Oscillation Camera Data Processing: Reflecting Range and Prediction of Partiality.
2. Monochromatized Synchrotron X-radiation from a Singly Bent Triangular Monochromator

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Abstract
Monochromatized synchrotron X-radiation from a singly bent triangular perfect-crystal monochromator system, uniformly adapted at LURE, DORIS and the SRS, offers considerable improvement in exposure times for protein single-crystal oscillation photography with improved resolution of data and better signal to noise. There are important differences in the diffraction geometry with such a system compared to a conventional laboratory experimental arrangement. In particular, an asymmetric beam cross fire, a spectral bandwidth that is variable over a wide magnitude and a correlation between the direction of the incident ray and its photon wavelength. These new considerations considerably affect the data processing considerations in oscillation camera work since they affect the appropriate flagging of partial reflections. As in an earlier treatment for conventional sources [Greenhough & Helliwell (1982). J. Appl. Cryst. 15, 338-351], reflecting-range formulae are derived from first principles, leading to expressions from which partiality can be calculated. It is shown that the general equations for monochromatized synchrotron X-radiation reduce to those for conventional sources when the characteristics of the former are removed. The often observed difference between spot sizes on opposite sides of the oscillation film, under certain conditions in the synchrotron case considered, is explained. At the Guinier position the equations reduce to those for a conventional source with asymmetric beam cross fire [Greenhough & Helliwell (1982). J. Appl. Cryst. 15, 338–351] with spots comparably placed on either side of the film showing no size difference due to the incident radiation. In view of the interest in the possibility of polychromatic single-crystal studies [Arndt, Greenhough, Helliwell, Howard, Rule & Thompson (1982). Nature (London), In the press] the vertical-rotation-axis equations are also derived. In both cases large differences in reflecting range, when working away from the Guinier position, are predicted for reflections identically placed except for the sense of horizontal displacement, with the elongated spots containing variable-wavelength information along their length. In order to find suitable values for use in the diffraction equations, the properties of the monochromator are discussed; this also leads to the conclusion that the full sampling of absorption-edge fine structure is feasible as demonstrated by Helliwell et al. [Acta Cryst. (1981). A37, C316].

Introduction
Synchrotron X-radiation sources are having a considerable impact on protein crystallography, giving intense tuneable collimated X-ray beams for single-crystal diffraction experiments. To provide the highest possible intensity at the sample, optical systems have been constructed at LURE (Lemonnier, Fourme, Rousseaux & Kahn, 1978; Kahn, Fourme, Gadet, Janin & Andre, 1982), DORIS (Hendrix, Koch & Bordas, 1979) and the Daresbury SRS (Helliwell et al., 1981) based on singly bent triangular perfect-crystal monochromators. At DORIS vertical focusing is provided by a segmented quartz mirror system and at the SRS a single bent 60 cm platinum-coated quartz mirror is to perform a similar function. It is not considered relevant in this work to discuss the virtues of this type of optical system, instead we concentrate on the properties of such a system which are pertinent to the diffraction process and the single-crystal oscillation method.

1. The monochromator
The effect of the oblique cut in producing a compression effect on the width of the reflected beam from
the monochromator has been described by Lemonnier et al. (1978), Kahn et al. (1982), Hendrix, Koch & Bordas (1979) and Helliwell et al. (1981). By bending the monochromator to the arc of a circle of curvature radius \( R \) it is possible to image the horizontal source width \( h \) at the sample. The size of this image is a convolution of three contributions, the source-size effect, the crystal rocking width and the imperfection in the Rowland focusing geometry producing a finite image from a point source. Lemonnier et al. (1978) have demonstrated that the image size is dominated by the source-size effect at LURE; the same is true of the SRS and DORIS and the width is given by \( hp'/p \) where \( p' \) is the focal distance.

The spread of wavelengths in the reflected beam, \( (\delta \lambda/\lambda)_{TOT} \), is given by a convolution of the crystal rocking-curve contribution with the geometrical factors of the source size and the variation of the angle of incidence along the curved monochromator surface (Lemonnier et al., 1978). The acceptance width of the monochromator is given by

\[
\omega_{acc} = \omega_{sym} \left[ \frac{\sin(\theta - \alpha)}{\sin(\theta + \alpha)} \right]^{1/2},
\]

where \( \alpha \) is the oblique cut and \( \theta \) the monochromator Bragg angle (Lemonnier et al., 1978). For a perfect triangular Ge(111) crystal used on the Daresbury SRS protein crystallography work station, \( \omega_{sym} \) at 1.5 Å is \( \approx 16' \), and for a similar Si(111) crystal \( \omega_{sym} \approx 9' \) (Clarke, Gani, Helliwell & Tanner, 1981). The angular width of the diffracted beam is

\[
\omega_{diff} = \omega_{acc} \cot \theta = \omega_{sym} \left[ \frac{\sin i}{\sin r} \right]^{1/2},
\]

where \( i = (\theta + \alpha) \) and \( r = (\theta - \alpha) \) (Lemonnier et al., 1978).

At the focus, the narrow crystal-rocking-width contribution to \( (\delta \lambda/\lambda)_{TOT} \) is \( \omega_{acc} \cot \theta \), spread over a distance \( \omega_{diff} p' \), which is greater than \( \omega_{sym} p' \). The source-size contribution to \( (\delta \lambda/\lambda)_{TOT} \) is \( \delta \theta \cot \theta \) where \( \delta \theta = h/p \), and the monochromator curvature contribution is

\[
L \left| \frac{\sin r}{2} - \frac{\sin i}{p} \right| \cot \theta,
\]

where \( L \) is the monochromator length. Thus we have

\[
(\frac{\delta \lambda}{\lambda})_{TOT} = \left\{ \left( \frac{h}{p} + \frac{L \sin r}{2 p'} - \frac{\sin i}{p} \right)^2 + \omega_{acc}^2 \right\}^{1/2} \cot \theta
\]

(Lemonnier et al., 1978).

However, \( (\delta \lambda/\lambda)_{TOT} \) is not what the sample receives if the sample size is less than the focus width. In particular, there is a photon energy gradient across the focus due to the source-size contribution as shown in Fig. 1, where the effects of \( \omega_{diff} \), focusing aberration and error in selecting the Guinier position, all considered in detail below, are not represented. The total energy spread due to the finite source is \( h/p \cot \theta \), over the total focus width \( hp'/p \); a sample or slit of size \( x_H \) at the focus thus receives \( (x_H/hp'/p)(h/p)\cot \theta \), i.e. a spread of \( (x_H/hp')\cot \theta \). Hence, at the Guinier position (Fig. 1) \( (p' = p' = p \sin r/\sin i) \) a narrow slit \( (\ll hp'/p) \) can be used to provide a fine spectral resolution beam incident on the sample \( (\sim 5 \times 10^{-4}, \delta \lambda/\lambda) \). This is important for the full sampling of absorption-edge fine structure (e.g. white line) in optimized anomalous dispersion studies. This application of bent-crystal monochromators by Helliwell et al. (1981), using a cobalt metal foil (K edge) and a Dy_2O_3 powder (Lm edge), had not previously been reported.

