Comparison of Curved Monochromator Neutron Data with the Prediction of a Simple Model

BY G. ALBERTINI
Facoltà di Medicina, Università di Ancona, Italy
A. BOEUF
JRC Euratom, Ispra, Italy and Institut Laue–Langevin, 156X Centre de Tri, 38042 Grenoble Cédex, France
S. MAZKEDIAN
Facoltà di Ingegneria, Università di Ancona, Italy
S. MELONE
Facoltà di Medicina, Università di Ancona, Italy
V. ROZZI
Facoltà di Ingegneria, Università di Ancona, Italy
AND F. RUSTICHELLI
Facoltà di Ingegneria, Università di Ancona, Italy, JRC Euratom, Ispra, Italy and Institut Laue–Langevin, 156X Centre de Tri, 38042 Grenoble Cédex, France

(Received 2 June 1976; accepted 5 November 1976)

The data available in the literature on curved neutron monochromators, i.e. Ge and CaF₂, has been compared with the predictions of a simple model recently developed to evaluate the neutron diffraction properties of curved crystals.

Introduction

A simple physical model to predict the neutron diffraction properties of deformed crystals was presented in a previous paper (Albertini, Boeuf, Cesini, Mazkedian, Melone & Rustichelli, 1976). In particular, this model allows the derivation of simple analytical expressions for the neutron diffraction patterns of curved crystals in the Bragg case. The validity of the model has been checked by comparison with the neutron diffraction patterns obtained by a rigorous treatment based on the Takagi (1962, 1969) and Taupin (1964) differential equations, as was done by Klar & Rustichelli (1973). Furthermore, the model was used to interpret the experimental results obtained by Boeuf & Rustichelli (1974) on chemically curved Si crystals.

In this paper, the model will be compared with different available experimental results on curved neutron monochromators (Egert & Dachs, 1970; Kalus, Gobert & Schedler, 1973). These experimental results refer to germanium and calcium fluoride monochromators. The Ge crystals were mechanically bent whereas the CaF₂ crystals were thermally bent.

Theoretical considerations

The model has been developed for the Bragg case and allows the prediction of the neutron diffraction patterns obtained with curved nonabsorbing crystals. The case of absorbing crystals will be treated in detail in the Appendix. The notation of Klar & Rustichelli (1973) will be used. The radius of curvature $R$ of the crystal is related to a quantity $c$ by the following equation*

$$R = \frac{\pi V_c^2}{4 F^2 d^3} \cot \theta \cos \alpha$$

where $V_c$ is the volume of the elementary cell, $F$ the structure factor, $d$ the interplanar distance, $\theta$ the Bragg angle, and $\alpha$ the angle between the incident neutron beam and the surface of the crystal. The lattice-spacing variation, due to the curvature of the crystal, is neglected. This contribution is usually very small compared with the misorientation of the plane. The quantity $A$ is related to the thickness of the crystal by

$$t(\text{cm}) = \frac{V_c}{2 F d} A.$$  

Finally, $y$ is an angular deviation measured in units equal to one-half of the Darwin curve. The width $\Delta \theta$ of this curve is given in the asymmetric Bragg case by

$$\Delta \theta \text{ (Darwin)} = \frac{4 F d^2}{V_c \pi} \left( \frac{\sin \beta}{\sin \alpha} \right)^{1/2} \tan \theta$$

where $\beta$ is the angle between the reflected beam and the surface of the crystal. If $y(0)$ is the deviation from the Bragg law at the surface of the crystal

$$y = y(0) + c A.$$  

* This equation is an extension of equation (48) of Klar & Rustichelli (1973) to the general asymmetric case.
In the simple model of Albertini et al. (1976), the curved crystal is divided into a series of small crystals which are considered to be perfect and to have a thickness \( A = 2/c \). The reflectivity of a perfect non-absorbing crystal is then given by Zachariasen (1967) for X-rays:

\[
\begin{align*}
\sin 2\frac{A}{(y^2 - 1)} & \frac{1}{1 + \sin^2 \frac{A}{(y^2 - 1)}}.
\end{align*}
\]  

(5)

This equation can also be used to evaluate the neutron reflectivity of a perfect crystal. For a given orientation \( y(0) \) of the crystal at a fixed wavelength, the total reflectivity of the crystal is given by

\[
r = 1 - \left[ \prod_{k=1}^{n_1} (1 - r_{ik}) \right] \left[ \prod_{k=1}^{n_{II}} (1 - r_{IIk}) \right].
\]

(6)

Region II corresponds to the part of the curved crystal where the Bragg condition is fully respected. \( r_{II} \) represents the reflectivity of region II obtained from (5) with \( y = 0 \) and \( A = 2/c \). Regions I and III are decomposed into \( n_1 \) and \( n_{II} \) perfect crystals respectively of different orientations \( y \) and same thickness \( (A = 2/c) \). \( r_{ik} \) and \( r_{IIk} \) are the reflectivities of these perfect crystals and are obtained from (5) as well. For more detailed information on the model, one should refer to Albertini et al. (1976).

