An X-ray Spectrometer for Inelastic Scattering Experiments. II. Spectral Flux and Resolution

BY P. SUORTTI,* P. PATTISON AND W. WEYRICH

Fakultät für Chemie, Universität Konstanz, Postfach 5560, D-7750 Konstanz, Federal Republic of Germany

(Received 30 January 1986; accepted 16 May 1986)

Abstract

Using the calculated angular dependence of the X-ray reflectivities of elastically curved perfect crystals presented in paper I [Suortti, Pattison & Weyrich (1986). J. Appl. Cryst. 19, 336–342], it is possible to predict both the spectral flux and energy resolution for various curved-crystal geometries. This analysis is applied both to dispersive and scanning types of X-ray spectrometers. It is shown that, when using optimized monochromator and analyzer components, it should be possible to measure Compton profiles with good statistics and resolution in a time scale of one or two days per profile. Furthermore, it is argued that the scanning spectrometer has a number of advantages, particularly regarding the signal-to-noise ratio. Details of the design and performance of an X-ray spectrometer of this type will be given in paper III, which follows [Pattison, Suortti & Weyrich (1986). J. Appl. Cryst. 19, 353–363].

1. Introduction

The problem of optimizing the design of X-ray spectrometers using focusing X-ray optics has been the subject of a large number of studies, ranging from the early work on curved crystals in the 1930's by Johann (1931) and Johansson (1933) to the recent intensive efforts to produce in-house facilities for measuring X-ray absorption fine structure using conventional Bremsstrahlung sources (see e.g. Stern, 1980). Almost all of these studies have concentrated on the geometrical aspects of reflection from the crystals and have paid little attention to the actual properties of the crystals. It has been rather a lucky coincidence that the dimensions of feasible constructions have matched the reflectivity curves of the most common monochromator and spectrometer crystals, so that many efficient designs have been put forward [see e.g. the review by Witz (1969)].

The aim of the present work is to optimize simultaneously the geometry and reflectivity of the spectrometer. The instrument is intended for Compton and resonant Raman X-ray scattering experiments, which have until now been severely handicapped by low count rates and also by insufficient energy resolution.

There is a wide choice of spectrometer designs, and only a few possibilities can be studied in detail. In addition to fulfilling the requirements of large solid angles and moderate energy resolution, the spectrometer must be able to scan a range $\Delta E/E$ of about 1/10. A low level of background is essential, which means that each optical element should 'see' only the preceding one. The relatively large scanning range means that equatorial and axial focusing cannot be realized simultaneously, if fixed-radii crystals are used. Therefore, only cylindrical geometries are studied, which have also the advantage that line-focus X-ray sources can be used. Another factor that favors this solution is decoupling of the equatorial and axial coordinates, which makes the geometrical aberrations inherently smaller.

2. Monochromator

(a) Johann and logarithmic spiral monochromators

The Johann geometry, where the crystal planes are bent to radius $\rho$ and the surface ground to radius $\rho/2$, satisfies the ideal Rowland-circle geometry. However, the present study deals with Johann and logarithmic spiral monochromators, which require only bending of a crystal wafer, and often are sufficiently close approximations of the ideal figure.

Details of the Johann geometry are shown in Fig. 1. The angular aberration due to the deviation from the Rowland circle is found to be

$$\delta = \frac{1}{2}x^2(\sec^2\theta - 1 - x^2)^{-1/2} \approx \frac{1}{2}x^2(\cot \theta). \quad (1)$$

The effect of this aberration has been shown in paper I (Fig. 4b) (Suortti, Pattison & Weyrich, 1986), and it is obvious that the Johann aberration sets an upper limit to the useful equatorial opening of the monochromator. It is seen from Table 1 that with Mo Kα radiation and low-order reflections of silicon and quartz relatively little is gained by equatorial openings larger than ±0.5°.

*Permanent address: Department of Physics, University of Helsinki, Siltavuorenpenegers 20 D, SF-00170 Helsinki 17, Finland.

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Table 1. Properties of SiO$_2$ and Si monochromators, which have typical bending radii

The meanings of the symbols are given in text. The primary flux, $dn/d\Omega$, corresponds to X-ray tube ratings of 60 kV, 30 mA for Mo K$_\alpha$ and 40 kV, 40 mA for Cu K$_\alpha$. The axial opening is $\Delta \psi = 8 \text{ mm}/2p_0$, which makes the height of the focal line equal to that of the source. The efficiency of the monochromator, $e_{LS}$ or $e_\pi$, is the average of those for the $\sigma$ and $\pi$ components of polarization.