Away from the Guinier condition when the curvature contribution to \( (\delta \lambda/\lambda)_{TOT} \) is significant, the direction of incidence of photons onto the sample is correlated with their energy (Fig. 2); the same is true for the neutron source case (Schoenborn, 1981). Again by introducing a narrow slit \( (\ll hp'/p) \) and by placing an Fe foil directly behind and a photographic film some distance away, Helliwell et al. (1981) produced streaks displaying the K absorption profile across their length (\( \approx 35 \) eV total, monochromator bending radius \( \approx 40 \) m). By considerably overbending a sym-
metric perfect crystal \((\alpha = 0\), bending radius \(\sim 1\) m), previous workers have obtained a bandwidth of \(\sim 1\) keV, useful for performing dispersive EXAFS (Matsushita, 1980; Frank, Fontaine, Lagarde, Lemonnier, Mimault, Radux & Sadoc, 1981). With respect to the Guinier position it is possible to 'underbend' the crystal (i.e. \(p' > p \sin r / \sin i\)), which in the limit of a flat crystal \((p' = -p)\), not considered here, with \(\alpha = 0\) gives an energy spread determined by the source divergence angle. In both the overbend and underbend cases, in addition to the correlation between energy and direction at the sample, there remains a variation of energy across the focus due to the finite source size.

The effects of \(\omega_{\text{diff}}\) and focusing aberration, along with the precision to which the Guinier position is known are now considered as they determine the photon energy resolution of each ray incident at the sample. Detailed experimental results are given in Helliwell et al. (1981).

**Photon energy resolution at any point within the focus width**

(A) Effect of the rocking width of the monochromator. In Fig. 3 the effect of the perfect-crystal rocking width is illustrated for an oblique cut and a finite polychromatic source. If we consider an incoming ray to the monochromator at \(\omega_{\text{acc}}\) about \(CY\) then the acceptance angular spread \(\omega_{\text{acc}}\) about \(CY\) produces an outgoing ray \(CY'\) with a broader emergent angular spread \(\omega_{\text{diff}}\) about \(CY'\). The wavelength spread across this outgoing fan of rays is \(\omega_{\text{sym}}\cot \theta\) (Lemonnier et al., 1978), but even for a Ge crystal where \(\omega_{\text{sym}}\) is quite large \((\sim 16°,\) Clarke, Gani, Helliwell & Tanner, 1981), this wavelength spread is extremely small. The incoming rays \(PC\) and \(QC\) at \(\omega_{\text{diff}}/2\) and \(-\omega_{\text{diff}}/2\) from \(CY\), respectively, also have acceptance angular spreads, \(\omega_{\text{acc}}\), producing emergent angular spreads \(\omega_{\text{diff}}\) about \(CP\) and \(CQ\), respectively, where \(P'CY' = O'C'Y' = P'CY = QCY = \omega_{\text{diff}}/2\). Hence, treating the bundles of rays centred on \(CP\), \(CY'\), \(CQ\) as each being essentially monochromatic in themselves, but with \(CP\) to \(CY'\) and \(CQ\) to \(CQ'\) separated in absolute wavelength by \(\sim (\omega_{\text{diff}}/2)\cot \theta\), the range of wavelengths passing along \(CY'\) is then \(\sim \omega_{\text{diff}}\cot \theta\), which defines the energy resolution along each ray. With \(h\) of the order of a few mm or more, this only remains true if \(CP\) or \(CQ\), traced back to the source, intercept the finite source size \(h\). For the Ge(111) 10.44° case where \(\omega_{\text{diff}} \sim 40°\) (Co K edge) then \(\omega_{\text{diff}} = 4\) mm. Since \(h_{\text{cule}} = 13.7\) mm (FWHM; \(h_{\text{meas}} = 9.87 \pm 0.28\) mm), for rays emanating from the edge of the source, \(CP\) (or \(CQ\)) will fall outside this FWHM range and be weak in flux. The energy resolution should therefore improve towards the edges of the focus, as one would expect since the total wavelength spread across the whole focus is given by \((\delta\lambda/\lambda)_{\text{TOT}}\) with \(\omega_{\text{acc}}\) and not \(\omega_{\text{diff}}\) the relevant angle (Kohra, Ando, Matsushita & Hashizume, 1978).

Since

\[
\omega_{\text{diff}} = \omega_{\text{sym}} \left[ \frac{\sin i}{\sin r} \right]^{1/2},
\]

then \(\delta E/E = \omega_{\text{diff}}\cot \theta\) would be improved by using a symmetric-cut crystal \((i = r)\) but in this case \(p_{\text{c}} = p\) and the focal width is too large \((9.87\) mm in this case) to be useful for protein samples. All other things being equal, Si(111) improves the \(\delta E/E\) resolution, because of its narrower rocking width, compared to Ge(111), by about \(\frac{1}{3}\) at the expense of flux. At 1.743 Å (Fe K edge), \(\delta E = 5.1\) eV for the Ge(111) crystal \([\alpha = 10.44°,\) \((\delta E/E)_{\text{sym}} = 3.2 \times 10^{-4}\)], and \(\delta E = 1.9\) eV for Si(111) \([\alpha = 10.29°,\) \((\delta E/E)_{\text{sym}} = 1.2 \times 10^{-4}\)]. (Here we assume that \(\omega_{\text{sym}}\) measured for Cu K\(\alpha\) is applicable to the slightly larger Fe K\(\alpha\) wavelength.)

(B) Focusing aberration. In Fig. 4 we consider the Rowland-circle mounting taking account of the oblique cut, \(\alpha\). Rays reflected by the monochromator from a source lying on a circle of radius \(R/2\), where

\[
\frac{2}{R} = \frac{\sin(\theta - \alpha)}{p'} + \frac{\sin(\theta + \alpha)}{p},
\]

form tangents to a circle (the caustic circle) of radius

![Fig. 3. Effect of rocking width on point-to-point energy resolution.](image)

![Fig. 4. Focusing aberration (CF) and depth of focus (\(\sim AB\)) for Rowland-circle mount.](image)
R \cos(\theta - \alpha). At the focus, FC represents the focusing aberration and is given by

$$FC \approx \frac{\rho L^2}{8R^2}$$

(Martin & Cacak, 1976), and since

$$\rho = R \cos(\theta - \alpha)$$

we get

$$FC \approx \frac{L^2 \cos(\theta - \alpha)}{8R}.$$ 

For \(L = 200 \text{ mm}, \) with Ge(111) (\(\alpha = 10.44^\circ\)) and at the Guinier condition (\(R = 50 \text{ mm}\) for \(\lambda = 1.60811 \text{ Å} \) (Co K edge), \(FC \approx 0.1 \text{ mm}\). This value can be reduced by reducing \(L\) so that with \(L = 100 \text{ mm}\), \(FC \approx 25 \mu \text{ m}\) and can be considered negligible compared with \(\omega_{\text{diff}}\rho'\) (0.65 mm for \(p_G\)), the crystal rocking-width contribution to the spread. If \(L = 200 \text{ mm}\) then we must convolute \(\omega_{\text{diff}}\) with \(FC/p'.\) Even if \(FC\) is as large as 0.1 mm then \(\delta E = \frac{\omega_{\text{diff}}}{p'} + \frac{(FC/p')^2}{2} \approx 40.5^\circ\) instead of 40°. For Co, \(h_{\text{Ge}}/p' = 2 \text{ mm FWHM (from } h_{\text{calc}}\text{)}, energy width \(20 \text{ eV}; 0.65 \text{ mm is equivalent therefore to } 6.5 \text{ eV and compares with the } 5.1 \text{ eV calculated previously (vide infra).}

Both the rocking width and focusing aberration limit the energy resolution in the over/underbend cases and at the Guinier condition.

