Phase-sensitive x-ray diffraction in the expanded distorted-wave approximation

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Based on an expanded distorted-wave approximation, analytical intensity expressions are derived for phase-sensitive reference-beam x-ray diffraction in both the transmission and the reflection cases. Results from this approach are compared with the rigorous n-beam dynamical theory calculations and are shown to be very accurate if the crystal is in the thin kinematic limit. The method represents a unified theoretical approach for both the incoherent standing-wave effect and the phase-sensitive three-beam diffraction of the internal wave fields. The simple formulas for thin crystals are particularly useful for phase determination in biological crystals as well as in semiconductor thin-film structures.

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I. INTRODUCTION

It has been recognized that multiple or three-beam x-ray diffraction can be used to extract crystallographic phase information based on the interference effect between two or more simultaneously diffracted waves in a crystal.¹ ¹⁰ Among the various techniques for the three-beam diffraction is the recently developed reference-beam x-ray-diffraction method, which incorporates the phase-sensitive three-beam diffraction into an oscillating-crystal experimental setup.¹¹ ¹³

Using reference-beam diffraction one can simultaneously collect a large number of three-beam interference profiles in a relatively short time period. Therefore this technique is particularly useful for structural analyses of biological crystals since the short exposure times can significantly reduce the possible radiation damage to the specimens.

Phase information contained in an n-beam interference profile can be rigorously interpreted by the full n-beam dynamical diffraction theory.³ In this theory, one calculates the diffracted intensities by first solving for all possible eigenstate wave vectors inside the crystal and followed by matching the boundary conditions for each eigenwave. These can be done only through numerical solutions. Although it provides a powerful and precise theoretical description of n-beam diffraction, because of its complexity it is usually difficult to use such a computational procedure to efficiently analyze a large number of interference profiles measured from an experiment. Furthermore, the explicit phase dependence is not immediately apparent during the computations.

In order to gain physical insights to the multibeam interference effect and to overcome the difficulties in data analyses of n-beam diffraction, considerable efforts have been made to find approximate analytical expressions of the diffracted intensities. The most widely used approaches, including dynamical approximations⁷ ¹⁴ ¹⁷ and a second-order kinematical approximation,¹⁸ are based on the perturbational principles. In these methods one evaluates first the diffracted wave for a primary (main) reflection H, and then calculates the effect of a secondary (detour) reflection G as a form of perturbation. Consequently, these perturbational theories are valid only when G is far from the Ewald sphere and not fully excited.

A significantly different approach, based on a distorted-wave Born approximation, has been developed recently to solve the three-beam diffraction problem.¹⁹ ²⁰ It represents an extension to the conventional distorted-wave theory that has been widely used in grazing-incidence diffraction from surface and multilayer structures,²¹ ²⁶ and thus has been termed an expanded distorted-wave approximation (EDWA). The EDWA theory involves two steps. First, it treats the average charge density plus a single Fourier component as the distorting component and uses a standard two-beam dynamical theory to solve for a distorted wave inside a bulk crystal. Second, it calculates the rescattering of the distorted wave by the remaining other Fourier components of the charge density, using a first-order Born approximation. This second step is standard in conventional distorted-wave scattering theory.

Unlike the perturbational approaches mentioned above, the EDWA approach evaluates the diffraction by the secondary reflection G first, giving rise to a distorted wave inside the crystal, which is then used as the new incident wave for calculating the diffracted intensity of a primary reflection H using a first-order Born approximation. This procedure closely resembles the typical arrangement of a reference-beam diffraction experiment, in which a single secondary reflection G is used as a reference beam common to all primary reflections measured in a data set on an area detector.¹¹ Because of the order of the calculations used in the EDWA, i.e., secondary reflection G first and primary reflection H second, the final analytical formula is valid over the entire angular range of the G reflection, and thus is ideally suited for analyzing the reference-beam diffraction profiles.

The principles of the EDWA theory for reference-beam diffraction have been published previously for the semi-infinite Bragg case¹⁹ and the thin crystal Laue case.²⁰ The purposes of this article are to provide a unified and more detailed description for both the Laue and the Bragg cases, and to evaluate the validity of the EDWA theory for various crystal thicknesses. Through comparisons with the full n-beam dynamical calculations, we show explicitly that the EDWA theory can provide accurate results for thin crystals.
where a crystal thickness is small compared to the Pendellösung or extinction length. Since most biological crystals fall into the thin-crystal category, the EDWA approach can be used directly in reference-beam diffraction experiments on proteins or other macromolecules to extract reflection phases from the measured interference profiles.

