absorption edge for yttrium, the intensity at two points on the observed lines at a mutual distance of 1 mm. is caused by radiation with a difference in wavelength of about 0·7 X.U.; 15 X.U. thus correspond to 20 mm. on the interference line.

The influence of crystal perfection, shape and size of the diamond on the intensity of the lines

In order to establish whether the degree of crystal perfection would influence the intensity of the inter-

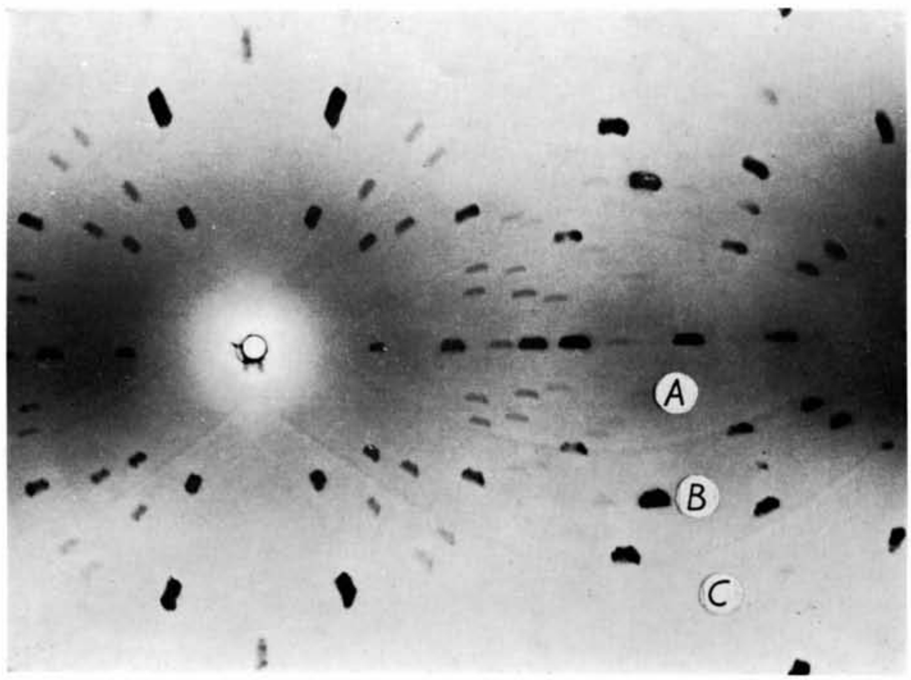

Fig. 2. Diamond. Incident beam in the [110] direction, [1$\bar{1}$0] vertical. Zr-filtered Mo radiation, 38 kVP., 15 mA. Cylindrical camera, diameter 60 mm. Exposure time: 3 hr.