<table>
<thead>
<tr>
<th></th>
<th>SiO$_2$(101)</th>
<th></th>
<th>Si(111)</th>
<th></th>
<th>Si(220)</th>
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<tr>
<td>$\rho$ (mm)</td>
<td>800</td>
<td>600</td>
<td>800</td>
<td>600</td>
<td>500</td>
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<tr>
<td>$p_0 = q_0$ (mm)</td>
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<td>$\delta \theta$ (mrad)</td>
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<tr>
<td>$\Delta p_0$ (mrad)</td>
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<tr>
<td>$P_\pi (10^{-3})$</td>
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<td>0.14</td>
<td>0.33</td>
<td>0.10</td>
<td>0.31</td>
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<tr>
<td>$\gamma (^\circ)$</td>
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<td>0.40</td>
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<td>0.01</td>
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<tr>
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<td>0.53</td>
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<td>3.8</td>
<td>8.7</td>
<td>4.1</td>
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<td>8.0</td>
<td>2.9</td>
<td>5.5</td>
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<td>$x_{\omega} = x_{\omega}/p_0$</td>
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<td>0.02</td>
<td>0.02</td>
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<tr>
<td>$e_{LS}$</td>
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<td>0.53</td>
<td>0.42</td>
<td>0.34</td>
</tr>
<tr>
<td>$e_\pi$</td>
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<td>0.24</td>
<td>0.15</td>
<td>0.18</td>
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<td>8.7</td>
<td>23.7</td>
<td>7.6</td>
<td>17.4</td>
<td>8.3</td>
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<tr>
<td>$n_\sigma (10^9 \text{ s}^{-1})$</td>
<td>4.6</td>
<td>9.0</td>
<td>3.4</td>
<td>6.1</td>
<td>4.5</td>
</tr>
<tr>
<td>$n(x_{\omega})/n(x_{\pi})$</td>
<td>0.21</td>
<td>0.01</td>
<td>0.10</td>
<td>0.00</td>
<td>0.10</td>
</tr>
</tbody>
</table>

An estimate for the shortest feasible source-to-monochromator distance $p_0$ is given by the requirement that the $K\alpha_2$ component is rejected,

$$\delta \theta = \theta(x_2) - \theta(x_1) \geq \Delta \theta + \Delta x/p_0 \approx 2\Delta x/p_0. \quad (2)$$

With the previous values of $x$ this gives $p_0 > 80$ mm for Si(111), Si(220) and SiO$_2$(101), and this agrees closely with the minimum feasible distance in a monochromator construction.

The limit imposed by the Johann aberration can be overcome by bending the monochromator crystal to a logarithmic spiral with the source at the focus (see Fig. 2). All rays make the same angle with the reflecting planes, as in the case of a Johansson crystal, but the rays do not focus back to a point or line. However, there is a pseudo-focus on the caustic, and with reasonable values of $x'$ this is sufficiently sharp.
(b) Monochromatic flux

The empirical relation between the flux of electrons incident on the tube target, \( n_{\text{el}} \), and the flux of characteristic K X-rays, \( n_{\text{phot}} \), can be written as

\[
\frac{n_{\text{phot}}}{n_{\text{el}}} = C_k \frac{(E - E_k)/1 \text{ keV}}{1.63 \exp(-0.095 Z)}, \quad (3)
\]

where \( E \) is the electron energy, \( E_k \) that of the K absorption edge, and \( Z \) the atomic number of the target (Coisson, 1980). The formula is based on measurements by Green (1963), whose estimate for the constant \( C_k \) is about \( 3 \times 10^{-4} \). Some other determinations of \( C_k \) give quite different results, and in the following it is considered as a parameter, which is to be measured independently. A tube current of 1 A produces a flux of electrons, \( n_{\text{el}} \), on the target of 6.242 \( \times \) \( 10^{18} \) s\(^{-1}\). The electrons do not penetrate into the target more than a few thousand A, so that the self-absorption of the X-rays is negligible except at very small take-off angles \( \gamma \).

Self-absorption is covered by a function \( f_1(\gamma, E, Z) \). The X-ray flux per unit solid angle is

\[
\frac{dn}{d\Omega} = \frac{1}{4\pi} f_1(\gamma, E, Z) f_2(\lambda, Z) n_{\text{phot}}, \quad (4)
\]

where \( f_2(\lambda, Z) \) is the fraction of wavelength \( \lambda \) of the total characteristic K spectrum. For instance, if an Mo tube is run at 60 kV, \( f_2(4^\circ) = 0.40 \) and \( f_2(6^\circ) = 0.60 \).

The transmission of the monochromator can be written

\[
T = \int S(x, x') R(x, x') dx dx', \quad (5)
\]

where \( S(x, x') \) is the source function, and \( R(x, x') \) the reflectivity of the monochromator, illustrated in Fig. 3. Without a detailed knowledge of the source function it was approximated by

\[
S = \frac{dn/d\Omega}{2r_m}, \quad (6a)
\]

where \( r = (x/p_0) - x' + tx^2 \) and \(-r_m \leq r \leq r_m \). Here the term \( tx^2 \) is due to the Johann aberration,

\[
tx^2 = \frac{1}{2}(\cot \theta)x^2 = \frac{1}{2}(x/p_0)^2 \cot \theta. \quad (6b)
\]

For the logarithmic spiral, \( t = 0 \). The calculated reflectivity curves shown in paper I (Fig. 9) suggest the form

\[
R(x, x') = P(\eta) \eta \exp[-\eta(x' - x)], \quad (6c)
\]

Here \( P(\eta) = \int R(\theta) d\theta \), \( \eta' = (x/p_0) + r_1 \), where \( r_1 \) depends on the angular setting of the monochromator (for maximum flux \( r_1 \approx r_m \)), and \( \eta \) is related to the half-width \( d\theta \) through \( \eta = \ln 2/d\theta \).

