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Physical Estimation of Triplet Phases – Effects of Different Radiation Sources and Modes for Profile Scans

F. Mo,[a]* B. C. Hauback,[a]† R. H. Mathiesen,[a] Å. Kvick[b] and E. Weckert[c]

[a]Institutt for Fysikk, Norges Teknisk-Naturvitenskapelige Universitet (NTNU), N-7034 Trondheim, Norway, [b]Experiments Division, ESRF, BP 220, F-38043 Grenoble CEDEX, France, and [c]Institut für Kristallographie, Universität Karlsruhe, Kaiserstrasse 12, D-76128 Karlsruhe, Germany. E-mail: fmo@phys.ntnu.no

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A comparative study has been made of the intensity profiles from three-beam experiments to estimate triplet phases using radiation from a conventional sealed-tube X-ray source and two different synchrotron sources. Synchrotron radiation, with its much smaller angular divergence, narrower spectral bandwidth and higher flux, distinctly improves the experimental conditions for physical phase estimation. Pure ψ scans about the primary diffraction vector, such as can be made with a six-circle diffractometer, further improve the conditions compared with combined ω/ψ scans with a four-circle instrument, where the rotation in ψ is accomplished by combining rotations about the three axes ω, χ and φ. Interference profiles collected by pure ψ scans and unfocused synchrotron radiation have FWHM values reduced by factors in the range 20–35 relative to those obtained with combined ω–$2\theta/\psi$ scans and radiation from a conventional source. As a consequence of these changes, which also involve greatly increased peak amplitudes, the $0/\pi$-type asymmetry of the profiles is exposed much more pronounced closer to the three-beam point, enabling unambiguous phase assignment for all triplets that were studied. The superiority of the pure ψ scan will be even more important in studies of general phases for which the phase information lies also in the relative heights of the (sharp) interference maxima for a triplet and the Friedel-related triplet.

Keywords: three-beam diffraction; physical phase estimation; beam divergence effects; scan mode effects.

1. Introduction

In a previous X-ray study of a centrosymmetric organic compound, the phase sums for several multiple-diffraction cases involving beams H, L and $H-L$ were estimated from modified ψ step-scans over the parent three-beam interaction maxima (Mo *et al.*, 1988). The sum of the phases, $\varphi_{-H} + \varphi_L + \varphi_{H-L}$, forms a three-phase structure invariant (SI). A total of 22 intensity profiles were recorded, of which 13 showed significant asymmetry, corresponding to a unique value of the triplet phase. In four other profiles the asymmetry was less clear, allowing only a tentative phase to be assigned. For the remaining five triplets, no phase could be determined. Of the former 17 (13 + 4) triplets that were used subsequently for the solution of the unknown structure, it was found later that one triplet of the group of four had been given an incorrect phase (Hauback & Mo, 1988).

This study was performed using a standard four-circle diffractometer and radiation from a sealed X-ray tube. The angular divergence and spectral bandwidth of such a beam cause line broadening imposing a greatly reduced asymmetry in the intensity profile farther away from the three-beam point. The much smaller angular divergence and narrower energy band that can be readily obtained with synchrotron radiation are, together with a vastly increased flux, favourable properties in physical phase measurements. To establish a more quantitative reference between the two types of radiation source, we have repeated some of the triplet-phase measurements on a crystal of the same organic compound with synchrotron radiation (NSLS, Brookhaven) using a similar scan technique on a four-circle diffractometer. Later, a six-circle instrument became available (ESRF, Grenoble) for the measurements. With this instrument, pure ψ scans can be made over the interference profiles, thus allowing a comparison of different scan modes using synchrotron radiation.

2. Theoretical background

The wavefield created in a crystal when n beams interact under mutual exchange of energy can be described by the Takagi–Taupin equations or by the fundamental equation of the plane-wave dynamical diffraction theory. In the formalism of the latter theory one obtains equation (1) [see equation (A12) of Batterman & Cole, 1964],

† Present address: Fysikkavdelingen, IFE, Pb 40, N-2007 Kjeller, Norway.

$$[\mathbf{k}^2(1-\Gamma F_0)-\mathbf{K}_H^2]\mathbf{E}_H-\mathbf{k}^2\Gamma\sum_{L\neq H}F_{H-L}\mathbf{E}_L+(\mathbf{K}_H.\mathbf{E}_H)\mathbf{K}_H=0. \quad (1)$$

