5. Concluding remarks

The investigation we have carried out demonstrates that the multiple diffraction of X-ray spherical waves (six-beam in particular) is a very complicated phenomenon. More systematic studies, both theoretical and experimental, are necessary. The intensity distribution on the topograph depends to a great extent on the distance between the source and the film. Since nonmonochromatic radiation has been used in the experiments it is impossible to compare the topographs in the forward-transmitted beam with the experimental topographs. Nevertheless, there is qualitative agreement. In order to reveal the fine structure of the experimental topographs it is desirable to use monochromatic radiation.

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Modified Two-Beam Description of X-ray Fields and Intensities near a Three-Beam Diffraction Point. Second-Order Solution

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Abstract

The previously developed first-order modification of two-beam diffraction near a third reciprocal-lattice point [Juretschke (1984). *Acta Cryst.* **A40**, 379-389] is extended to second order. To this order a modified two-beam description is still retainable, and the normal modes persist in their original polarization, but now with respect to two-beam asymptotes rotated and with a Bragg angle altered relative to the original two-beam case. Integrated intensities are evaluated for modified strong and weak primary two-beam symmetric Bragg reflections, in the Bragg–Bragg and Bragg–Laue configurations, and some implications of the results are discussed.

1. Introduction

A compact analytical description of X-ray modes and intensities in the neighborhood of multiple-interaction regions would have practical as well as theoretical uses. Among other things, it would allow the evaluation of the influence of additional nearby reflections on a particular reflection of interest without having to invoke the computer machinery of a full n-beam solution. Equally well, it would lead to easy identification of particular features of diffraction, such as the traditional *Aufhellung*, or *Umweganregung*, using relatively simple rules.

The first-order formulation of such a description (Juretschke, 1982, 1984; Høier & Marthinsen, 1983), based on a systematic perturbation treatment of the n-beam equations, has already been applied successfully to clarify some old problems (Juretschke & Barnea, 1986; Juretschke & Wagenfeld, 1986), as well as to predict additional results (Juretschke, 1986*a*, *b*). In the course of these investigations, however, a number of instances emerged in which this first-order formulation led to null results, or where it applied in only a very limited domain, so that the dominant

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asymptotic effects were expected to be of second or higher order.

This paper presents a systematic formulation of such second-order asymptotic modifications of a two-beam case. Surprisingly, it can continue to be cast in the same formalism as the first-order effects, at least in the formulation so far that is restricted to three-beam cases. In contrast to first-order theory, where an n-beam interaction is the sum of $n-2$ three-beam cases, second-order effects introduce additional crosslinks for $n>3$ that obscure the basic systematics of the solution. Such higher-n cases, even though they probably follow the same pattern as $n=3$, will require a separate study.

The essential results of the first-order treatment are that in the presence of the perturbing influence of a reciprocal-lattice point **L** approaching the Ewald sphere, a two-beam case **0H**, with structure factor F_H, continues to be describable in the immediate neighborhood of the **0H** reflection as a two-beam case, but with a modified structure factor F_{HL} and centered around a shifted Lorentz point. The σ and π polarizations remain as normal modes, but the modifications of the two modes are not the same. As a result of this particular form of the perturbation, the full arsenal of the existing two-beam literature can be adapted directly to describe the influence of an approaching **L** merely by inserting the appropriate modifications mentioned above.

The underlying rationale of the method parallels that of the Bethe treatment of dynamic potentials in electron diffraction (Bethe, 1928) using second-order perturbation theory. As adapted to X-rays, such a perturbation theory becomes a series solution in powers of $1/\xi_L$ where ξ_L is the distance of **L** from the Ewald sphere (excitation error). The existing first-order treatment contains all terms of order $1/\xi_L$. It also retains some terms of order $(1/\xi_L)^2$, in order to 'complete squares' and thus maintain the compactness of the solution. The second-order formulation presented here includes all terms of order $(1/\xi_L)^2$, but, following the same spirit of elegance, will also retain some contributions of higher order.

Contrary to some initial expectations, it has turned out that the same conceptual picture of the first-order solution carries over to the next order: the problem is still reducible to a modified two-beam case, with the normal modes retaining their original polarizations. The new modifications add further corrections to F_{HL}, and to the shift of the Lorentz points. In addition, they introduce a rotation in the two-beam asymptotes, as well as a change in the Bragg angle. Once these modifications have been identified, the same adaptation of the existing two-beam literature utilized in first order can then be extended to include all second-order effects. In retrospect, these qualitatively new modifications arising in second order are not unexpected, since they begin to describe the transition from the two-beam Lorentz point to the correct three-beam Lorentz point in increasing detail.

Once known, the second-order contributions will also shed new light on the restrictions imposed on a converging perturbation series, and perhaps on identifying the proper mode combinations required to deal with degeneracies.

Various higher-order corrections to two-beam diffraction have played an important role in high-energy electron diffraction for many years [*e.g.* see the summary in Herzberg (1971)], and have also been developed systematically based on Bethe's approach (Ichikawa & Hayakawa, 1977). Apart from the additional complexities of vector rather than scalar fields, the treatment presented here emphasizes those aspects of dynamical solutions important for X-rays, in particular modifications of structure factors as well as of absorption, and as much as possible within the familiar conceptual framework of traditional two-beam theory.

The exposition below relies heavily on the formulation and notation introduced by Juretschke (1984), most of which will not be reproduced here. For brevity, that paper will from now on be designated by (I).

2. Second-order solution – modes and dispersion relation

The second-order solution builds on the first-order solution of (I) by a systematic inclusion of all terms of order $(1/\xi_L)^2$ not already included – for the sake of compactness – in the first-order results. For convenient reference, we reproduce here the exact formulation of the three-beam problem that led to the modified two-beam result in first order.

