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Multiple Scattering of X-Rays and Neutrons II. Neutron Multiple Scattering by an Aluminum Single Crystal*

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Some experiences of multiple diffraction of neutrons by an aluminum crystal are described in this paper. The experimental conditions needed to assure that the results are treatable by the analytical methods developed earlier [1,2] are discussed.

It was found that the Soller collimators are not adequate in this technique, as they do not limit the vertical divergence of the incident beam; a new type of collimator, described in the text, had a notable result in the peak resolution.

It is seen that the simultaneous differential equations[2] cease to be valid, as expected, when the crystal approaches perfection, that is, in this context, when the mosaic angular spread η is lower than about 0.01°. Then, the problem has to be treated dynamically.

§1 Introduction

The number of references to the phenomenon of multiple scattering of neutrons is very limited, probably due to the experimental difficulties normally found in this field. Borgonovi and Caglioti in 1962 [3] and Moon and Shull in 1964 [4] dealt specifically with the problem. Spencer and Smith (1960) [5], O'Connor and Sosnowski (1960)[6] and Blinowski and Sosnowski (1960) [7] when studying problems of neutron spectroscopy found multiple scattering of neutrons as causing intensity variations in the reactor spectrum. In the latter three a direct neutron beam from the reactor is used, thus permitting the use of collimators with a few minutes of angular divergence without fearing the decrease in intensity, which is still high enough for the measurements. Consequently they attain a rather good resolution: interaction peaks with half maximum intensity width of 10' to 1', which compare favourably with those obtained by Borgonovi and Caglioti (1962)[3] and by Moon and Shull (1964)[4] ranging from 0.5° to about 4°; these were obtained with a monochromatized beam of a much lower intensity and presumably for this reason could not use such a good collimation. However, in these papers, no details are given, neither as to the type of collimators used nor on their angular divergences.

In this paper, we measure the mosaic spread of the crystal by means of neutron multiple scattering and try to ascertain whether it is significantly different when measured in different directions in the crystal. Resolution then becomes important since it affects simultaneously the intensity of the peaks, from which the value of η is obtained, as well as position and width of the peaks, which are used in the determination of the crystal parameters and their errors as described in Part IV.**

§2 Discussion of Some Experimental Conditions

Since use will be made of the intensity solution found for a crystal plate by Caticha-Ellis (1969)[1] and of the recurrent Taylor series solution given in Part I, the experiments had to be performed so as to satisfy the conditions implicit in those derivations:

1. The mosaic angular distribution function is approximately gaussian and isotropic with standard deviation η.

2. η is much larger than the half maximum intensity width s of the reflection by a single crystallite.

3. The incident neutron beam is approxi-

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mately monochromatic and its angular divergence δ is much smaller than η and larger than s

$$\eta \gg \delta > s$$

4 The cross section of the incident beam should be smaller than the area of the entrance crystal face so that eq (I 8) is applicable

Hypothesis 1 and 2 depend entirely on the crystal under study and will be further discussed on the light of the results obtained Hypothesis 3 is not easy to satisfy in neutron diffraction experiments since it implies in the use of high collimation with serious loss of intensity This item accounts for some of the major experimental difficulties encountered in practice and shall be treated apart under the heading collimation Hypothesis 4 is not well verified in neutron diffraction experiments since in order to obtain a resonable flux of neutrons one must use a beam with a cross section of several square centimeter Whenever this hypothesis is well verified the effective path lengths of the different beams are given by T/γ_i T being the thickness of the plate and γ_i the direction consines with respect to the normal or are limited by absorption according to eq (I 8)

Since the cross section of the incident beam here is comparable with the dimensions of the crystal the path lengths of the different beams depend on all the dimensions of the sample The values calculated which appear in Table I were obtained graphically In this case the coefficient of linear absorption being very low $\mu \simeq 0\ 05\ cm^{-1}$ and the longest path length of about 3 5 cm the value of μl_i is always less than 0 20 As for the values $\bar{Q}_{ij} l_i$ observing that for the reflection (111) of aluminum $Q \simeq 0\ 001$ and that $\bar{Q} = Q/\eta\sqrt{2\pi}$ for values of $\eta \simeq 0\ 05$ one obtains $\bar{Q} = 0\ 45$ and then $\bar{Q} l_0 = 1\ 6$ which does not verify the conditions (I 7) for rapid convergence of the series