Here, $\mathbf{k}$ and $\mathbf{K}_H$ are wave vectors, with $\mathbf{k}$ along the incident beam (vacuum) and $\mathbf{K}_H$ along the primary diffracted beam (crystal), $\mathbf{E}_H$ is the electric field vector corresponding to vector $\mathbf{K}_H$, $\Gamma = r_e\lambda^2/\pi V$, where r_e is the classical electron radius and V is the volume of the unit cell, F_H is the structure factor of reflection H. In the X-ray case, (1) can be further reduced to

$$[\mathbf{k}^2(1-\Gamma F_0)-\mathbf{K}_H^2]\mathbf{E}_H-\mathbf{k}^2\Gamma\sum_{L\neq H}F_{H-L}\mathbf{E}_L=0, \quad (2)$$

either by exact elimination for the σ polarization state or as an approximation of the order of 10^{-5} for the π polarization state. Each polarization state is treated separately, which requires that a field vector $\mathbf{E}_H$ is decomposed into two mutually orthogonal components corresponding to the polarization directions σ and π,

$$\mathbf{E}_H=E_H^\sigma\hat{\sigma}_H+E_H^\pi\hat{\pi}_H, \quad (3)$$

where $\hat{\sigma}$ and $\hat{\pi}$ are the unit vectors in the σ and π directions perpendicular to $\mathbf{K}_H$. Insertion into (2) gives, for the σ and π states, respectively,

$$[\mathbf{k}^2(1-\Gamma F_0)-\mathbf{K}_H^2]E_H^\sigma-\mathbf{k}^2\Gamma\sum_{L\neq H}F_{H-L}[E_L^\sigma\hat{\sigma}_L\hat{\sigma}_H+E_L^\pi\hat{\pi}_L\hat{\sigma}_H]=0, \quad (4a)$$

$$[\mathbf{k}^2(1-\Gamma F_0)-\mathbf{K}_H^2]E_H^\pi-\mathbf{k}^2\Gamma\sum_{L\neq H}F_{H-L}[E_L^\sigma\hat{\sigma}_L\hat{\pi}_H+E_L^\pi\hat{\pi}_L\hat{\pi}_H]=0. \quad (4b)$$

In the case of two beams, $\mathbf{K}_O$ and $\mathbf{K}_H$, equation (4) reduces to four scalar equations, two for each polarization state. The solution of the corresponding eigenvalue problem is then straightforward and can be found together with its physical interpretation in various references (*e.g.* von Laue, 1941; Batterman & Cole, 1964).

More than two beams must be excited simultaneously to generate multibeam phase effects. The simplest n-beam case involves three beams (n = 3). To obtain an analytical solution of (4) for $n \geq 3$ requires the introduction of approximations. A viable approximation for this purpose was developed originally by Bethe (1928) for electron diffraction, but has been extended later to the X-ray case (Marthinsen, 1981). This approach distinguishes between weak and strong beams (waves). The following assumptions are made:

(i) Only two waves, the incident and the primary diffracted waves, O and H, respectively, are considered strong. All the other waves (L) are weak as they are not fully excited.

(ii) The weak waves can be represented by (4), but only terms corresponding to the strong waves (O,H) are retained in the sums.

(iii) For each of the strong waves, the contribution from the other strong wave is taken separately. All (L) waves remain in the sums and are expressed by their contributions from (O,H).

(iv) The weak beams, being weakly coupled to the strong beams, are treated in the kinematical approximation.

In the X-ray case, a further approximation which depends on the diffraction geometry is:

(v) All terms coupling σ and π polarization are neglected, *i.e.* cross terms $\sigma_i.\pi_j = 0$.