The field equations for the **0H** fields $E_0^\sigma, E_H^\sigma, E_0^\pi, E_H^\pi$ are, from (I-17),

$$\begin{aligned}
&2\tilde{\xi}_0^\sigma E_0^\sigma + F_{H\bar{L}}^\sigma E_H^\sigma \\
&\quad +\tfrac{1}{2}(F_{\bar{L}}/\tilde{\xi}_L)(\Pi_2 F_L E_0^\pi - \Pi_3 F_{L-H} E_H^\pi) = 0,\\
&F_{HL}^\sigma E_0^\sigma + 2\tilde{\xi}_H^\sigma E_H^\sigma \\
&\quad +\tfrac{1}{2}(F_{\bar{L}-\bar{H}}/\tilde{\xi}_L)(\Pi_2 F_2 E_0^\pi - \Pi_3 F_{L-H} E_H^\pi) = 0,\\
&2\tilde{\xi}_0^\pi E_0^\pi + F_{H\bar{L}}^\pi E_H^\pi \\
&\quad +\tfrac{1}{2}(F_{\bar{L}}/\tilde{\xi}_L)(\Pi_2 F_L E_0^\sigma + \Pi_2 F_{L-H} E_H^\sigma) = 0,\\
&F_{HL}^\pi E_0^\pi + 2\tilde{\xi}_H^\pi E_H^\pi \\
&\quad -\tfrac{1}{2}(F_{\bar{L}-\bar{H}}/\tilde{\xi}_L)(\Pi_3 F_L E_0^\sigma + \Pi_3 F_{L-H} E_H^\sigma) = 0.
\end{aligned} \tag{1}$$

The reduced variables $\tilde{\xi}_0^\sigma$, $\tilde{\xi}_H^\sigma$; $\tilde{\xi}_0^\pi$, $\tilde{\xi}_H^\pi$ appearing in (1) are related to the actual excitation errors ξ_0, ξ_H in the plane of incidence by

$$\begin{aligned}\tilde{\xi}_0^\sigma &= \xi_0/k\Gamma - \Pi_1 F_L F_{\bar{L}}/(4\tilde{\xi}_L),\\ \tilde{\xi}_H^\sigma &= \xi_H/k\Gamma - \Pi_1 F_{L-H} F_{\bar{L}-\bar{H}}/(4\tilde{\xi}_L),\\ \tilde{\xi}_0^\pi &= \xi_0/k\Gamma - \Pi_4 F_L F_{\bar{L}}/(4\tilde{\xi}_L),\\ \tilde{\xi}_H^\pi &= \xi_H/k\Gamma - \Pi_5 F_{L-H} F_{\bar{L}-\bar{H}}/(4\tilde{\xi}_L),\end{aligned} \tag{2}$$

with $1/\tilde{\xi}_L = k\Gamma/\xi_L$ as the perturbation parameter.

The effective structure factors in (1) are given by [including correction of a printing error in (I-16b)]:

$$\begin{aligned}F_{HL}^\sigma &= F_H - \Pi_1 F_L F_{\bar{L}-\bar{H}}/(2\tilde{\xi}_L),\\ F_{HL}^\pi &= PF_H - \Pi_6 F_L F_{\bar{L}-\bar{H}}/(2\tilde{\xi}_L),\end{aligned} \tag{3}$$

and the coefficients $\Pi_1, \ldots, \Pi_6$ in (1) to (3) are geometrical factors for properly resolving the vector fields defined in (I-15), while $P = \cos 2\theta_B$.

The first-order solutions of (I) are obtained by neglecting the final set of brackets in each of the four equations (1). In second order, these brackets have to be included, but in the nature of corrections, which will allow certain simplifications.

In general, we expect (1) to lead, at a given angle of incidence, to distinct first-order solutions ($\tilde{\xi}_0^\sigma$, $\tilde{\xi}_H^\sigma$) and ($\tilde{\xi}_0^\pi$, $\tilde{\xi}_H^\pi$). We can then relate the σ and π fields of a particular solution by explicitly expressing the concluding brackets in (1) in terms of the variables of that solution. Thus, for the σ solution we solve the last two equations of (1) for E_0^π and E_H^π, and then form their combination appropriate to the concluding brackets in the first two lines of (1). This yields

$$\begin{aligned}&(1/2\tilde{\xi}_L)(\Pi_2 F_L E_0^\pi - \Pi_3 F_{L-H} E_H^\pi)\\ &\quad = -(2/\tilde{\xi}_L^2)[(F_L E_0^\sigma + F_{L-H} E_H^\sigma)\\ &\qquad \times (4\tilde{\xi}_0^\pi \tilde{\xi}_H^\pi - F_{HL}^\pi F_{\bar{H}\bar{L}}^\pi)]^{-1}\\ &\qquad \times [\Pi_3^2 F_{L-H} F_{\bar{L}-\bar{H}} \tilde{\xi}_0^\pi + \Pi_2^2 F_L F_{\bar{L}} \tilde{\xi}_H^\pi\\ &\qquad + \tfrac{1}{2}\Pi_2\Pi_3 (F_L F_{\bar{L}-\bar{H}} F_{\bar{H}\bar{L}}^\pi + F_{\bar{L}} F_{L-H} F_{HL}^\pi)].\end{aligned} \tag{4a}$$

Here the ($\tilde{\xi}_0^\pi$, $\tilde{\xi}_H^\pi$) are those related to the solution ($\tilde{\xi}_0^\sigma$, $\tilde{\xi}_H^\sigma$) *via* (2). Similarly, for the π-solution we obtain

$$\begin{aligned}&(1/2\tilde{\xi}_L)(F_L E_0^\sigma + F_{L-H} E_H^\sigma)\\ &\quad = (-2/\tilde{\xi}_L^2)[(\Pi_2 F_L E_0^\pi - \Pi_3 F_{L-H} E_H^\pi)\\ &\qquad \times (4\tilde{\xi}_0^\sigma \tilde{\xi}_H^\sigma - F_{HL}^\sigma F_{\bar{H}\bar{L}}^\sigma)^{-1}]\\ &\qquad \times [F_{L-H} F_{\bar{L}-\bar{H}} \tilde{\xi}_0^\sigma + F_L F_{\bar{L}} \tilde{\xi}_H^\sigma\\ &\qquad - \tfrac{1}{2}(F_L F_{\bar{L}-\bar{H}} F_{\bar{H}\bar{L}}^\sigma + F_{\bar{L}} F_{L-H} F_{HL}^\sigma)].\end{aligned} \tag{4b}$$