II. GENERAL FORMULATION

In a standard distorted-wave Born approximation for x rays, the electric susceptibility \( \chi(\mathbf{r}) \) of a scattering system is divided into two parts: \( \chi(\mathbf{r}) = \chi_1(\mathbf{r}) + \chi_2(\mathbf{r}) \), with \( \chi_1(\mathbf{r}) \) being the distorting component and \( \chi_2(\mathbf{r}) \) the remaining component. The procedure is to obtain an exact solution for \( \chi_1(\mathbf{r}) \) only, resulting in a distorted wave \( \mathbf{D}_1 \) in the near field. This distorted wave is then subsequently scattered by \( \chi_2(\mathbf{r}) \), which can be evaluated using a first-order Born approximation:

\[
\mathbf{D}(\mathbf{r}) = \frac{\exp(-ik_0r)}{4\pi r} \int d\mathbf{r}' \exp(-ik_0 \mathbf{u} \cdot \mathbf{r}') \nabla' \times \nabla' \times \left[ \chi_2(\mathbf{r}') \mathbf{D}_1(\mathbf{r}') \right],
\]

where the integral is evaluated over the volume of the scattering specimen, \( k_0 = 2\pi/\lambda \) is the wave number of the incident beam, \( \lambda \) the wavelength, and \( \mathbf{D}(\mathbf{r}) \) is the electrical displacement of the final scattered wave along direction \( \mathbf{u} \).

For a crystal, the electric susceptibility \( \chi(\mathbf{r}) \) can be expanded into a Fourier series: \( \chi(\mathbf{r}) = -\Gamma S^{H} \exp(-i\mathbf{H} \cdot \mathbf{r}) \), where \( \Gamma = r_e^2/(4\pi V_c) \), \( r_e \) is the classical radius of an electron, and \( V_c \) the unit-cell volume. When Eq. (1) is used to investigate diffuse scattering in grazing-incidence conditions from surfaces and multilayer systems, the distortion component \( \chi_1(\mathbf{r}) \) includes only the homogeneous average susceptibility for a substrate or each layer, \( -\Gamma F_0 \), which gives rise to Fresnel waves as the distorted wave, and \( \chi_2(\mathbf{r}) \) in general covers any inhomogeneous variations such as surface and interfacial roughnesses.

The expanded distorted-wave approximation developed by our group extends the conventional distorted-wave approach in a such a way that it can be used to evaluate the phase-sensitive reference-beam diffraction in bulk crystals. A key revision made in the EDWA approach is that a sinusoidal Fourier component \( -\Gamma S^{L} F_L e^{-iL \cdot r} \) related to the reference reflection \( \mathbf{G} \) is added to the homogeneous average electric susceptibility \( -\Gamma F_0 \) in the distorting component \( \chi_1(\mathbf{r}) \). The distorted wave \( \mathbf{D}_1(\mathbf{r}) \) due to the new distortion \( \chi_1(\mathbf{r}) \) is calculated by the Bragg reflection cases. The distorted wave in these cases consists of two beams, \( \mathbf{K}_0 \) and \( \mathbf{K}_G \), and the scattering of these distorted waves by the crystal along \( \mathbf{k}_H \) is calculated by the EDWA using a first-order Born approximation.

The subsequent scattering of \( \mathbf{D}_1(\mathbf{r}) \) due to the remaining susceptibility \( \chi_2(\mathbf{r}) = -\Gamma S^{L} S^{H} F_L e^{-iL \cdot r} \) \((L \neq 0, \pm G)\) of the other Fourier components is obtained by Eq. (1). In the evaluation of the volume integral in Eq. (1), it is helpful to realize that compared to the phase factors \( K_0 \cdot \mathbf{r} \) and \( K_G \cdot \mathbf{r} \), the distorted-wave amplitudes \( D_0 \) and \( D_G \) in Eq. (2) are slow-varying functions in space since they generally depend on the extinction length or Pendellosung length which is a factor of \( 1/(|\Gamma| F_{G}^{j}) \) larger than the x-ray wavelength. Therefore these amplitudes can be factored out of the integral and the gradient operations, and the two Bloch waves in Eq. (2) can be treated as two plane waves for this step. Using a similar mathematical procedure as in Ref. 18 and omitting an insignificant prefactor, the scattered wave \( \mathbf{D}_H \) of a primary reflection \( \mathbf{H} \) has the following simple form:

