Observations of Borrmann-Lehmann Interference Patterns with Synchrotron Radiation

BY A. R. LANG AND G. KOWALSKI*

H. H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, England

A. P. W. MAKEPEACE

Department of Physiology, School of Medicine, University of Bristol, Bristol BS8 1TD, England

AND M. MOORE

Department of Physics, Royal Holloway and Bedford New College, Egham Hill, Egham, Surrey TW20 0EX, England

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Abstract


* On leave from Institute of Experimental Physics, University of Warsaw, Poland.

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surface of (100) type and the Bragg-diffracting planes and internally reflecting Bragg surface oriented orthogonal to the entrance surface. Fringes of high visibility were obtained, even from specimens showing significant X-ray topographic evidence of lattice imperfections, but the fringe spacings observed were extremely sensitive to lattice distortions. Measurements of $F_{220}$ from Borrmann-Lehmann fringe spacings gave values much in error, worse with the shorter wavelength. Borrmann-Lehmann fringe-spacing measurement is advocated as a sensitive test of lattice perfection.

1. Introduction

The roles played by the bounding surfaces of a crystal in dynamical and in kinematical diffraction theory may be contrasted. According to kinematical theory, the crystal shape determines the variation of diffracted intensity with glancing angle (the diffraction line profile) but does not affect the integrated intensity (which depends only on the crystal volume, so long as absorption can be neglected). On the other hand, when dynamical diffraction theory applies, the strength of Bragg-diffracted rays is strongly dependent upon the geometrical shape of the crystal: partial or total reflection may occur, and the intensity of Bragg-diffracted rays generally varies in an oscillatory manner depending upon the trajectory between X-ray entrance and exit surfaces. The best known oscillations are those of Pendellösung fringes. Such fringes arise from dispersion-surface interbranch interference. Less familiar are intrabranch interference phenomena dependent upon the geometry of the crystal surfaces. Those in the latter class first to be explored experimentally are named after their discoverers (Borrmann & Lehmann, 1963; Lehmann & Borrmann, 1967).

In the early studies of Borrmann-Lehmann fringes the diffraction conditions corresponded to the high-absorption case, with $\mu t > 10$, $\mu t$ being the normal X-ray linear absorption coefficient and $t$ the crystal thickness. Thus, in the first experiments (Borrmann & Lehmann, 1963) the value of $\mu t$ was 18 and in later experiments (Lehmann & Borrmann, 1967) it was 76. Accordingly, observations were confined to a very narrow central bundle of rays within the energy-flow triangle contained between the directions $K_s$ and $K_g$ subtending from the point of incidence on the crystal: only those rays travelling nearly parallel to the Bragg planes and for which anomalous transmission was strong could be photographed.

In the present work absorption was moderately low ($\mu t = 1-6$) or small ($\mu t = 0.47$), and rays spanning the whole opening angle of the energy-flow triangle were observed. We have made use of the continuous spectrum of synchrotron radiation to record fringe patterns at selected wavelengths, and the property of linear polarization of synchrotron radiation has been exploited to produce patterns with either $\sigma$-mode or $\pi$-mode polarizations alone. The latter was done both for the purpose of eliminating overlap of $\sigma$-mode and $\pi$-mode patterns and also to enable us to control the strength of scattering via the factor $|\cos 2\theta_i|$. The specimens we used were polished parallelepipsed of natural diamonds. One of these was deemed to be of sufficiently high long-range lattice perfection to be suitable for testing the usefulness of Borrmann-Lehmann fringe-spacing measurements to calculate $F_s$. The numerical values derived were disappointingly bad, but they pointed to a useful application of Borrmann-Lehmann fringe measurements in a quite different direction, that of serving as an extremely sensitive indicator of lattice distortion.