### (C) Errors in the Guinier position

When setting the Guinier condition an error in the value of the oblique cut \(\alpha\) can cause an error in \(p_G\). We have

$$p_G = \frac{\sin(\theta - \alpha)}{\sin(\theta + \alpha)}p,$$

hence

$$\delta p'_G = \frac{-p \sin 20 \delta \alpha}{\sin^2(\theta + \alpha)}.$$

Such an error in \(p'_G\) will cause a contribution to \(\delta \lambda / \lambda\) by the curvature component

$$\frac{L}{2} \left[ \frac{\sin(\theta - \alpha)}{p'} - \frac{\sin(\theta + \alpha)}{p} \right] \cot \theta,$$

giving an increase from zero at \(p'\) to

$$\delta \left(\frac{\delta \lambda}{\lambda}\right) = \frac{L}{2} \left[ \frac{\cos(\theta - \alpha)}{p'} + \frac{\cos(\theta + \alpha)}{p} \right] \delta \alpha.$$

Table 1 shows the effect of an error \(\delta \alpha\) if we had assumed that \(\alpha = 10.5^\circ\) (exactly) and \(p_G = 2507 \text{ mm [Ge(111), 1.5 Å]. Measurement of four pairs of perfect Si and Ge crystals with different angles of cut has revealed differences of up to } 0.25^\circ\) between the nominal and measured \(\alpha\) values (Helliwell, 1981). The errors in measuring \(\alpha\) were estimated at \(\pm 0.05^\circ\). For \(\delta \alpha = 0.05^\circ, \delta p'_G = 50 \text{ mm and } \delta (\delta \lambda / \lambda) \text{ is } 1 \text{ eV, compared with } \delta p'_G = 250 \text{ mm and } \delta (\delta \lambda / \lambda) = 6 \text{ eV for } \delta \alpha = 0.25^\circ.\)

### (D) Summary

What is the best practical energy resolution at a point within the focus width? At \(p_G, \) Fe K\(\alpha\) wavelength with a 10.5° cut Si(111) monochromator, then \(\delta E\) summed over the \(\omega_{\text{diff}}\cot \theta\) (1.9 eV), \(\delta \alpha = 0.05^\circ\) (1 eV) and depth-of-focus (3 eV) terms for \(L = 200 \text{ mm}\) gives 5.9 eV; for \(L = 100 \text{ mm}\) we get 3.9 eV and for Ge(111) we get 9.1 and 7.1 eV respectively.

For the over/underbend cases, \(\delta \alpha\) and the depth of focus affect only the total \(\delta \lambda / \lambda\) achievable, so if \(\delta (\delta \lambda / \lambda)_{\text{TOT}} = 35 \text{ eV then the effect of these terms is to increase or decrease this by } \delta E\) or so. With the focusing aberration contribution considered negligible we are left only with \(\omega_{\text{diff}}\cot \theta.\)

Resolutions with \(\delta E / E \leq 5 \times 10^{-4}\) are in fact perfectly adequate to explore X-ray absorption edges for optimized anomalous dispersion studies. The monochromator can be set to explore absorption edges or not depending on the problem in hand, and the total \(\delta \lambda / \lambda\) across the focus should not be confused with what the (smaller) sample sees.

### The diffraction model

Consider the general case of a sample of horizontal size \(x_{ph} (\approx p')\) bashed in the focused beam from the monochromator not at the Guinier position (Fig. 5).

In the horizontal direction the sample receives rays
converging at an angle $\beta_H \approx (L/p') \sin \Gamma$ with a range of energies, due to the curvature, correlated with the direction of incidence. The total range of energies is known to an accuracy limited by the knowledge of $x$ and the depth of focus, but the correlation direction/energy is maintained. On each and every ray, however, is a spread of energies $\delta E/E \approx \omega_{\text{diff}} \cot \theta$; this is akin to the $K\alpha_1-K\alpha_2$ splitting with a conventional source dealt with in an earlier paper (Greenhough & Helliwell, 1982a). Even though each point in the focus receives a convergence angle $\beta_H$, there is a slight progressive change in the mean direction of incidence from one side of the focus to the other. In the case of the sample at the focus, which has the advantage of maximizing the flux intercepted by the sample (Helliwell et al., 1981), this is indicated by $\gamma_H$ where

$$\beta_H p' \approx \gamma_H (p' - z)$$

and

$$z \approx x_H/\gamma_H,$$

hence

$$\gamma_H \approx \beta_H + \frac{x_H}{p'}.$$

The finite size of sample also accepts only a fraction of the energy gradient across the focus, with the sample seeing $\delta \theta \approx x_H/p'$, giving

$$\left( \frac{\delta \lambda}{\lambda_{\text{loc}}} \right) \approx \frac{x_H}{p'} \cot \theta.$$  

If absorption-edge fine structure is of interest then $x_H$ may be artificially limited by using a slit. Thus, as we move horizontally across the sample, a set of incoming rays converge at each point at an angle $\beta_H$, each ray carrying a spread $\omega_{\text{diff}} \cot \theta$, but with a change in energy with direction due to the curvature component, with a slight rotation of $\beta_H$ and a change in mean energy (due to the gradient across the focus) from point to point. This is shown in Figs. 6(a), (b) and (c) for overbent, Guinier and underbent conditions, respectively, where the means of the extreme rays of three convergent fans are shown. The energy gradient at the focus maintains the same direction and in each case

$$\lambda_2 - \lambda_1 \approx \lambda_m \frac{x_H}{p'} \cot \theta$$

and with $\lambda_2 + \lambda_1 \approx 2\lambda_m$ we get

$$\lambda_2 - \lambda_1 \approx \lambda_m \frac{x_H}{2p'} \cot \theta.$$  

In terms of the diffraction process, transferring all rays to intercept the origin of reciprocal space as in Figs. 7(a), (b) and (c), the incoming rays at the origin have a total convergence angle $\gamma_H$. In order to clarify which wavelengths impinge in which directions a plot of incoming angle versus wavelength for each fan may be constructed. These are shown in Figs. 8(a), (b) and (c). Since there are actually rays impinging at $\pm \gamma_H/2$, and since $\beta_H$ is only marginally less than $\gamma_H$, in order to derive a practical model for the diffraction process the shaded regions containing no incoming rays are assumed to do so, thus giving the plots the form of a parallelogram. About each incoming ray we then have a spread of

$$\left( \frac{\delta \lambda}{\lambda_{\text{loc}}} \right) + \left( \frac{\delta \lambda}{\lambda_{\text{loc}}} \right) \approx \frac{x_H}{p'} + \omega_{\text{diff}} \cot \theta.$$  

For the overbent case (Fig. 8a) this spread is about a mean varying from $\lambda_2$ to $\lambda_1$. 

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![Fig. 5. Horizontal beam cross fire at the focus showing the effect of the finite sample size $x_H$.](image_url)
\[ \lambda_m - \frac{(\delta \lambda)c}{\beta H} \frac{x_H}{2p'} = \lambda_{\min} \text{ at } + \frac{\gamma_H}{2} \]

to

\[ \lambda_m + (\delta \lambda)c + \frac{(\delta \lambda)c}{\beta H} \frac{x_H}{2p'} = \lambda_{\max} \text{ at } - \frac{\gamma_H}{2} \]

(from Fig. 8a), giving a spread

\[ \delta \lambda = \frac{\gamma_H}{\beta H} (\delta \lambda)c; \]

therefore we have a direction-correlated term

\[ \left( \frac{\delta \lambda}{\lambda} \right)_{corr} \approx \frac{\gamma_H}{\beta H} \left( \frac{\delta \lambda}{\lambda} \right)_c \]

and a conventional-source-type term

\[ \left( \frac{\delta \lambda}{\lambda} \right)_{conv} \approx \left( \frac{x_H}{p'} + \omega_{diff} \right) \cot \theta. \]

At the Guinier position (Fig. 8b) completion of the rectangle simply gives

\[ \left( \frac{\delta \lambda}{\lambda} \right)_{corr} = 0 \]

with the conventional term as before, and in the underbent case (Fig. 8c) the correlated component becomes as for the overbent case.

