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The Study of Multiple Reflections with the Aid of a Microfocus X-ray Source

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A microfocus X-ray tube having a projected focus size *circa* 30×3 microns used with a simple photographic recording arrangement allows clear observation of *Aufhellungen* in single-crystal reflections and extinction lines in the directly transmitted beam. Multiple reflections, such as an apparent '200' in diamond generated by successive reflection from two octahedral planes, can be readily identified by their special shape. It is pointed out that uncertainty of coherent-domain boundary conditions presents a serious obstacle to the derivation of information on relative phases through the study of simultaneous X-ray reflections.

It is nowadays appreciated that the conditions necessary for the occurrence of multiple reflections are quite frequently fulfilled during the course of recording of diffraction data from single crystals. However, such reflections only become obvious when they appear as 'forbidden' reflections, (see, for instance, Bland (1954), and Silcock (1956)), for as long as the reciprocal-lattice points representing allowed reflections themselves form a Bravais translation lattice, a multiple reflection will always be superimposed upon a normal allowed reflection. One of the simplest structures in which multiple reflections can be observed alone is the diamond: with this crystal, for example, an apparent '200' reflection can be generated by successive reflection from two octahedral planes, $111+1\bar{1}\bar{1}=200$. Using a microfocus X-ray tube as source of X-rays, together with an appropriately designed (but quite simple) photographic recording technique, this forbidden 200 reflection can be observed with surprisingly high intensity. The interest of such an experiment resides in the fact that the precise definition of diffraction geometry made possible by a point X-ray source provides a clear demonstration of the relations between a number of associated diffraction phenomena, such as *Aufhellungen*, *Umweganregungen*, and intersections of extinction lines in the directly transmitted X-ray beam (Lang, 1955*a*). This information in turn throws light on the debated question of under what conditions, if any, can interference of X-ray reflections be observed in a manner likely to provide a clue to relative phases.

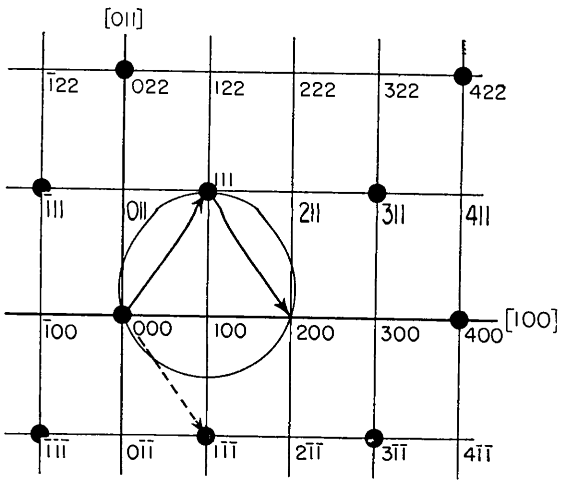

Fig. 1. Diamond reciprocal-lattice section.

In the case of the diamond '200', the reciprocal-lattice construction of Fig. 1 shows how this forbidden reflection can be generated by setting the crystal so that the Ewald sphere passes through the reciprocal-lattice points 111 and 200. The principle of the recording technique (Lang, 1955*b*) is illustrated in Fig. 2. X-rays diverge from a point source S and are limited by an aperture to form a beam of rectangular section. The beam is transmitted by the crystal under investigation, its intersections with the faces of the crystal are indicated schematically by the dashed rectangles A and A'. The sheet B represents the section of the crystal cut by those rays in the divergent incident beam which satisfy the conditions for Bragg reflection of the characteristic radiation by a particular plane. The transmitted Bragg reflection, as received on the photographic film, forms an image C which is a projection of the crystal section B. The fine structure of the image C can be studied to obtain point-by-point information on reflecting power and macromosaic misorientations within the volume of the crystal. An image, D, of the directly transmitted beam can also be recorded. In the present instance, C represents the 111 reflection of diamond. Now *if* the 200 reflection were allowed, then, with the crystal setting of Fig. 1, another sheet of X-rays, E, would make the correct angle with the crystal to produce the reflection 200. The projection of E would appear on the film as the image F. However, part of the beam reflected by the (111) planes can, as it passes through the crystal, be reflected again, this time by the $(1\bar{1}\bar{1})$ planes. Points in the crystal at which the once-