2. Diffraction geometry

Consider a crystal bounded by plane facets. Let $K_0$ and $K_s$ be the wave vectors of incident waves and of waves Bragg-diffracted by reflection $g$, respectively. These vectors refer to wave directions outside the crystal. For present purposes their directions need not be distinguished from those of the corresponding crystal waves, $k_0$ and $k_s$, which are linked precisely by $k_s = k_0 + g$.) For each facet, identified by its unit normal $n$, a negative value of $(K_0, n)(K_s, n)$ defines a 'Bragg surface', and a positive value a 'Laue surface' (and independently of whether $n$ is defined as inward or outward drawn). When $(K_0, n)(K_s, n)$ is positive at both X-ray entrance and exit surfaces of the specimen we have Laue-Laue diffraction, as in the familiar situation of a plate-shaped specimen when both $K_0$ and $K_s$ pass through the plate from one major surface to another. However, if the point of incidence of X-rays is moved towards an edge of the plate the energy-flow triangle may be cut by a lateral surface whose normal $n$ makes $(K_0, n)(K_s, n) < 0$. We then have Laue-Bragg diffraction, at least in part of the energy-flow triangle (Saka, Katagawa & Kato, 1972a). Two Laue-Bragg situations have to be considered, depending upon whether the lateral surface cuts into the $K_s$ side or the $K_0$ side of the energy-flow triangle. They are shown in Figs. 1(a) and (b), respectively. The diagrams are drawn with the simplifying assumption that the section of the crystal by the plane of incidence is rectangular, and that the Bragg-diffracting planes are normal to the major faces of the plate (the 'symmetrical Laue case' for Laue-Laue diffraction). The patterns reproduced below were all obtained with such simple specimen geometry. An important parameter is the ratio of $a$, the distance of the point of incidence of X-rays from the edge of the crystal, to $t$, the crystal thickness. Obviously, Laue-Bragg diffraction conditions only come into play when $(a/t) < \tan \theta_i$. 

A property of Bragg surfaces is that when the Poynting vector $s$ of a Bloch wave in the crystal impinges upon a Bragg surface a significant fraction of its energy is reflected back into the crystal. Thus, in the situations shown in Fig. 1 [and more fully explained in Fig. 4(a)], rays may reach the Laue exit surface to the right of the corner $E$ by two paths, one direct from the point of incidence of X-rays and the other via reflection at the lateral Bragg surface. The optical path difference between these rays produces the Borrmann–Lehmann fringes which can be observed in both the $K_0$ and the $K_\pi$ beams issuing from the crystal surface to the right of $E$. A complete theoretical treatment of the Laue–Bragg case has been presented by Saka, Katagawa & Kato (1972a). They call the diffraction geometry of Fig. 1(a) ‘Type I’, and that of Fig. 1(b) ‘Type II’. We suggest that a more descriptive and easily remembered terminology is desirable. The significant action of the Bragg surface is partial reflection of the Poynting vector $s_i$ impinging on it from within the crystal. Calling the reflected Poynting vector in the crystal $s_r$, we have $s_r = s_i + \Delta s$. In Fig. 1(a) $\Delta s \cdot g$ is negative, and in Fig. 1(b) $\Delta s \cdot g$ is positive. Accordingly, we propose that ‘Type I’ be termed the ‘negative case’, and ‘Type II’ the ‘positive case’.

Some characteristics of Borrmann–Lehmann fringes are illustrated by the section topographs in Figs. 2 and 3. These were obtained with a conventional radiation source (Cu Ka$_\alpha$) before the Synchrotron Radiation Source at Daresbury became operational. Fig. 2 is the $K_\pi$-beam pattern taken in the negative setting, Fig. 1(a). Borrmann–Lehmann interference fringes can be observed in both $K_\omega$- and $K_\pi$-beam images of the Laue exit surface to the right of the edge $E$. Crystal settings: (a) ‘Type I’, ‘negative’, (b) ‘Type II’, ‘positive’.

Fig. 1. Diffraction geometry for observing Borrmann–Lehmann interference phenomena. Parallelepiped-shaped crystal with faces normal to the plane of incidence. The Bragg-diffracting planes are normal to the Laue entrance surface and parallel to the lateral Bragg surface. Diffraction angles correspond to the 220 reflection of Cu Ka$_\alpha$ by diamond; $2\theta_\alpha = 75^\circ$. $P_\omega$ and $P_\pi$ are photographic plates set normal to the $K_\omega$ and $K_\pi$ beams, respectively. The screen $S$ is movable as indicated by the double-headed arrows. With $S$ withdrawn, the directly transmitted narrow ribbon incident beam is recorded on $P_\omega$. Borrmann–Lehmann interference fringes can be observed in both $K_\omega$- and $K_\pi$-beam images of the Laue exit surface to the right of the edge $E$. Crystal settings: (a) ‘Type I’, ‘negative’, (b) ‘Type II’, ‘positive’.

Fig. 2. $K_\pi$-beam section topograph showing Borrmann–Lehmann fringes, negative setting (cf. Fig. 1a), Cu Ka$_\alpha$ radiation, 220-type reflection. Arrow points to the image of the crystal edge $E$. Laue–Bragg conditions to the left of the arrow, Laue–Laue conditions to its right. Crystal thickness $t = 1$ mm ($\mu t = 1.6$), $a = 0.18$ mm, width of section topograph image $= 1.25$ mm, full crystal height $= 5$ mm, but the image height reproduced here is 1.4 mm. (All topographs in the figures were recorded on Ilford L4 nuclear plates, emulsion thickness 25 $\mu$m.)
internal, are to be blamed for fringe-spacing inconstancy.