Since the right-hand sides of (4a) and (4b) are proportional to $1/\tilde{\xi}_L^2$, they were properly negligible in the first-order solution. Now they must be retained, but being already explicitly of second order, the individual contributions within them can be used, if necessary, to lowest order. To this order, for example, for a particular σ-like or π-like solution $\tilde{\xi}_0^\sigma$ and $\tilde{\xi}_0^\pi$ are interchangeable. Hence, since (4a) applies to a σ-like solution, we can write the denominator as

$$\begin{aligned}4\tilde{\xi}_0^\pi \tilde{\xi}_H^\pi - F_{HL}^\pi F_{\bar{H}\bar{L}}^\pi &\simeq F_{HL}^\sigma F_{\bar{H}\bar{L}}^\sigma - F_{HL}^\pi F_{\bar{H}\bar{L}}^\pi\\ &= D/2\end{aligned} \tag{5a}$$

and correspondingly in (4b), which applies to a π-like solution, the denominator can be approximated by

$$\begin{aligned}4\tilde{\xi}_0^\sigma \tilde{\xi}_H^\sigma - F_{HL}^\sigma F_{\bar{H}\bar{L}}^\sigma &\simeq F_{HL}^\pi F_{\bar{H}\bar{L}}^\pi - F_{HL}^\sigma F_{\bar{H}\bar{L}}^\sigma\\ &= -D/2.\end{aligned} \tag{5b}$$

The same kind of interchange of $\tilde{\xi}$'s can be applied to terms in the numerators of (4), and we can also use, where appropriate, relations, such as in (4a),

$$\tilde{\xi}_H^\pi E_H^\pi = -\tfrac{1}{2} F_{HL}^\sigma E_0^\sigma \quad \text{or} \quad \tilde{\xi}_0^\pi E_0^\sigma = -\tfrac{1}{2} F_{\bar{H}\bar{L}}^\sigma E_H^\sigma.$$

The object of these manipulations is to rewrite the first two and the last two lines of (1) separately in a two-beam-like form. Indeed, this is achieved by introducing the modified field modes

$$\begin{aligned}\bar{E}_0^\sigma &= E_0^\sigma - \Pi_3^2 (F_{L-H} F_{\bar{L}-\bar{H}})(F_{\bar{L}} F_{L-H}) E_H^\sigma/(2D\tilde{\xi}_L^2)\\ \bar{E}_H^\sigma &= E_H^\sigma - \Pi_2^2 (F_L F_{\bar{L}})(F_L F_{\bar{L}-\bar{H}}) E_0^\sigma/(2D\tilde{\xi}_L^2)\\ \bar{E}_0^\pi &= E_0^\pi - \Pi_2\Pi_3 (F_{L-H} F_{\bar{L}-\bar{H}})\\ &\qquad \times (F_{\bar{L}} F_{L-H}) E_H^\pi/(2D\tilde{\xi}_L^2)\\ \bar{E}_H^\pi &= E_H^\pi - \Pi_2\Pi_3 (F_L F_{\bar{L}})(F_L F_{\bar{L}-\bar{H}}) E_0^\pi/(2D\tilde{\xi}_L^2).\end{aligned} \tag{6}$$

In terms of these fields $\bar{E}$, and using the simplifications discussed above, the four equations (1) decouple completely, to second order:

$$\begin{aligned}&\{2\tilde{\xi}_0^\sigma[1 - (F_L F_{\bar{L}}/2D\tilde{\xi}_L^2)(\Pi_3^2 F_{L-H} F_{\bar{L}-\bar{H}} + \Pi_2^2 F_L F_{\bar{L}})]\\ &\quad + [\Pi_2^2 (F_L F_{\bar{L}})^2/D\tilde{\xi}_L^2](\tilde{\xi}_0^\sigma - \tilde{\xi}_H^\sigma)\\ &\quad + (F_L F_{\bar{L}}/2D\tilde{\xi}_L^2)(\Pi_2^2 f^\sigma - \Pi_2\Pi_3 f^\pi)\}\bar{E}_0^\sigma\\ &\quad + \tilde{F}_{\bar{H}\bar{L}}^\sigma \bar{E}_H^\sigma = 0,\\ &\tilde{F}_{HL}^\sigma \bar{E}_0^\sigma + \{2\tilde{\xi}_H^\sigma[1 - (F_{L-H} F_{\bar{L}-\bar{H}}/2D\tilde{\xi}_L^2)\\ &\quad \times (\Pi_3^2 F_{L-H} F_{\bar{L}-\bar{H}} + \Pi_2^2 F_L F_{\bar{L}})]\\ &\quad + [\Pi_3^2 (F_{L-H} F_{\bar{L}-\bar{H}})^2/D\tilde{\xi}_L^2](\tilde{\xi}_0^\sigma - \tilde{\xi}_H^\sigma)\\ &\quad + (F_{L-H} F_{\bar{L}-\bar{H}}/2D\tilde{\xi}_L^2)(\Pi_3^2 f^\sigma - \Pi_2\Pi_3 f^\pi)\}\bar{E}_H^\sigma = 0,\end{aligned} \tag{7a}$$

$$\begin{aligned}&\{2\tilde{\xi}_0^\pi[1 + (\Pi_2^2 F_L F_{\bar{L}}/2D\tilde{\xi}_L^2)(F_L F_{\bar{L}} + F_{L-H} F_{\bar{L}-\bar{H}})]\\ &\quad - [\Pi_2^2 (F_L F_{\bar{L}})^2/D\tilde{\xi}_L^2](\tilde{\xi}_0^\pi - \tilde{\xi}_H^\pi)\\ &\quad - [(F_L F_{\bar{L}})/2D\tilde{\xi}_L^2](\Pi_2^2 f^\sigma - \Pi_2\Pi_3 f^\pi)\}\bar{E}_0^\pi\\ &\quad + \tilde{F}_{\bar{H}\bar{L}}^\pi \bar{E}_H^\pi = 0,\\ &\tilde{F}_{HL}^\pi \bar{E}_0^\pi + \{2\tilde{\xi}_H^\pi[1 + (\Pi_3^2 F_{L-H} F_{\bar{L}-\bar{H}}/2D\tilde{\xi}_L^2)\\ &\quad \times (F_L F_{\bar{L}} + F_{L-H} F_{\bar{L}-\bar{H}})]\\ &\quad - [\Pi_3^2 (F_{L-H} F_{\bar{L}-\bar{H}})^2/D\tilde{\xi}_L^2](\tilde{\xi}_H^\pi - \tilde{\xi}_0^\pi)\\ &\quad - [(F_{L-H} F_{\bar{L}-\bar{H}})/2D\tilde{\xi}_L^2](\Pi_3^2 f^\sigma - \Pi_2\Pi_3 f^\pi)\}\\ &\quad \times \bar{E}_H^\pi = 0.\end{aligned} \tag{7b}$$