In each case \((\delta \lambda/\lambda)_c\) is the spread due to the curvature component + depth of focus, and with the depth-of-focus term assumed negligible and with \(\gamma\) precisely known we are simply left with the modified monochromator curvature component. In order to include the degree of bending in the diffraction model the modulus signs will be removed from the \((\delta \lambda/\lambda)_c\) term (vide infra) making \((\delta \lambda)_c\) positive for the overbent case and negative for the underbent case. Thus in the SRS configuration we always have the correlated component of wavelength \(\lambda - (\delta \lambda/2)_{corr}\) impinging at

Fig. 7. Transfer of the rays in Fig. 6 to meet at the origin of reciprocal space \(O\) for (a) overbend, (b) Guinier position and (c) underbend. Use of the same symbols in each diagram is not intended to denote equality.

Fig. 8. Plot of wavelength \(\nu\) versus incoming angle to the origin of reciprocal space for (a) overbend, (b) Guinier position and (c) underbend. \((\delta \lambda/\lambda)_{corr} = (x_H/p')\cot \theta; (\delta \lambda/\lambda)_{R} = \omega_{diff}\cot \theta. \delta \lambda_p\) is the curvature component of the spectral spread. \(\delta \lambda_{Roc}\) and \(\delta \lambda_{Rk}\) are grossly enlarged with respect to \(\delta \lambda_c\). Use of the same symbols in each diagram is not intended to denote equality.
Table 2. Beam cross-fire angles and contributions to the spectral dispersion for a sample 0.3 x 0.3 mm at the focus, for \( \lambda = 1.5 \) Å and Ge(111) monochromator (\( \alpha = 10.37^\circ \)) focusing at various distances.

The monochromator length \( L \) is 200 mm accepting 4 mrad of the SR horizontal divergence, the vertical SR divergence is 0.25 mrad from a source height 0.43 mm, and \( p \) is 20.9 m. A precisely known \( \alpha \) and zero depth of focus are assumed. \( \theta = 13.27^\circ \).

<table>
<thead>
<tr>
<th>( p'(m) )</th>
<th>( \frac{\delta \lambda}{\lambda}_{\text{corr}} )</th>
<th>( \omega_{\text{diff}} \cot \theta )</th>
<th>( x/u \cot \theta )</th>
<th>( L \sin r )</th>
<th>( x_H )</th>
<th>( \gamma_H = \frac{x_v + h_v}{p' + p'} )</th>
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\( \omega_{\text{diff}} = \omega_{\text{symm}} \left( \frac{\sin i}{\sin r} \right)^{1/2} = 45^\circ = 0.22 \) mrad (\( \alpha_{\text{symm}} = 16^\circ \)).

\[
\frac{\delta \lambda}{\lambda}_{\text{conv}} \approx \frac{\delta \lambda}{\lambda}_{\text{corr}} \cot \theta + \frac{x_H}{p'} \cot \theta, \quad \frac{\delta \lambda}{\lambda}_{\text{corr}} \approx \frac{\gamma_H}{\beta_H} \cot \theta \frac{L}{2} \left( \frac{\sin r}{p'} - \frac{\sin i}{p} \right).
\]

\( \gamma_H \approx L \sin r \frac{\sin r}{p'} - \frac{x_H}{p'}; \beta_H \approx (L \sin r)/p'. \)

With a vertical focusing mirror 11 m from the source, at the Guinier position we get \( \gamma_v = 11.0/12.466 \approx 0.22 \) mrad.

\[ + \gamma_H/2 + \lambda + (\delta \lambda/2)_{\text{corr}} \text{ at } -\gamma_H/2 \text{ where } + \gamma_H/2 \text{ is on the left of the central ray viewed along the direction of this ray. In terms of the Ewald sphere the direction-correlated wavelengths in the horizontal plane give rise to Ewald spheres of radius 1 along the central ray (\( d^* = \lambda/d \)), of radius \( \lambda/\left( \lambda - (\gamma_H/\beta_H) \delta \lambda/2 \right) \text{ at } +\gamma_H/2 \) (left) and \( \lambda/\left( \lambda + (\gamma_H/\beta_H) \delta \lambda/2 \right) \text{ at } -\gamma_H/2 \) (right). The sense of left and right needs to be reversed in the alternative source–monochromator–sample configuration, as at DORIS and LURE. Thus we have a direction correlated component \( \frac{\delta \lambda}{\lambda}_{\text{corr}} \approx \left( \frac{x_H}{p'} \cot \theta + \frac{x_H}{p'} \right) \cot \theta \) with each ray having a spread of \( (\delta \lambda/\lambda)_{\text{conv}} \) in the horizontal plane.

In the absence of a focusing mirror the angular spread in the vertical direction is determined by the SR divergence \( \alpha_v \) and the source height \( h_v \) (0.43 mm FWHM calculated at the Daresbury SRS). The total divergence at the focus is thus \( \alpha_v \) spread over a distance \( F_v \approx \alpha_v (p + p') - h_v \) with a sample (or slit) of height \( x_v \) at the focus, the accepted divergence is then

\[ \gamma_v \approx (x_v + h_v)/(p + p') \]

giving approximately 6.5" or 0.03 mrad for a 0.3 mm sample at the Daresbury SRS. The introduction of a focusing mirror with 1:1 focusing will reproduce \( \alpha_v \) at each point of the focus in the vertical plane giving

\[ \gamma_v \approx \alpha_v, \]

which is 0.25 mrad at the Daresbury SRS, around 0.014°. This quantity will vary dependent on the type of vertical focusing used.

For the purposes of the diffraction model we assume no correlation between wavelength and direction in the vertical sense, such that any ray impinging on the sample will have the same mean wavelength as the ray it coincides with when projected to the horizontal plane.

Tables 2 and 3 take the case of a Ge(111) monochromator, 200 mm long as used at the Daresbury SRS as an example, giving the magnitudes of the various contributions to \( (\delta \lambda/\lambda)_{\text{conv}} \) and \( (\delta \lambda/\lambda)_{\text{corr}} \), along with \( \gamma_H \) and \( \gamma_v \). Table 2 is for a fixed wavelength \( (1.5/\lambda) \) but variable focusing distance \( p' \), with Table 3 having fixed \( p' \) and variable wavelength. The Guinier position at 1.5 Å is \( \approx 2.566 \text{ m} \). The more complicated geometry away from the Guinier position suggests that data is best collected at the Guinier position when possible, assuming of course that \( \alpha \) has been determined accurately since errors in \( p' \) will otherwise occur. Kahn et al. (1982) also suggest that data be collected at the Guinier position to minimize diffraction spot sizes (to improve unit-cell resolution). However, experiments to date requiring different wavelengths have usually been performed away from the Guinier position; there are several reasons for this as follows.

The possibility of accessing 'white lines' in absorption edges has been demonstrated by Helliwell et al. (1981) with the monochromator at the Guinier...
position; data has been obtained previously only near to and not at the absorption edge, with the monochromator usually set at overbend giving a large $\delta \lambda/\lambda$, with no attempt being made to use the absorption-edge fine structure. Alternatively, data is collected at short wavelengths to reduce absorption effects in high-angle reflections or at longer wavelengths to increase scattering power for small samples when absorption is not a problem. In these instances the Guinier position may not be accessible for the angle of cut and optical bench lengths used. At the Daresbury SRS a range of crystals with different angles of cut are available.

It is clear therefore that a treatment of the diffraction geometry in the general case is required. A considerable amount of oscillation film data has and will be collected. It is likely that much of this data has been or will be collected away from the Guinier position, and unless due allowance is made for the actual geometry used then systematic errors in the flagging of fully and partially recorded reflections will occur. As we shall see the direction-correlated component $(\delta \lambda/\lambda)_{\text{corr}}$ gives rise to widely varying reflecting ranges over a film, and the asymmetric cross fire produces results very different from those where a symmetrical approximation is used even at the Guinier position. In the case of virus photographs where the calculated degree of partiality is used initially and then refined, it is important for a sensible and practical model to be established for a meaningful refinement to be made.