To illustrate the positive setting, Fig. 1(b), both a $K_\sigma$-beam image (Fig. 3a) and a $K_\rho$-beam image (Fig. 3b) are shown. (The features arrowed are identified in the figure captions.) The same edge $E$ of the same specimen appears in both Figs. 2 and 3. The average Borrmann–Lehmann fringe spacing in Fig. 3 is larger than in Fig. 2 because $a$ is smaller, but the general patterns of fringe-spacing inconstancy are recognizably similar in the two settings. In $K_\sigma$-beam section topographs, the intensity tails off to zero at the margin of the pattern remote from the ribbon incident beam, as predicted by theory (Kato, 1960). This weak and featureless side of the section image has not been included in the field reproduced. Noteworthy in Fig. 3(b) is the strong image of the incident beam reflected from the Bragg surface. On the original plates it is seen that the strength and intensity profile of this image closely resemble those of the intense margins of a normal $K_\sigma$-beam section in the low and moderately low absorption cases. Consequently, it is easy to measure accurately the distance ($A$) on the plates between this Bragg-reflected beam and the Laue-margin of the section topograph. Thus $a$ may be accurately measured, via $a = \frac{1}{2}A \sec \theta_B$ with the present simple geometry. Comparison of $K_0$ and $K_\rho$ beam pairs in both positive and negative settings confirms that Borrmann–Lehmann fringe maxima in the $K_0$ and $K_\rho$ beams coincide in position along the exit surface, and that a minimum occurs at the edge $E$, in agreement with theory (Lehmann & Borrmann, 1967; Saka, Katagawa & Kato, 1972a, b).

3. Simple theory

Fig. 4(a) shows the ray paths involved in Borrmann–Lehmann fringe formation. These are similar for the Type I, negative, and Type II positive settings; the latter is depicted in Fig. 4(a). For calculating fringe

![Ray paths involved in Borrmann-Lehmann interference in the Laue-Bragg case, positive setting (Type II).](image-url)
spacings due to interference between waves following trajectory \([1]+[2]\) and those following trajectory \([3]\) the simple analysis given by Borrmann & Lehmann (1963) suffices. Fig. 4(b) shows the relevant wave points on the dispersion surface. For the high-absorption case Borrmann and Lehmann needed to consider only branch (1) and waves in the \(\sigma\)-polarization state. Here both branches (1) and (2) are considered, but only for a single polarization mode, in conformity with conditions applying in the synchrotron radiation experiments. For each ray drawn in Fig. 4(a), i.e. those identified as \([1],[2],[3]\) and \([E]\), the relevant wave points \(P^{(1)}\) and \(P^{(2)}\) on the dispersion surface branches, the Poynting vector \(s\), the angle \(\Theta\) between \(s\) and the Bragg plane, and the energy-flow parameter \(p = \tan \Theta / \tan \theta_b\) are identified by subscripts. The angle \(\Theta\) is conventionally taken as negative when \(s\) lies between the Bragg plane and \(K_0\), so that, with \(FO = a\) and \(EW = b\), tan \(\Theta_F = -a/t\) and tan \(\Theta_s = (b-a)/t\), for example. In the following elementary calculation the rectangular components \(X\) and \(Z\) of the difference between crystal wave vectors and the vector \(k_F^\parallel = k_0^\parallel + g\) at the Lorentz point \(O\) in Fig. 4(b) are employed. Since the Bragg surface is parallel to the Bragg reflecting plane, both \(X\) and the \(g\) component of the ray path reverse exactly after reflection. Hence to find the phase difference, \(S\), between trajectory \([1]+[2]\) and trajectory \([3]\) one can simply compute the phase difference between trajectory \([3]\) and trajectory \([1]\) continued to \(W^*\) on an assumed extended exit face. \(\left[\text{This neglects the reversal of phase of the} k^\parallel \text{wave on reflection, needed to satisfy the boundary conditions on the Bragg surface} \right.\left(\text{Lehmann \& Borrmann, 1967; Saka, Katagawa \& Kato, 1972a}.\right)\]

For branch-(1) waves at \(W\)

\[
S^{(1)} = 2\pi |(Z_1^\parallel)^2 - |Z_1^\parallel^2| - 2\pi (a - b) |X_3^\parallel| + 2\pi (a + b) |X_1^\parallel|.
\]

From the symmetry of Fig. 4(b) it is apparent that the magnitudes of \(S^{(1)}\) and \(S^{(2)}\) are the same. Hence superscripts can be dropped, and calculation continued for branch (1) alone. In the coordinate system of Fig. 4(b) the dispersion-surface hyperbolae are given by

\[
Z^2 = X^2 \tan^2 \theta_b + (2\xi_g)^{-2}.
\]