The two structure-factor-like coefficients in (7) are

$$\tilde{F}^{\sigma}_{HL}=F^{\sigma}_{HL}-(\Pi_2\Pi_3/2D\tilde{\xi}^2_L)F_LF_{\bar{L}-\bar{H}}f^{\pi},$$
$$\tilde{F}^{\pi}_{HL}=F^{\pi}_{HL}+(\Pi_2\Pi_3/2D\tilde{\xi}^2_L)F_LF_{\bar{L}-\bar{H}}f^{\sigma}, \tag{8}$$

where, in both (7) and (8), we have introduced the abbreviations

$$f^{\sigma}=F_{\bar{L}}F_{L-H}F^{\sigma}_{HL}+F_LF_{\bar{L}-\bar{H}}F^{\sigma}_{\bar{H}\bar{L}}$$
$$f^{\pi}=F_{\bar{L}}F_{L-H}F^{\pi}_{HL}+F_LF_{\bar{L}-\bar{H}}F^{\pi}_{\bar{H}\bar{L}}. \tag{9}$$

While this procedure has transformed the four equations of (1) into two sets of two, these still lack the familiar two-beam form. However, once it is recognized how this form is modified if the two-beam equations are written with respect to the wrong origin, a rotated coordinate system, and the wrong Bragg angle, a further simplification is possible.

For example, Fig. 1 shows a set of two-beam dispersion surface asymptotes for Bragg angle θ_B and crossing at the Lorentz point at the origin $(k_x, k_z)=(0,0)$, so that a tiepoint is defined by ξ_0, ξ_H. Let the Lorentz point be shifted to the location (Δ_x, Δ_z), the coordinate system be rotated counterclockwise by an amount $\frac{1}{2}(\chi_2-\chi_1)$ and the Bragg angle be altered by $\frac{1}{2}(\chi_2+\chi_1)$, as indicated in Fig. 1. If all shifts are small, then to first order in the shifts the same tiepoint is described by excitation errors $\bar{\xi}_0$, $\bar{\xi}_H$ relative to the new asymptotes that are related to the original set (ξ_0, ξ_H) by

$$\bar{\xi}_0=\xi_0(1-\tan\theta_B\chi_1)+(\chi_1/\sin 2\theta_B)(\xi_0-\xi_H)$$
$$+\cos\theta_B(\Delta_x-\cot\theta_B\Delta_z)$$
$$\bar{\xi}_H=\xi_H(1-\tan\theta_B\chi_2)+(\chi_2/\sin 2\theta_B)(\xi_H-\xi_0)$$
$$+\cos\theta_B(\Delta_x+\cot\theta_B\Delta_z). \tag{10}$$

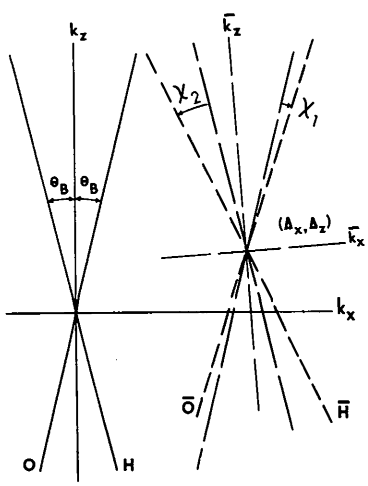

Fig. 1. Two-beam asymptotes **0**, **H** through the Lorentz point at $(k_x, k_z)=(0,0)$, and the corresponding asymptotes $\bar{\mathbf{0}}$, $\bar{\mathbf{H}}$ displaced to the Lorentz point (Δ_x, Δ_z), and rotated respectively by angles χ_1, χ_2 from the original Bragg angle θ_B. $\bar{\mathbf{0}}$, $\bar{\mathbf{H}}$ refer to a new coordinate system $\bar{k}_x$, $\bar{k}_z$ rotated counterclockwise by $\frac{1}{2}(\chi_2-\chi_1)$, and a new Bragg angle shifted by $\frac{1}{2}(\chi_2+\chi_1)$.

By inspection, (7*a*,*b*) have coefficients of nearly this form and they can be brought into this form exactly, to second order, by multiplying both equations by the appropriate factor that establishes the proper relation between the first and second correction terms in (10). This results in the following shifts:

$$\tfrac{1}{2}(\chi_2-\chi_1)=\pm(\sin 2\theta_B/4D\tilde{\xi}^2_L)$$
$$\times[\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2-\Pi_2^2(F_LF_{\bar{L}})^2], \tag{11a}$$

$$\tfrac{1}{2}(\chi_2+\chi_1)=\pm(\sin 2\theta_B/4D\tilde{\xi}^2_L)$$
$$\times[\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2+\Pi_2^2(F_LF_{\bar{L}})^2],$$
$$\Delta_x=\pm(1/8\cos\theta_BD\tilde{\xi}^2_L)$$
$$\times[(\Pi_3^2F_{L-H}F_{\bar{L}-\bar{H}}+\Pi_2^2F_LF_{\bar{L}})f^{\sigma}$$
$$-\Pi_2\Pi_3(F_{L-H}F_{\bar{L}-\bar{H}}+F_LF_{\bar{L}})f^{\pi}], \tag{11b}$$

$$\Delta_z=\pm(\tan\theta_B/8\cos\theta_BD\tilde{\xi}^2_L)$$
$$\times[(\Pi_3^2F_{L-H}F_{\bar{L}-\bar{H}}-\Pi_2^2F_LF_{\bar{L}})f^{\sigma}$$
$$-\Pi_2\Pi_3(F_{L-H}F_{\bar{L}-\bar{H}}-F_LF_{\bar{L}})f^{\pi}],$$

with the upper and lower signs applying to the σ- and π-like solutions, respectively.