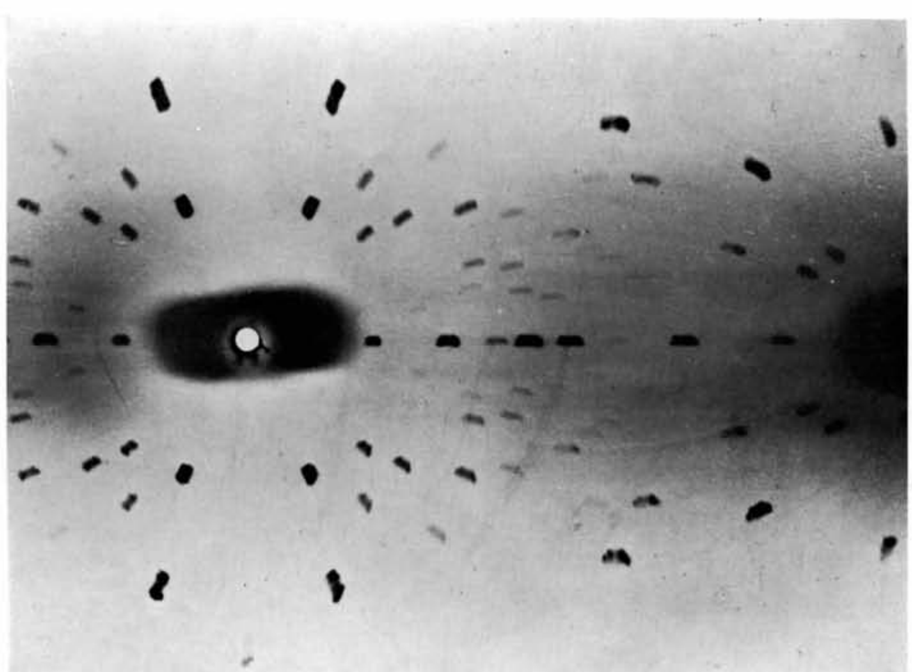

Fig. 3. Diamond. Incident beam in the [110] direction, [1$\bar{1}$0] vertical. Zr-filtered Mo radiation, 38 kVP., 15 mA. Cylindrical camera, diameter 60 mm. Exposure time: 12 hr. Cylindrical Y_2O_3 filter between crystal and film.

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ference pattern, eleven different diamonds were investigated. Nine of them were so-called 'coated stones', probably rather imperfect crystals. The other two seemed to be perfect octahedra with edges 1·2 mm. Four of the coated stones were cubes of edge approximately 1 mm., the other five were incomplete cubes with dimensions ranging between 3 and 6 mm. (Figs. 2 and 3 represent the X-ray pattern of an incomplete cube about 4 mm. in cross-section.)

Each diamond was examined for different orientations of the crystal and for exposure times varying between $\frac{1}{2}$ and 40 hr. The cross-section of the incident X-ray beam was 1 mm. Thus it was possible to compare diagrams from various parts of the same crystal. More than 100 exposures were made.

All the coated stones gave interference patterns. The intensity of the lines varied considerably from crystal to crystal. On the other hand the dependence on crystal orientation was small. The large incomplete cubes gave excellent patterns whereas only one of the smaller cubes showed strong interference lines. For the most imperfect diamond the Laue diagram consisted of clusters of diffuse spots. This diamond gave interference lines only when the incident beam passed through the centre of the crystal where the mosaic structure was probably less pronounced.

None of the perfect diamonds showed the interference effect. It is very unlikely that their octahedral shape can explain why no interference lines are observed. The reason must lie in the high perfection of these diamonds.

To establish the influence of the crystal size on the interference pattern, one of the incomplete cubes was split. Whereas a fraction $2 \times 0{\cdot}5$ mm. in certain directions gave a strong pattern, no lines could be observed when the dimension was reduced to about $0{\cdot}5 \times 0{\cdot}5$ mm. This suggests a certain minimum size of the diamonds in order that lines can be seen.

The fine structure of the lines

It is of interest to determine the fine structure of the Compton interference lines. Intersecting a line from a point between two parallel lines (i.e. point B in Fig. 2), it looks as if the intensity first rises above the background, then drops below it and finally reaches the background level. Several attempts were made to achieve conclusive microphotometer traces (I am very much indebted to Dr A. Taylor for his help). It was not until the intensity variations were enhanced by repeated reproductions on high-contrast films that the photometer curves in Fig. 4 were obtained. The upper curve represents the section AB (see Fig. 2), the lower curve the section BC. The density measurements were accomplished with a non-recording microphotometer, and readings were made every 0·05 mm. The curves correspond to the average of three independent readings.

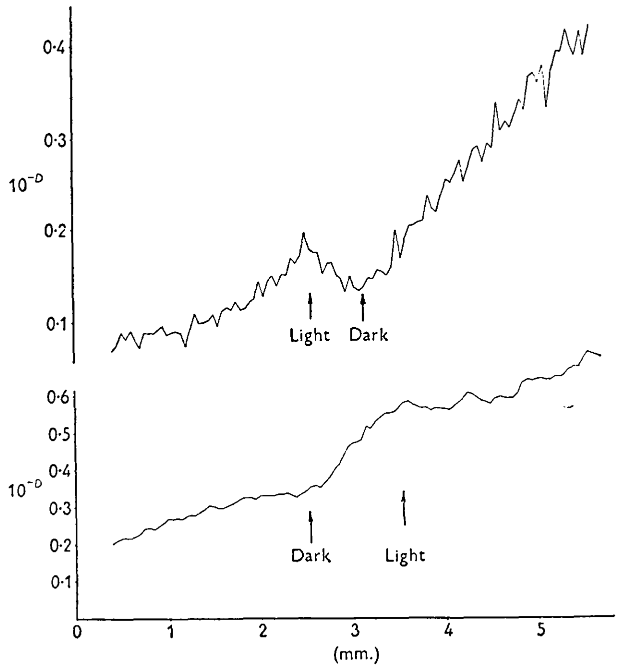

Fig. 4. Microphotometer traces of the interference lines. Upper curve represents section AB in Fig. 2, lower curve section BC. Ordinate scale 10^{-D}, where D is film density.