II. Reflecting range and calculation of partiality

In § I the correlated and conventional components of the spectral spread have been derived. In the conventional-source case we have shown that symmetric spectral dispersion can simply be added to the sample mosaic spread in the form

$$L\left(\eta d^* \cos \theta + \frac{\delta \lambda}{\lambda} d^* \sin \theta\right)$$

to give the reflecting range in the absence of beam cross fire, and also that each of the effects in turn can be reproduced by a suitable reciprocal-lattice volume element (Greenhough & Helliwell, 1982a), interacting with the remaining effects. Rather than combining sample mosaic spread and $(\delta \lambda/\lambda)_{\text{corr}}$ to begin with, the reflecting range due to the cross fire $(\delta \lambda/\lambda)_{\text{corr}}$ is first found, this is then modelled by a suitable reciprocal-lattice volume element whose interaction with $(\delta \lambda/\lambda)_{\text{corr}}$, extending the reflecting range, is then investigated. Since the sense of left and right is of importance, the conventions outlined in § I are used with a signed $(\delta \lambda/\lambda)_{\text{corr}}$. The horizontal form of the nest of origins of Ewald spheres present is evident from Fig. 8, bearing in mind that distances to the reciprocal-lattice origin are given by $1/\lambda$. The derivations are carried out with figures suggesting an overbent monochromator, i.e. with $(\delta \lambda/\lambda)_{\text{corr}}$ positive; the results apply equally well to all positions. Thus from Fig. 8(a), representing the origin of reciprocal space as a distance unity from the central mean $\lambda$, we extend this figure to show the nest of Ewald-sphere origins as in Fig. 9.

Horizontal rotation axis

In order to find the reflecting range of a general reciprocal-lattice point, we must first model the correlated spectral dispersion and beam cross fire in terms of a geometric surface about the centre of the Ewald sphere $C$. Letting $C$ represent the centre of the unit-radius Ewald sphere corresponding to $\lambda$ and $d^*$ (Greenhough & Helliwell, 1982a) with zero relative divergence, the horizontal extremes of the surface will then be defined by points $1 + \delta$ and $1 - \delta$ from the origin of reciprocal space, with divergence angles $+\gamma/2$ and $-\gamma/2$ respectively from the central reference ray as shown in Fig. 10. The value of $\delta$ is $\sim 1/2(\delta \lambda/\lambda)_{\text{corr}}$ (Greenhough & Helliwell, 1982a). In Fig. 10 let $AC = \gamma_{He}/2$, $BC = -\gamma_{He}/2$ and $AB = \gamma_{He}$, and setting $\gamma_{He}/4 \approx \gamma_{He}/4$, the approximation that $A$, $B$ and $C$ are collinear can thus be made with

$$\frac{\gamma_{He}}{2} \approx \frac{\gamma_{He}}{2} \approx \frac{\gamma_{He}}{2} \approx \frac{1}{2}(4\delta^2 + \gamma_{He}^2)^{1/2}$$

and the vertical projection of the surface becomes a line of length $(4\delta^2 + \gamma_{He}^2)^{1/2}$ symmetrical about $C$. The effect of this approximation is to introduce a slight

<table>
<thead>
<tr>
<th>$\lambda$ (Å)</th>
<th>$\theta$ (°)</th>
<th>$(\delta \lambda/\lambda)_{\text{corr}}$</th>
<th>$\omega_{\text{diff}} \cot \theta$</th>
<th>$x_H \cot \theta$</th>
<th>$L \sin \theta$</th>
<th>$x_H \cot \theta$</th>
<th>$\gamma_{He}$</th>
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<td>0.125</td>
<td>0.03</td>
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</tbody>
</table>
asymmetry to $\gamma_H$ itself such that $\gamma_H/2$ and $-\gamma_H/2$ become only approximately equal.

For the vertical-beam cross fire all incoming rays at a given horizontal displacement from the central reference ray are of the same wavelength. Thus in the vertical plane containing this reference ray the geometric surface is defined by a line of length $\sim \gamma_V$ as shown in Fig. 11.

An elliptical surface placed about $C$ with major and minor axes $\gamma_H/2$ and $\gamma_V/2$ respectively will thus approximate to the surface which contains the centres of the Ewald spheres which are present (Fig. 11), leaving $(\delta/\lambda)_{\text{conv}}$ for the moment. Placing a circle of radius $\gamma_H/2$ about this ellipse as in Fig. 12, viewed from the source side, gives effective horizontal and vertical cross fires of $(\gamma_H/2)\cos \psi$ and $(\gamma_V/2)\sin \psi$ respectively on the perimeter of the ellipse. In order to find the effective $\delta$, $\delta_e$, at $\psi$ we return to the approximated vertical projection as in Fig. 13, where we have $S'C = (\gamma_H/2)\cos \psi$ and hence $\delta$ and $\delta_e$ may be found in terms of $\beta$ and $S'C$ resulting in an excellent approximation for $\delta_e$:

$$\delta_e \approx \delta \cos \psi. \quad (2)$$

A further result necessary for our derivations is the angle $\chi$ in Fig. 13 from which we get

$$\cos \chi \approx \gamma_H/\gamma_{He}. \quad (3)$$

It has been shown (Greenhough & Helliwell, 1982a) that sample mosaic spread $\eta$ gives a spherical cap which can be represented as a spherical reciprocal-lattice volume element of radius $\varepsilon$ where, as in previous derivations (Greenhough & Helliwell, 1982a), we use

$$\sin \theta_e = \frac{d^*}{2(1 + \delta_e)} \quad (4)$$

where $\delta_e$ is $\delta \cos \psi$ and $d^* = \xi/d = 2 \sin \theta$ with Ewald-sphere radii $(1 + \delta_e)$. This gives an 'effective' $\varepsilon$, $\varepsilon_e$, as

$$\varepsilon_e \approx \frac{\eta}{2} \frac{d^*}{\cos \theta_e}, \quad (5)$$

neglecting for the moment the $(\delta/\lambda)_{\text{conv}}$ contributions which smear out the ellipse.

Fig. 9. The nest of Ewald-sphere centres in the overbend case.

Fig. 10. Vertical projection of extreme incoming rays.

Fig. 11. The correlation ellipse for a horizontal rotation axis showing the Ewald-sphere centres due to beam cross fire and direction-correlated wavelength.

Fig. 12. Geometrical description of the correlation ellipse, view from $C$ towards $O$.