The diameter \(V_1 V_2\) is the reciprocal of the extinction distance \(\xi_g\) \(\left(\text{the }\text{Pendellösung} \text{ period parallel to the Bragg planes}\right): \xi_g = (e/mc^2)(Fg/CA)/(\pi V \cos \theta_b),\) where \(C\) is the polarization factor and \(V\) the volume of the unit cell. For any Poynting vector \(s\), tan \(\Theta = -dZ/dX\) and, from (2), tan \(\Theta = -X \tan^2 \theta_n / Z\). With the assumption that \(b\) is small compared with \(a\) it is convenient to refer phases to that of ray \([E]\) and to introduce \(\Delta X = X_3 - X_1, 2\Delta Z = Z_1 - Z_3\), with \(\Delta Z = -\tan \theta_b \Delta X = (a/t) \Delta X\). Substitution in (1) gives simply

\[
S = 4\pi \xi_g X_g.
\]

The proportionality of \(S\) to \(b\) shows that the fringes are equispaced along the exit surface close to \(E\). Their spacing, \(\Delta_a\), on the exit surface is

\[
\Delta_a = (2X_g)^{-1}.
\]

If the deviation parameter \(\omega = 2\xi_g \tan \theta_b X\) and the relation \(\omega^2 = p^2/(1 - p^2)\) are introduced, the right-hand side of (4) can be expressed in terms of \(p = a/(t \tan \theta_b)\) and \(\xi_g\). When \(p^2\) is sufficiently small that one may put \(\omega^3 = p^3\),

\[
\Delta_a = \xi_g \tan^2 \theta_b (t/a),
\]

or

\[
\Delta = \xi_g \sin \theta_b \theta_n (t/a)
\]

when the fringe spacing \(\Delta = \Delta_a \cos \theta_b\) on the photographic plate is measured. Equations (5) reproduce Borrmann & Lehmann's finding. Without the approximation applicable when \(p^2\) is small,

\[
\Delta_a = \xi_g \tan^2 \theta_b (t/a)(1 - p^2)^{1/2}.
\]

The expression used for calculating \(F_\xi\) from \(\Delta\) is

\[
F_\xi = \frac{(mc^2\pi V)}{e^2 \sin \theta_p (1 - p^2)^{1/2} \frac{1}{a\Delta}}.
\]

In their spherical-wave analysis of the Laue-Bragg case Saka, Katagawa & Kato (1972a) show that the Bragg-reflected waves behave as if they were diverging from a virtual source outside the crystal. In our simple geometry the virtual source is located at \(O'\) on \(OF\) prolonged, with \(O'F = FO\). With this model of two energy-flow triangles overlapping, the similarity of contributions to the \(K_\xi\)-beam image from rays \(OR\) and \(PR\) in the positive setting [noted above in regard to Fig. 3(b)] is understandable. Furthermore, under low- and moderate-absorption conditions the Pendellösung fringe systems of the two energy-flow triangles overlap between \(E\) and \(R\). In fact, reference to Fig. 4(b) shows that for each polarization mode four periodicities should be recognizable in patterns produced by a perfect lightly-absorbing crystal: the Borrmann-Lehmann fringes, the two Pendellösung fringe systems, and the beat between the latter pair. In the patterns shown here, the calculated beat period between the Pendellösung fringe system was large close to \(E\) compared with the period of the first few Borrmann-Lehmann fringes measured. From the expressions given by Kato [1961, equation (55); 1974, equation (4.21b)] the Pendellösung fringe spacing at any point on the base of the energy-flow triangle in the symmetrical Laue-Laue case is easily calculated. At \(W\), for example, the spacing on the exit surface due to the source at \(O\) is

\[
\xi_g \tan^2 \theta_B (1 - p^2) \frac{1}{a - b}.
\]
At $E$ the fringe spacing of both Pendellösung fringe systems is
\[ \xi_n \tan^2 \theta_n (1 - p_{2n}^2)^{1/2} (t/a), \]
just the same as the Borrmann-Lehmann fringe spacing, equation (6). The reason for this identity is apparent from Fig. 4(b). When $W$ is close to $E$ the difference between $[k_{g_1} - k_{g_2}]$ at $E$ and $W$ is $2P_{E}P_{W}^{(1)} = P_{E}^{(1)}P_{W}^{(3)}$. Of course whether the two Pendellösung fringe systems will reinforce or cancel each other in the vicinity of $E$ depends upon the value of $a$.