§3 Experimental

The experimental arrangement is a classic one it makes use of a lead crystal monochromator (220) three Soller collimators and a BF_3 detector A five axis goniometer of a design due to Prof R A Young * who kindly provided the drawings was built with some minor modifications The additional $\sum$ axis can always be directed in the direction of the scattering vector with obvious advantage when making multiple scattering experiments The internal diameter of the χ-circle (22 5 cm) is big enough to hold crystals of considerable size However when possible it was preferred to mount the crystal so that the ϕ-axis is coincident with the scattering vector and with

*R A Young Private communication (1968)

Table I Effective path lengths and mosaic spread values of an aluminum crystal

Secondary peak (s)	Type	Azimuth ω	l_0 cm	l_1 cm	l_2 cm	l_3 cm	Γ		η
$0\bar{2}0$ $1\bar{3}1$	Trans Trans	28 9	3 63	1 85	3 30	3 01	0 771 ± 9	0 135	+10 −10
$00\bar{2}$ $11\bar{3}$	Trans Trans	31 1	3 60	1 86	3 34	2 97	0 770 ± 9	0 132	+10 −10
$2\bar{2}2$	Refl	34 6	3 58	2 29	2 90	—	0 881 ± 10	0 155	+27 −18
$1\bar{1}1$	Trans	50 1	3 25	1 81	3 15	—	0 892 ± 9	0 147	+25 −17
$13\bar{1}$ $02\bar{2}$	Refl Trans	53 3	3 34	1 74	2 37	3 85	0 811 ± 9	0 142	+22 −12
$13\bar{3}$	Trans	54 8	3 12	1 66	3 75	—	0 934 ± 12	0 170	+30 −25
$33\bar{1}$	Trans	118 7	3 42	1 74	3 66	—	0 916 ± 12	0 132	+32 −25
020	Refl	130 1	3 70	1 79	2 67	—	0 839 ± 10	0 140	+12 −12

the $\sum$ axis This preference is due both to the fact that the mechanical precision of the ϕ axis in this apparatus is higher than that of the $\sum$ axis and to its angular range being 360 the $\sum$ axis has a range limited to 25 or 30 degrees to each side of the vertical position when it starts interfering with the neutron beams

The diffractometer has been semi automatized and is controlled by means of Selsyn motors

The electronic detecting system is formed by two single channels one connected to the diffracted beam and the other to the monitor of the direct beam both being controlled by an electronic programmer Each channel actuates one pen of a double pen recorder thus providing permanent graphic records of the intensities of the incident and diffracted beams The counts obtained in a given interval can be printed by a high speed printer. A detailed account of the mechanical electrical and electronic constructions and controls has been given by Parente (1973) [8]

The aluminum single crystal used in the experiments has dimensions $3\times3\times1$ inches The (111) plane which coincides with the 3×3 face was oriented with its normal along the ϕ axis which was then made to coincide with the $\sum$ axis The data shown in graph form in Figs 1 4 and 5 were measured with the ϕ axis turning at an angular speed of 1 / hour and the counts accumulated every 3 minutes or 0 05 were printed automatically by a high speed printer Each measurement was then attributed to the middle of the angular interval

§4 Collimation

The collimators of the arrangement were initially of the Soller type The horizontal divergence of the first collimator within the reactor can be changed by sliding alternate plates in its body between a maximum value of about 3 and a minimum of 20 In the third collimator where horizontal divergences of 1 11 and 36 are possible the lower value had to be used throughout The divergence of the second collimator is essential for the resolution of the experiment it could be changed from 27 to 8 and then to 4 by sliding additional plates in the same body In Fig 1 the multiple scattering intensity graphs obtained for a crystal of lead of 16 mosaic spread set to reflect (111) when the second collimator has vertical divergences of 27 and 8 are displayed the latter shows as expected a marked increase in resolution corresponding to the fact that $\eta>\delta$ while in the former $\eta<\delta$ A similar run with the 4 collimator did not show an appreciable change interaction peaks of about 0 8° half maximum intensity width with the 8 collimator attained about 0 6° when measured with 4 horizontal divergence at the cost of a substancial loss in intensity Since the mosaic angular spread of the sample, about 16 was 4 times bigger than the hori