Using (11) in (10), the explicit excitation errors with respect to the new coordinate system are, for σ-like modes,

$$\bar{\xi}^{\sigma}_0=\tilde{\xi}^{\sigma}_0[1-\sin^2\theta_B\Pi_2^2(F_LF_{\bar{L}})^2/D\tilde{\xi}^2_L]$$
$$+[\Pi_2^2(F_LF_{\bar{L}})^2/2D\tilde{\xi}^2_L](\tilde{\xi}^{\sigma}_0-\tilde{\xi}^{\sigma}_H)$$
$$+(F_LF_{\bar{L}}/4D\tilde{\xi}^2_L)(\Pi_2^2f^{\sigma}-\Pi_2\Pi_3f^{\pi})$$
$$\bar{\xi}^{\sigma}_H=\tilde{\xi}^{\sigma}_H[1-\sin^2\theta_B\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2/D\tilde{\xi}^2_L]$$
$$+[\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2/2D\tilde{\xi}^2_L](\tilde{\xi}^{\sigma}_H-\tilde{\xi}^{\sigma}_0)$$
$$+(F_{L-H}F_{\bar{L}-\bar{H}}/4D\tilde{\xi}^2_L)(\Pi_3^2f^{\sigma}-\Pi_2\Pi_3f^{\pi}) \tag{12}$$

and the corresponding π-like coordinates follow by replacing D by $-D$.

In terms of (12) we finally obtain the fully reduced two-beam equations

$$2\bar{\xi}^{\sigma}_0\bar{E}^{\sigma}_0+\bar{F}^{\sigma}_{\bar{H}\bar{L}}\bar{E}^{\sigma}_H=0,$$
$$\bar{F}^{\sigma}_{HL}\bar{E}^{\sigma}_0+2\bar{\xi}^{\sigma}_H\bar{E}^{\sigma}_H=0,$$
$$2\bar{\xi}^{\pi}_0\bar{E}^{\pi}_0+\bar{F}^{\pi}_{\bar{H}\bar{L}}\bar{E}^{\pi}_H=0,$$
$$\bar{F}^{\pi}_{HL}\bar{E}^{\pi}_0+2\bar{\xi}^{\pi}_H\bar{E}^{\pi}_H=0, \tag{13}$$

where the modified structure factors are

$$\bar{F}^{\sigma}_{HL} = \{1 + (P/4D\tilde{\xi}^2_L)[\Pi^2_3(F_{L-H}F_{\bar{L}-\bar{H}})^2 + \Pi^2_2(F_LF_{\bar{L}})^2] + [(\Pi^2_2 + \Pi^2_3)/4D\tilde{\xi}^2_L](F_LF_{\bar{L}})(F_{L-H}F_{\bar{L}-\bar{H}})\} \times [F^{\sigma}_{HL} - (\Pi_2\Pi_3/2D\tilde{\xi}^2_L)F_LF_{\bar{L}-\bar{H}}f^{\pi}],$$
$$\bar{F}^{\pi}_{HL} = \{1 - (P/4D\tilde{\xi}^2_L) \times [\Pi^2_3(F_{L-H}F_{\bar{L}-\bar{H}})^2 + \Pi^2_2(F_LF_{\bar{L}})^2] - [(\Pi^2_2 + \Pi^2_3)/4D\tilde{\xi}^2_L](F_LF_{\bar{L}})(F_{L-H}F_{\bar{L}-\bar{H}})\} \times [F^{\pi}_{HL} + (\Pi_2\Pi_3/2D\tilde{\xi}^2_L)F_LF_{\bar{L}-\bar{H}}f^{\sigma}]. \quad (14)$$

To obtain solutions of (13) we must also establish the relation between $\bar{\xi}_0$ and $\bar{\xi}_H$ imposed by the boundary conditions that all waves vary exponentially in amplitude along the surface normal. For the symmetric Bragg case, with which we will be concerned explicitly, this condition was expressed in (I-5), and the same equation must continue to hold because the actual surface has not been altered. For convenience it is repeated here:

$$\xi_0 + \xi_H = \delta = k\Gamma[-(\Delta\theta/\Gamma)\sin 2\theta_B + iF''_0]. \quad (15)$$

Applying the consecutive transformations $\xi \to \tilde{\xi} \to \bar{\xi}$, (15) takes the form, using the new (reduced) excitation errors,

$$\{1 + [(1+P)/2D\tilde{\xi}^2_L][\Pi^2_3(F_{L-H}F_{\bar{L}-\bar{H}})^2 - \Pi^2_2(F_LF_{\bar{L}})^2]\}\bar{\xi}_0 + \bar{\xi}_H = \bar{\delta} \quad (16)$$

where $\bar{\delta}$ is the new (reduced) relation to the angular scale, and includes a shift of the reference angle, given by

$$\bar{\delta} = (\delta/k\Gamma)[1 + (P/2D\tilde{\xi}^2_L)\Pi^2_3(F_{L-H}F_{\bar{L}-\bar{H}})^2 - (\Pi^2_2/2D\tilde{\xi}^2_L)(F_LF_{\bar{L}})^2] - (\Pi_1/4\tilde{\xi}_L)(F_LF_{\bar{L}} + F_{L-H}F_{\bar{L}-\bar{H}}) + (1/4D\tilde{\xi}^2_L)[(\Pi^2_3F_{L-H}F_{\bar{L}-\bar{H}} + \Pi^2_2F_LF_{\bar{L}})f^{\sigma} - \Pi_2\Pi_3(F_LF_{\bar{L}} + F_{L-H}F_{\bar{L}-\bar{H}})f^{\pi}]. \quad (17)$$

As they stand, (16) and (17) apply to the σ-like solutions. For the π-like solutions we let D change sign everywhere, and in addition replace the first-order correction in (17) by

$$-(1/4\tilde{\xi}_L)[\Pi_4F_LF_{\bar{L}} + \Pi_5F_{L-H}F_{\bar{L}-\bar{H}}].$$

The form of (16) labels it as describing an *unsymmetric* Bragg reflection. This is to be expected, since the coordinate system that diagonalizes (10) is rotated with respect to the actual surface. In fact, the rotation expressed in (16) is precisely that given by (11*a*).