4. Observations with synchrotron radiation

4.1. Comparison of patterns obtained with Cu $K\alpha_1$ and with synchrotron radiation

For the first few fringes, close to $E$, the fringe visibility is 100% in the $K_\sigma$ beam with the positive setting (Type II), and also 100% in the $K_\rho$ beam in the negative setting (Type I). It is experimentally easier to record $K_\rho$-beam patterns since the precisely positionable screen $S$ is not required. In synchrotron-radiation experiments the Bragg reflection utilizes a much smaller fraction of the total energy of the X-ray spectrum of the source than when a characteristic radiation such as Cu $K\alpha_1$, produced in a conventional X-ray tube with a clean target and run at optimum kilovoltage, is being reflected. A practical consequence in experiments with white synchrotron radiation is that at diffraction angles close to $K\alpha_1$ there is relatively more unwanted flux from scattering by air and slit jaws than in an equivalent conventional-source experiment. Therefore, recording of synchrotron-radiation $K_\rho$-beam Borrmann-Lehmann fringe patterns has not been attempted to date, and experiments have been confined to the positive setting in order to obtain best fringe contrast in the $K_\rho$ beam. Some experiments were performed with the same arrangement in plan as in Fig. 1(b) but with the specimen oriented so that the Bragg surface was not normal to the plane of incidence. In this case $a$ varied linearly with distance normal to the plane of incidence. This geometry produced interestingly complex patterns, but the simple geometry of Fig. 1(b), with constant $a$ for the whole crystal region imaged, was preferred for making fringe-spacing measurements.

The diamond which produced the images seen in Figs. 2 and 3 was the most perfect suitably faceted specimen available for the present study; but in these topographs departures from long-range lattice perfection were strongly evident. However, when set in a different orientation, using a [110] Bragg surface orthogonal to that effective in Figs. 2 and 3, highly regular patterns of both Pendellösung and Borrmann-Lehmann fringes were obtained which encouraged hope that reliable values of $F_\rho$ might be extracted from the Borrmann-Lehmann fringe-spacing measurements. Typical patterns obtained in the new orientation are shown in Fig. 5: Cu $K\alpha_1$, in (a), synchrotron radiation, $\lambda = 1.5$ Å, in (b). In this orientation the height of the crystal normal to the plane of incidence was 2 mm, and this full height appears in Fig. 5. The fringes are closely parallel over almost the whole height of the topographs. The values of $|p_\rho|$ were relatively high, so that the Borrmann-Lehmann fringe spacing was small. In both topographs about ten Borrmann-Lehmann fringes of high visibility run parallel to the left-hand margin of the image. They are shown in more detail in Figs. 6(a) and (b) where a 445 $\mu$m high segment of the left-hand part of each topograph is reproduced at higher magnification. Note that the overall image height in Fig. 5(a) is about 5% greater than in Fig. 5(b). This is due to the different ratios of distances between source and specimen, and specimen to plate, applying in the two experiments. In Fig. 5(a) the focus of the X-ray tube was only 450 mm from the specimen. In the synchrotron-radiation experiments the specimen was 80 m from the tangent point on the Storage Ring at
the SERC Daresbury Laboratory, Warrington, Cheshire. There, with a specimen-to-plate distance of 30 mm, the vertical magnification departed from unity by less than 4 parts in 10⁴. The slits close to the crystal defining the narrow ribbon incident beam had apertures between 10 and 15 μm in width in all experiments. At Daresbury the device designed for taking section topographs with synchrotron radiation (Lang, 1983) was used, mounted on the specimen axis of the white radiation camera (Bowen, Clark, Davies, Nicholson, Roberts, Sherwood & Tanner, 1982).

Another comparison of Cu Kα₁ and synchrotron-radiation fringe patterns is shown in Figs. 7(a) and (b). In this pair the values of α are closely similar, and are less than in the pair described above. Little difference between the Borrmann-LEhmann fringe patterns in Figs. 7(a) and (b) can be discerned. Images of internal lattice defects (e.g. the feature with mean height coordinate 0.78 in the left half of the image) and damage to the specimen surface (e.g. the patch centred on height coordinate 0.14 and fairly close to the left-hand margin) show identically in the two topographs. The dynamical feature that identifies the image taken with unpolarized Cu Kα₁ radiation is the periodic fading of Pendellösung fringe visibility evident between R' and R and due to the different fringe periods for the o- and z-polarization modes (Hart & Lang, 1965; Hattori, Kuriyama & Kato, 1965).