Several authors have studied the effects of beam polarization on X-ray multiple diffraction (*e.g.* Juretschke, 1982, 1986; Luh & Chang, 1991; Weckert & Hümmer, 1997; Larsen & Thorkildsen, 1998). For the unpolarized beam from a conventional X-ray source, the coupling terms $\sigma_i.\sigma_j$ and $\pi_i.\pi_j$† are in general equally important. With synchrotron radiation, which is nearly linearly polarized in the orbit plane, the polarization mode is σ or π for a vertical or horizontal primary diffraction plane, respectively. It was pointed out by Weckert & Hümmer (1997) that some scalar products between polarization vectors can become negative, depending on the actual diffraction geometry. If sufficiently large, these terms may invert the asymmetry of the three-beam interference profile, leading to an incorrect assignment of the phase. In their study of finite perfect crystals, Larsen & Thorkildsen (1998) have shown that such anomalous inversion may occur both with σ- and π-polarized incident beams. With synchrotron radiation monochromated at $\lambda \simeq 1.0$ Å by an Si(111) crystal, the magnitudes of the vectors in a $\sigma_i.\pi_j$ cross term will be approximately $0.95^{1/2}$ and $0.05^{1/2}$. For these terms to become important relative to the leading $\pi_i.\pi_j$ (or $\sigma_i.\sigma_j$) coupling term, the vectors representing the incident ($\mathbf{K}_O$), the primary diffracted ($\mathbf{K}_H$) and the secondary diffracted ($\mathbf{K}_L$) beams must approach orthogonality, which implies high-resolution reflections. In practice, neglecting the cross terms is a valid approximation when both the H and the L reflections in a triplet have angle 2θ within a limited angular range at modest resolution. In the present case, 2θ for H and L were in the range 6–40° (λ = 1.0000 Å) for all the triplets studied. For this value of λ the $\sigma_i.\pi_j$ cross terms of any of the triplets were $\leq$4% (synchrotron) or <8% (conventional source) of the parent polarization term, *e.g.* p_{HL}. This result confirms the validity of approximation (v).

Assumptions (i)–(v) allow three-beam diffraction to be treated as a perturbed two-beam case. The first-order approximation to the solution can be expressed as a function of s_L, the distance of the secondary reciprocal lattice node (rln), L, from the Ewald sphere,

$$\begin{aligned} I_H &= QZ|F_H|^2[1-2Q^{1/2}PR_F\cos\Phi_3(1/s_L)+QP^2R_F^2(1/s_L)^2] \\ &= QZ|F_H|^2[y_B], \end{aligned} \quad (5)$$

where $Q = (1/2k\Gamma)^2$, $P = (p_{OL})(p_{HL})/(p_{OH})$, in P, *e.g.* the polarization term $p_{OH} = \hat{\mathbf{x}}_O.\hat{\mathbf{x}}_H$, where $\mathbf{x}$ is either both σ or

† The π component of the polarization vectors are defined to lie in the plane of the incident and the primary diffracted beams, *i.e.* in the primary diffraction plane.

both π, $Z = (p_{OH})^2$, $\Phi_3 = \varphi_{-H} + \varphi_L + \varphi_{H-L}$, a three-phase structure invariant, $R_F = |F_{H-L}||F_L|/|F_H|$, s_L is the distance of the secondary rln L from the Ewald sphere, where, by definition, $s_L > 0$ for L outside the sphere.

The terms within square brackets can be regarded as a first-order dynamical correction to the kinematical result, valid for the centrosymmetric case. The information about an invariant triplet phase, Φ_3, is contained in the term with s_L^{-1}. For a centrosymmetric crystal, the sign of $\cos\Phi_3$ is projected out as a characteristic asymmetry in the backgrounds of the corresponding three-beam interaction extremum. Several authors have developed expressions which relate an observed asymmetry uniquely to the correct value of the three-phase SI (Juretschke, 1982; Chang, 1982; Thorkildsen & Mo, 1983; Hümmer & Billy, 1982, 1986). We prefer here to define the distance of the secondary rln L from the Ewald sphere as the leading parameter s_L (Thorkildsen, 1983). Introducing

$$x_s = s_L/Q^{1/2}PR_F = As_L, \tag{6}$$

one obtains for y_B (see also Juretschke, 1982),

$$y_B = [1 - 2\cos\Phi_3(1/x_s) + (1/x_s)^2]. \tag{7}$$

This equation reveals unambiguously the relation between a triplet phase, say $\Phi_3 = \pi$, and the intensity levels for $s_L \neq 0$. In a perfect crystal the sharp minima in y_B will occur for $s_L \simeq 10^{-6}$ Å^{-1}.