These rotations, and the shifts of the Lorentz point contained in (17), as well as the change in Bragg angle in (14*a*), are all indicative of the transition of the Lorentz point from the two-beam to the three-beam case, which now includes not only translations but also rotations towards the three-beam symmetry. It is important to note that in this process (17) also includes a change of scale between $\bar{\delta}$ and the angular deviation $\Delta\theta$.

3. Second-order solution – fields and intensities

The successful diagonalization of (13) indicates that even in second order the modes remain characterized by σ or π polarization. Therefore we expect that the modes $(\bar{E}^{\sigma}_0, \bar{E}^{\sigma}_H)$ and $(\bar{E}^{\pi}_0, \bar{E}^{\pi}_H)$ are separately excitable. *Via* (I-12) they also give rise to accompanying L fields, as given below:

$$E^{\sigma}_L = -(P_x/2\tilde{\xi}_L)(F_L + F_{L-H}E^{\sigma}_H/E^{\sigma}_0)E^{\sigma}_0 + (P_{0x}/2\tilde{\xi}_L) \times (F_L - F_{L-H}E^{\pi}_H/E^{\pi}_0)E^{\pi}_0$$
$$E^{\pi}_L = (P_{xz}/2\tilde{\xi}_L)(F_L + F_{L-H}E^{\sigma}_H/E^{\sigma}_0)E^{\sigma}_0 - (1/2\tilde{\xi}_L) \times [(P_{0z} - P_{0xz})F_L + (P_{0z} + P_{0xz})F_{L-H}E^{\pi}_H/E^{\pi}_0]E^{\pi}_0. \quad (18)$$

Since to second order such L fields must also be included in the boundary conditions at the surface, the Bragg-Bragg case ($\theta_L > 0$) and the Bragg-Laue case ($\theta_L < 0$) require separate discussion.

In the Bragg-Bragg configuration, the L fields give rise to beams leaving the crystal in the $\mathbf{K}_L$ direction. No additional fields are needed to satisfy any boundary condition. In fact, of the three modes existing at any given angle of incidence, or $\bar{\delta}$, for each polarization, only one, namely that following from (13), is a mode characterized by normal absorption. The other two modes diverge towards the opposite surface of the crystal and are not excited.

In the Bragg-Laue configuration, however, the L fields (18) move into the crystal, and since no such fields exist above the surface, they must be accompanied by another L field in order that the net L fields vanish at the interface. Here, indeed, there exists another set of predominantly L fields, designated by $E^{\sigma}_L(L)$, $E^{\pi}_L(L)$, at each polarization, that shows normal absorption. In turn, these fields give rise to 0 and H fields. In the same approximation of large $\tilde{\xi}_L$, the latter can be derived from (I-12), and are

$$E^{\sigma}_0(L) = -[F_{\bar{L}}/2\tilde{\xi}_0(L)][P_xE^{\sigma}_L(L) - P_{xz}E^{\pi}_L(L)],$$
$$E^{\sigma}_H(L) = [F_{\bar{L}-\bar{H}}/2\tilde{\xi}_0(L)][P_xE^{\sigma}_L(L) - P_{xz}E^{\pi}_L(L)],$$
$$E^{\pi}_0(L) = -[F_{\bar{L}}/2\tilde{\xi}_0(L)][-P_{0x}E^{\sigma}_L(L) + (P_{0z} - P_{0xz})E^{\pi}_L(L)],$$
$$E^{\pi}_H(L) = [F_{\bar{L}-\bar{H}}/2\tilde{\xi}_0(L)][P_{0x}E^{\sigma}_L(L) + (P_{0z} + P_{0xz})E^{\pi}_L(L)], \quad (19)$$

with

$$\tilde{\xi}_0(L) \simeq -\tilde{\xi}_H(L) = (\sin\theta_B/\sin\theta_L)\tilde{\xi}_L.$$

For each exciting field $\bar{E}_0^\sigma$ or $\bar{E}_0^\pi$, $E_L^\sigma(L)$ and $E_L^\pi(L)$ can be chosen such as to cancel the fields (18) at the surface. According to (19), this then gives rise to additional field contributions in the $\mathbf{K}_0$ and $\mathbf{K}_H$ directions. With these corrections taken into account, the ratios of the net reflected to incident fields become, to order $1/\tilde{\xi}_L^2$,

$$\begin{aligned}
&E_H^\sigma(\text{net})/E_0^\sigma(\text{inc})\\
&\quad=[1+(\Pi_1 \sin\theta_L/4\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times(F_LF_{\bar{L}}+F_{L-H}F_{\bar{L}-\bar{H}})]\bar{E}_H^\sigma/\bar{E}_0^\sigma\\
&\qquad+(\Pi_1 \sin\theta_L/4\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times[F_LF_{\bar{L}-\bar{H}}+F_{\bar{L}}F_{L-H}(\bar{E}_H^\sigma/\bar{E}_0^\sigma)^2],\\
&E_H^\pi(\text{net})/E_0^\pi(\text{inc})\\
&\quad=[1+(\sin\theta_L/4\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times(\Pi_4F_LF_{\bar{L}}+\Pi_5F_{L-H}F_{\bar{L}-\bar{H}})]\bar{E}_H^\pi/\bar{E}_0^\pi\\
&\qquad+(\Pi_6 \sin\theta_L/4\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times[F_LF_{\bar{L}-\bar{H}}+F_{\bar{L}}F_{L-H}(\bar{E}_H^\pi/\bar{E}_0^\pi)^2].
\end{aligned}\tag{20}$$

The relative reflected intensities follow directly from (20). Since, in any integration, contributions proportional to odd powers of (E_H/E_0) average more or less to zero, we need only write down the terms that do contribute. Using (20), the reflected intensity in the Bragg–Laue case becomes

$$\begin{aligned}
&|E_H^\sigma(\text{net})/E_0^\sigma(\text{inc})|^2\\
&\quad=[1+\Pi_1 \sin\theta_L/2\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times\text{Re}\,(F_LF_{\bar{L}}+F_{L-H}F_{\bar{L}-\bar{H}})]|\bar{E}_H^\sigma/\bar{E}_0^\sigma|^2\\
&|E_H^\pi(\text{net})/E_0^\pi(\text{inc})|^2\\
&\quad=[1+\sin\theta_L/2\tilde{\xi}_L^2 \sin\theta_B)\\
&\qquad\times\text{Re}\,(\Pi_4F_LF_{\bar{L}}+\Pi_5F_{L-H}F_{\bar{L}-\bar{H}})]|\bar{E}_H^\pi/\bar{E}_0^\pi|^2.
\end{aligned}\tag{21}$$

Since there are no such corrections in the Bragg–Bragg configuration, in that case (21) applies without a prefactor. For L beams parallel to the surface, $\theta_L=0$, and the prefactor in (21) becomes unity, so that there is a continuous transition between the Bragg–Laue and Bragg–Bragg cases.

