Complete determination of x-ray polarization using multiple-beam Bragg diffraction

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Phase-sensitive interference and polarization mixing in multiple-beam diffraction lead to a method of measuring the polarization of electromagnetic waves in the x-ray regime. With this technique one can determine all three components of the Poincaré polarization vector and therefore characterize the polarization completely. Experimental results using GaAs(442) multiple reflections are presented.

Many frontier studies in x-ray physics, material science, and crystallography, such as nonresonant and resonant magnetic scattering,\textsuperscript{1-3} magnetic circular dichroism,\textsuperscript{4} and anisotropy of anomalous scattering,\textsuperscript{5} require an accurate knowledge on the polarization of an incident x-ray beam and can benefit, in many cases, from a polarization analysis on the scattered or fluorescent photons. It is therefore important to characterize the x-ray polarization in a general and convenient way, especially for an elliptically polarized x-ray beam. For linear polarization analyses, elastic scattering or Bragg reflections at a scattering angle of 90° can be used as a high-precision polarimeter\textsuperscript{6,7} in a wide energy range. For circular polarization analyses, optical mirrors as phase shifters have been used in the ultraviolet and soft x-ray regions,\textsuperscript{8} while magnetic Compton scattering has been used to characterize x rays at relatively high energies.\textsuperscript{9} In this paper we introduce a technique of measuring both linear and circular polarization components in an x-ray beam. This method is called multiple-beam diffraction (MBD), and is based on the interference effect among simultaneously excited Bragg reflections inside a crystal.\textsuperscript{10-13} We will show that the interference and polarization mixing that occur in a MBD process can lead to complete polarization analysis, including the determination of partial elliptical polarization.

The intensity and polarization of an x-ray plane wave can be completely characterized by the four Stokes parameters:\textsuperscript{14} \(s_0 = \langle D_x^2 \rangle + \langle D_y^2 \rangle, \ s_1 = \langle D_x^2 \rangle - \langle D_y^2 \rangle, \ s_2 = 2(D_x \times D_y \cos \delta), \) and \(s_3 = 2(D_x D_y \sin \delta),\) where \(D_x\) and \(D_y\) are projections of the electric-field vector along two orthogonal directions \(x\) and \(y,\) \(\delta\) is the phase between them, and the angular brackets indicate a time averaged quantity. The Stokes parameters are directly related to measured intensities in an experiment: \(s_0\) is the total intensity, \(s_1, s_2,\) and \(s_3\) are the intensity differences between the 0° and the 90° linear, the 45° and the −45° linear, and the two senses of circular polarization, respectively. For a general x-ray plane wave, \(s_0 \geq (s_1^2 + s_2^2 + s_3^2)^{1/2},\) where the equality holds for a completely polarized beam. If we are interested only in the polarization and not in the absolute intensity, we can then use the three normalized Stokes parameters \(P = (P_1, P_2, P_3)\) with \(P_i = s_i/s_0\) \((i = 1, 2, 3).\) The vector \(P\) is called the Poincaré vector of polarization.\textsuperscript{5,14} Its length \(|P| = 1\) indicates the x-ray wave is totally polarized and \(|P| < 1\) indicates it is partially polarized. As we will show, the interference intensity in a MBD process depends on all three Poincaré components and therefore can be used to completely characterize the x-ray polarization.

An MBD process occurs when two or more atomic planes satisfy Bragg’s law simultaneously inside a crystal. A convenient way to achieve such a condition is to rotate the diffracting crystal around the scattering vector \(Q,\) described by an azimuthal angle \(\phi,\) while always keeping one Bragg reflection \(H = Q\) excited.\textsuperscript{15,16} When the crystal is in an orientation such that another reciprocal node \(L\) is on the sphere of reflection (Ewald sphere), a multiple-beam diffraction will occur which may give rise to a secondary peak in the intensity of the \(H\) reflection. The reflection \(H\) is called the main reflection and the reflection \(L\) is the detoured reflection. A third reflection \(H - L\) is necessary to bring the detour-diffracted beam back into the \(H\)-diffracted direction and is called the coupling reflection. It has been known\textsuperscript{11} that the detour-diffracted (through \(L\) and \(H - L\)) and the direct-diffracted (through \(H\)) beams can interfere with each other and produce an intensity enhancement and reduction pattern in the neighborhood of the multibeam excitation. In general, the two diffracted beams have different phase shifts according to the structure factors involved, \(F_H, F_L,\) and \(F_{H - L},\) with phases \(a_H, a_L,\) and \(a_{H - L},\) respectively. The interference between the two waves depends, therefore, on the relative phase shift \(\delta_{H - L} = a_L + a_{H - L} - a_H,\) and can be used to extract lost phase information in a diffraction experiment.\textsuperscript{11-13}