4.2. Variations of wavelength and polarization factor

Experiments have been performed using continuous-spectrum synchrotron radiation to investigate the way diffraction contrast due to various types of crystal defect varies with wavelength in the range 0.57 to 2.5 Å when recorded on transmission topographs (Lang, Makepeace, Moore & Machado, 1983). The results highlighted the useful properties of radiation of wavelength about 1 Å, for which no strong conventional sources are available. Choice of this wavelength for Borrmann-LEhmann fringe observations was suggested by other considerations too. Firstly, the interest lay in Borrmann-LEhmann interference phenomena in the low-absorption case. This precluded use of a wavelength appreciably longer than 1-5 Å. Secondly, it was desired to retain a wide-section topograph image (to give a good field in which both Borrmann-LEhmann and Pendellösung fringes might be resolved) without having to increase the crystal thickness. This suggested setting 2θₚ not less than ~45°, and hence λ not less than ~1 Å. (In the symmetrical Laue-Laue case, the section topograph image width is 2t sin θₛ.)

As regards changing the polarization factor, use was made of the facility offered by the white radiation camera at Daresbury for bodily rotation of collimation system, specimen axis and detector axis all together about the axis of the synchrotron beam line. With this facility, one can switch quickly between o-polarization mode alone (plane of incidence on specimen normal to the storage-ring orbit plane) and π-polarization mode alone (plane of incidence parallel to the orbit plane) without changing any other adjustments.

Figs. 8(a) and (b) show Borrmann-LEhmann fringe patterns with the same specimen and similar diffraction geometry as in Figs. 5 and 7, but now taken with λ = 1 Å, and with σ polarization in (a) and π polariz-

Fig. 6. High-magnification illustrations of Borrmann-LEhmann fringes. Micrograph field width = 400 μm. (a) Part of Fig. 5(a), spacing of fringes near left-hand margin = 11 μm. (b) Part of Fig. 5(b), spacing of fringes near left-hand margin = 9-4 μm.

Fig. 7. Borrmann-LEhmann fringe patterns typical of moderately low values of pₛ. (a) Cu Kα₁ radiation, |pₛ| = 0.27, a = 0-21 mm. (b) Synchrotron radiation, λ = 1.5 Å, σ polarization, |pₛ| = 0.26, a = 0-20 mm.
In comparing Fig. 8(a) with Fig. 8(b), note that there is only a small difference, about 12 μm, between the values of \( a \) in (a) and (b), so that differences between the patterns are mainly attributable to change of \( \Delta \) rather than of \( a \) in the product \( a\Delta \). There is an obvious difference between Figs. 8(a) and (b) in the pattern of irregularity of intensity and spacing of the Borrmann-Lehmann fringes. This is attributable more to a different pattern of interaction with lattice irregularities than of interaction with the Pendellösung fringe systems. Note also that there is no great difference in \( p_E \) between the patterns of Fig. 7 (mean \( |p_E| = 0.26 \)) and of Fig. 8 (mean \( |p_E| = 0.29 \)), and one might expect comparable fringe quality in these two figures. However, there is definitely a greater irregularity of Borrmann-Lehmann fringe spacing at the shorter wavelength. Inspection of the original plates also shows that the mean intensity in the image close to \( E \) relative to that in the region \( R'R \) is less in the case of the shorter wavelength.

The effects of changing from \( \sigma \) to \( \pi \) polarization when \( |\cos 2\theta_p| \) falls low are strikingly shown by Fig. 9. Here \( |p_E| = 0.22 \), not greatly different from its value in Fig. 7. The calculated periods of both Pendellösung and Borrmann-Lehmann fringes are increased fourfold in Fig. 9 compared with Fig. 7(b). The consequent easier resolution of the Pendellösung fringes on the right-hand side of the image is evident. But in Fig. 9 the perturbation of the Borrmann-Lehmann fringes by lattice imperfections is so great as to render impossible the assignment of any definite period to these fringes. Also, the mean intensity between \( p = p_E \) and \( p = 0 \) is very weak relative to that in the rest of the pattern, and in comparison with the region near \( E \) in Figs. 5 and 7.