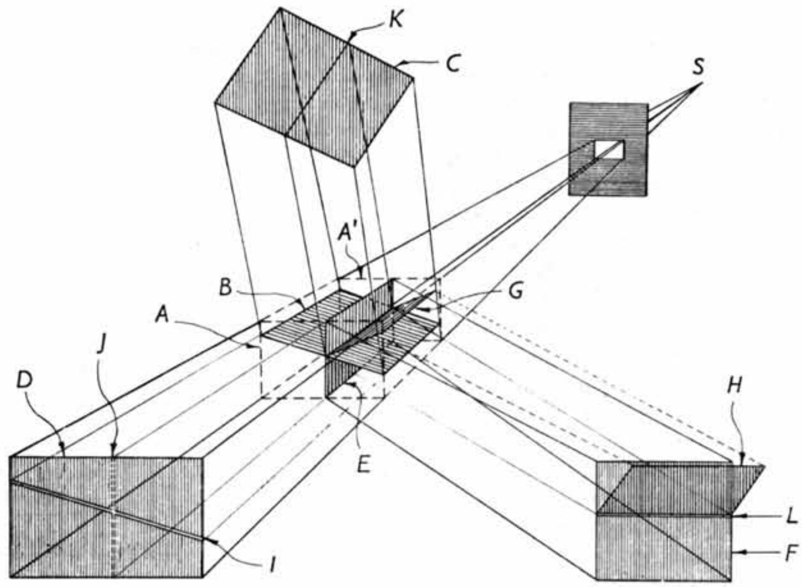

Fig. 2. Geometry of incident and reflected X-ray beams.

reflected rays make the correct angle for reflection by $(1\bar{1}\bar{1})$ planes lie on the sheet G. The twice-reflected beam leaves the crystal in the same direction as a 200 reflection, and the projected image of the crystal section G thus appears on the film at H. It will be seen that H will generally be smaller than F, but it need not fall wholly within the limits of F. Fig. 2 represents the case of a 'good' crystal. If the crystal were a 'bad' one, containing low-angle boundaries or warped lattice planes, then the sheets B and E would be faulted and dislocated, the sheet G would be incomplete, and the forbidden reflection would make an irregular image on the film, of area less than H. If conventional X-ray tube and collimating arrangements were used the basic geometry would of course be the same, but all details would be obscured. The Hilger microfocus tube produces a focal spot of effective dimensions about $30\mu \times 3\mu$, when the target is viewed at a take-off angle of 6°. The precise definition of the images provided in these circumstances not only allows immediate recognition of a double reflection but also makes possible the observation under high resolution of *Aufhellungen* and extinction lines.

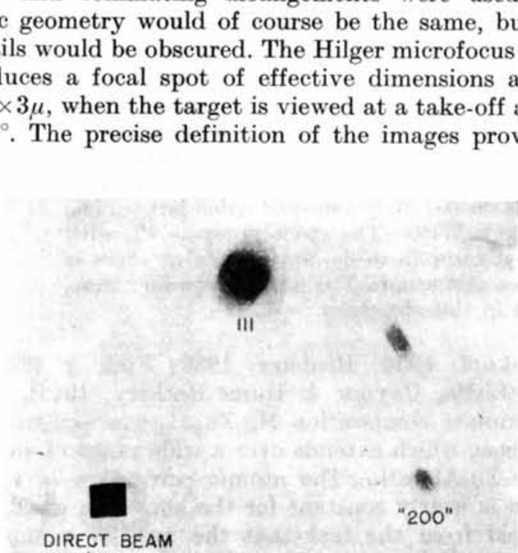

Fig. 3. Diamond diffraction pattern showing forbidden 200 reflection.