Besides the interference, another effect that can exist in an MBD process is the polarization state mixing. It occurs when the diffraction direction \(L\) is not in the plane normal to the incident wave vector \(k_0.\) When that happens, the polarization component that is normal (\(\sigma\)) to the original diffraction plane can produce a component in the \(H\)-diffracted beam that is parallel to \(\sigma\) to this plane, and vice versa. It can be shown that the polarization dependence in the detour-diffracted wave is identical to that of a double Thomson scattering process by two electrons.\textsuperscript{17} This polarization mixing effect, together with the phase-probing capability of MBD can allow for determination of the general polarization parameters, including the phase between the \(\sigma\) and the \(\pi\) components.

The polarization and phase dependence of MBD can be
explicitly shown by the perturbation theory of scattering which takes into account the multiple-beam (MB) effect.\textsuperscript{11} Based on this theory, the diffracted wave field for reflection \( H \) in the neighborhood of multiple reflection \( L \) is given by

\[
D_H = N r \frac{F_{1} F_{H - L}}{F_{H}} \frac{e^{-ik_{H}r}}{r} \cdot n 
\times \left[ n \times \left( D_0 - \Gamma \frac{F_{1} F_{H - L}}{F_{H}} \left[ e^{i\delta_{H}} k_{L} \times (k_{L} \times D_0) \frac{k_{L}}{k_{H} - k_{L}} \right] \right) \right].
\]  

(1)

In this expression, \( \Gamma = r_{e} \lambda^2 / \pi V_{c} \), \( \lambda \) is the incident x-ray wavelength, \( V_{c} \) is the unit-cell volume, \( N \) is the number of unit cells in the crystal, and \( r_{e} \) is the classical radius of an electron. The incident wave is assumed to be \( D_{0e} = k_{e} e^{i\Gamma} \).

\[
[A] = \begin{bmatrix}
A_{o\sigma} & A_{o\pi} \\
A_{e\sigma} & A_{e\pi}
\end{bmatrix} = \Gamma \frac{F_{1} F_{H - L}}{F_{H}} \begin{bmatrix}
k_{L} - (L \cdot \sigma_{o})^2 \\
-(k_{L} \cdot \pi)(L \cdot \sigma_{o})
\end{bmatrix}
\]

where \( \sigma_{o} \) and \( \pi_{o} \) are the unit polarization vectors normal and parallel to the diffraction plane for the incident beam, and \( \sigma \) and \( \pi \) are those for the \( H \)-diffracted beam, respectively. The existence of the nondiagonal elements in matrix \([A]\), and thus \([M_{H,H}]\), represents the polarization mixing which can occur in a MBM process. The necessary condition for it to happen is that \( L \cdot \sigma_{o} \neq 0 \), i.e., \( L \) does not lie in the diffraction plane.

In the vicinity of \( \phi = \phi_{1} \), where the \( L \) reflection is excited, \([A]\) varies slowly and may be considered constant when compared to \( B(\phi) = -k_{o}/[2 \cos \theta (L \cdot \sigma_{o})] \times (\phi - \phi_{1}) \), which changes its sign as \( \phi \) crosses \( \phi_{1} \). This phase change by \( \pi \) on either side of \( \phi = \phi_{1} \) of the detour-diffracted amplitude, when added to the amplitude contributed by the main reflection \( H \), is the cause of the asymmetric interference intensity pattern that is usually observed in an MBM experiment.\textsuperscript{11} The function \( B(\phi) \) can be called the orientation parameter for a given \( L \). The geometry matrix \([A]\) evaluated at \( \phi = \phi_{1} \) is a pure function of crystal geometry, augmented by the structure factors of the detour reflection. The third member in the second term of Eq. (3) is the phase factor \( e^{i\delta_{H}} \), which is determined by the structure factors of the crystal. Therefore we see that the multiple-beam interactions are completely determined by the structure, the geometry, and the orientation of the crystal.