The integrated flux for an angular opening \( \Delta \psi \) (axial), \( 2x_m = 2x_m/p_0 \) (equatorial) is

\[
n = \Delta \psi \int_{-x_m}^{x_m} dx/p_0 \int_{-x_a}^{x_a} S(x, x') R(x, x') dx', \quad (7a)
\]

where the limits of integration are (taking \( r_1 = r_m \))

\[
x_a = (x/p_0) - (x' - 2r_m - tx^2),
\]

\[
x_m = (x/p_0) + r - tx^2 = x' - tx^2. \quad (7b)
\]

Substitution of \( S(\eta, r_m, t) \) gives, when \( \Delta \chi = 2x_m \),

\[
n = \Delta \psi \frac{dn}{d\Omega} P(\eta) \frac{1 - \exp(-2\eta r_m)}{2\eta r_m} \int \exp(-\eta tx^2) dx
\]

\[
= e(\eta, r_m, t) \Delta \psi \Delta \chi \frac{dn}{d\Omega}. \quad (8)
\]

Here \( e(\eta, r_m, t) \) is the efficiency of the monochromator. The numerical values can be taken from paper I (Figs. 7, 8 and 9), and the results are collected in Table 1. The distances \( p_0 \) and the take-off angles \( \gamma \) are chosen to be compatible with \( d\theta \approx 2r_m \) and \( x_m \) is selected such that the Johann aberrations are equal for the three reflections being considered. In the case of Mo K\( \alpha \) radiation, \( \text{SiO}_2(101) \) gives a slightly better flux than \( \text{Si}(111) \) or \( \text{Si}(220) \), but unfortunately also quite a lot of the K\( \alpha_2 \) component is transmitted. Therefore, \( \text{SiO}_2(101) \) will be excluded from subsequent comparisons. On the other hand, for Cu K\( \alpha \), \( \text{SiO}_2(101) \) seems to be the best choice. \( \text{Ge}(111) \) and \( \text{Ge}(220) \) were also included in the calculations for Cu K\( \alpha \), but, because of the larger absorption, \( P_\gamma \) and \( P_\chi \) were only about 60% of the values for \( \text{SiO}_2(101) \). As mentioned before, the Johann aberration limits the flux, so that the maximum useful equatorial opening, \( \Delta \chi = 2x_m \), is about 0.04 for \( \text{SiO}_2(101) \) or \( \text{Si}(111) \) and 0.05 for \( \text{Si}(220) \). The flux from a logarithmic spiral monochromator increases linearly with \( x_m \), but the broadening of the pseudo-focus sets a limit for \( x_m \). This will be discussed under the next heading.

The dependence of the flux on the bending radius was studied in the case of \( \text{Si}(111) \) and Mo K\( \alpha \) radiation. If the take-off angle is kept constant \( \gamma = 4^\circ \), \( 2\eta r_m = (\Delta x)\ln 2/(\sin \theta) = 0.686 \), independently of \( \rho \). The flux depends on the axial opening \( \Delta \psi \), and Fig. 4 illustrates the flux to a constant \( \Delta \psi \) through a constant axial slit. The dependences on \( \rho \) are not very strong, and practical considerations usually favor a...
compact construction, unless an almost complete rejection of $Kz_2$ is required.

The above calculations are rather approximate, because a rectangular source function is assumed, and the effects of the vertical divergence and other deviations from the ideal cylindrical geometry are ignored. Nevertheless, they give valuable guidelines for the selection of the parameters of the monochromator.

An ideal monochromator would reflect all Mo $Kz_j$ radiation which is emitted to $\Omega = \Delta \varphi / \Delta x_j$, i.e. $R(\theta)$ should be unity over the width of the source, $2 \varrho_m$. The efficiency of the monochromator, $e(\eta, \varrho_m, t)$, is the average value of $R(\theta)$ (in reality less than unity) in this interval, and Table 1 indicates that with Mo $Kz$ radiation and a monochromator bent to a logarithmic spiral efficiencies up to 50% should be achieved. The numerical values of the flux in Table 1 are based on Green's (1963) value of $C_K$ [see (3)], which may be only of the right order of magnitude, but the purpose of giving these numbers is to demonstrate that large fluxes of monochromatic X-rays are available from conventional X-ray tubes. The details of construction are given in paper III of this series (Pattison, Suortti & Weyrich, 1986), and we will show that the measured flux is reasonably close to the estimates given above.