Note that (5) and (7) diverge as $s_L \to 0$, and cannot be used to estimate I_H at or very close to the exact three-beam point, $s_L = 0$. This is no constraint in the centrosymmetric case as the phase information resides in the backgrounds of the intensity profile, away from $s_L = 0$. For $\Phi_3 \simeq +\pi/2$ or $-\pi/2$ the diffraction power is symmetric in s_L, and the phase information lies in the relative heights of the profile extrema at $s_L = 0$. Expressions for general phase values have been developed by Hümmer & Billy (1986) from higher-order approximations to dynamical plane-wave theory, and by Thorkildsen (1987) employing the Takagi–Taupin equations for a finite perfect crystal. Note that the former authors define a triplet as $-H$, L, $H - L$, while in the definition of Thorkildsen (1987) this is H, $-L$, $L - H$, *i.e.* with inverted indices. The conclusions on the phase signature in the interference profiles for non-centrosymmetric triplets, most pronounced for $\Phi_3 \simeq \pm\pi/2$, are therefore reversed.

3. Experimental

3.1. *Measurements with a conventional X-ray source*

Incident radiation unpolarized; primary diffraction plane horizontal. One single crystal of the organic compound *N*,*N*′-diphenyl-*N*-(pyridinyl)urea hemihydrate (EHM III) was used for all the phase measurements which were carried out on a four-circle diffractometer with Nb-filtered Mo radiation from a standard sealed X-ray tube. EHM III has the chemical formula $C_{18}H_{15}ON_3.1/2H_2O$, $M_r = 298.35$, and crystallizes in space group *I*2/*a* with unit-cell volume $V = 2953.2$ (4) Å^3 (at 86 K) and $Z = 8$ (Hauback & Mo, 1988). Data for 22 three-beam interaction maxima were collected by repeated $\omega/2\theta$ scans over the primary diffracted intensity and stepping in ψ over the exact three-beam position. On a four-circle instrument a rotation about the primary diffraction vector **H**, a ψ scan, is accomplished by small rotations in the angles ω, χ and φ. Details of the experimental procedure are given elsewhere (Mo *et al.*, 1988).

3.2. *Measurements at the National Synchrotron Light Source (NSLS)*

Incident radiation $\geq 95\%$ polarized in the horizontal plane; primary diffraction plane vertical. One single crystal of the compound was used for all the phase measurements. They were made on a four-circle instrument on beamline X-7B. A preliminary investigation had been made with the conventional beam set-up by ω scans over several reflections using a narrow (0.025°) horizontal slit. The FWHM values, 0.04–0.055°, were very similar to those obtained for the crystal that was used in the first phase study. With synchrotron radiation, however, FWHM values from ω scans were in the range 0.007–0.008°.

Five different three-phase SIs were reexamined. Two of them had been correctly phased in the conventional study, one had been tentatively assigned the value π, which proved to be incorrect, and the last two triplets belonged to the group of five for which no asymmetry could be found in the backgrounds. Even with the resolution available on the diffractometer, 0.0005° in ω, χ and φ, satisfactory ψ scans over the very sharp three-beam interaction maxima for this crystal are difficult to obtain. The profiles were therefore mapped out by carrying out small ω step-scans in the vertical plane for each step in ψ, a combined ω/ψ scan. This procedure also facilitates a more direct comparison with the results from the conventional study, as a similar technique was employed then (Mo *et al.*, 1988). In the study at NSLS, the steps in ψ were 0.0025° in the range $\psi_0 - 0.02°$ to $\psi_0 + 0.02°$, increasing to 0.005 and then to 0.01° towards the limits $\psi_0 \pm 0.05°$. In one case these limits were increased to $\psi_0 \pm 0.2°$. The scan range in ω was $\omega_c - 0.02°$ to $\omega_c + 0.02°$, in steps of 0.001°; ψ_0 corresponds to the exact three-beam position, ω_c is the local value calculated for ω on the exact ψ trajectory. Smaller steps could have been used in ψ and also in ω, but occasional instabilities in the beam intensity made it imperative to reduce the data-acquisition time. The wavelength used for one of the triplet phase measurements was 0.8070 Å; for the other triplets $\lambda = 0.9434$ Å. A slightly focused beam was used with a vertical beam divergence of 0.003° and energy resolution $\Delta\lambda/\lambda = 3.5 \times 10^{-4}$. As in the conventional-source study, $\mu t \leq 0.1$, where μ is the linear absorption coefficient and t is the average path length in the crystal. It was shown (Hümmer & Billy, 1982) that absorption effects in the intensity profiles recorded under multiple-diffraction conditions are negligible when $\mu t < 0.5$.