A convenient way of expressing a MBM intensity for a general polarization is to use the Stokes-Poincaré parameters and the density-matrix method. According to this method,\textsuperscript{13,14} the polarization state of an x-ray beam is completely characterized by the density matrix \([\rho]\), which can be expressed in terms of the Stokes-Poincaré parameters \((P_1, P_2, P_3)\) in the following way:

\[
[\rho] = \frac{1}{2} \begin{bmatrix}
1 + P_1 & P_2 - iP_3 \\
P_2 + iP_3 & 1 - P_1
\end{bmatrix}.
\]

The normalized diffracted intensity \( I \) can be obtained by a matrix trace operation\textsuperscript{3}

\[
I = \text{Tr} [M_{H,H} \rho M_{H,H}^\dagger],
\]  

(4)

where \( \delta_{H,H} \) is defined as \( k_{H} = k_{0} + H, k_{L} = k_{0} + L, \) and \( n = k_{H}/k_{H} \). The first term in Eq. (1) represents the direct-diffracted wave by \( H \) and the second term represents the detour-diffracted wave by \( L \) and \( H - L \) with a phase shift of \( \delta_{H,H} \) defined above.

In terms of the components normal \((\sigma)\) and parallel \((\pi)\) to the diffraction plane defined by \( H \) and \( k_{0} \), Eq. (1) can be expressed using a \( 2 \times 2 \) matrix \([M_{H,H}]\) as

\[
\begin{bmatrix}
D_{H\sigma} \\
D_{H\pi}
\end{bmatrix} = \frac{N r \lambda^{2} / \pi V_{c}}{F_{H}} \begin{bmatrix} A_{o\sigma} & A_{o\pi} \\
A_{e\sigma} & A_{e\pi}
\end{bmatrix} \begin{bmatrix}
D_{0\sigma} \\
D_{0\pi}
\end{bmatrix},
\]

where

\[
[M_{H,H}] = \begin{bmatrix}
1 & \cos 2\theta \\
0 & \cos 2\theta
\end{bmatrix} + B(\phi) e^{i\delta_{H}} \begin{bmatrix} A_{o\sigma} & A_{o\pi} \\
A_{e\sigma} & A_{e\pi}
\end{bmatrix},
\]  

(3)

with \( \theta \) = Bragg angle for \( H \), \( B(\phi) = k_{0}^{2} / (k_{H}^{2} - k_{L}^{2}) \), and

\[
\begin{bmatrix}
L \cdot \sigma_{o} \\
L \cdot \pi_{o}
\end{bmatrix} = \begin{bmatrix}
-(k_{L} \cdot \pi_{o})(L \cdot \sigma_{o}) \\
(k_{L} \cdot \sigma_{o})(L \cdot \pi_{o})
\end{bmatrix},
\]

where the superscript daggers indicates the Hermitian conjugate. Using Eq. (4), it is straightforward to show that the interference intensity \( I_{int} \) between the two terms of \([M_{H,H}]\) in Eq. (3) is given by

\[
I_{int} = B(\phi) [A_{o\sigma} \cos \delta_{H}(1 + P_{1}) + A_{e\sigma} \cos \delta_{H} P_{2} - A_{e\sigma} \sin \delta_{H} P_{3}],
\]  

(5)

\[
I_{int} = B(\phi) \cos 2\theta [A_{o\sigma} \cos \delta_{H}(1 - P_{1}) + A_{e\sigma} \cos \delta_{H} P_{2} + A_{e\sigma} \sin \delta_{H} P_{3}].
\]

Here \( I_{int} \) and \( I_{int} \) refer to the H-diffracted \( \sigma \) and \( \pi \) components, respectively. Because Eq. (5) depends on all three components of \( P \), the interference intensity from a known crystal structure can be used to determine \( P \). Furthermore, by choosing a specific multiple-beam combination, \( H \) and \( L \), thus a specific set of \([A]\) and \( \delta_{H,H} \), one can make the multiple-beam interference more sensitive to a particular Poincaré component than others. For example, a detector reflection with \( \delta_{H,H} = \pm 90^\circ \) and a nonzero \( A_{e\sigma} \) will be sensitive to \( P_{3} \), the circular component in the incident beam. Such a condition for \( \delta_{H,H} \) is most easily achieved by using a noncentrosymmetric crystal such as GaAs.

To demonstrate the ability of measuring x-ray polarization with the MBM, we have performed an experiment using a GaAs crystal at the D-1 station of CHESS. This station receives synchrotron radiation emitted by clockwise (when looking from above the ring) circulating positions with a bending radius of 32 m and a particle energy of 5.4 GeV. In the experiment we started with the radiation about 0.11 mrad below the orbital plane (observation angle \( \psi = -0.11 \) mrad) which should have a certain degree of right-handed elliptical polarization. The x-ray beam was monochromated to \( \lambda = 1.3 \) Å by a pair of vertically diffracting Si(111) crystals and was slit down to a size of \( 0.25 \times 0.25 \) mm\(^2\) at 15 m from the source. The higher-order harmonics in the beam were substantially suppressed by detuning the second monochromator crystal by 70% with respect to the first.
FIG. 1. Multiple-beam interference profiles measured on the GaAs(442) main reflection. The three detour reflections are (511) in (a) and (d), (151) in (b) and (e), and (153) in (c) and (f). Data in (a)–(c) were taken using the synchrotron radiation emitted at 0.11 mrad below the positron orbital plane and those in (d)–(f) were using radiation at 0.11 mrad above the orbital plane. The experimental data are represented by squares and are normalized to the two-beam (442) intensity at azimuthal positions far away from any detour reflections. The solid curves are theoretical calculations fit to the data by adjusting polarization parameters \( P_1, P_2 \), as described in the text. The polarization ellipses thus determined are shown in the top insets.