It must also be noted that if the starting Bragg reflection **0H** is not symmetric, then (15) is modified, and changes such as in (16) and (17) must be superimposed on the unsymmetric starting configuration. Similarly, the Bragg–Bragg to Bragg–Laue transition will then not necessarily take place when $\theta_L=0$. In such a situation, though, the boundary conditions and all results dependent on them include the azimuthal angle explicitly, and its changes may also have to be taken into account in other terms besides ξ_L.

4. Integrated intensities – strong and weak symmetric Bragg reflections

For many purposes, such as the description of Renninger scans, we are interested in the integrated intensity of an **0H** reflection and its modification by the perturbation of **L**. This modification is generally expected to differ for σ and π polarizations, for the Bragg–Bragg and the Bragg–Laue configurations, and for strong and weak two-beam reflections. We obtain here these various results for a reflection that in its unperturbed state is a symmetric Bragg reflection, in consonance with the boundary conditions in (15) through (17).

All integrated intensities are of the form

$$\int |[E_H(\text{net})/E_0(\text{inc})]|^2 \text{d}(\Delta\theta),$$

and here they can all be brought into a product of two prefactors and an integral:

$$\begin{aligned}
&(1+A/\tilde{\xi}_L^2)(1-B/\tilde{\xi}_L^2)\\
&\qquad\times\int |[\bar{E}_H(\text{net})/\bar{E}_0(\text{inc})]|^2 \text{d}(\Delta\bar{\theta}),
\end{aligned}\tag{22}$$

where A measures the Bragg–Laue correction, as in (21), and B takes into account the change of scale $\text{d}(\Delta\theta)/(\Delta\bar{\theta})$ that follows from (17),

$$B=\pm\text{Re}\,\{(1/2D)[P\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2-\Pi_2^2(F_LF_{\bar{L}})^2]\}.\tag{23}$$

The remaining integral in (22) has the standard form of a two-beam integrated intensity for an unsymmetric Bragg case, which is known for both strong and weak reflections (Afanas'ev & Perstnev, 1969).

A. *Strong reflections*

Here the standard integrated intensity is

$$\begin{aligned}
&(8\Gamma/3\sin 2\theta_B)\{|\gamma_H/\gamma_0|^{1/2}|F_H|\\
&\qquad-(3\pi/4)[(\gamma_0+|\gamma_H|)/2\gamma_0]|F_0''|\}
\end{aligned}\tag{24}$$

with γ_0, γ_H the direction cosines between $\mathbf{K}_0$ and $\mathbf{K}_H$ and the inner surface normal.

For the unsymmetric case described by Fig. 1 and (11), the angular factors in (24) are (primes stand for real parts)

$$\begin{aligned}
\gamma_0&=(1+\chi_1'\cot\theta_B)\sin\theta_B,\\
\gamma_H&=-(1+\chi_2'\cot\theta_B)\sin\theta_B,
\end{aligned}\tag{25}$$

$$\sin 2(\theta_B+\Delta\theta_B)=[1+(\chi_1'+\chi_2')\cot 2\theta_B]\sin 2\theta_B$$

so that (24) becomes

$$\begin{aligned}
&(8\Gamma/3\sin 2\theta_B)[1+\tfrac{1}{2}(\chi_2'-\chi_1')\cot\theta_B-(\chi_2'+\chi_1')\cot 2\theta_B]\\
&\qquad\times|F_H|[1-(3\pi/4)|F_0''/F_H'|].
\end{aligned}\tag{26}$$

By expressing the prefactor in (26) in terms of (11*a*), and combining with the second prefactor of (22), we finally obtain for the Bragg–Bragg case (*i.e.* $A=0$), the integrated intensities for the σ and π modes:

$$I^{\sigma}_{HL}(\mathrm{BB}) = (8\Gamma/3\sin 2\theta_B)\,\mathrm{Re}\,\{1-[(3P-1)/4D\tilde{\xi}^2_L]\times[\Pi_2^2(F_LF_{\bar{L}})^2+\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2]\}\times|\bar{F}^{\sigma}_{HL}|[1-(3\pi/4)|\bar{F}^{\sigma\prime\prime}_0/\bar{F}^{\sigma\prime}_{HL}|] \quad (27)$$

$$I^{\pi}_{HL}(\mathrm{BB}) = (8\Gamma/3\sin 2\theta_B)\,\mathrm{Re}\,\{1+[(3P-1)/4D\tilde{\xi}^2_L]\times[\Pi_2^2(F_LF_{\bar{L}})^2+\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2]\}\times|\bar{F}^{\pi}_{HL}|[1-(3\pi/4)|P\bar{F}^{\pi\prime\prime}_0/\bar{F}^{\pi\prime}_{HL}|].$$

The structure factors $\bar{F}^{\sigma}_{HL}$ and $\bar{F}^{\pi}_{HL}$ appearing in (27) are defined in (14), and $\bar{F}''_0$ is contained in the shift, to second order, in (17), in analogy with the appearance of F''_0 in (15).