\[
\mathbf{D}_H = \mathbf{u} \times \left[ \mathbf{u} \times (F_H \mathbf{D}_0 + F_{H,G} e^{i\alpha_G} e^{-iK_G \cdot r}) \right] e^{-ik_0r} /
\]

where the Bragg’s condition \( k_0 \mathbf{u} = K_0 + \mathbf{H} \) has been applied. This equation is a general expression for EDWA, with the first and the second terms representing a singly scattered wave (O→H) and a doubly scattered wave (O→G→H), respectively.

Although Eq. (3) is valid for all cases of incident-beam polarization, in the next sections we consider only situations where the incident polarization is \( \sigma \) polarized, i.e., perpendicular to the diffraction plane defined by \( \mathbf{K}_a \) and \( \mathbf{K}_G \). These are the most common cases when a linearly polarized syn-
The incident beam amplitude has been taken as unity. Using Eqs. (6) and (7), it follows that the total distorted waves, with wave vectors \( \mathbf{K}_0 = (\mathbf{K}_{0a} + \mathbf{K}_{0b})/2 \) and \( \mathbf{K}_G = (\mathbf{K}_{Ga} + \mathbf{K}_{Gb})/2 \), combined over the two branches of the dispersion surface, are given by

\[
D_0(z) = \frac{\Delta k z}{2} \left[ 1 + \frac{\sin(\Delta k t)}{\Delta k t} \right] - \frac{\tau \sin \delta}{2b(\eta_G^2 + 1)} \left[ 1 - \frac{\sin(\Delta k t)}{\Delta k t} \right] \frac{1 - \cos(\Delta k t)}{\Delta k t} \]

and

\[
D_G(z) = \frac{i}{\sqrt{b}} \frac{1}{\lambda} \frac{\Delta k z}{2} \sin \left( \frac{\Delta k z}{2} \right) \sin \left( \frac{\Delta k z}{2} \right) \,
\]

Equation (9) is an analytical expression for calculating the reference-beam diffraction intensity as a function of the reflection rocking angle through \( \eta_G \), with an explicit dependence on triplet phase \( \delta \). Note that here \( I_H \) is already equivalent to the integrated intensity in three-beam dynamical calculations, normalized to the two-beam value. Thus the EDWA is a tremendously simplified method, from which an interference profile can be conveniently generated.

In Fig. 2, we show a numerical example of the three-beam interference profiles for \( \mathbf{G} = (004) \) and \( \mathbf{H} = (317) \) of GaAs calculated using Eq. (9) and compared to the full dynamical diffraction results. The geometry is assumed to be a symmetric Laue case \( (b=1) \), with a crystal thickness of \( t = 0.5 \mu m \) in Fig. 2(a) and \( t = 0.2 \mu m \) in Fig. 2(b). The structure factors involved in the three-beam case are \( |F_{004}| = 163.3, \ |F_{317}| = 68.7, \ |F_{313}| = 109.9, \) while the invariant triplet phase \( \delta \) is artificially assigned by four representative values 0, 90, 180, and 270° in order to show the phase sensitivity of the EDWA approach. In all four cases, we see that the EDWA results are in excellent agreement with the dynamical calculations over the entire angular range for \( \mathbf{G} \), even on the small intensity modulations which are essentially the thickness-dependent Pendellö sung fringes arising from terms \( \cos(\Delta k t) \) in Eq. (9). Similar oscillatory behavior also exists in three-beam diffraction calculations on a finite crystal using a Takagi-Taupin dynamical approach.30