To measure the polarization of this beam, especially its circular component, we mounted a GaAs single crystal on the center of a standard four-circle diffractometer and measured three multiple-beam interference profiles, \( L = \{511\}, \{151\}, \) and \( \{153\} \), on the main reflection \( H = \{442\} \). In these figures each data point (square) represents an integrated intensity over a (442) rocking curve, normalized to the (442) two-beam intensity obtained away from any multiple reflections. The asymmetric intensity profiles that are due to the multiple-beam interference effect are clearly seen on all three reflections. Table I lists all the relevant geometric and structural parameters for these three multiple-beam reflections. The interference intensity \( I_{\text{int}} \) can be directly obtained from the experiment, by taking the intensity difference \( \Delta I = I(\phi_L + \Delta \phi) - I(\phi_L - \Delta \phi) \) on either side of the excitation point \( \phi_L \) of the detour reflection \( L \). It is easy to show that \( \Delta I \approx 2I_{\text{int}} \). If we plot \( \Delta I \) vs \( 1/\Delta \phi \) we should get a straight line with its slope \( n = \Delta I/(1/\Delta \phi) \) proportional to the quantities in the square brackets of Eq. (5). By measuring this slope, we can determine a linear combination of \( (P_1, P_2, P_3) \), which defines a plane in Poincaré space. With three measurements on three different detour reflections, a point in Poincaré space can, in principle, be uniquely determined. In Fig. 2 the data in Figs. (1a)–(1c) are plotted after the transformation by the method just described. The solid lines are the least-square fits to the data. The three slopes obtained from the measured reflections are \( n(511) = 0.7208 \pm 0.0634 \) \( \mu \text{rad} \), \( n(151) = 0.3582 \pm 0.0310 \) \( \mu \text{rad} \), and \( n(153) = -0.6119 \pm 0.1492 \) \( \mu \text{rad} \). Because of the experimental uncertainties on the slope values, instead of directly solving the linear equations for \( P_1, P_2, P_3 \) from the three measured slope values, we use a fitting routine to minimize the standard \( \chi^2 \) function by adjusting \( P = (P_1, P_2, P_3) \). Using our measured slopes, we found that the \( \chi^2 \) minimum occurs at \( P = (0.77 \pm 0.18, 0.05 \pm 0.40, -0.57 \pm 0.07) \), which yields a total polarization of \( |P| = 0.96 \pm 0.14 \). The error bars were estimated from the points that give twice the \( \chi^2 \) value at the minimum. The upper bounds on all three components are eventually limited by \( |P| = 1 \). The large uncertainties on \( P_1 \) and \( P_2 \)

<table>
<thead>
<tr>
<th>( (hkl) )</th>
<th>( \phi_L )</th>
<th>( A_{\text{ew}} / \Gamma )</th>
<th>( A_{\text{sew}} / \Gamma )</th>
<th>( A_{\text{esw}} / \Gamma )</th>
<th>( A_{\text{sew}} / \Gamma )</th>
<th>( \delta_{\text{ML}} )</th>
<th>( \lambda L \cdot \sigma_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(511)</td>
<td>102.67°</td>
<td>613</td>
<td>503</td>
<td>-182</td>
<td>232</td>
<td>83.09°</td>
<td>-0.66</td>
</tr>
<tr>
<td>(151)</td>
<td>77.33°</td>
<td>613</td>
<td>-503</td>
<td>182</td>
<td>232</td>
<td>83.09°</td>
<td>0.66</td>
</tr>
<tr>
<td>(153)</td>
<td>101.86°</td>
<td>482</td>
<td>-480</td>
<td>26</td>
<td>62</td>
<td>-81.92°</td>
<td>0.71</td>
</tr>
</tbody>
</table>

TABLE I. Geometric and structural parameters for the GaAs(442)/(hkl) multiple-beam reflections. The azimuthal angle \( \phi = 0° \) is defined by the convention that the reciprocal vector \((-1,1,0)\) is lying on the diffraction plane, with a projection that is antiparallel to the incident wave vector \( k_0 \). The positive rotation corresponds to the situation that the Ewald sphere rotates around the main reflection \( H \) according to the right-hand rule.
are due to the fact that all three multiple reflection combinations have their phases $\delta_{\text{ML}}$ close to $\pm 90^\circ$, which were purposely chosen originally to yield high sensitivity on the circular polarization $P_3$.