(c) Distribution at the focus

The performance of the monochromator is not totally characterized by the flux only. In the following, the spatial, angular and wavelength distributions at the focus, as well as the degree of polarization, will be studied.

The line source of the target of the X-ray tube is focused back to a line in the Rowland-circle geometry, and, to a good approximation, the angular distributions are determined by the opening of the monochromator. On the other hand, the spatial width of the X-ray beam at the focus is due to the source size and geometrical aberrations. The effects of the axial divergences and beam penetration are common to Johansson, Johann and logarithmic spiral crystals, but the equatorial divergence causes broadening only in the last two cases. These questions will be treated in detail for the analyzer, when the energy resolution is estimated, but here the distributions of discrete energy components are of interest.

The penetration of the X-ray beam into the monochromator causes an asymmetric broadening to the low-angle side. The average penetration is related to the width of the reflectivity curve. It can be seen from Fig. 1 that the displacement at the focus is

$$\Delta x_f = \rho (\sin \theta) \Delta \theta = c \sin \theta.$$  

(9)

The constant $c$ has turned out to be 0.25 mm for both 111 and 220 reflections of Si for Mo $Kz$ radiation. The displacements of focus are thus 0.03 and 0.05 mm, respectively, which are of the same order as the projected widths of the source.

Aside from beam penetration into the crystal Johann and Johansson geometries are symmetrical, and the focus is the image of the source. The effect of the Johann aberration is to limit the useful equatorial divergence to $x'_m = 3(\cot \theta)^{-1/2}$, as seen in (8) and Table 1. This is also illustrated in Fig. 1, where $\Delta x_f$ must remain smaller than the width of the source. The axial divergences increase the length of the focal line, and the intensity distribution is determined by the slits. On the other hand, reflection from the logarithmic spiral is not symmetrical, and the equatorial divergence broadens the pseudo-focus on the caustic. For a symmetrically cut crystal this broadening is

$$\Delta x_{f,m} = -q_0 x'_m \cot \theta;$$  

(10)

i.e. towards the low-angle side (de Wolff, 1951; Witz, 1969). This is twice the Johann error, as is intuitively clear. It is seen from Table 1 that with $x'_m = 0.01$, $\Delta x_{f,m}$ is 0.08 mm for Si(111), when Mo $Kz$ radiation is used. This is about twice the projected width of the source, and thus the useful equatorial opening of the logarithmic spiral monochromator may be less than that of a Johann crystal, if a sharp focal line is required. With the same equatorial opening the flux from the logarithmic spiral monochromator is larger, because the ‘window’ of acceptance in the $x-x'$ phase space has straight edges, so that the source distribution $S(x, x')$ and the reflectivity $R(x, x')$ overlap more completely (see Fig. 3).

A logarithmic spiral monochromator separates $z_2$ and $z_3$ as well as a Johann monochromator, while the curved phase space window of the Johann crystal may transmit more of the unwanted component. The contribution of $z_2$ can be estimated from (8), and with $r_2 = r_m$ the Johann monochromator is no worse than the logarithmic spiral. The ratio $z_2/z_1 \approx 0.5 \exp(-\eta \Delta \theta)$, and it was seen earlier that the bending

![Fig. 4. Flux from logarithmic spiral (LS) and Johann (J) Si(111) monochromators when the bending radius $\rho$ is varied. The distance from the source is $p_0 = \rho \sin \theta$. Curves (a) correspond to a constant axial opening $\Delta \varphi = 0.044$, and curves (b) to a constant axial slit $d \varphi = 8$ mm at the focus. The Mo $Kz$ tube is run at 60 kV and 30 mA, and the take-off angle is 4°. The dotted line shows the contribution of $Kz_2$ in the exit beam.](image-url)
radius, and thus $\eta_1$ can be increased without too large losses, if a better rejection of $\alpha_2$ is required.

The degree of polarization is obtained from the integrated reflectivities that are calculated separately for the perpendicular ($\sigma$) and parallel ($\pi$) components of the incident radiation. It is seen from paper I (Fig. 7) that the ratio of the reflectivities first increases over the dynamical value, $|\cos 2\theta|$, and then decreases to the kinematical value, $\cos^2 2\theta$, when the bending radius decreases. This behavior is similar to the polarization properties of mosaic crystals, where the ratio may exceed $|\cos 2\theta|$ because of large secondary extinction (Jennings, 1984).

3. Analyzer

The above conclusion that the bent-perfect-crystal monochromator comes very close to the ideal solution suggests a similar construction for the analyzer. The monochromator focuses the radiation on a line, which is only slightly wider than the projected source. The sample is placed near or at the focal line. The analyzer is ideally a Johansson crystal, which is usually approximated by a Johann crystal, while the logarithmic spiral is not so suitable, since it is tuned to a specific wavelength. The operation of the analyzer was shown in paper I (Figs. 4a and b), which illustrate the dispersive (stationary) mode and the monochromatic (scanning) mode as phase-space diagrams. It is obvious that the average flux is the same in both cases, and in the following a detailed calculation of the flux and resolution is given.