As customary in dynamical diffraction theory, we use a normalized Pendellösung thickness \( A \) to characterize whether a crystal is thin or thick, where \( A \) is defined by Zachariassen29 as the following:

\[
A = \frac{1}{2} \frac{\Delta k}{\eta_G} = \frac{1}{2} \eta_G \quad = \frac{1}{2} \frac{\Delta k}{\eta_G} = \frac{1}{2} (\Delta k) \,
\]
The calculations shown in Fig. 2 for crystal thicknesses of $t = 0.5 \mu m$ and $t = 0.2 \mu m$ correspond to effective thicknesses of $A = \pi t |F_G|/|\lambda \cos \theta_G| = 0.12$ and 0.05, respectively, which are therefore thin crystal cases since $A \ll 1$. As the thickness increases, the kinematic approximation involved in calculating the scattered intensity $I_H$ of the distorted wave becomes less accurate and therefore the EDWA results can exhibit significant departures from the $n$-beam dynamical calculations. We will discuss this point in more detail in Sec. V.

Assuming Pendello"sung thickness $A$ is small, Eq. (8) for the distorted-wave amplitudes can be simplified to the following expressions: 

\[
\begin{align*}
D_0(z) &= \cos(A \eta_G) + i \sin(A \eta_G) \\
D_{G}(z) &= i \sqrt{b} \sin(A \eta_G)/\eta_G
\end{align*}
\]  

(11)

where $A = A \cdot z/t$, and for symmetric Laue cases $b = 1$, Eq. (9) is reduced to

\[
I_H(\eta_G) = 1 - \tau \sin \delta \frac{\sin^2(A \eta_G)}{A \eta_G} + \frac{\tau}{\eta_G} \left( \cos \delta + \frac{\tau}{2 \eta_G} \right) \\
\times \left( 1 - \frac{\sin(2A \eta_G)}{2A \eta_G} \right).
\]

(12)

For the examples shown in Fig. 2, Eq. (12) gives rise to the exact same results as those using Eq. (9). The extension of Eq. (12) to the $b \neq 1$ cases is trivial by replacing $\tau$ with $\tau \sqrt{|b|}$.

One of the characteristics that can be seen from Fig. 2 is the effect of "size broadening" in the case of three-beam interference. As long as the thin-crystal limit applies, the angular range in which the interference effect occurs becomes wider for thinner crystals, while its peak interference intensity decreases. In fact, with the help of Eq. (12) we can show that this is generally true, and in the case that the product of $A$ and the structure-factor ratio $\tau$ is also small, the interference angular range is inversely proportional to crystal thickness $A$ while the interference peak intensity is linear with the thickness $A$. To prove this, we start with Eq. (12) when $\delta = 0^\circ$, and take the derivative $dI_H/d\eta_G$ and set it to zero in order to find the extrema locations $\eta_m$. Expanding $\sin(2A \eta_G)$ to a fifth-order Taylor series, we obtain a quadratic equation, $3(A \eta_m)^2 + 4\tau \eta_m = 5$. This equation for $\eta_m$ can be easily solved, and in the case of $A \tau \ll 1$, we have

\[
\eta_m = \sqrt{5/3}/A = \pm 1.29/A,
\]

(13)

\[
I_H(\eta_m) \approx 1 \pm 0.57 A \tau.
\]

(14)

Equations (13) and (14) show explicitly that the angular location $\eta_m$ of the interference extrema is proportional to $1/A$ and the interference intensity $I_H(\eta_m) - 1$ is proportional to $A$. In practice, $\eta_m$ can be converted back to a real angular dimension:

\[
\Delta \theta_m = \pm 1.29 d_G/(\pi t) = \pm 0.41 d_G/t,
\]

(15)

where $d_G$ is the $d$ spacing of reflection $G$ and $t$ is the crystal thickness. For the $t = 0.5\mu m$ example given in Fig. 2(a), these equations yield that $\Delta \theta_m \approx 0.12$ mrad, $I_H(\text{max}) \approx 1.11$ and $I_H(\text{min}) \approx 0.89$, as compared to $\approx 0.14$ mrad, 1.13 and 0.88, as shown in Fig. 2(a) for the $\delta = 0^\circ$ curve. These excellent agreements demonstrate that the simple Eqs. (13)–(15) can be used conveniently in thin-crystal cases to accurately estimate the angular range of an ideal three-beam profile and the maximum observable interference effects in a reference-beam diffraction experiment.