The curves in Fig. 4 show that the fine structure of the Compton interference lines is complementary to the fine structure of a Kossel line, where the light portion is always found on the convex side of the cone. The lines may be favourably compared with the separate deficiency and reflection lines in a divergent-beam pattern where the X-ray source is outside the crystal. As illustrated in Fig. 5, the X-rays are reflected towards R whereas the radiation is deficient in intensity in the directions E. The pattern in Fig. 5

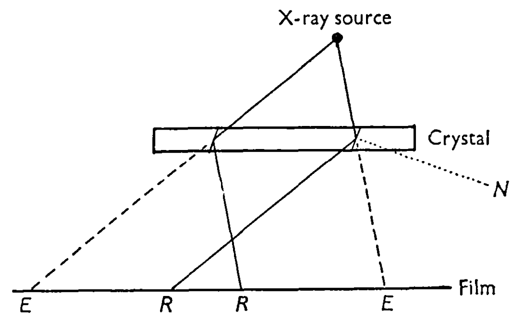

Fig. 5. The formation of reflection and deficiency lines in divergent-beam photographs.

can be described by two types of cones, of which the deficiency cone lies inside the reflection cone. The separation between the two lines depends on the distance between X-ray source and crystal and the thickness of the crystal, and can be made very small.

Lonsdale (1947) ascribes the divergent-beam pattern to secondary extinction. The experiments seem to show that the Compton interference lines must also be due to secondary extinction. Diamonds showing the interference effect are probably perfect over very small regions. The interaction between the Compton radiation emitted from an atom and the surrounding atoms within a perfect region is thus small and probably without importance for the appearance of the interference line. The fact that perfect diamonds do not give interference diagrams is further evidence, that secondary extinction, and not the nature of the wave-field in a perfect crystal, is the cause of the interference pattern. Since the crystal must be large enough for the secondary extinction to be effective, it is understandable why small diamonds do not show interference lines.

When radiation is reflected in a certain direction from the lattice planes, the intensity of the incident beam is reduced. Reflection from the opposite side of the planes will in the same directions decrease and increase the intensity respectively. From energy considerations we would expect the sum of the excess and defect of intensity compared to the general background to be zero in the two directions. With $[1\bar{1}0]$ vertical and [111] in the direction of the incident beam, three of the (111) lines pass close to the undeviated beam. The value of φ for Compton radiation in such a direction is close to zero, and considerably smaller than for the Compton radiation reflected in the same direction by the lattice planes. As expected, the lines appear dark. The corresponding light lines in the background are also seen.

Closing remarks

It has been shown that the interference pattern observed for diamond is due to Compton radiation emitted from the atoms within the crystal and reflected from the lattice planes. The fine structure of the lines is explained by assuming secondary extinction. Such interference diagrams may also be obtainable with other mosaic-type crystals. Like diamond, they should have low absorption and certain strongly reflecting planes.

The present work was carried out at the Polytechnic Institute of Brooklyn, and I am indebted to Prof. Fankuchen for use of the laboratory and X-ray equipment. My thanks are due to Prof. Ewald for suggesting the problem, for obtaining the necessary diamonds, through the courtesy of Dr Grenville-Wells, and for his advice during the investigation. My work in Brooklyn was made possible through a grant from the International Cooperation Administration, for which I want to express my gratitude.

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The Utilization of Relationships between Sign Relationships

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It is shown that sign relationships are interrelated in such a way that the failure of one particular sign relationship may inevitably lead to the failure of a number of others. A method of utilizing these interrelationships is illustrated by the determination of signs for projections of the (known) structures of purpurogallin and α-glucose.

For the application of direct methods of sign determination it is often convenient to represent the unknown signs by alphabetical symbols. In this paper the nomenclature suggested by Woolfson (1957) will be followed, and two-dimensional reflexions (say of type $hk0$) are divided into four groups

(*a*) h even k even;
(*b*) h odd k odd;
(*c*) h odd k even;
(*d*) h even k odd.

The signs of the reflexions of group (*a*) are represented