(a) Flux

The Rowland-circle geometry for the dispersive case is illustrated in Fig. 5. The optimum location of the sample is determined by the length of the analyzer crystal and the range of wavelengths to be covered. For one wavelength the solid angle is proportional to the width of the scattering volume of the sample, but this is exactly compensated by the inverse proportionality of the intensity of scattering. For simplicity, we assume in the following calculation that the incident power is distributed uniformly in the scattering volume.

For a given wavelength, the flux is obtained from (4) to (8). The Johann aberration varies with wavelength as different parts of the analyzer are effective, which we ignore for the time being. In (4), $n_{\text{phot}}$ is now the incident monochromatic photon flux $n_0$ times the scattering probability of the process being studied. This is usually given by the scattering coefficient $\sigma$ per unit path length $l$. Therefore, the observed flux per unit wavelength $\lambda'$ and solid angle can be written as

$$\frac{dn}{d\Omega} = n_0(f_4A_1)d^2\sigma/d\lambda'd\Omega.$$  \hspace{1cm} (11)

Here $A_1$ is the effective thickness of the sample, and the factor $f_4$ takes into account the absorption in the sample, unless this is included in $A_1$. If the projected width of the scattering volume, as seen from the analyzer, is $w$, then $2r_w = w/a_1$ and $2x_w = w/a_2$, and scattering into the solid angle $d\Omega'$ is given by the spectral flux

$$\frac{dn}{d\lambda} = n_0(f_4A_1)d\psi \frac{d^2\sigma}{d\lambda'd\Omega} \frac{a_1}{a_2} \int R(\theta)d\theta.$$ \hspace{1cm} (12)

When this is integrated over $\lambda'$ using the average values for the other factors and noting that $a_1/a_2 = (\chi_2 - \chi_1)/(\theta_2 - \theta_1) = \Delta \chi/(\theta_2 - \theta_1)$,

$$n = n_0(f_4A_1) \frac{A\psi A\chi}{\theta_2 - \theta_1} P \frac{d\sigma}{d\Omega}.$$ \hspace{1cm} (13)

Here $\theta_2$ is the largest and $\theta_1$ the smallest analyzer angle for the wavelength range being reflected.

The operation of the analyzer in the scanning mode can be described on the basis of Figs. 3 and 6. In the phase-space picture the window of acceptance, i.e. the reflectivity curve $R(\lambda', x, x')$, sweeps through the source distribution $S(\lambda', x, x', y, y')$ projected on the surface of the analyzer crystal. The wavelength of the scattered radiation, $\lambda'$, and the distance between the sample and the analyzer, $p_0$, are related, and in the following they

![Fig. 5. Rowland-circle geometry for a dispersive Johansson analyzer. C is the sample, F1 and F2 the virtual source points, A1 and A2 the respective reflecting points on the analyzer, and D1 and D2 the focal points at the detector. From the triangles F1CF2 and A2CA1, (a2/a1)δ = χ1 - χ2 and θ2 - θ1 = δ.](image)

![Fig. 6. Perspective drawing of a scanning Johann-type analyzer. Two aberrations are shown: a ray with the axial divergence $y' = \psi$ is reflected when $\epsilon > 0$, and one with the equatorial divergence $x' = \chi$ when $\epsilon < 0$. F1 and F2 are the source points for wavelengths $\lambda_1'$ and $\lambda_2'$, respectively.](image)
Table 2. Compton (C) and resonant Raman scattering (RRS) from a few substances

<table>
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<tr>
<th>Substance</th>
<th>Mo Kα₁</th>
<th>Mo Kα₂</th>
<th>Mo Kα₃</th>
<th>Cu Kα₁</th>
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<td>(dσ/dΩ)₀C (cm⁻¹ sr⁻¹)</td>
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<td>n (s⁻¹)</td>
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</tbody>
</table>

Here, μ₀ is the linear absorption coefficient for the incident radiation, σ the scattering cross section per unit length for the process in question, dσ/dΩ that per unit solid angle at scattering angle θ (170° for Compton scattering), and fₐΔl the effective path length of the incident radiation in the sample. The average value of dσ/dΩ is σ/4π. The following values are used in the flux calculations of Compton scattering: n₀ = 5 × 10⁹ s⁻¹, ΔψΔχ = 0.010, P = 0.64 × 10⁻⁴ [for Si(440)], and θ₂ - θ₁ = 0.058, which corresponds to a scan of 2ΔEₑ = 2.2 keV down from E₀ = 17.48 keV (Mo Kα₁). In the case of RRS the corresponding figures are: n₀ = 8 × 10⁹ s⁻¹, ΔψΔχ = 0.004, P = 1.2 × 10⁻⁴ [for SiO₂(10₁)], and θ₂ - θ₁ = 0.044, for a scan of 1.0 keV.

The average count rate is the same as that from (13), because θ₂ - θ₁ = ω(t₂ - t₁).