3.3. *Measurements at the European Synchrotron Radiation Facility (ESRF)*

Incident radiation $\geq$ 95% polarized in the horizontal plane; primary diffraction plane horizontal. The same crystal in an identical mount as in the study at NSLS was used for the measurements on the Swiss–Norwegian beamline (SNBL) at the ESRF. A total of 14 Friedel-related three-phase SI pairs were measured on a six-circle diffractometer (Hümmer *et al.*, 1989). With this instrument an arbitrary scattering vector can be brought down in the equatorial plane, aligned with the ψ axis, and a ψ scan performed by rotating only one axis. This is in principle a superior procedure to one in which three axes are involved in a compound rotation. The instrument at SNBL had an angular resolution of 0.0002° in ω, χ, φ and ψ, allowing very precise ψ scans to be made over even very narrow profiles. The 14 profile pairs were scanned about the diffraction vector $\mathbf{H}$ from $\psi_0 - 0.06°$ to $\psi_0 + 0.06°$ in steps of 0.0003°. The exact three-beam point can be checked experimentally as the orientation in which the primary (H) and the secondary (L) reflections are simultaneously at their peak positions. ω scans were made over all 28 primary reflections. The range of FWHM values, 0.0055–0.009°, can be taken to represent the crystal mosaicity. The wavelength in all cases was 1.0000 Å. An unfocused beam was used, the vertical and the horizontal beam divergencies were 0.001° and $\Delta\lambda/\lambda = 1.4 \times 10^{-4}$.

The wavelength is critical for determining the frequency of n-beam situations. For a crystal in random orientation, the number of rln lying within a shell of thickness 2Δ about the Ewald sphere is

$$N \simeq 8\pi\Delta(V/\lambda^2). \tag{8}$$

The parameter Δ depends on the width of the diffraction lines, which again is determined by several factors related to properties both of the crystal and of the radiation source. Setting an effective $\Delta = 5 \times 10^{-4}$ Å^{-1} shows that N can be large even for a crystal with a small unit cell with the wavelengths used in the present study. Therefore, one must ascertain that there are no other *strong* three-beam interactions that can interfere with the one being selected for study.

4. Results

The profile scans of all the three-beam interaction maxima collected with synchrotron radiation provided unambiguous indications of phase in agreement with the correct result. Three of the measured profiles are reproduced in Fig. 1. The profiles measured with radiation from a conventional source (NTH) are shown as 1(a), 1(b) and 1(c). The corresponding profiles from the synchrotron radiation studies appear below as 2(a), 2(b) and 2(c) (X-7B, NSLS) and 3(a), 3(b) and 3(c) (SNBL, ESRF). Each point in series 2 represents the average *integrated* intensity from four to six ω scans; in series 3 these points are the average intensities from 8 to 15 ψ scans. The phase sum, Φ_3, for triplet (a), $233/\bar{4}00/2\bar{3}\bar{3}$, was correctly assigned the value 0 in the original study; for the two triplets $545/3\bar{2}\bar{3}/\bar{8}22$, ($b$), and $433/0\bar{3}\bar{3}/\bar{4}00$, ($c$), the phase could not be determined. The intensities in all plots have been normalized, 100 corresponds to the two-beam intensity level. Along the horizontal axis, ψ_L gives the rotation in ψ, but is signed according to s_L. Thus, $\psi_L > 0$ corresponds to the rln L lying outside the Ewald sphere; $\psi_L = 0$ is the three-beam point. Note that the ψ_L scale in the plots for series 1 (NTH) is a factor of ten larger than for the two series with synchrotron radiation. The crucial feature of a $0/\pi$-type triplet profile is the sign of an asymmetry in the background near ψ_0 [see equation (5)]. An asymmetry ratio can be defined as $r_A = I_{\rm min}/I_{\rm two\text{-}beam}$, where $I_{\rm min}$ is the minimum intensity close to the three-beam point. The interference maxima from series 2 and 3 have been truncated to give a better view of r_A in the synchrotron radiation studies; the ordinate scale for the two series are identical for the same triplet. Table 1 gives pertinent data for the five interference profiles that were measured in all the series: amplitudes, FWHM and r_A values.

The FWHM values of the interference maxima measured at NSLS are in the range 0.013–0.021°. This range was 0.140–0.160° from the conventional-source study. The reduction in peak width, about an order of magnitude, is a very important improvement. It is caused primarily by the much smaller vertical angular divergence of the synchrotron beam, which was here 0.003° compared with about 0.12° with the conventional beam set-up. The narrowing of the maxima reveals a more pronounced asymmetry in the backgrounds at shorter distance from the three-beam point. All profiles recorded at NSLS show asymmetry in the backgrounds with the low level for $\psi_L > 0$ which identifies a triplet phase $\Phi_3 = 0$; the r_A values are in the range 0.987–0.897. The measured increase in amplitude over the two-beam level in the NSLS study is about seven to 18 times larger than with the conventional source. This enhancement can also be ascribed to the small beam divergence, but also to a contracted bandwidth. Both factors ensure that the three-beam interference takes place over an angular range reduced by a factor of about ten relative to that observed with the conventional source.