The corresponding Bragg–Laue expressions are obtained, utilizing (22), by adding the prefactor containing A. With the two A's given in (21), we obtain

$$I^{\sigma}_{HL}(\mathrm{BL}) = [1+(\Pi_1\sin\theta_L/2\sin\theta_B\tilde{\xi}^2_L)\times\mathrm{Re}\,(F_LF_{\bar{L}}+F_{L-H}F_{\bar{L}-\bar{H}})]I^{\sigma}_{HL}(\mathrm{BB})$$

$$I^{\pi}_{HL}(\mathrm{BL}) = [1+(\sin\theta_L/2\sin\theta_B\tilde{\xi}^2_L)\times\mathrm{Re}\,(\Pi_4F_LF_{\bar{L}}+\Pi_5F_{L-H}F_{\bar{L}-\bar{H}})]I^{\pi}_{HL}(\mathrm{BB}). \quad (28)$$

B. Weak reflections

Here the standard integrated intensity is, to lowest order,

$$(\pi\Gamma/4\sin 2\theta_B)[2|\gamma_H|/(\gamma_0+|\gamma_H|)]|F_H|^2/|F''_0|. \quad (29)$$

Applying the same procedure as above to expand the angular factors, we find exactly the same prefactors as in (A). Hence we obtain immediately the modification of the weak reflection to second order for Bragg–Bragg:

$$I^{\sigma}_{HL}(\mathrm{BB}) = (\pi\Gamma/4\sin 2\theta_B)\{1-[(3P-1)/4D\tilde{\xi}^2_L]\times[\Pi_2^2(F_LF_{\bar{L}})^2+\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2]\}\times|\bar{F}^{\sigma}_{HL}|^2/|\bar{F}^{\sigma\prime\prime}_0|$$

and (30)

$$I^{\pi}_{HL}(\mathrm{BB}) = (\pi\Gamma/4\sin 2\theta_B)\,\mathrm{Re}\,\{1+[(3P-1)/4D\tilde{\xi}^2_L]\times[\Pi_2^2(F_LF_{\bar{L}})^2+\Pi_3^2(F_{L-H}F_{\bar{L}-\bar{H}})^2]\}\times|\bar{F}^{\pi}_{HL}|^2/|\bar{F}^{\pi\prime\prime}_0|.$$

The Bragg–Laue cases follow from (28).

5. Discussion

The outcome of all the consecutive mathematical manipulations in the above sections is a relatively simple conceptual understanding as to what happens in second order, and a set of compact expressions, (27) to (30), for describing the second-order corrections to the integrated intensity of a two-beam symmetric Bragg reflection.

Both are the result of systematically retaining also many higher-order terms in order to preserve the general two-beam formalism. This is accompanied, however, with considerable interior complexity, as exemplified in (14) for the effective structure factors. The usefulness of the results remains to be tested in particular instances. Detailed application of the theory to pertinent instances identified in recently completed studies of first-order effects (Juretschke, 1986*a*, *b*) will be presented shortly.

In addition, however, there remain problems of interpretation of some of these second-order results. In particular, as defined in (11), the rotations of the asymptotes and the change in Bragg angle are actually complex, and, similarly, (17) predicts that the effective average absorption coefficient $\bar{F}''_0$ depends on the angle of incidence $\Delta\theta$. Hence to the extent that the (usually small) imaginary contributions to these transformations are included, the representation of all results in terms of a modified two-beam framework, in which the effective average absorption coefficient is expected to be constant, begins to be breached. This variation of F''_0 with $\Delta\theta$ has been ignored in the results given in § 4, since it is outside the standard two-beam treatments.

It is worth noting that the second-order formulation presented here is built entirely on a perturbation solution of (1). It can therefore also be considered to be a first-order correction to the first-order theory, and is thus in the nature of a Wigner rather than a Schrödinger perturbation development. Using this point of view, it becomes clear from (5), for example, that the solution will not converge when the first-order structure factors F^{σ}_{HL} and F^{π}_{HL} become alike, so that it cannot be used to resolve the question of how the crossings of the dispersion surface edges in Fig. 7 of (I) are changed in a more exact treatment. At the same time, since it pinpoints the difficulty, this formulation may serve as a starting point for removing the degeneracy of the solutions of that figure. By the same argument, D in (5) will always be large in cases of anomalous asymmetry (Juretschke, 1986*a*) where F^{σ}_{HL} increases and F^{π}_{HL} decreases as **L** is neared, so that here the second-order treatment should be valid for exploring beyond the limits of first-order modifications.

Finally, one may also have to be concerned about the extent to which corrections of the size included here will be augmented by corrections of similar size arising from retaining some dynamical contributions to ξ_L. Since this is clearly outside any two-beam framework, it would require separate study. In the same spirit, it would also be worthwhile to try to connect the results obtained here with the much more intricate exact monumental three-beam treatment by Ewald & Héno (1968), whose consequences have remained largely unexplored.

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Direct Measurements of the Complex X-ray Atomic Scattering Factors for Elements by X-ray Interferometry at the Daresbury Synchrotron Radiation Source

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Abstract

A scanning X-ray interferometer system [Hart & Siddons (1981). *Proc. R. Soc. London Ser. A*, **376**, 465-482] has been rebuilt for operation at the SERC Daresbury Synchrotron Radiation Source (SRS). The SRS permits an increase in energy resolution by one decade and *simultaneously* an intensity gain of one thousand times, though in practice the solid-state detector employed limited the peak intensity utilized so that experiments which hitherto demanded one month of counting time are now performed at higher spectral resolution in 2-4 h. Absolute measurements are reported of f' and f'' for the K edges of most elements between ^{34}Se and ^{26}Fe and for L edges of ^{79}Au and ^{74}W over energy ranges of about ±2 keV near absorption edges and scans with better than 1 eV energy resolution of X-ray absorption near edge structure (XANES) and extended X-ray absorption fine structure (EXAFS) spectra near the edges. Over wide energy ranges the results are compared with the most recent calculations of Cromer & Liberman [*Acta Cryst.* (1981), **A37**, 267-268], which are now easily available to workers in the field.

Introduction

The two-beam scanning X-ray interferometer system which we described earlier (Hart & Siddons 1981) has now been rebuilt and is operating routinely at the Science and Engineering Research Council's Synchrotron Radiation Source (SRS) at Daresbury. A few minor changes were required to match the characteristics of the SRS; the interferometer now disperses radiation in the vertical plane. The mechanical response of the scanning system has been improved and the sequencing of the experiment has been altered to match the characteristics of the more intense source.

The interferometry experiment window on line 7 is 53 m from the effective X-ray source which is 12 mm wide and 300 μm high (Hart & Siddons 1982*a*). With a slit height of 300 μm, the angular divergence of radiation on the interferometer is about 2″ which

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