As pointed out in the Introduction, the energy resolution of the analyzer will be optimized for detailed studies of Compton profiles and the spectrum of resonant Raman scattering, and these are used as examples for estimates of the flux.

The scattering cross section can be written as

\[
\frac{dσ}{dΩ} = r^2 M₀ Kp₀ f(θ),
\]

where \(r^2\) is the scattering cross section of an electron, \(M₀\) the number of scattering units (atoms, unit cells) per unit volume, \(Kp₀\) the polarization factor and \(f(θ)\) the scattering factor. This is integrated over the wavelength distribution of the process in question, and it depends on the scattering angle θ. Theoretical values of dσ/dΩ are given in Table 2. The average effective thickness of the sample is \(fₐΔl\). For a plate of thickness \(T\),

\[
fₐΔl = \left(\frac{μ₀ + μ_{\text{cos} α}}{μ_{\text{cos} β}}\right)^{-1} \times \left\{1 - \exp \left[-\left(\frac{μ₀ + μ_{\text{cos} α}}{μ_{\text{cos} β}}\right)T\right]\right\}_{1}^{θ₂} \frac{d²n}{dλ' dx'} dx' \int_{θ₁}^{θ₂} dΩ Δφ(λ').
\]

The resolution function \(s(p₀, x') \ast r(λ', x')\) will be studied in the following section, while, for the calculation of the integrated scattered intensity, it is sufficient to note that the function is normalized to unity. If the angular velocity of the analyzer is ω, the observed total scattered intensity in the time interval \(t₂ - t₁\) is

\[
\int_{t₁}^{t₂} n dt = \frac{1}{ω} \int_{θ₁}^{θ₂} \frac{d²n}{dλ' dx'} dx'.
\]
\[
\int_{A} \Delta l = \frac{\exp(-\mu T/\cos \beta)}{\mu_0 - \mu \cos \alpha/\cos \beta} \\
\times \left\{ 1 - \exp\left[ -\left( \frac{\mu_0}{\cos \alpha} - \frac{\mu}{\cos \beta} \right) T \right] \right\}, \quad (18c)
\]

which approaches \((T/\cos \alpha) \exp(-T/\cos \beta)\), when \(\mu_0/\cos \alpha \approx \mu/\cos \beta\).

In these formulae, \(\mu_0\) is the attenuation coefficient for the incident radiation, \(\mu\) the average of that for the scattered radiation, and \(\alpha\) and \(\beta\) are the angles between the plate normal and the incident and exit beams, respectively.

The examples of Table 2 include Compton scattering from weakly absorbing samples and resonant Raman scattering from the surface layer of a thick sample. Schematic illustrations of the spectra are given in Fig. 7. The numbers demonstrate that the available flux through the analyzer is adequate for routine measurements of Compton scattering, as \(10^6\) to \(10^7\) counts are collected in one day. In this respect the stationary and scanning modes are equivalent, at least in principle. The differences will be discussed later, and it will become apparent that, in most cases, the scanning mode is to be favored.

The response function of the analyzer is the product of the geometrical part and the integrated reflectivity, \(S_2(p_0)P(\gamma')\). The first factor depends on the details of the experimental set up, but the second can be calculated using the lamellar model. Some results are shown in Fig. 8, and it is clearly necessary to include the effects in the analysis of the measured distribution \(dn/d\gamma'\).

(b) Resolution

In the following, most of the discussion is based on the scanning mode. As seen above, the angular resolution of the analyzer is due to the width of the normalized reflectivity curve \(r(\gamma', \gamma')\) and to the size of the effective source, \(s(p_0, \gamma')\). The latter also includes the effects of non-ideal geometry, i.e. departures from the Rowland circle, off-axis rays, error in the distance between the sample and the analyzer. As far as these factors are independent, the total resolution function is the convolution of the individual angular functions,

\[
G(\epsilon) = s(p_0, \gamma') \ast r(\gamma', \gamma') = g_1(\epsilon) \ast \cdots \ast g_N(\epsilon), \quad (19)
\]

where \(\epsilon = \theta - \theta_0\) is the deviation from the reading of the analyzer angle. The effective size of the source and the active volume of the analyzer can be limited by the receiving slit in the front of the detector, but, as in the calculation of the flux, the slit is assumed to be wide enough to transmit the whole beam. This was also the condition for projection of the axial distributions on the equatorial plane.

Examples of the reflectivity curves \(R(\epsilon) = g_1(\epsilon)\) of bent crystals were given in paper I (Fig. 9), and an approximate analytic form for \(g_1(\epsilon)\) is given by \((6c)\). The projected width of the scattering volume depends on the thickness of the sample, absorption, scattering angle and the distance \(p_0\). The angular distribution of scattering from the sample, as seen from the analyzer, is in the Bragg case (see Fig. 9)

\[
g_2(\epsilon) = \int_0^T I(x) \exp \left[ -\left( \frac{\mu_0}{\cos \alpha} + \frac{\mu}{\cos \beta} \right) T \right] dT \quad (20a)
\]

where

\[
\epsilon = (T \sin 2\theta + x)/p_0 \cos \alpha. \quad (20b)
\]

This function was calculated for \(I(x) = \text{constant}\), when \(|x| < w/2\), where \(w\) is the width of the incident
beam, and the result is given together with other aberration functions in Fig. 10 and the associated Table 3. The width of \( g_2(\epsilon) \) can be made small by asymmetric scattering from a strongly absorbing sample, but only at the expense of intensity.