Two primary reflections were examined by ω scans at the three-beam point in the NSLS study, giving FWHM values of 0.0073° for $8\bar{4}4$ and 0.0070° for 433. The corresponding FWHM values in ψ from the combined ω/ψ step-scans are 0.0211 and 0.0156°, respectively, an increase of two or three times in magnitude. It has been shown by Thorkildsen (1987) that movement of rln L relative to the Ewald sphere during measurement of the integrated intensity will reduce and broaden the interference extremum. Our results are in consonance with the theoretical prediction. In our study the small ω scans will also move H out of the exact Bragg position which will tend to smear the profile further. This smearing is avoided in a pure ψ scan. With the use of unfocused synchrotron radiation the major contribution to line broadening now comes from the crystal mosaicity.

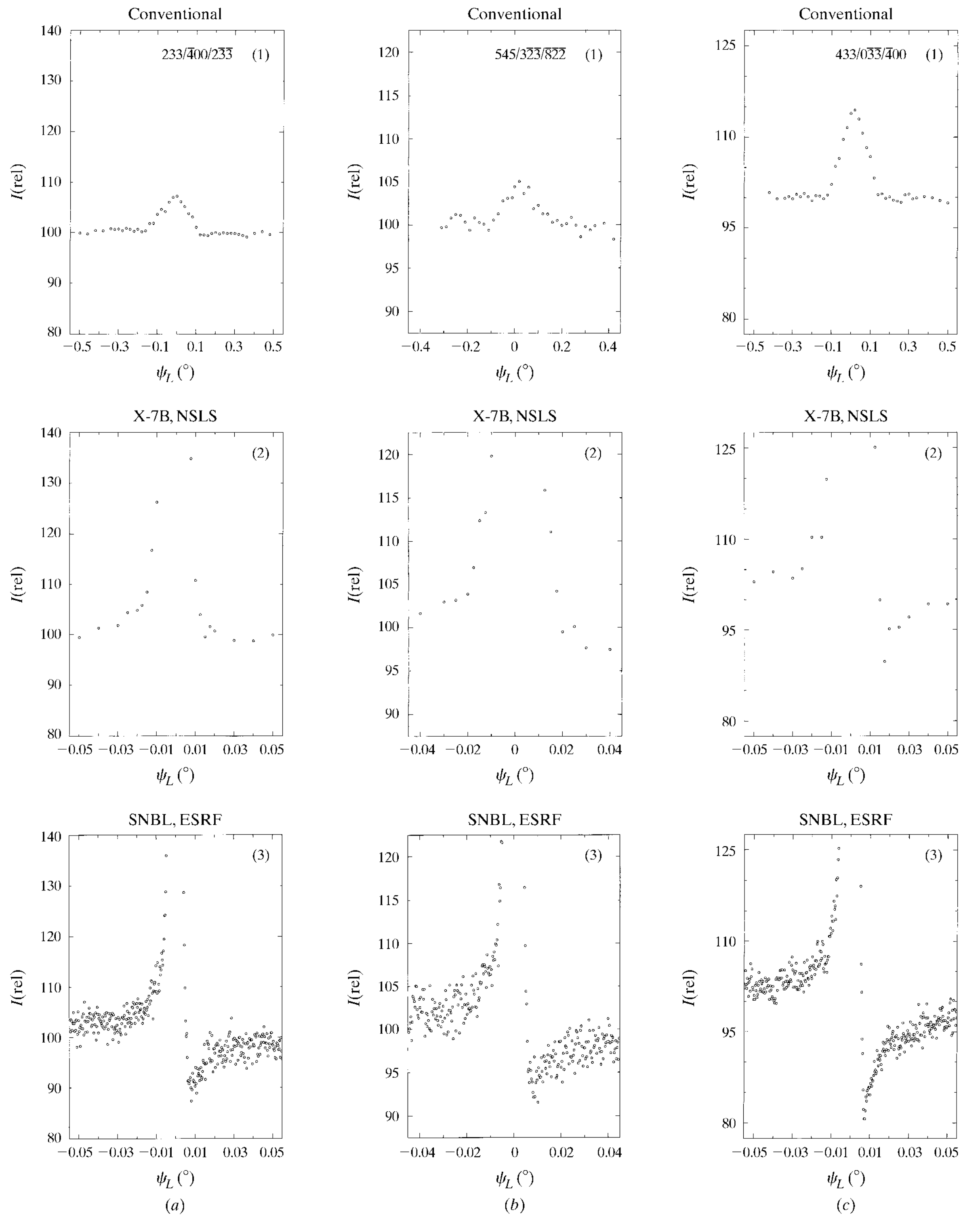

Figure 1

Profiles of the primary diffracted intensity I_H in three different three-beam interactions, $-H, L, H-L$. The triplets are (*a*) $233/\bar{4}00/2\overline{33}$, (*b*) $545/3\overline{23}/\overline{822}$ and (*c*) $433/0\overline{33}/\bar{4}00$. Profiles have been collected with radiation from a conventional source, λ = 0.71073 Å, and combined ω–$2\theta/\psi$ scans (series 1), synchrotron radiation, λ = 0.9434 Å, and combined ω/ψ scans (series 2), synchrotron radiation, λ = 1.0000 Å, and pure ψ scans (series 3). Intensities are given relative to the two-beam level (= 100), ψ_L is the signed rotation of the secondary reciprocal lattice node L from the exact three-beam point, $\psi_L = 0$. Measurements in series 2 and 3 were made with the same crystal specimen in the identical mount. Note the change in the ψ_L scale between plots from series 1 and those from the two series with synchrotron radiation.

Table 1
Characteristic parameters for five three-beam interference profiles.

Series 1: with conventional source; series 2: with synchrotron radiation at X-7B, NSLS; series 3: with synchrotron radiation at SNBL, ESRF; $r_A = I_{\rm min}/I_{\rm two\text{-}beam}$, where $I_{\rm min}$ is the minimum intensity near the three-beam point.