Comparison between theoretical results and available data

Germanium mechanically bent crystals

The model previously described will now be used to interpret data already published in the literature and concerning dynamical neutron diffraction by a curved monochromator. In particular, Egert \& Dachs (1970) have published some experimental data on the integrated reflectivity of Ge crystals as a function of the curvature. The 0.1 cm thick Ge crystals were mechanically bent till a radius of curvature between 500 and 140 cm was obtained. In a first series of experiments, the crystals were tested in the symmetric Bragg condition, the (200) plane being cut parallel to the surface. In Fig. 1, the theoretical curve calculated with the model \( (R_{\text{theor}}) \) is compared with the data of Egert \& Dachs (1970) obtained at the neutron wavelength \( \lambda = 1.71 \text{ Å} \). The two intersecting straight lines which appear in the figure are the theoretical evaluations of the authors. Fig. 2 represents the same comparison for the experimental data obtained at \( \lambda = 1.08 \text{ Å} \) and \( \lambda = 0.85 \text{ Å} \). Fig. 3 shows the comparison for the asymmetric Bragg conditions investigated by the authors. The reflexion 351 has been investigated at \( \lambda = 1.04 \text{ Å} \) which corresponds to an incidence angle \( \alpha = 15^\circ 89' \) and to \( \beta = 60^\circ 13' \). The reflection 531 was investigated at the same neutron wavelength, corresponding to \( \alpha = 60^\circ 13' \) and \( \beta = 15^\circ 89' \). It appears that, in general, the agreement between the theoretical previsions of the model and the experimental data is satisfactory. The biggest discrepancy corresponds to the symmetric Bragg case at \( \lambda = 1.08 \text{ Å} \) and \( \lambda = 0.85 \text{ Å} \). The experimental reflectivity is lower than the theoretical. This discrepancy could not be explained by the fact that the absorption was neglected in the model, its contribution being estimated to be lower than 4%. Although it cannot be completely ruled out that the discrepancy is due to the approximations of the model, it seems more likely that the low experimental reflectivity
arises from a lack of homogeneity in the curvature and therefore the presence of primary extinction.

**Calcium fluoride thermally bent crystals**

Kalus, Gobert & Schedler (1973) have published some experimental data concerning the reflectivity of calcium fluoride thermally bent crystals. The 333 and 440 reflexions were investigated in the Bragg symmetric case. The thickness of the CaF₂ crystal was 0.5 cm and the radius of curvature obtained was included between 9 m and infinity for the 333 reflexion and 4.3 m and infinity for the 440 reflexion. All three measurements reported here have been performed at a neutron wavelength \( \lambda = 1.37 \text{ Å} \).

Fig. 4 shows the experimental rocking curve obtained by the authors and the comparison with the theoretical rocking curve deduced from the model for the 440 reflexion and for a crystal curvature of 4.3 m. The dotted straight line represents the theoretical peak reflectivity evaluated by the authors. It appears that the experimental peak reflectivity is lower than the theoretical by a factor of about two, and the experimental width is larger than the theoretical by about the same factor so that the integrated reflecting powers are nearly equal. The discrepancy can be due to a broadening effect arising from the finite width of the primary beam or, more probably, to the non-ideal behaviour of the crystal which, already at the radius of curvature infinity, shows a peak reflectivity of 0.24 instead of the theoretical value 1.

Fig. 5 shows the theoretical diffraction pattern predicted by the model for the 440 reflexion at three different curvature radii (4.3, 5.4, 6.7 m). The full straight line represents the experimental peak reflectivities (detailed rocking curves are not available) and the dotted straight line the theoretical peak reflectivities evaluated by the authors.

Table 1 reports all the possible comparisons between the model calculations and the experimental results. It appears that the experimental peak reflec-

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**Table 1. Comparison between the model calculations and experimental results**

<table>
<thead>
<tr>
<th>Reflexion</th>
<th>( F_{\text{abt}} )</th>
<th>Radius of curvature</th>
<th>Kalus et al. (1973) Model predictions</th>
</tr>
</thead>
<tbody>
<tr>
<td>333</td>
<td>1.96</td>
<td>( \infty )</td>
<td>1.06 0.26 1</td>
</tr>
<tr>
<td>333</td>
<td>10^{-12}</td>
<td>0.41</td>
<td>0.54 0.74 0.53</td>
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<td>333</td>
<td>19.4</td>
<td>0.25</td>
<td>0.60 0.87</td>
</tr>
<tr>
<td>333</td>
<td>13.3</td>
<td>0.46</td>
<td>0.68</td>
</tr>
<tr>
<td>333</td>
<td>13.9</td>
<td>0.47</td>
<td>0.67</td>
</tr>
<tr>
<td>440</td>
<td>6.36</td>
<td>( \infty )</td>
<td>1.04 0.24 1</td>
</tr>
<tr>
<td>440</td>
<td>10^{-12}</td>
<td>0.30</td>
<td>0.99 3.3</td>
</tr>
<tr>
<td>440</td>
<td>5.4</td>
<td>0.67</td>
<td>0.98 3.1</td>
</tr>
<tr>
<td>440</td>
<td>4.3</td>
<td>0.61</td>
<td>0.92 3.7</td>
</tr>
</tbody>
</table>