We would like to point out that the results calculated using Eq. (12)–(15) are universal since all parameters are expressed through the dimensionless quantities such as $A$ and $\eta_G$. The scaling rules (13)–(15) further suggest that in the case of thin crystals, all interference profiles with the same triplet phase $\delta$ are in fact self-similar and have the same scalable shape. In this limit, the figure-of-merit of an observable interference profile can be defined as the angular spread $\eta_m$ multiplied by the peak interference intensity $I_H(\eta_m) - 1$: $\eta_m[I_H(\eta_m) - 1] = 0.74 \tau$, which implies that a stronger coupling reflection $H \cdot G$ or a weaker primary reflection $H$ would provide a stronger interference effect. This is true until the structure factor ratio $\tau$ becomes so large that the quadratic terms omitted in Eq. (14) start to take effects, as discussed in Ref. 20.
IV. BRAGG REFLECTION GEOMETRY

In a Bragg reflection geometry shown in Fig. 1(b), the boundary conditions at both the entrance surface $z=0$ and the exit surface $z=t$ have to be used in order to solve for the distorted-wave amplitudes:

\[
\begin{align*}
D_{0\alpha} + D_{\beta} &= 1 \\
D_{G\alpha} e^{-i \mathbf{k}_{\alpha} \cdot \mathbf{z}} + D_{G\beta} e^{-i \mathbf{k}_{\beta} \cdot \mathbf{z}} &= 0,
\end{align*}
\]

where $\mathbf{z}$ is a unit vector along the $z$ axis. Using the same procedure as in the previous section, we obtain the following expression for the distorted-wave amplitudes $D_0(z)$ and $D_G(z)$:

\[
\begin{align*}
D_0(z) &= \frac{\eta_G \sin [\Delta k (t-z)/2] + i \sqrt{\eta_G - 1} \cos [\Delta k (t-z)/2]}{\eta_G \sin (\Delta k t/2) + i \sqrt{\eta_G - 1} \cos (\Delta k t/2)} \\
D_G(z) &= \frac{-\sqrt{\eta_G} \sin [\Delta k (t-z)/2]}{\eta_G \sin (\Delta k t/2) + i \sqrt{\eta_G - 1} \cos (\Delta k t/2)}.
\end{align*}
\]

with $\Delta k = k_0 \Gamma |\mathbf{F}_G| \sqrt{\eta_G - 1}/\gamma_0 |\gamma_G|$. Inserting Eq. (17) into Eq. (5), the diffracted intensity for reflection $\mathbf{H}$ integrated over the crystal thickness can be calculated numerically by a simple computer routine using complex variables. For an analytical expression, care has to be taken when evaluating $\sqrt{\eta_G - 1}$ which can be imaginary depending on whether $|\eta_G| > 1$ or $|\eta_G| < 1$. Nonetheless, the following expressions for $I_H$ can be derived:

(i) for $|\eta_G| > 1$,

\[
I_H = \frac{2}{|\zeta|^2} \left( \eta_G |b| + \tau^2 - 2 \eta_G \tau \sqrt{|b|} \cos \delta \right) \left[ 1 - \sin (\Delta k t) \right] \\
+ \frac{2 (\eta_G^2 - 1)}{|\zeta|^2} \left[ \frac{\sin (\Delta k t)}{\Delta k t} \right] \\
+ \frac{4 \tau}{|\zeta|^2 \sqrt{|b|}} \sqrt{\eta_G - 1} \sin \delta \cos (\Delta k t) - 1 \Delta k t,
\]

with $|\zeta|^2 = 4 \eta_G^2 - 2 - 2 \cos (\Delta k t)$, and

(ii) for $|\eta_G| < 1$,

\[
I_H = \frac{e^{2 \Delta k' t} - 1 - 2 \Delta k' t e^{\Delta k' t}}{\Delta k' t |\zeta'|^2 |b|} \left( \tau^2 + \eta_G^2 |b| - 2 \eta_G \tau \sqrt{|b|} \cos \delta \right) \\
+ \frac{e^{2 \Delta k' t} - 1 + 2 \Delta k' t e^{\Delta k' t}}{\Delta k' t |\zeta'|^2} (1 - \eta_G^2) \\
- \frac{2 (e^{\Delta k' t} - 1)^2}{\Delta k' t |\zeta'|^2 \sqrt{|b|}} \tau \sqrt{1 - \eta_G^2} \sin \delta,
\]

with $|\zeta'|^2 = e^{2 \Delta k' t} + (2 - 4 \eta_G^2) e^{\Delta k' t} + 1$ and $\Delta k' = k_0 \Gamma |\mathbf{F}_G| \sqrt{1 - \eta_G^2} \sqrt{|y_0|} |\gamma_G|$. These equations are the final analytical expressions of the Bragg-case reference-beam diffraction intensities for the entire range of $\mathbf{G}$ reflection rocking angle. Although Eqs. (18a) and (18b) have different mathematical forms, interference profiles generated from these two equations are always continuous and smooth across the degenerate points $\eta_G = \pm 1$.