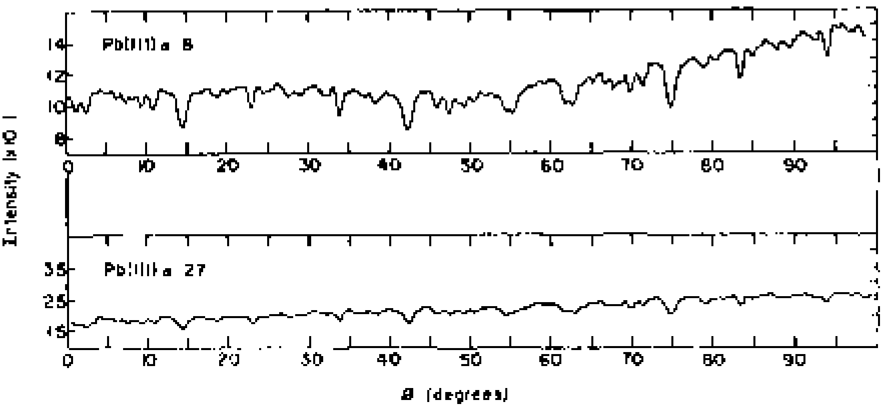

Fig 1 Comparison of resolution obtained with two Soller collimators of 27 and of 8 horizontal divergence on the same reflection (111) of a Pb single crystal with a mosaic spread of about 16 The first graph illustrates the case $\eta>\delta$ and the second $\eta<\delta$ which is a necessary condition in these experiments

zontal angular divergence of the incident beam hypothesis 3 seemed reasonably well satisfied However the resolution attained was not considered to be quite satisfactory Moreover a copper crystal tested under the same conditions showed not entirely consistent angular separations of its interaction peaks A further decrease of the horizontal divergence was impractical but the vertical divergence which had been kept constant at the value of about 4° throughout the measurements could be modified Vertical divergence which is relatively unimportant in simple scattering experiments becomes important in multiple scattering since secondary reflections take place in any orientation within the crystal

In order to limit simultaneously the vertical and the horizontal divergence of the incident beam a new type of collimator was designed and built this consists in a body with four alternate sections of vertical and horizontal plates as shown in Fig 2 The different sections were dimensioned in such a way that plates of the same type in different sections are the exact prolongation of each other The lengths of the different sections are then calculated so that no ray can pass through non consecutive channels Simple geometrical considerations made on Fig 3, followed by the condition of having the same vertical and horizontal divergence allowed

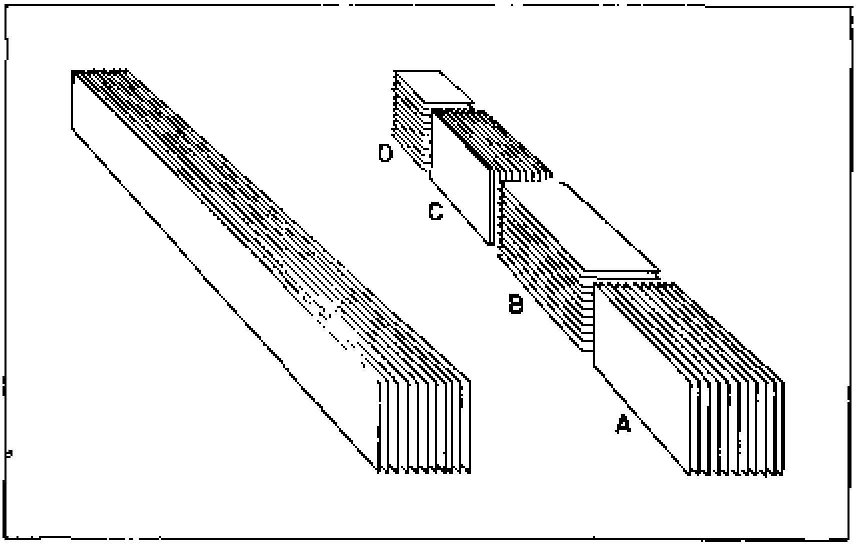

Fig 2 Scheme of a 4-section special collimator which limits simultaneously both horizontal and vertical beam divergences compared with a common Soller collimator

