DIRECT DETERMINATION OF X-RAY REFLECTION PHASE RELATIONSHIPS THROUGH SIMULTANEOUS REFLECTION*

M. Hart and A. R. Lang
H. H. Wills Physics Laboratory, University of Bristol, Bristol, England

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This Letter reports a successful direct experimental determination of x-ray reflection phase relationships through coherent dynamical interactions in simultaneous x-ray reflection. When a crystal is so oriented with respect to the incident beam that both reflections h ($=hkl$) and h' ($=h'k'l'$) are excited, the region in reciprocal space concerned is in the vicinity of the point of intersection of the three spheres Σ_0, Σ_h, and $\Sigma_{h'}$ centered, respectively, on reciprocal lattice points O, h, and h', the radii of the spheres being $\kappa = nK$ where n is the crystal refractive index and K the free-space wave number for the radiation used. In this region the dispersion surface is made up of three sheets, and in the crystal there are nine wave vectors $\vec{k}_g^{(i)}$ ($g=O, h, h'$; $i=1, 2, 3$). (For diffraction at low angles, polarization effects may be neglected; otherwise one would have to consider six sheets.) Clearly the situation is very complicated when all the diffracted waves are comparable in amplitude. However, we have achieved our result in a straightforward way by three steps.

(1) High-resolution x-ray diffraction topographs[1,2] are used to provide adequate definition of angular and spatial relations.

(2) A valuable simplification is gained by restricting attention to an investigation of the modification of the dispersion surface of the h reflection as one proceeds along the line of intersection of Σ_0 and Σ_h and approaches the intersection with $\Sigma_{h'}$.

(3) For studying small modifications of the dispersion surface we make use of the phenomenon of Pendellösung interference. It has been shown[3,4] that in x-ray projection topographs[2] of wedge-shaped crystals in the Laue case the Pendellösung fringe pattern is determined by the minimum diameter of the dispersion surface hyperbolae, which is proportional to the structure amplitude $|F_h|$. The fringe spacing, for a given wedge angle, is inversely proportional to this diameter. In the present investigation, displacements of Pendellösung fringes on the topograph provide the measure of modifications of the dispersion surface.

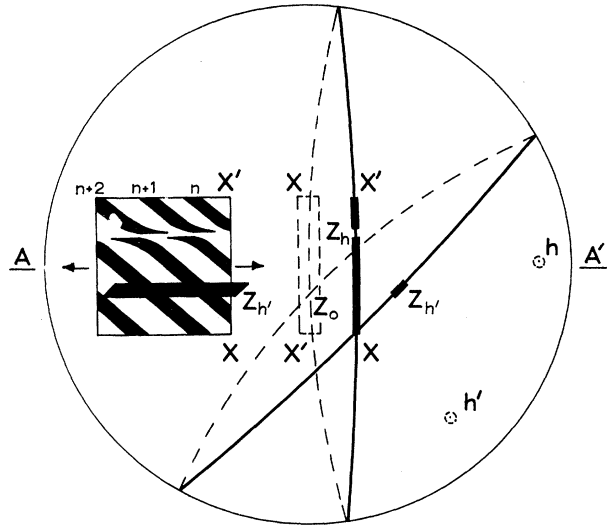

FIG. 1. Diffraction geometry for simultaneous reflection.

The geometry of the experiments is explained in the stereographic projection, Fig. 1. The crystal (not shown) is at the center of the sphere. Diffracted beams lie in the hemisphere above the paper, whereas the incident beam lies in the hemisphere below the paper. Continuous and interrupted lines are used, respectively, on the hemispheres above and below the paper. The line AA' represents the horizontal and the crystal is illuminated by a narrow vertical ribbon of x rays. The angular range of the incident-ray bundle is represented by the interrupted rectangle, much exaggerated in the drawing. The x-ray source is effectively a point and the vertical range of incident rays is determined by the height of crystal illuminated. There is thus a one-to-one correspondence between position on the crystal and the vertical angle of the ray incident upon it. The plane normals h and h' are shown, together with the cones generated by all rays making the appropriate Bragg angle with these planes for the radiation used (Ag $K\alpha_1$). The plane normal h lies horizontal and the whole of the incident beam straddles the h cone. A diffracted beam the full height of the crystal is thus obtained, incident rays X and X' giving rise to diffracted rays X and X', respectively. Next, the crystal is rotated about plane normal h to bring the h' cone into the incident beam, but in this case the incident beam cuts only a small part of the cone. The incident ray common to both the h cone and h' cone is at Z_0, the small bundle of the h' diffracted beam is at $Z_{h'}$, and in the extended h diffracted beam we find an Aufhellung[5] in the direction Z_h corresponding to the incident ray Z_0. The crystal has the shape of a gently tapering wedge, with the thickness contours making a moderate angle with the horizontal. The projection topograph obtained by traversing the crystal back and forth appears as sketched in the left-hand part of Fig. 1. The thin end of the wedge is in the upper right and the Pendellösung fringe order is in the sequence indicated. (The diffracted beam $Z_{h'}$ may possibly pass through the diffracted beam screening slits and appear as a streak as shown.)