Fig. 3 shows the diffraction pattern obtained with the geometry of Figs. 1 and 2, using a rather imperfect, strongly reflecting diamond. The crystal, of 3·15 carats, had the habit of a distorted octahedron, elongated along [011]. Ag $K\alpha$ radiation was used and the tube kilovoltage was kept below 44 kV. so that the harmonic $\frac{1}{2}\lambda$ was not excited. When the kilovoltage was raised just above 44 the '200' double reflection appeared partly framed in a weak 400 Laue spot. Using unfiltered Ag $K\alpha$ radiation, a tube current of 300 μA. and a tube kilovoltage of about 42 kV., the 'forbidden' reflection was strongly visible after only a few minutes exposure. The weak reflection seen on the upper right of Fig. 3 is a 311 Laue spot. Where the sheet B, extended, cuts D there will be found an extinction line, I, crossing the image of the direct beam (not visible with the magnification and degree of exposure of Fig. 3). If a reasonably strong direct 200 reflection were possible, then another extinction line J would be found where the extension of the sheet E cuts D, and the diffraction spots C and F would be crossed by *Aufhellungen* at K and L, respectively. Extinction lines, and *Aufhellungen* in various reflections have been observed with diamonds, but they are much more conspicuous in the case of LiF, NaCl and aluminum crystals.

If the 200 were a weak reflection, then the double reflection H would be seen superimposed upon it. The question has been raised (for example, by Lipscomb, 1949) as to whether information on relative phases can be obtained from such superimpositions. The answer is clear from Fig. 2. Since the diffracted beams F

and H arise from reflections from different crystal sections, E and G, there is no coherence between them, and no interference can be expected. It is only for the one incident ray, that which passes through the intersection of sheets B and E and which is *simultaneously* reflected by more than one plane, that interference effects are possible. This fact suggests two experimental approaches. Either one may employ a closely collimated incident beam, restricted to lie in one of the sheets B or E, and study the form of the maxima and minima in reflections when simultaneous reflection occurs (Williamson & Fankuchen, 1955, 1956), or one could use the present technique to study the fine-structure of intersections of extinction lines in the directly transmitted beam, attempting to do with X-rays what Miyake & Kambe have done for electrons (Kambe, 1954). In either case there are serious obstacles to be overcome, even supposing the techniques were developed to give adequate resolution. The volume of crystal traversed by the X-ray beam may be expected to contain many coherent domains, and the configuration of internal domain boundaries will be unknown. Hence the form and strength of the dispersion surface for each domain may be different, all effects observed would be the resultant of contributions from a number of domains, and could hardly be expected to be interpretable. Only if a sufficiently thin slice of a highly perfect crystal were used as specimen would there be a good possibility of obtaining defined boundary conditions. Unfortunately such crystals are not likely to be encountered among those for which it is a matter of practical interest to gain information on relative phases of reflections.

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The Crystal Structure of the Metallic Phase Mg_{32} (Al, Zn)$_{49}$*

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A complete determination of the crystal structure of the ternary phase (λ or T phase) in the magnesium–aluminum–zinc system with composition approximately $Mg_3Zn_3Al_2$ has been carried out with use of intensity data from single-crystal Weissenberg photographs. The refinement of the structural parameters was carried out by use of Fourier projections, followed by application of the method of least squares. The unit of structure, based on the body-centered cubic lattice, has $a_0 = 14 \cdot 16$ Å, as previously reported by Laves, Löhberg & Witte. The space group is T_h^5, with 162 atoms per unit cube. The structure shows the correct formula to be Mg_{32}(Al, Zn)$_{49}$; there is some disorder in occupancy of positions by aluminum and zinc atoms. The icosahedron and truncated tetrahedron are prominent coordination polyhedra in this structure.

Introduction

Phase diagram studies of the magnesium–aluminum–zinc system have shown that there exists a ternary phase, which has been given the name λ phase or T phase (Egar, 1913; Köster & Wolf, 1936; Köster & Dullenkopf, 1936; Riederer, 1936; Fink & Willey, 1937; Little, Raynor & Hume-Rothery, 1943). The approximate composition $Mg_3Zn_3Al_2$ was assigned to the phase, which extends over a wide range of values of the Zn/Al ratio. The atomic percentage of magnesium is nearly constant for the alloys, as would be expected from the fact that the metallic radius of magnesium is about 15% greater than those of aluminum and zinc.

Laves, Löhberg & Witte (1935) made an X-ray

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