### 4.3. Measurement of structure factor

All the factors multiplying \( (a\Delta)^{-1} \) in (7) are known, or are easily determinable when specimen and wavelength dependent. However, before we present the values of \( F_E \) obtained, a few remarks on instrumental factors which might possibly affect the observed resolution and spacing of fringes are called for. Firstly, one can dismiss variations of magnification factor normal to the plane of incidence (as discussed above in §4.1) as being of any importance, since stretch of the image parallel to the image of the edge \( E \) has no effect on the fringe spacing. Next consider a standard calculation for geometrical resolution on a white-radiation topograph. The X-ray source dimensions at the tangent point on the storage ring are nominally 0.3 and 12 mm normal and parallel to the orbit plane, respectively. With the very large ratio of source-to-specimen (80 m) to specimen-to-photographic plate (30 mm) distances employed, the
calculated geometrical-optical blurrings of the X-ray image of a point on the crystal, in the two directions given, are 0-1 and 4-5 μm, respectively. Although the latter dimension is large from the point of view of X-ray topographic imaging of defects, it could only influence Bormann–Lehmann fringe visibility in the π-polarization patterns, and is still small compared with the smallest fringe periods measured. Lastly, there is a significant difference to be noted in the effect the width of the beam-defining slit has on the visibility of Pendellösung compared with Bormann–Lehmann fringes. Suppose this slit width is δ. Then, for symmetrical Laue-case diffraction conditions, the Pendellösung fringe visibility will fall to zero within such proximity to the section topograph margins that |\frac{\pi}{4} \tan \theta_{|p|} p^{-1} (1 - p^2)^{1/2} - \delta|, applying the expression quoted in § 3 for Pendellösung fringe spacing on the X-ray exit surface. On the other hand, in Bormann–Lehmann fringe patterns, the effect of finite slit width is to introduce uncertainty in the value of δ, whereby the fringe visibility becomes impaired only when the fringe order is comparable with a/δ: fringes with periods smaller than δ can be well observed (e.g. Fig. 6b).

In Table 1 the values of the 220 structure factor given in the experiments here reported are listed in the column headed F(BLF). The last column contains the ratio F(BLF)/F(D) where F(D) is an accepted accurate value of F_{220}, 15.39, given by Dawson (1967). The table includes the measurable synchrotron radiation patterns illustrated in the figures, plus three more deemed of measurable quality. In the advent of powerful synchrotron sources of continuous radiation extends the range of sizes and shapes of specimens with which Bormann–Lehmann fringe spacings is an extremely sensitive way of revealing lattice distortions. It deserves serious consideration as a practical crystal assessment technique, simpler to perform than double-crystal rocking-curve measurements. Uragami (1971) has shown section topographs combining Laue-Laue and Laue–Bragg–Laue diffraction from a wedge-shaped silicon crystal with absorption paths ranging down to low values, but his complicated image geometry precludes easy comparison of observed with expected fringe periods. The evidence from our experiments is that comparing observed with calculated fringe spacings. Uragami (1971) has shown section topographs combining Laue-Laue and Laue–Bragg–Laue diffraction from a wedge-shaped silicon crystal with absorption paths ranging down to low values, but his complicated image geometry precludes easy comparison of observed with expected fringe periods. The evidence from our experiments is that comparing observed with calculated Bormann–Lehmann fringe spacings is an extremely sensitive way of revealing lattice distortions. It deserves serious consideration as a practical crystal assessment technique, simpler to perform than double-crystal rocking-curve measurements. The advent of powerful synchrotron sources of continuous radiation extends the range of sizes and shapes of specimens with which Bormann–Lehmann interference phenomena can be studied. The effects of epitaxial growth or ion implantation on the Bragg surface could be monitored non-destructively, possibly in real time.

For comparison with the synchrotron radiation patterns, the findings on two Cu Kα patterns illustrated are included. In these the dominant apparent fringe periodicity is that of the π-polarization mode, and the value C = 1 is adopted in the calculation of F(BLF). Examination of the plates shows that the F(BLF) values are much more in error than the values of F_{220} that would be derived from Pendellösung fringe-spacing measurements. Even in the case of Fig. 9, where no Bormann–Lehmann fringe period can be assigned with any confidence, the Pendellösung fringes in the right-hand part of the image give an F_{220} value comparable with the best in the table.

5. Concluding remarks

In their pioneer experiments on a 12 mm thick silicon crystal with Mo Kα radiation, Bormann & Lehmann (1963) obtained agreement within ~25% between observed and calculated fringe spacings. Uragami (1971) has shown section topographs combining Laue-Laue and Laue–Bragg–Laue diffraction from a wedge-shaped silicon crystal with absorption paths ranging down to low values, but his complicated image geometry precludes easy comparison of observed with expected fringe periods. The evidence from our experiments is that comparing observed with calculated Bormann–Lehmann fringe spacings is an extremely sensitive way of revealing lattice distortions. It deserves serious consideration as a practical crystal assessment technique, simpler to perform than double-crystal rocking-curve measurements. The advent of powerful synchrotron sources of continuous radiation extends the range of sizes and shapes of specimens with which Bormann–Lehmann interference phenomena can be studied. The effects of epitaxial growth or ion implantation on the Bragg surface could be monitored non-destructively, possibly in real time.