The electron diffraction dynamical theory of Kambe and Miyake,[6,7] modified to the x-ray case, shows that in the vicinity of $\Sigma_{h'}$ the minimum diameter of the h reflection dispersion surface hyperbolae is proportional not to $|F_h|$ but to quantities $|F_+|$ just inside $\Sigma_{h'}$, and $|F_-|$ just outside $\Sigma_{h'}$, to a good approximation. Whether $|F_+|$ is greater or less than $|F_-|$ depends upon whether $\cos\Phi_{hh'}$ is positive or negative, $\Phi_{hh'}$ being a function of the individual phase angles, α, of the reflections h, h', and $h'-h$,

$$\Phi_{hh'} = \alpha_h - \alpha_{h'} + \alpha_{h'-h}.$$

In a centrosymmetric cyrstal, $\Phi_{hh'}$ can only be zero or π; hence $\cos\Phi_{hh'}$ is then restricted to the values ±1. Now wave vectors of rays incident between X and Z_0 (and hence diffracted between X and Z_h) lie outside $\Sigma_{h'}$, whereas those incident between X' and Z_0 lie inside $\Sigma_{h'}$. Thus, on the topograph, one expects the effective diameter of dispersion surface hyperbolae to be proportional to $|F_+|$ just above the Aufhellung, and to $|F_-|$ just below the Aufhellung. Hence if $|F_+|$ is greater than $|F_-|$ the Pendellösung fringe spacing is decreased above the Aufhellung and increased below; and vice versa. In the former case the fringe discontinuity appears as sketched. The experiment is quite a sensitive one, since at the fringe of order 5, a change of only 10% in dispersion surface diameter will shift the fringes by half a period.

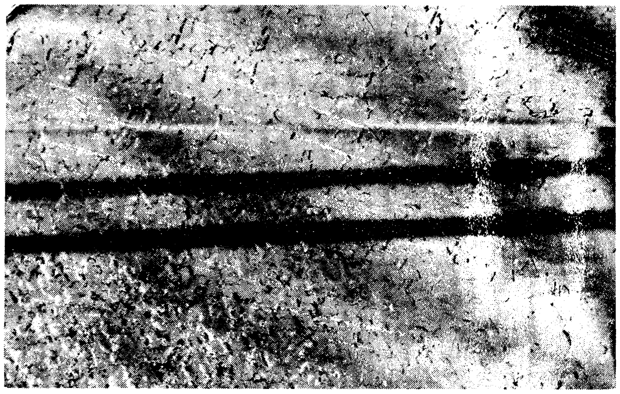

FIG. 2. Diffraction topograph showing dislocation of Pendellösung fringes at an Aufhellung.

A topograph corresponding to the sketch in Fig. 1 is shown in Fig. 2. The crystal is germanium, the reflection h is $2\bar{2}0$ and the reflection h' is $3\bar{1}\bar{1}$. The Aufhellung appears across the field and the streaks due to the h' reflection (both $K\alpha_1$ and $K\alpha_2$) appear across the lower half. Although the crystal thickness variation is not as smooth as could be desired, and numerous dislocations are scattered over the field, the form of the shift of the Pendellösung fringes clearly shows that $|F_+|$ is greater than $|F_-|$.

Since the structure is centrosymmetric, the observation shows that $\cos\Phi_{hh'} = 1$, and hence $\Phi_{hh'} = \alpha_h - \alpha_{h'} + \alpha_{h'-h} = 0$. This result is in agreement with that calculated for the known structure. A direct experimental determination of phase relationships is thus effected.

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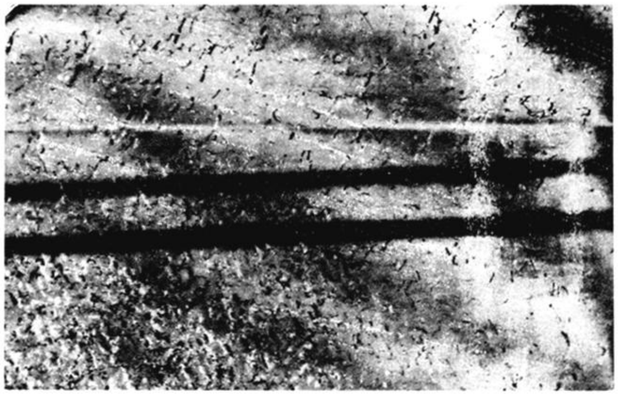

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