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**Fig. 4. Experimental neutron diffraction pattern of a CaF₂ curved crystal \( r_{\text{exp}} \) and the prediction of the model \( r_{\text{theor}} \) at \( \lambda = 1.37 \text{ Å} \). Radius of curvature 4.3 m.**
tivities are always much lower than the theoretical values predicted by the model. As already mentioned by the authors, these discrepancies are probably due to the imperfections of the crystals which prevent the observation of dynamical effects. The theoretical values evaluated by the authors are closer to the experimental results than the values obtained by our model. We do not believe that this is due to the greater reliability of their model, which is quite approximate and is also based on an ideal behaviour of the crystal. In fact, the validity of our model was checked by comparison with rigorous dynamical theory calculations and with the experimental data of Albertini et al. (1976). By assuming the reliability of our model, the theoretical data presented here gives quantitative information about the non-ideal behaviour of the monochromator used and constitutes a reference for further work on more ideal monochromators.

Conclusions

Available data on curved neutron monochromators were compared with the predictions of a recent simple model. The agreement was satisfactory for the data concerning Ge monochromators at different curvature radii, neutron wavelengths and reflecting planes. The agreement was poor in the case of CaF₂ monochromators. This was explained by the fact that the monochromators used were not perfect. The theoretical data gives useful information for the design of further monochromators of this kind with a higher degree of perfection. Furthermore the model was modified to include neutron absorption and should be easily extended to the design of curved monochromators for the continuous X-ray spectrum of synchrotron radiation.

APPENDIX

Neutron diffraction by an absorbing curved crystal

Albertini et al. (1976), expressed the reflectivity $r_c$ of a curved crystal in the most general form as

$$ r_c = 1 - \prod_{j=1}^{n} (1 - r_j) \quad (A1) $$

where $r_j$ is the reflectivity of a generically perfect crystal layer, which they defined and which approximates a region of the deformed crystal, and $n$ is the number of layers. In the following, we will correct (A1) for absorption in the case of a curved crystal. For a curved crystal, (A1) is equivalent to (6): in (A1) $j$ represents the index of the generically perfect crystal of region I or II or III of thickness $A = 2/c$. Instead of (A1) which gives the reflectivity of a deformed crystal as a product, one could deduce another expression which gives the reflectivity as a sum of the different contributions from the different layers. The expression obtained by following this procedure is

$$ r_c = r_1 + \sum_{j=1}^{n} r_{j+1} \prod_{k=1}^{j} (1 - r_k). \quad (A2) $$

The generic term appearing in (A2) consists of the factor $\prod_{k=1}^{j} (1 - r_k)$ which represents the probability for a neutron to reach the layer $j+1$, and of the factor $r_{j+1}$ which is the reflectivity of layer $j+1$. It is easy to verify that the two expressions (A1) and (A2) are equivalent.

In order to introduce absorption in (A2), the reflectivity $r_m$ of the layer $m$ of the monochromator must be multiplied by an attenuation factor $a_m$ because of the absorption in the path between the surface and the layer $m$ itself and
between it and the surface. If one assumes an average path of the neutrons, which are all supposed to be reflected at the median depth of the \( m \)th layer, one obtains, in the asymmetric Bragg case, an entrance path given by

\[
\frac{t_0}{\sin \alpha} \left[ (m-1) + \frac{1}{2} \right],
\]

and an emerging path given by

\[
\frac{t_0}{\sin \beta} \left[ (m-1) + \frac{1}{2} \right]
\]

where \( t_0 \) is the thickness of each one of the perfect crystal layers of the model. Then, by calling \( \mu \) the linear absorption coefficient of the crystal material at the given neutron wavelength, one obtains the expression for the attenuation factor,

\[
a_m = \exp \left[ -\mu t_0 (m-\frac{1}{2}) \right] \left[ \frac{1}{\sin \alpha} + \frac{1}{\sin \beta} \right]. \quad (A3)
\]

Therefore, when the neutron absorption is included (42) becomes

\[
r_{ac} = r_1 a_1 + \sum_{j=1}^{n-1} a_{j+1} r_{j+1} \prod_{k=1}^{j} (1-r_k) \quad (A4)
\]

where \( a_1 \) and \( a_{j+1} \) are deduced from (A3). An analogous expression should be valid also for X-ray diffraction by a curved absorbing crystal and could be useful in the design of monochromators for the continuous spectrum of synchrotron radiation.

References