Triplet	Series	Peak amplitude relative to two-beam level (= 100)	FWHM (°)	r_A
$233/\bar{4}00/2\bar{3}\bar{3}$	1	107.2	0.160	99.3
	2	210	0.0129	98.7
	3	410	0.0049	87.4
$33\bar{2}/\bar{4}00/1\bar{3}2$	1	348.1	0.150	94.0
	2	1783	0.0148	89.8
	3	6430	0.0057	60.0
$8\bar{4}4/02\bar{2}/\bar{8}2\bar{2}$	1	135.0	0.160	
	2	727	0.0211	90.3
	3	1882	0.0090	82.8
$545/3\bar{2}\bar{3}/\bar{8}\bar{2}\bar{2}$	1	105.1	0.140	
	2	147	0.0200	97.4
	3	291	0.0057	91.5
$433/0\bar{3}\bar{3}/\bar{4}00$	1	114.5	0.153	
	2	249	0.0156	89.7
	3	456	0.0048	80.7

FWHM values in ψ from our measurements at the ESRF are in the range 0.0045–0.0090°, which is a further reduction of about a factor of three from the NSLS study. The decrease in peak width is significant, revealing very clearly the short-range asymmetry in the backgrounds (see Fig. 1 and Table 1). As FWHM values from ω scans of the reflections $8\bar{4}4$ and 433 at the three-beam point are quite similar to the values from the NSLS study, 0.0077 and 0.0059°, respectively, the contraction in ψ is ascribable largely to the scan mode, with a contribution from the smaller beam divergence at the SNBL (ESRF). The enhancement of the peak heights relative to those from the NSLS study is in the range 2–4. As the same crystal in the identical mount was used in both synchrotron radiation studies and the wavelengths were very similar, the effects of the beam path-lengths on the intensity profiles should be nearly identical. However, because of orthogonal primary diffraction planes in the two experiments, the polarization effects are different, with a larger general reduction of intensity for the π-polarized incident beam in the study at SNBL. The results demonstrate and quantify the superiority of the pure ψ scan for extracting phase information from three-beam interference profiles. The advantage of this scan mode becomes even more important in studies of general triplet phases, in particular of the $\pm\pi/2$ type, where the phase signature lies predominantly in the relative heights of the (sharp) interference extrema for a triplet and the Friedel-related triplet.

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