The degree of circular polarization can be calculated for off-axis radiation, based on the properties of synchrotron radiation, particle source size, the slit sizes used in the experiment, and the observation angle $\psi$. The reduction of $\pi$ polarization due to the vertically diffracting monochromator crystals also needs to be taken into account. The circular polarization calculated this way yields a value of $P_3 = -0.75$. The 70% detuning between the two monochromator crystals, that was employed to reduce the harmonics, further lowers this value to $P_3 = -0.53$, which agrees well with our measured value $P_3 = -0.57 \pm 0.07$.

Based on the measured value of $P$, we calculated the MBD intensity using Eq. (4) for three multiple reflections, and the results, convoluted with a Gaussian instrumental width function ($\sigma = 0.003^\circ$), are shown in Fig. 1 as solid curves. These curves show good agreement with experimental data. In fact, one could fit the data in Fig. 1 directly using Eq. (4), but measured peak values need to be taken into account. At the MB peak position, Eq. (1) needs to be modified to include an absorption correction in the denominator of the MB resonant term, and that correction can be used as an adjustable parameter to match the measured peak intensities. This direct fitting procedure may be necessary if two or more multiple reflections are close together in the azimuthal scan.

The MBD method is sensitive to the handedness of a circular polarization. This point is demonstrated by another set of measurements on the same three MB reflections using the synchrotron radiation emitted 0.11 mrad above the orbital plane, $\psi = 0.11$ mrad, where we have equal total intensity as in the previous measurements but the circular component in the beam should change from right to left handed. The results are shown in Figs. 1(d)–1(f). As one can see, the sense of the asymmetry is reversed, as compared to Figs. 1(a)–1(c). An exact reversal in the asymmetry would be expected if $\delta_{\text{ML}} = \pm 90^\circ$ when the circular polarization changes its rotation sense. By simply changing the third Poincaré parameter from $P_3 = -0.57$ to $+0.57$ and leaving $P_1 = 0.77$ and $P_2 = 0.05$ unchanged, we produce theoretical calculations [solid curves in Figs. 1(d)–1(f)] that agree very well with the experimental data. This implies, as expected, that the synchrotron spectrum is symmetric with respect to the electron-positron orbital plane.

In summary, we have shown that MBD in a crystal can be used to completely determine the Poincaré polarization vector $P$ of an x-ray beam. The phase-dependent interference effect and the ability to mix the $\sigma$ and $\pi$ polarizations in a MBD process are the two important features that make the diffracted intensity sensitive both to the magnitudes of $\sigma$ and $\pi$ components and to the phase between them. The sensitivity of this new MBD polarimeter can be tuned to a specific type of polarization by choosing a proper MB combination. For example, a MB combination with the relative phase $\delta_{\text{ML}} = \pm 90^\circ$ in a noncentrosymmetric crystal is sensitive only to circular polarization $P_3$ and thus serves as a circular polarimeter. The MBD method can be applied to a wide x-ray energy range. With an ordinary inorganic crystal such as GaAs we expect it to work at energies as low as $\sim 3$ keV, which is simply limited by the largest possible Bragg angle for a low-order reflection. This lower limit can be decreased if an organic crystal of good quality is utilized. The upper energy limit is essentially determined by the density of defect reflections, which increases with energy and scales with the unit-cell size of the crystal. For GaAs the upper energy limit is $\sim 20$ keV. This wide energy range covers most $K$ and $L$ absorption edges that are of interest for magnetic studies. Compared to measuring circular polarization with magnetic Compton scattering, this MBD method can provide a much greater signal-to-noise ratio because it uses Bragg reflections from electric charge scattering. With this strong signal one can characterize an x-ray incident beam on a relatively fast time scale. It may also be possible to measure polarization of a magnetically diffracted beam from magnetic materials or detect circularly polarized fluorescence x rays near an atomic absorption edge. This kind of circular polarimetry on secondary x-ray photons may provide more direct insight on magnetic interaction of x rays with matter.

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19. K.-J. Kim, X-Ray Data Booklet (Lawrence Berkeley Laboratory, Berkeley, CA, 1986), Sect. 4.