Fig. 13. Derivation of the effective wavelength at a given point on the correlation ellipse and angular disposition $\chi$ of the correlation ellipse.
In Fig. 14(a) a general reciprocal-lattice point $P$ is shown approaching the nest of Ewald spheres centred on the ellipse, and just meeting this nest at $x_o$, contacting first the Ewald sphere centred at $S_0$. Extreme contacts with the nest of spheres will in general occur for Ewald spheres centred on the ellipse perimeter. Careful preservation of angular sense is required for interpretation of the results. From Fig. 14(a) we get

$$\cos(x_o - \beta_o) \approx \frac{d'^2 - 2\zeta(y_H/2)\cos \psi_o - 2e_0(1 + \delta_o) - \epsilon_o^2}{2\xi(1 + \delta_o)}.$$  (6)

The only proviso with respect to $\psi_o$ for the point shown with positive $x$ at entry, giving rise to reflection to what we will call the bottom of the film, is that $0 \leq \psi_o \leq 180^\circ$. This is entirely self evident since the other perimetal Ewald sphere of radius $1 + \delta \cos \psi_o$ is displaced away from $P$ in the $-x$ direction. Thus $\beta_o$ is in the same sense as $x_o$ and we again set $\gamma_o \approx 1$, and with the sign of $\beta_o$ determined by $\sin \psi_o$ we have

$$\beta_o \approx \frac{(y_H/2)\sin \psi_o}{(1 + \delta \cos \psi_o)}.$$  (7)

If the reciprocal-lattice point $P$ is now rotated by $\varphi_R$ about $y$ until the volume element about $P$ is completely within the nest of Ewald spheres except for a single remaining point of contact, the centre of this final contacting sphere of radius $(1 + \delta \cos \psi_f)$ must lie such that $180^\circ \leq \psi_f \leq 360^\circ$ for our 'bottom'-of-film reflection. Hence, we get from Fig. 14(b),

$$\beta_f \approx \frac{(y_H/2)\sin \psi_f}{(1 + \delta \cos \psi_f)}$$  (8)

and

$$\cos(x_o - \beta_f - \varphi_R) \approx \frac{d'^2 - 2\zeta(y_H/2)\cos \psi_f + 2e_1(1 + \delta_f) - \epsilon_f^2}{2\xi(1 + \delta_f)}.$$  (9)

In order to establish the relationship between $\psi_o$ and $\psi_f$, the reflecting ranges $\varphi_o$, from $x_o$ to the position where $P$ is on the unit-radius Ewald sphere centred at $C$, and $\varphi_f$, the remainder of $\varphi_R$, need to be established. When the point $P$ is on the unit sphere centred at $C$ we have

$$\cos(x_o - \psi_o) = \frac{d'^2}{2\xi} = \cos(x_f + \psi_f)$$  (10)

(Greenhough & Helliwell, 1982a) as may be seen from Fig. 14.

By expansion of (10), substitution from (6) and (9) and application of the half-tangent formulae (Greenhough & Helliwell, 1982a), we get, after recasting the equations into the form containing $E_0$ and $E_f$ (Greenhough & Helliwell, 1982a), using the excellent approximations given there and substituting for $\delta_o$, $\delta_f$, $\beta_o$ and $\beta_f$ from (2), (7) and (8),

$$\varphi_o \approx \frac{(y_H/2)\sin \psi_o - Ld'^2/2 + (\zeta L\gamma_H/2)\cos \psi_o}{1 + \delta \cos \psi_o}$$

$$+ \frac{2e_0L}{2} + \frac{Ld'^2}{2}$$

and

$$\varphi_f \approx \frac{(-y_H/2)\sin \psi_f + Ld'^2/2 - (\zeta L\gamma_H/2)\cos \psi_f}{1 + \delta \cos \psi_f}$$

$$+ \frac{2e_1L}{2} - \frac{Ld'^2}{2}.$$  

Assuming that $\epsilon$ is invariant with respect to $\psi$, i.e. $\cos \theta_f \approx \cos \theta$ in (5), taking $d\varphi_o/d\psi_o$ and $d\varphi_f/d\psi_f$, and setting to zero will give the values of $\psi_o$ and $\psi_f$ corresponding to the outside and inside conditions. Thus we get, neglecting a $\partial y$ term in each case,
and since $\psi_o \neq \psi_i$

$$\psi_i \simeq \psi_o + \pi.$$  \hspace{1cm} (12)

Thus $\beta_i$ in (8) becomes

$$\beta_i \simeq \beta_o - \gamma_i \sin \psi_o.$$  \hspace{1cm} (13)

Substituting from (12) and (13) into (9) and combining the result with (6) by the usual half-tangent procedure gives, with a re-arrangement of the $\xi$ term,

$$\tan \left( \frac{\phi_R - \gamma_i \sin \psi_o}{2} \right) = \pm \left[ \left( \frac{4\xi^2 - A^2}{A + B} \right)^{1/2} - \frac{4\xi^2 - B^2}{A + B} \right]^{1/2},$$  \hspace{1cm} (14)

where the negative sign between the roots represents a single stimulation and where

$$A = \frac{d^* - \gamma_i H \cos \psi_o - 2\epsilon_o(1 + \delta \cos \psi_o) - \epsilon_i^2}{(1 + \delta \cos \psi_o)}$$  \hspace{1cm} (15)

and

$$B = \frac{d^* + \gamma_i H \cos \psi_o + 2\epsilon_i(1 - \delta \cos \psi_o) - \epsilon_i^2}{(1 - \delta \cos \psi_o)}$$  \hspace{1cm} (16)

with full stimulation not possible if the blind-region condition $4\xi^2 < B^2$ holds. The value of $\psi_o$ is obtainable from (11) being

$$\psi_o \simeq \tan^{-1} \frac{\gamma_i}{L d^* \delta + L \gamma_i H},$$  \hspace{1cm} (17)

where the ± corresponds to that in (14) and where $0 \leq \psi_o \leq 180^\circ$ for our bottom-of-film reflection (+x) and $180 \leq \psi_o < 360^\circ$ for the top. This removes all ambiguity in $\psi_o$, with the positive sign in (14) and (17) corresponding to the bottom and the negative sign to the top. Values for $\epsilon_o$ and $\epsilon_i$ may be obtained by reference to (5).

In transforming (14) to the more practical form of a variable-width rocking curve or a variable-size reciprocal-lattice volume of radius $E$ (Greenhough & Helliwell, 1982a), we begin by equating both $A$ and $B$ [(15) and (16)] with $E'$. Solving for $E'$ we then get

$$E' \simeq \frac{\delta d^*}{2} \cos \psi_o + \frac{\gamma_i H}{2} \cos \psi_o + \varepsilon.$$  \hspace{1cm} (18)

Now we may use the approximation to (14) (Greenhough & Helliwell, 1982a) giving

$$\phi_R \simeq 2L \left[ \pm \left( \frac{\delta d^*}{2} \cos \psi_o + \frac{\gamma_i H}{2} \cos \psi_o + \varepsilon \right) + \frac{\gamma_i}{2L} \sin \psi_o \right]$$  \hspace{1cm} (19)

with the ± signs still referring to ±x and the restrictions on $\psi_o$ still holding. Equation (19) is now in the form $\phi_R \simeq 2EL$ with a reciprocal-lattice volume element of radius $E$ interacting with a single-unit-radius Ewald sphere. The correlated spectral dispersion/beam cross-fire effects have thus been accounted for in (19), it now seems reasonable to place any uncorrelated symmetric effects onto the unit-radius Ewald sphere producing a contribution to $E$ of $\varepsilon'$ due to $(\delta \lambda/\lambda)_{\text{conv}}$. Combining with the symmetric $\varepsilon$ we have

$$\varepsilon_S = \varepsilon + \varepsilon' = \frac{1}{2} \left[ \eta + \frac{(\delta \lambda/\lambda)_{\text{conv}}}{\tan \theta} \right].$$  \hspace{1cm} (20)

which we write as

$$\varepsilon_S = \Delta_S \frac{d^* \cos \theta}{2}$$  \hspace{1cm} (21)

(Greenhough & Helliwell, 1982a), showing that the result is the same whether the uncorrelated symmetric effects are included in $\varepsilon$ to begin with or finally.