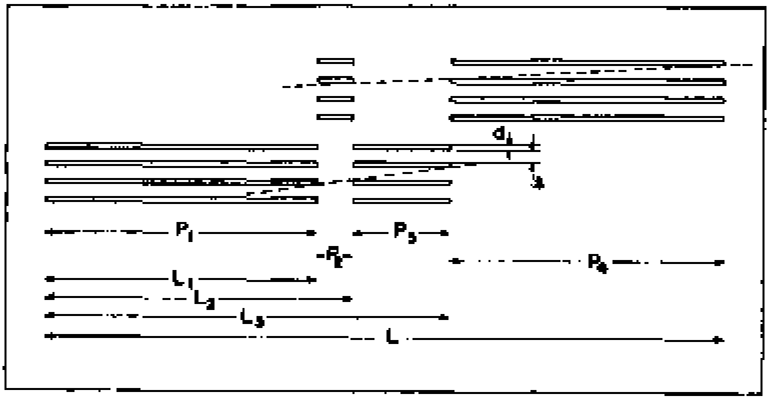

Fig 3 Basic geometrical considerations for the dimensioning of the 4-section collimator No ray can pass through non consecutive channels

the calculation of the dimensions of the 4 sections The conditions imposed from the out set were total length of the collimator (L = 810 mm) thickness of the plates (d = 0 3 mm) and free separation between plates (S = 0 878 mm) The final dimensions of the sections turned out P_1 = 330 4 mm P_2 = 38 1 mm P_3 = 111 1 mm and P_4 = 330 4 mm

The total length of the horizontal plates includes in this design the intermediate space between the corresponding two sections which is however occupied by a vertical plate section Thus the total vertical and horizontal lengths are equal

$$L_v = P_2 + P_3 + P_4 = 479\ 6\ \mathrm{mm}$$

$$L_h = P_1 + P_2 + P_3 = 479\ 6\ \mathrm{mm}$$

The horizontal and vertical divergences have the common value

$$\alpha_h = \alpha_v = 0\ 878/479\ 6 = 1\ 8 \times 10^{-3}\mathrm{rad} \simeq 6\ 3$$

A collimator with only two sections one vertical and one horizontal in order to have the same divergences should be 959 2 mm long The difference between the lengths of a 4 section and a 2 section collimator is a function of $(S+d)/d$ becoming negligible when the plate thickness is $d \ll S$ but it may be considerable if a still lower divergence should be required

The use of this new collimator brought about a considerable improvement in the resolution as shown in Fig 4 which reproduces two runs of a copper crystal taken respectively with the old Soller 4 collimator and the new one for the primary reflection (111) The interaction peak with plane (11$\bar{1}$) for instance had its width reduced from about 50′ to 15 while the total intensities were reduced only in about 20 to 25 % Figure 5 shows a multiple diffraction diagram of an aluminum crystal of 3 × 3 × 1″ oriented so that the (111) plane is continuously reflecting the neutron beam taken with the new collimator while the crystal turned around the (111) vector at an angular speed of 1 /hour

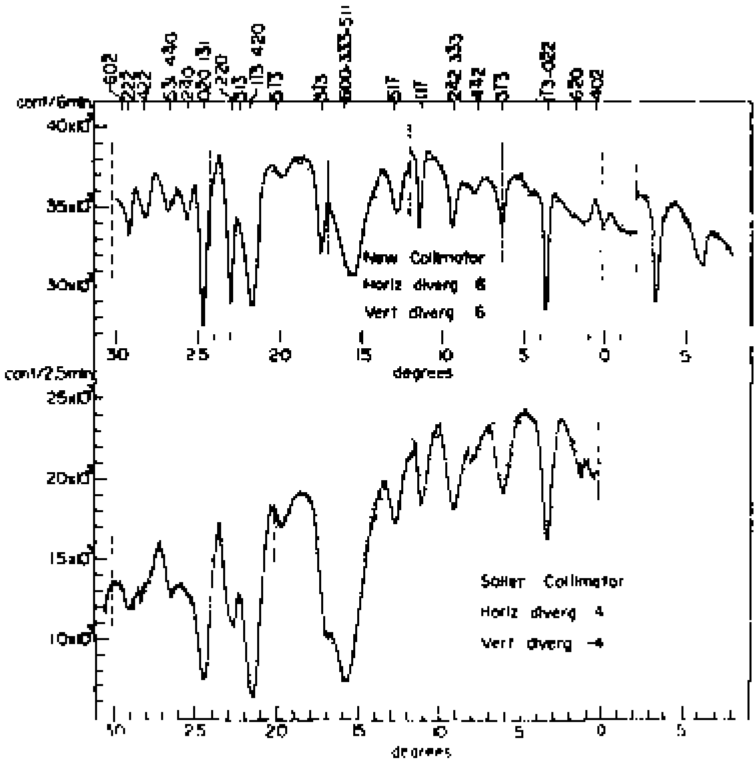

Fig 4 Comparison of the resolution obtained with a Soller collimator of 4 horizontal and about 4 vertical divergence and the special 4 section collimator with both horizontal and vertical divergences of about 6 The sample was a copper crystal of 18 mosaic spread set to reflect (111) permanently