The aberrations owing to the diverging rays were studied by Johann (1931) and Johansson (1933). The equatorial Johann aberration allows rays that have \( \epsilon < 0 \) to be reflected; see (1). The intensity distribution is

\[
g_3(\epsilon) = |\epsilon|^{-1/2}
\]

when the source distribution is uniform. The axial divergence \( \psi = \varphi' \) decreases the reflection angle, so that at a given \( \theta_A \) and \( \lambda' \) rays with \( \epsilon(\psi) > 0 \) are reflected. The relationship for small \( \psi \) is

\[
\epsilon = \frac{1}{2} \psi^2 \tan \theta = \frac{1}{2} (h/\rho \sin \theta)^2 \tan \theta,
\]

where \( h = p_0 \psi \). The intensity distribution would be the same as that for the equatorial aberration, but now the distribution of \( \psi \) is triangular, and the result is

\[
g_4(\epsilon) = \epsilon^{-1/2} - \epsilon_m^{-1/2},
\]

where \( \epsilon_m \) is the maximum deviation from (22). It is seen from (1) and (22) that \( g_3 \) and \( g_4 \) become important at small Bragg angles, and these aberrations may limit the choice of analyzer reflections.

The above aberrations usually dominate, but small effects arise from anticlastic bending of the crystal. If a rectangular plate is bent by two equal and opposite momenta, the surface of the crystal is not a cylinder, but the inner side of a torus. The radius of the transverse bend is

\[
p_N = p/\nu,
\]

where \( \nu \) is the Poisson ratio. The effects on the scattering angle can be ignored in the present case, but the toroidal shape may become critical in sagittal focusing (Sparks, Ice, Wong & Batterman, 1982). If the crystal is clamped to the support by the edges, it may assume a different shape, and these details will be discussed in the experimental part.

The alignment errors may become the most important factors of geometrical aberrations. If the line-shaped source of scattered radiation is not perpendicular to the plane of the Rowland circle, the effective width of the source is increased. The source function \( g_2(\epsilon) \) is convoluted by the projection of the axial distribution which, in the case of a uniform distribution, is

\[
g_2(\epsilon) = p_0 (2\zeta h_m)^{-1}; |\epsilon| \leq \zeta h_m/p_0,
\]

where \( \zeta \) is the tilt angle as seen from the analyzer. The effect of an error in distance \( p_0 \) is seen from Fig. 5 and
by comparing the phase-space diagrams shown in paper I (Fig. 4). The angular range seen by the analyzer is \( \theta_2 - \theta_1 = (a_2/a_1)\Delta \chi \), and so

\[
g_\theta(c) = p(2\Delta \chi |p - p_0|)^{-1}, |c| \leq \Delta \chi |p - p_0|/p. \tag{26}
\]

The wavelength or energy resolution is obtained from the Bragg law

\[
|\Delta \lambda'/\lambda'| = |\Delta E/E| = \Delta \epsilon \cot \theta_A \tag{27}
\]

where \( \Delta \epsilon \) is the width of the angular resolution function \( G(\epsilon) \).

Results for a few typical cases are collected in Table 3. The optimum combination is the one which gives the largest detected flux at the given energy resolution. It is not possible to survey all solutions, but three different choices are included in the table. The first two are based on a Johann monochromator, which provides a narrow focal line on the sample, and the intensity loss is compensated by the use of low-order reflections of the analyzer. The third solution is based on the use of a logarithmic spiral monochromator, which makes the scattering volume quite large, but this is balanced by the larger distances and Bragg angle of the analyzer. It is obvious that a Si(220) analyzer is not suitable for high-resolution Compton profile measurements, while it should be able to reveal the details of the resonant Raman Compton profile. When Cu K\( \alpha \) is used as the exciting radiation. The resolution provided by Si(440) is sufficient for Compton profile measurements, and also Si(400) could be used, if \( T \) and \( \Delta \chi \) are reduced. The loss of intensity could be compensated by increasing \( \Delta \psi \).

The dependence of the energy resolution on the wavelength was studied in the case of the Si(440) analyzer. The results are shown in Fig. 11. \( \Delta E \) is approximately proportional to \( E^3 \) and \( \Delta \lambda' \) to \( \lambda^{-1} \), which is to be expected as the major angular aberrations are proportional to \( \cot \theta_A \), \( \cosec \theta_A \) or \( \theta_A^{-1} \). The Compton peak is located at 0.757 \( \AA \), when the scattering angle \( \theta \) is 170°, and at this wavelength the relative resolution is \( \Delta E/E = 3.5 \times 10^{-3} \). The change of \( \Delta E \) or \( \Delta \lambda' \) is quite large over the range of the Compton profile. This makes the deconvolution procedure more complicated, although it does not pose a serious difficulty as long as \( G(\epsilon) \) can be measured and/or calculated.