It should be noted that the derivative of $\phi_R$ with respect to $\psi_o$ in (19) again gives (17) for the required value of $\psi_o$. If we replace $L$ by $1/|x_c|$ (Wonacott, 1977) then we can rewrite (19), with $\varepsilon$ replaced by $\varepsilon_S$, since by taking + + for positive and --- for negative; we can then take the alternative sign into $|x|$ to give, with $\phi_R$ having ± values,

$$\phi_R \simeq \frac{2}{x_c} \left[ \frac{\delta d^*}{2} \cos \psi_o + \frac{\gamma_i H}{2} \cos \psi_o + \varepsilon_S \right]$$  \hspace{1cm} (22)

where (17) becomes

$$\psi_o \simeq \tan^{-1} \frac{x_c \gamma_i}{d^* \delta + \gamma_i H},$$  \hspace{1cm} (23)

with, as before, $0 \leq \psi_o \leq 180^\circ$ for positive $x$ (bottom) and $180 \leq \psi_o \leq 360^\circ$ negative $x$ (top). The value of $x_c$ is most properly taken as that which places $P$ on the unit Ewald sphere. The calculation of fraction recorded may be accomplished via $E$ or $\Delta_S$ as outlined by Greenhough & Helliwell (1982a), provided $\psi_o$ is selected correctly from (17) and the correct signs are chosen. It is perhaps easier, however, to substitute in (22) for $\cos \psi_o$ and $\sin \psi_o$ from (23) giving, choosing the correct signs, and replacing $|x_c|$ by $1/L$ and removing the sign from $\phi_R$,

$$|\phi_R| \simeq + \left[ L^2 (\delta d^*/2 + \gamma_i H)^2 + \gamma_i^2 \right]^{1/2} + 2\varepsilon_S L,$$  \hspace{1cm} (24)

and hence

$$E \simeq \frac{1}{2} \left[ (\delta d^*/2 + \gamma_i H)^2 + \gamma_i^2 \right]^{1/2} + \varepsilon_S$$  \hspace{1cm} (25)
and
\[ \Delta_\delta \approx \frac{1}{d* \cos \theta} \left( (\delta d*^2 + x_H) + \frac{\gamma^2}{L^2} \right)^{1/2} + \Delta_S, \quad (26) \]

where \( \varepsilon_S \) and \( \Delta_S \) are given by (20) and (21). It is now immediately apparent from (24) that reflections placed identically except for the sense of \( x \) (vertical displacement) will have the same reflecting range, while a change in the sign of the horizontal coordinate \( \zeta \) will give different results.

At the Guinier position we have \( \delta = 0 \) and the rocking curve \( \Delta_\delta \) becomes, replacing \( d* \cos \theta \) by \( (x^2 + y^2)^{1/2} \) (Greenhough & Helliwell, 1982a), \( \zeta \equiv y \), and with \( \tan \alpha = x_c/y \),
\[ \Delta_\alpha \approx (\gamma^2 \sin^2 \alpha + \gamma_H^2 \cos^2 \alpha)^{1/2} + \Delta_S \quad (27) \]
or
\[ \Delta_\alpha \approx \Delta_a + \Delta_S, \quad (28) \]
which is exactly as derived for asymmetric beam cross fire in the conventional case (Greenhough & Helliwell, 1982a) with \( \gamma_H = \gamma_r \) and \( \gamma_H = \gamma_c \).

Replacing \( \Delta_S \) according to (20) and (21), and using \( \varphi_R \approx 2Ell \) and \( \Delta_S = 2E/d* \cos \theta \) (Greenhough & Helliwell, 1982a) we get
\[ \varphi_{R0} \approx L \left[ \eta + \frac{\delta \lambda}{\lambda_{\text{conv}}} \tan \theta + \Delta_a \right] d* \cos \theta \quad (29) \]
\[ E_0 \approx \frac{1}{2} \left[ \eta + \frac{\delta \lambda}{\lambda_{\text{conv}}} \tan \theta + \Delta_a \right] d* \cos \theta \quad (30) \]
\[ \Delta_\alpha \approx \eta + \frac{\delta \lambda}{\lambda_{\text{conv}}} \tan \theta + \Delta_a \quad (31) \]
and the conventional source equations for asymmetric beam cross fire (Greenhough & Helliwell, 1982a) are reproduced at the Guinier position.

**Vertical rotation axis**

By following the methods outlined for the horizontal rotation axis, but by retaining the reference \( \psi \) in our Ewald-sphere-centre ellipse as \( \psi = 0 \) along the rotation axis \( (+\zeta) \) such that \( \delta \approx \delta \sin \psi \) we get
\[ \varphi_R \approx \frac{2}{x_c} \left[ \frac{\delta d*^2}{2} \sin \psi_o + \frac{\gamma_H}{2} \cos \psi_o + \varepsilon_S \right. \]
\[ + \left. \frac{x_c \gamma_H}{2} \sin \psi_o \right] \quad (32) \]
as the equivalent to (22), and
\[ \tan \psi_o \approx \frac{x_c \left( \gamma_H + d*^2 \delta/|x_c| \right)}{\gamma_H} \quad (33) \]
as the equivalent of (23) but with the new \( \psi_o \) restrictions according to \( \zeta \), with \( \psi_o \) now restricted as \(-\pi/2 \leq \psi_o \leq \pi/2 \) for \(+\zeta \) (top-of-film reflection) and \( \pi/2 \leq \psi_o \leq -\pi/2 \) for \(-\zeta \) (bottom). It is evident from (32) and (33) that for a particular value of \( \zeta \) the values of \( \psi_o \) for \( \pm x_c \) will give different values for \( \varphi_R \) giving different reflecting ranges on the left and right of the film. If, however, reflections with the same value of \( x \) are considered, then that with \(+\zeta \) will give \( \psi_o \) with the value for \(-\zeta \) being \(-\psi_o + 180^\circ \) placing it in the correct quadrant according to the restrictions. Equation (32) will then yield
\[ \varphi_R(\text{top}) = -\varphi_R(\text{bottom}) \]
as for the horizontal case. It should be noted that the \( \psi_o \) restrictions allow the various contributions to \( \varphi_R \) to work against each other as we move across the film. It transpires that for positive \( x \), i.e. left of film, assuming that \( \delta \) is positive (overbend) that \( \psi_o \) is in the range \( 0-\pi/2 \) for \(+\zeta \)\((\pi/2-\pi \) for negative \( \zeta \) making all terms additive, while for negative \( x \) there are various cross-over points. This is well illustrated in the zero level, \( \zeta = 0 \), giving \( \psi_o = \pm \pi/2 \) from (33) and hence
\[ \varphi_R \approx \frac{2}{x_c} \left[ \pm \left( \frac{\delta d*^2}{2} + \varepsilon_S + \frac{x_c \gamma_H}{2} \right) \right] \quad (34) \]
which for positive \( x \) gives all additive contributions but for negative \( x \) we get the \( \delta \) term working against the \( \varepsilon_S + \gamma_H \) terms. This is shown in Fig. 15, and by investigation of this figure we arrive at
\[ x_c = \frac{-\delta d*^2}{2 \sin \gamma_H/2} \approx -\frac{\delta d*^2}{\gamma_H} \quad (35) \]

![Fig. 15. Reflecting ranges \( \varphi_R \) for zero-level reflections and a vertical rotation axis.](image-url)
Table 4. Reflecting ranges (°) for \( r' = 2.4 \) m and horizontal rotation axis with the monochromator set at overbend (\( \lambda = 2.008 \) Å) and at the Guinier condition (\( \lambda = 1.474 \) Å)

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<th>Parameters from Table 3. Sample mosaic spread set at 0.1° (Left-of-film reflection has +( \zeta ))</th>
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<th>( \lambda = 1.474 ) Å</th>
<th>( \lambda = 2.008 ) Å</th>
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which is the cross-over condition as seen in (34) with the reflecting range determined only by \( \epsilon_s \).