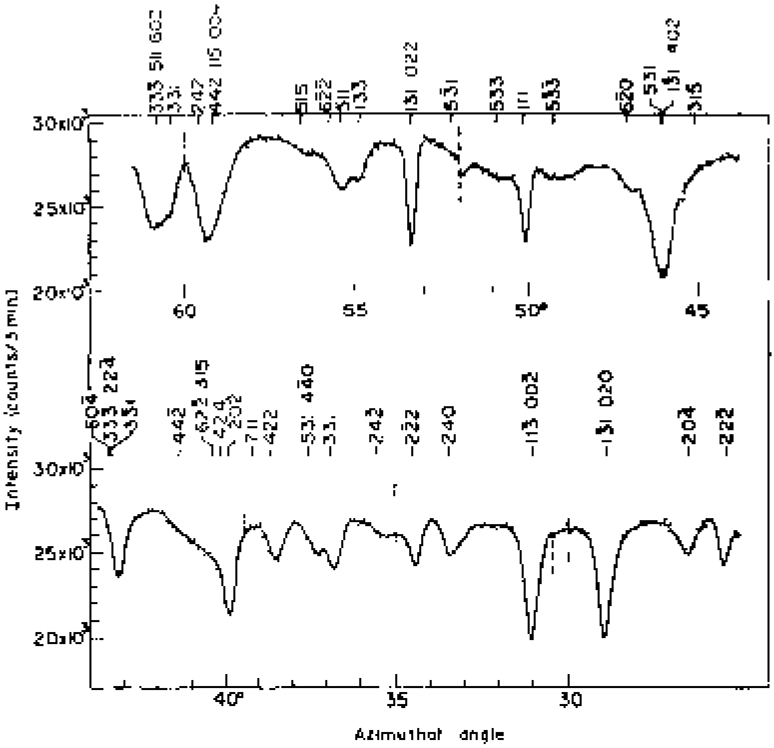

Fig 5 Multiple scattering of neutrons by an aluminum crystal of 3 × 3 × 1 set to reflect (111) Rotation of 1 /hour Every point corresponds to the counts accumulated in three minutes Vertical traces separate data measured in different days

By comparison with previous works in this field the resolution attained here is considered to be excellent It is also an experimental demonstration of the importance of the vertical divergence of the beam in multiple scattering experiments In normal single scattering work this is not so and common Soller collimators used to limit horizontal divergence are enough to produce a good resolution

As expected from the convolution theorem the improvement in resolution brought about by the lowering of the incident beam divergence had two important consequences better localization of the peak positions and more reliable peak intensities From the accurate difference in position of some interaction peaks and very accurate lattice parameters measured by the pseudo Kossel X ray divergent beam method an extremely precise measurement of the neutron wavelength effectively scattered in a multiple way was possible (Parente and Caticha Ellis Part IV) * The relative intensities of the interaction peaks is subsequently used to determine the mosaic spread η of the crystal

*C B R Parente and S Caticha Ellis to be submitted to Japan J appl Phys

§5 Determination of the Mosaic Spread

The value of η can be accurately obtained (Caticha Ellis 1969) from the ratio Γ between the intensity p_1^1 of the single reflection that is measured in the two beam case (incident and the diffracted primary beam) and the intensity p_1^2 obtained when another plane is simultaneously diffracting

$$p_1^2 = \Gamma\, p_1^1 \qquad (1)$$

The ratio Γ obtained experimentally is equated to the ratio of the expressions obtained in the theoretical treatment for both cases These expressions are taken as functions of η which is left as a variable As the actual solution of the equations is quite involved the reverse procedure was followed i e the value of Γ was calculated for different values of η with the help of a computer Use was then made of the recurrence formula of Parente and Caticha Ellis (Part I) [21] which permitted a rapid calculation