A numerical example of the EDWA theory, Eq. (18), for the Bragg case is shown in Fig. 3(a) and compared with the three-beam dynamical diffraction calculation. Here we have chosen the same GaAs (317) (004) case except that the ref-

FIG. 3. (a) Comparisons of EDWA calculations (lines) with the full three-beam dynamical theory (symbols) for a symmetric Bragg case: GaAs $\mathbf{G} = (004)$ and $\mathbf{H} = (317)$, x-ray wavelength $\lambda = 0.918$ Å, and crystal thickness $t = 0.5 \mu$m. (b) Same as in (a) except that the magnitude of $F_{317}$ is reduced by a factor of 10 to illustrate the Umweganregung effect.
ference reflection $G=(004)$ is now a symmetric Bragg reflection ($b=-1$). In the dynamical diffraction calculations, the present diffraction geometry is, more precisely, a Bragg-Laue case while the Laue case in the previous section is actually a Laue-Laue case according to the diffraction geometry of $H=(317)$. As can be seen in Fig. 3(a), the agreement between the EDWA theory and the dynamical calculation is again excellent over the entire angular range of the $G$-reflection rocking curve. Further calculations have shown that such agreements exist as long as the crystal thickness is considered thin as discussed in the next section. Good agreements between the EDWA and the $n$ beam have also been found for “mixed” three-beam diffraction geometries, i.e., Bragg-Laue and Laue-Bragg cases.

By comparing Fig. 3(a) of Bragg case with Fig. 2(a) of Laue case for the same thickness, we find an interesting phenomenon that the three-beam interference effect appears to be much stronger in the Bragg case than in the Laue case. This phenomenon can be understood from the concept of effective thickness characterized by parameter $A$. As defined in Eq. (10), $A$ is equal to $\pi T |F_G|/t(\lambda \cos \theta_G)$ in a symmetric Laue case and $\pi T |F_G|/t(\lambda \sin \theta_G)$ in a symmetric Bragg case. It is well known in dynamical diffraction that when $A$ is small compared to unity, the two-beam rocking curves in both the Bragg and the Laue cases are identical\(^{29}\) for the same value of $\theta_G$. For the cases considered here, since the Bragg angle for the GaAs (004) is $\theta_G=19^\circ$, the Bragg case is effectively much thicker than the Laue case by a factor of $\tan(\theta_G)$, thus giving rise to a stronger three-beam effect. In fact, we have verified through numerical calculations that the three-beam interference profiles appear identical to each other in the two cases as long as $A$ is the same. Nevertheless, for a same physical thickness the Bragg geometry is a more favorable setup for three-beam interference observations in experiments.

We would like to note that the EDWA theory not only provides a correct description of the phase-dependent interference effect, but also takes into account the energy flow balance among different reflections, namely the Umweganregung or the Aufhellung effects that depend only on the strengths of the structure factors involved.\(^{10}\) In Ref. 20, we have demonstrated these effects in the EDWA calculations for the Laue case. To see these effects in the Bragg-case example Fig. 3(a), we artificially reduce $|F_H|$ by a factor of 10 to adjust the ratio of $|F_H|/|F_G|$ from 0.4 to 0.04. With the reduced $|F_H|$ value, the $I_H$ intensity profiles calculated from Eq. (18) and from the dynamical diffraction theory are plotted in Fig. 3(b). Apparently, the intensity values of all four representative profiles are enhanced considerably as a result of the energy transfer from the strong reflection $G$ to the weak reflection $H$. Nonetheless, the EDWA results are again in excellent agreements with the $n$-beam dynamical theory, indicating that the energy-flow effects are fully accounted for in the EDWA approach.