(c) Comparison of dispersive and scanning analyzers

The efficiency of the dispersive mode is equal to that of the scanning mode, but in practice the two arrangements differ considerably. The principal differences are listed briefly in the following, while the practical considerations will be discussed in paper III.

The dispersive mode has some very attractive features. No moving parts are needed, and the whole spectrum is recorded at the same time, which eliminates the effects of time-dependent instabilities. There is no deterioration of resolution due to alignment errors of the sample. However, there are a few serious geometrical limitations. First, different wavelengths are reflected from different parts of the analyzer crystal. The reflectivity of the crystal may vary slightly, and, at any rate, \( \Delta \psi = \Delta \psi \Delta \lambda \) changes with \( \lambda' \). If a Johann crystal is used, the location of the focus is shifted towards smaller angles when \( \lambda' \) moves away from the mean value. On the other hand, \( \Delta \chi \) is small and the Johann aberration does not degrade the resolution. Otherwise the resolution functions are the same in both cases, except that the role of the detector in the dispersive mode is that of the source in the scanning mode. In addition to the position resolution of the detector, the resolution is limited by the parallax effect, because the detector must be placed perpendicular to the reflected beam. The equatorial angle \( \Delta \gamma = a_2 \Delta l \), within which one wavelength is reflected, can be minimized by placing the sample at the focus of the monochromator, but the parallax effect may still be the dominant factor in the resolution function.

It was seen above that the aberrations become large when the Bragg angle of the analyzer is small. These difficulties are overcome by the use of a Cauchois-type analyzer. This analyzer corresponds to the dispersive mode of the reflecting analyzer only (see Fig. 12). The beam enters the cylindrically bent crystal on the convex side and exits on the concave side (Laue case). If the crystal is cut asymmetrically, the same kind of enhancement of the integral reflection as in the Bragg case is obtained owing to bending. For weakly absorbing crystals, the reflectivity curve is almost flat-topped (Albertini et al., 1977). The Cauchois analyzer has a few advantages: the ‘Johann’ aberration is small, and, if the sample is brought close to the crystal,
small analyzer crystals can be used. On the other hand, the crystal should be quite thin, of the order of 0.1-0.2 mm, which nevertheless limits its use to short wavelengths. More serious may be the fact that the distance between the sample and the detector is large, so that the solid angle subtended is small. Shielding is also a problem, because the radiation incident on the analyzer crystal cannot be confined as effectively as in the Bragg case. Finally, as in the dispersive mode of the reflecting analyzer, a higher background of the incoherently scattered radiation from the crystal is to be expected. For these reasons, we have not studied the Cauchois-type analyzer in detail, although this construction has already been used in studies of Compton scattering with synchrotron radiation (Loupias & Petiau, 1980).

The scanning analyzer lacks the advantages of the stationary construction. On the other hand, if the equatorial opening $\Delta \chi$ is kept constant, the same area of the crystal is always used, as discussed in the preceding section. The range of the scan can be limited to the region of interest, and, perhaps most important, an efficient detector behind a narrow receiving slit gives a good signal-to-noise ratio. Many of the aberrations are common to both spectrometer constructions, and extensive test measurements and calculations of corrections are required for a sufficient characterization and optimization of the instrument.

4. Summary

The inelastic scattering processes of X-rays are probes of the various excitations of the solid state. However, this area has remained largely unexplored because of the small cross sections of these processes. It is not surprising that some of the most promising applications of synchrotron radiation are in this field. The purpose of the present work was to demonstrate that conventional X-ray sources are adequate for many types of experiments and may even compete with the synchrotron sources. Most inelastic scattering is quite diffuse, so that large solid angles (or large volumes of reciprocal space) can be used, and the present spectrometer is designed to take maximum advantage of this situation.

The calculations of this paper were used as guidelines when designing an optimized scanning spectrometer for measurements of Compton profiles using Mo $K\alpha_1$ radiation from a commercial fine-focus tube run at 1.8 kW. A detailed description of the design and performance of this spectrometer will be presented in paper III. We will show that it is quite feasible to measure Compton profiles with good resolution and statistics within a measuring time of about one day per profile, provided the photoabsorption cross section of the sample is not too large.

The authors are grateful to the Deutsche Akademische Austauschdienst and the Academy of Finland for supporting visits and to the Fonds der Chemischen Industrie for continuing financial assistance.

References


Fig. 12. Cauchois-type X-ray spectrometer. $S$ is the sample, $A$ the asymmetrically cut analyzer crystal, which is bent to the radius $\rho$ (=diameter of the Rowland circle). PSD is the position sensitive detector, which is placed at the focus of the analyzer.