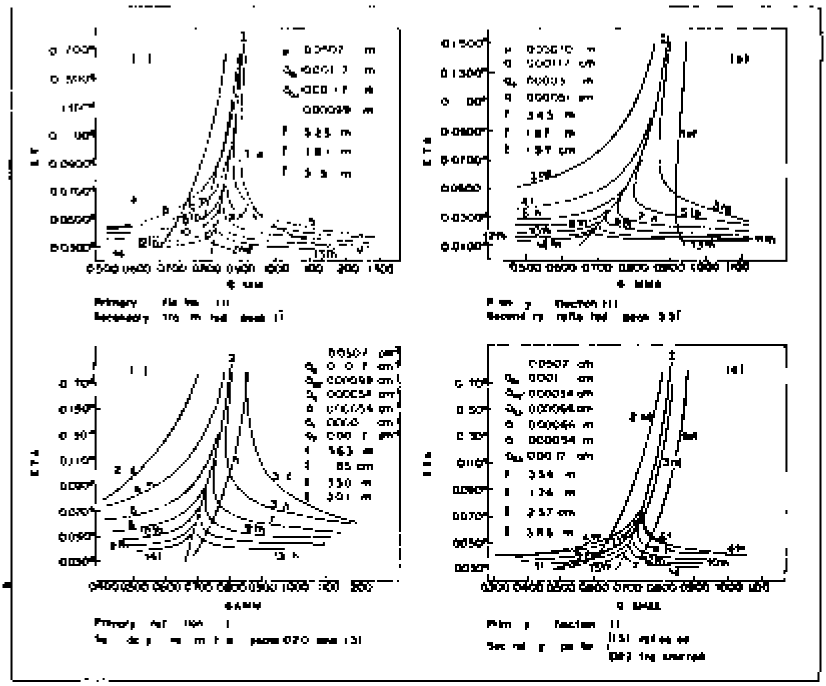

Fig 6 $\eta(\Gamma)$-curves for four different interaction cases in an aluminum crystal with the primary reflection (111)

6a Interaction with a transmitted (Laue case) peak ($\bar{1}\bar{1}1$)

6b Interaction with a reflected (Bragg case) peak (331)

6c Simultaneous interaction with two transmitted peaks (020) and (131)

6d Simultaneous interaction with one reflected ($13\bar{1}$) and one transmitted ($02\bar{2}$) peak

of the successive orders in the Taylor s expansion series In this case a maximum of 14 terms in p_1^n and p_2^n were calculated this being more than enough to obtain a very good approximation

Some of the Γ curves obtained are depicted in Fig 6 The successive curves labelled with the order of the approximation used in their calculation converge towards a central curve obtained by numerical integration of the simultaneous differential equations (1 2) The curves presented here illustrate several different cases Transmitted (Fig 6a) and reflected (Fig 6b) secondary beam two transmitted secondary beams (Fig 6c) and one transmitted and one reflected secondary beams (Fig 6d) Any combination can indeed be treated accurately by using this method of calculation

Table 1 shows the results of several determinations of η for the aluminum crystal from data taken from different interaction peaks In this calculations use were made of point to point measurements of the intensities taken at angular intervals of 0 025° over the peak and at 0 2 otherwise with a counting statistical standard deviations of about 0 4% The heights and widths of the peaks were obtained from adjustment by means of gaussian curves

§6 Conclusions

The analysis of the $\eta(\Gamma)$ curves shown in Fig 6 is very instructive and several conclusions can be drawn from them

1 The convergence of the series is quick for large values and slow for low values of η

2 A few terms perhaps 3 to 6 can give a good approximation for values of $\eta > 0\ 10$ say

3 The curves due to approximations of even and of odd order generally lie on different sides of the central curve This is unimportant from the viewpoint of the accuracy except for very low values of η that is for nearly perfect crystals where the mathematical treatment to establish the set of differential equations (1 2) is no longer valid

4 In this nearly perfect crystal region the curves become nearly horizontal so that the

value of Γ for a given low value of η becomes undetermined This is not entirely due to the series termination since even the central exact curve shows here a tendency to become horizontal It is interesting to note that at this low limit of angular misfit below about 0 10 for this crystal dynamical treatment should certainly be more accurate

5 The alternance of the successive approximations may in some cases be used to obtain a better approximation by taking the average between two successive low orders

6 The results presented in Table I for an aluminum crystal show values of η over 0 10 , which are thus well within the region where a few terms can provide a good approximation The errors quoted, depending on the inaccuracy of the experimental values of Γ are of about the same order of magnitude as the differences found in the values of η for different directions

7 The more steep the central curve the less sensitive is the measurement of η Among the four cases considered in Fig 6 the one of Fig 6b for the reflections (111) and (33$\bar{1}$) is the more sensitive

No theoretical study of the sensitivity of the method has so far been made Such a study would eventually discover some criterion for the selection of the reflections to use when precise values of η are required

Acknowledgements

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