Just like in the Laue case, the expressions (17) and (18) can be greatly simplified if $A\ll1$, since $\cos(A\sqrt{\eta_G^{-1}-1}) \approx \cos(A\eta_G)$ and $\sin(A\sqrt{\eta_G^{-1}-1})/\sqrt{\eta_G^{-1}-1} \approx \sin(A\eta_G)/\eta_G$. With these approximations and keeping only the leading terms in $A$, Eq. (17) for the distorted waves can be reduced to

$$
\begin{align*}
D_0(z) &= \cos(z' A \eta_G) - i \sin(z' A \eta_G), \\
D_G(z) &= i \sqrt{|b|} \sin(z' A \eta_G)/\eta_G,
\end{align*}
$$

(19)

where $z'=1-z/t$. Inserting Eq. (19) into Eq. (5) and changing the dummy integral variable to $z'$, we realize that Eq. (19) gives rise to the same integral as in the Laue case except for a sign difference in $\eta_G$, which is due to its definition through $b$. Therefore we conclude that the final diffracted intensity $I_H$ in a thin Bragg case is identical to Eq. (12) in the thin Laue case. This conclusion of the EDWA provides an analytical proof to our earlier discussion that the three-beam interference profiles for thin crystals are indistinguishable in Laue and Bragg cases as long as the same effective thickness $A$ is used in the calculations. Furthermore, it shows that Eqs. (13)–(15) can also be applied to Bragg cases to estimate the angular range and the peak interference intensities of reference beam diffraction.

V. DISCUSSIONS AND CONCLUSIONS

We have shown in the previous sections that the EDWA is a simple phase-sensitive diffraction theory that can provide accurate three-beam profiles in excellent agreements with the $n$-beam dynamical calculations, in the case of thin crystals. As the crystal thickness increases, however, the EDWA results may exhibit significant departure from the $n$-beam dynamical calculations. In order to investigate at what thickness values the EDWA results start to deviate from the exact $n$-beam theory, we have performed Laue-case calculations of the diffracted intensities at the center of the $G$ reflection rocking curve ($\eta_G=0$) for the GaAs (317)/(004) $\delta=\pm90^\circ$ cases as a function of the effective thickness $A$, using both the EDWA and the $n$-beam approach. The results are shown in Fig. 4 with the lines for the EDWA and symbols for the $n$ beam. As a reference, the two-beam integrated intensity curve for the $G$ reflection is shown as the dotted curve. In Fig. 5, we show the detailed discrepancies of the four representative interference profiles at thickness $t=5\,\mu m$ which corresponds to $A=1.2$. All EDWA calculations shown in Figs. 4 and 5 are performed with Eq. (9) without the thin-crystal approximation.

First of all, we can see from Fig. 4 that the EDWA calculations exhibit the Pendelosung oscillations, qualitatively similar to the two-beam and $n$-beam dynamical calculations, with a correct period of $A=\pi$ that corresponds exactly to the two-beam Pendelosung distance of 12.7 $\mu m$ for the $G$ reflection. Quantitatively, however, the $n$-beam results are more complex, essentially due to the dynamical Pendelosung effect of the primary reflection $H$, which has been completely ignored in the EDWA approach. Therefore it is not surprising to see the substantial deviations between the two theories at larger $A$ values. In particular, we note that the two EDWA curves for $\delta=270^\circ$ and $\delta=90^\circ$ never cross over each other, namely, the intensity for $\delta=270^\circ$ is always greater than or equal to that for $\delta=90^\circ$. This is not true in the full $n$-beam calculations, where the curves do cross over.
that the agreement appears to be better for the
d shows a larger deviation for A
thin, as discussed in Ref. 10.
for thick Laue cases but is not a problem when the crystal is
5 thus reversing the peak intensities for the
d beam interference profiles.10 It is also noticeable in Fig. 4
offer reliable triplet-phase measurements from the three-
dynamical calculations for the Laue cases of
ference is caused by the three-beam interference that effec-
ts that the EDWA theory is a good approximation in the
thin-crystal regime of A
5, m
5

FIG. 4. Comparisons of the EDWA peak intensities I
(\(\eta_0\) = 0), as a function of effective crystal thickness A, with the full
dynamical calculations for the Laue cases of \(\delta = \pm 90^\circ\) in Fig. 2.
The apparent oscillations in the intensities are the Pendellösung
fringes that also exist in the standard two-beam dynamical calculation
(shown as dotted line) for the G reflection.

thus reversing the peak intensities for the \(\delta = 270^\circ\) and \(\delta = 90^\circ\) cases. This may complicate the phase determinations
for thick Laue cases but is not a problem when the crystal is thin, as discussed in Ref. 10.