By following the same procedure as for the horizontal rotation axis, substituting for \( \sin \psi_o \) and \( \cos \psi_o \) from (33) into (32) we arrive at

\[
\varphi_R \simeq + \left[ L (L \delta d^2 + \gamma_H) + L^2 \zeta^2 \gamma_V^2 \right]^{1/2} + 2 \epsilon_s L \quad (35)
\]

\[
E \simeq \frac{1}{2} \left[ (\delta d^2 + \gamma_H/L)^2 + \zeta^2 \gamma_V^2 \right]^{1/2} + \epsilon_s \quad (36)
\]

\[
\Delta_s \simeq \frac{1}{d^* \cos \theta} \left[ (\delta d^2 + \gamma_H/L)^2 + \zeta^2 \gamma_V^2 \right]^{1/2} + \Delta_s \quad (37)
\]

The choice between the (±) alternatives is again determined by the sign of \( x \), with (35) for example being written as

\[
\varphi_R \simeq + \left[ L (L \delta d^2 + x \gamma_H) + L^2 \zeta^2 \gamma_V^2 \right]^{1/2} + 2 \epsilon_s L \quad (38)
\]

The conventional source equations (Greenough & Helliwell, 1982a) are again reproduced at the Guinier position with \( \gamma_V = \gamma_x \) and \( \gamma_H = \gamma_y \).

### III. Results and conclusions

The equations derived here show that large left–right differences will occur in the reflecting range when working away from the Guinier position. Fig. 16 shows diffraction photographs of native glycerol-dehyd 3-phosphate dehydrogenase taken at the SRS with the standard horizontal rotation axis. Table 4 shows calculated reflecting ranges for these two photographs, and, as can be seen, the spot extension suggested by this table for left-of-film reflections is evident for the overbend case. Since methods of assigning partiality to reflections depend on quantities directly related to \( \varphi_R \), not accounting for \( \delta \) will cause serious errors in these assignments. Furthermore, the asymmetric beam cross fire, even at the Guinier position, causes large deviations from the conventional symmetrical cross-fire case. For example, in Table 4 at 4 Å resolution at the Guinier position, reflecting ranges due to the cross-fire term become 0.23, 0.24, 0.28, 0.41° instead of 0, 0.07, 0.15, 0.34° if the cross fire is treated as symmetrical, with \( \gamma_V \) reset to equal \( \gamma_H \) (4 mrad in this case). This asymmetry is particularly marked in the examples given since \( \gamma_V \) is virtually negligible in the absence of a vertical focusing arrangement. Thus for the Guinier position and reflections with low values of \( \zeta \) (i.e. \( \psi_o \rightarrow 90 \) or 270) the contribution of the cross-fire term to the reflecting range is very close to zero. Moving away from \( \zeta = 0 \) for the Guinier-position case the value of \( \varphi_R \) rapidly reaches 0 (or 180°) and the cross-fire contribution is dominated by \( \gamma_H \). For the overbend case there is a
particular value of $\zeta$ where the reflecting range is due solely to the conventional term (since $\gamma V \approx 0$), occurring when $\delta d^* = -\zeta \gamma H$. This is also evident in Table 4, and corresponds to $\psi_O = 90$ or $270^\circ$ [equation (45)]. For any particular value of $d^*$ the value of $\psi_O$ is very close to 0 or $180^\circ$ except for a very small range of $\zeta$ about the $\psi_O = 90$ or $270^\circ$ position. Thus, in general, all the left-of-film reflections, and most of the right, have the limits of their reflecting range defined by $\psi_O \approx 0, 180^\circ$. It is important to realise however that a significant value of $\gamma V$ will increase the number of reflections with $\psi_O$ not near 0 or $180^\circ$.

In deriving a guide to the size and inclination of the diffracted spots on the film we may neglect the contribution of $\varepsilon_S$ as it does not significantly affect the general trend. Where an accurate estimate of spot size is required more precise formulations will be needed including crystal size and mosaicity. As can be seen from the Guinier-position photograph the conventional-type components produce little variation in spot size over the whole film; plots such as those of Wonacott (1977) confirm this. It is not intended that a detailed description of the spot size be given here, this is dealt with elsewhere (Greenhough & Helliwell, 1982b). Only reflections with $\psi_O$ around 0 or $180^\circ$ are considered, $0^\circ$ for $+\delta$ (left) and $180^\circ$ for $-\delta$ (right). Thus from Fig. 17 the vertical extent of the spots for $+\delta$ is given by

$$V = cf (\tan \nu_V - \tan \nu_V')$$

where

$$\sin \nu_V \approx \frac{\zeta \sin(\varphi_O - \varphi_R)}{(1 - \delta)^2 - (\zeta + \gamma_H/2)^2}$$

and

$$\sin \nu_V' \approx \frac{\zeta \sin(\varphi_O)}{(1 + \delta)^2 - (\zeta - \gamma_H/2)^2},$$

where $\varphi_O$, the $\varphi$ at interaction with $\psi = 0^\circ$, and $\varphi_R$ are both signed [see equation (22)] giving a signed $\zeta$ term representing $x$. The horizontal extent of the spots may also be derived from Fig. 17 as

$$H \approx cf' [\tan \nu_H - \tan \nu_H']$$

where

$$\sin \nu_H \approx (\zeta - \gamma_H/2)/(1 + \delta)$$

$$\sin \nu_H' \approx (\zeta + \gamma_H/2)/(1 - \delta)$$

$$cf' \approx (cf^2 + M^2)^{1/2}.$$
Table 5. Predicted trends in reflecting ranges \((q_R)\) and spot sizes \((S)\) and inclinations \((\nu)\) for the Guinier (G) and overbent (O) photographs

Conventional component \(\varepsilon_2\) not included in calculations.

<table>
<thead>
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<th>Film position Vertical</th>
<th>Film position Horizontal (mm)</th>
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</thead>
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</tr>
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<tr>
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<tr>
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<td>1.45</td>
</tr>
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</table>

The Lorentz factor is given by

\[ L^{-1} = |x_c| \geq |\zeta\sin(q_o - q_R/2)|. \]

From these equations we may derive the spot sizes and inclinations in terms of film position as shown in Table 5. Bearing in mind the presence of partials (compare \(q_R\) with the 1.5° rotation range) and the neglect of \(\varepsilon_3\), the trends in Table 5 are mirrored in the photographs where the given inclination \(\nu\) of the spot is to the horizontal for the top of the film; the angular sign of \(\nu\) is reversed in the bottom of the film with \(S\) unchanged. It is notable that even though the diffraction spots in the overbent case are much elongated in relation to those on the Guinier position photograph, order-to-order resolution is generally maintained due to their inclination.

One final result which is of interest is that the spots on the film are formed by different-wavelength incoming rays interacting with the mosaic-spread reciprocal-lattice volume according to \(\psi\), with different wavelengths being recorded at different points within the diffraction spot. There is thus an energy distribution within each spot giving rise to the possibility of a polychromatic diffraction experiment allowing a complete sampling of an absorption edge for optimized anomalous dispersion studies to solve the phase problem (Arndt, Greenhough, Helliwell, Howard, Rule & Thompson, 1982). Where the spots are drawn out into streaks there will be an energy gradient represented along the length of the streak. The situation is complicated somewhat by \((\delta/\lambda)_{\text{rot}}\) and mosaic spread but is simplified by an insignificant \(\gamma_v\), since a full plot of each diffraction spot, in terms of contributing energy and position, must use a spherical cap for \(\eta\) interacting with the nest of Ewald spheres due to \(\delta\), \((\delta/\lambda)_{\text{rot}}, \gamma_p\) and \(\gamma_v\). Further considerations of spot size and shape and the energy profile are given elsewhere (Greenhough & Helliwell, 1982b).

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References


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