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Table 1. Measurements of F_{220} from Bormann–Lehmann fringe spacing

<table>
<thead>
<tr>
<th>Plate no.</th>
<th>Fig. no.</th>
<th>Radiation (A, A)</th>
<th>Polarization mode</th>
<th>a (mm)</th>
<th>Δ (μm)</th>
<th>CxΔ (Å²)</th>
<th></th>
<th>F(BLF)</th>
<th>F(BLF)/F(D)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3/5</td>
<td>5(a)</td>
<td>Cu Kα</td>
<td>π+π</td>
<td>0.38</td>
<td>11</td>
<td>420</td>
<td>0.40</td>
<td>0.87</td>
<td>25-7</td>
</tr>
<tr>
<td>3/8</td>
<td>7(a)</td>
<td>Cu Kα</td>
<td>π+π</td>
<td>0.21</td>
<td>23</td>
<td>480</td>
<td>0.27</td>
<td>0.96</td>
<td>24-6</td>
</tr>
<tr>
<td>40</td>
<td>5(b)</td>
<td>Syn. (1-5)</td>
<td>π</td>
<td>0.44</td>
<td>9-4</td>
<td>410</td>
<td>0.56</td>
<td>0.83</td>
<td>24-9</td>
</tr>
<tr>
<td>41</td>
<td>6(b)</td>
<td>Syn. (1-5)</td>
<td>σ</td>
<td>0.33</td>
<td>13</td>
<td>430</td>
<td>0.42</td>
<td>0.91</td>
<td>26-3</td>
</tr>
<tr>
<td>42</td>
<td></td>
<td>Syn. (1-5)</td>
<td>σ</td>
<td>0.23</td>
<td>21</td>
<td>480</td>
<td>0.29</td>
<td>0.95</td>
<td>24-8</td>
</tr>
<tr>
<td>44</td>
<td>7(b)</td>
<td>Syn. (1-5)</td>
<td>σ</td>
<td>0.20</td>
<td>23</td>
<td>460</td>
<td>0.26</td>
<td>0.97</td>
<td>26-1</td>
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<tr>
<td>45</td>
<td>8(b)</td>
<td>Syn. (0-0)</td>
<td>σ</td>
<td>0.18</td>
<td>14</td>
<td>250</td>
<td>0.42</td>
<td>0.91</td>
<td>28-7</td>
</tr>
<tr>
<td>46</td>
<td></td>
<td>Syn. (1-0)</td>
<td>σ</td>
<td>0.12</td>
<td>18</td>
<td>220</td>
<td>0.28</td>
<td>0.96</td>
<td>34-9</td>
</tr>
<tr>
<td>48</td>
<td>8(b)</td>
<td>Syn. (0-0)</td>
<td>π</td>
<td>0.13</td>
<td>23</td>
<td>210</td>
<td>0.30</td>
<td>0.95</td>
<td>37-1</td>
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Observations of Borrmann-Lehmann Interference Patterns

Determinations of Phase Using Multiple-Beam Effects*

By J. Z. Tischler
Solid State Division, Oak Ridge National Laboratory,† Oak Ridge, TN 37831, USA

AND B. W. Batterman
School of Applied and Engineering Physics and Cornell High Energy Synchrotron Source, Cornell University, Ithaca, New York 14853, USA

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Abstract

It is shown that the absolute phase of a weak reflection can be obtained from an experimental integrated intensity measurement by taking into account multiple-beam effects. The absolute structure factor of the basis-forbidden 622 reflection in Ge has been measured. The contribution from weakly excited reflections whose phases are known acts as a reference wave for the reflection in question. It is further shown that, even in the limit of weak excitation of multiple beams, the intensity distribution in reciprocal space is determined by dynamical, not kinematical, theory. The remarkable result is demonstrated that several hundred beams are required to fully calculate X-ray multiple-beam effects, and an efficient approximation is presented for multiple-beam dynamical calculations with weak reflections. The intensity of weak multiple-beam effects, at least in the diamond structure, is roughly proportional to Z\(^2\), where Z is the atomic number.

I. Introduction

X-ray scattering measurements can provide important information about the structure of crystalline materials. However, an unambiguous determination of structure requires knowledge of both the intensity and the phase of the scattered Bragg beams.

Generally, indirect phase information can be obtained through the use of a reference beam which is coherently related to the Bragg reflection being considered. One type of reference beam is created by the simultaneous excitation of a series of Bragg beams whose phases are known. The interaction of these multiple beams with the principal Bragg beam allows one to extract the unknown phase.

The phases of the F's, the structure factors, for the allowed diamond-structure reflections are easily calculated from the known atomic positions. However, for the basis-forbidden \( h + k + l = 4n + 2 \) it is the shape of the electron distribution around the nucleus that determines the value of \( F \). For a spherical distribution, \( F \) is exactly zero. Thus for a

* The experimental work was part of a doctoral dissertation at Cornell Univ. See Tischler & Batterman (1984).