Despite the discrepancies at larger A values, Fig. 4 indicates that the EDWA theory is a good approximation in the
thin-crystal regime of \(A < 1\) as the EDWA basically agrees with the n-beam results. This is also the regime where the
\(\delta = 270^\circ\) and \(\delta = 90^\circ\) peak intensities do not cross and thus offer reliable triplet-phase measurements from the three-
beam interference profiles.10 It is also noticeable in Fig. 4 that the agreement appears to be better for the \(\delta = 90^\circ\) case,
meaning beyond \(A = 1\), while the \(\delta = 270^\circ\) case seems to show a larger deviation for \(A < 1\). We believe that this difference is caused by the three-beam interference that effec-
tively makes the H reflection weaker (stronger) in the \(\delta = 90^\circ(270^\circ)\) case, thus better (less) suited for the kinematic approximations involved in the EDWA approach.

It is useful to note that expression (12) for thin crystal
cases is a function of rocking angle \(\eta_0 \propto \Delta \theta\) through three
parameters, \(\tau, A\), and \(\delta\). If the diffraction geometry is asym-
metric, the first two parameters become \(\tau \sqrt{|b'|}\) and \(A \sqrt{|b'|}\), respectively. For a given three-beam combination G, H, and
H-G but unknown geometry and/or thickness, as usually the case in experiments with protein crystals, these three parameters
are independent of each other through thickness \(t\), asymmetry \(b\), and triplet phase \(\delta\). Therefore, in a real data
analysis, quantities \(\tau\) (or \(A\)), \(A\), and \(\delta\) in Eq. (12) should be
treated as three independent variables (fitting parameters),
where \(A\) \(\delta\) determines the peak interference intensity, \(A\) controls
the effective width of the interference profile, and \(\delta\) is
the triplet phase of the three reflections involved.

The EDWA approach not only can be used to reproduce the n-beam dynamical calculations of reference-beam inter-
ference profiles, but is also helpful in systematic investiga-
tions of the effects of various practical parameters on the
observable interference intensities in thin-crystal cases. Us-
ing Eqs. (11), (19), and (12), one can study the influence of such parameters as crystal size and mosaicity, x-ray wave-
length, unit-cell dimensions, and relative strengths of primary (H), detour (G), and coupling (H-G) reflections. Some of
these effects have been discussed in the previous sections
as well as in Ref. 20, and we would just like to provide one
more example. Using Eqs. (10) and (14), one can easily con-
clude that the interference intensity \(I_H(\eta_0)\) \(-1\) is roughly
proportional to x-ray wavelength \(\lambda\), indicating that the three-
beam effect is more observable with lower energy x-rays if
absorption effect is neglected. This conclusion agrees with
the previous experimental observations8 and numerical
calculations.10

The successful application of the distorted-wave scattering
to the case of reference-beam diffraction demonstrates the
usefulness of the distorted-wave theory in general. In the
area of x-ray scattering and diffraction, while most previous
distorted-wave applications in the literature21–26 involve only
Fresnel optical theory for distorted waves in grazing-
incidence conditions, a two-beam dynamical theory has been
used previously to obtain Bloch waves as the distorted
waves described in this article are essentially the same wave
fields that form x-ray standing waves27 in crystals and thin
films. Thus the analytical expressions presented here, es-
pecially Eqs. (11) and (19) for thin crystals, can be applied in
the x-ray standing wave studies of bulk crystals, thin films,
and superlattices when overall absorption can be
neglected.32,33 In this connection, the EDWA approach
provides a natural unified transition in x-ray theory from the
incoherent standing wave yield to the coherent scattering of
the distorted internal waves that gives rise to the three-beam interference. It is therefore expected that our EDWA results may stimulate other research efforts in distorted-wave appli-
cations, especially in the area of multiple-beam x-ray crys-
tallography, x-ray standing waves, and three-beam structural
studies of thin films, surfaces, and interfaces.34–36

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