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Crystal Mosaic Spread Determination by Shape Fit Analysis

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Abstract: The crystal mosaic spread determination method is based upon the least square fit of the wavelength distribution of the transmitted neutrons through a slab of a single crystal with the measured one. The method was applied for a crystal cut along ($h_ck_cl_c$) lattice plane when it is oriented by an angle ψ with respect to the incident neutron beam direction and satisfying a single Bragg reflection from a certain (hkl) plane. A computer code CRYSHAPE is developed for calculation and fit shape analysis. The deduced crystal angular mosaic spread value was found to be within accuracy of 1.2 min of arc. The method was applied to determine the mosaic spread value of lead single crystal cut along its (311) plane and zinc (002) plane. The measurements of the transmitted neutrons were performed using both time-of-flight and fixed-angle scattering spectrometers, installed in front of the of the ET-RR-1 reactor horizontal channels.

Key words: Crystal mosaic spread, Pb, Zn, single crystals

INTRODUCTION

Large single crystals are intensively used for neutron diffraction work and especially for monochromatization[1]. Now-a-days single crystals have found a wide application as thermal neutron band pass filters for fission reactors[2-5]. For such purpose one may make use of a high quality single crystal, i.e. the standard deviation η of the angular distribution of the subgrain structure from an average direction over all grains (mosaic spread) of such crystal must be a few minutes of arc.

X-ray methods are commonly used for the mosaic spread determination of single crystals[6]. However the X-ray methods are not efficient for studying the mosaic structure of light elements. The FWHM of neutron rocking curve for single crystal gives an estimate of the mosaic spread value. However crystal surface roughening and the geometrical spread of the collimating system limits the application of such method[7,8].

Adib *et al.*[9] proposed a neutron transmission method for the crystal mosaic spread determination. The method is based on measuring the FWHM of the wavelength distribution of the neutrons transmitted through a crystal at different angles ψ, where ψ is the angle between the perpendicular to the crystal surface and the incident neutron beam. The relation between the FWHM of the distribution and $\sin^2\psi$ was found to be a straight line whose slope could yield the crystal mosaic spread. This method has been successfully applied to determine the mosaic spread of pyrolytic graphite crystal[9]. However the method is restricted for crystals having small neutron absorption cross-sction, as in case of pyrolytic graphite crystal, since the attenuation of the incident neutron beam when passing through such crystals will be more pronounced and thus the shape of the neutron transmission curve through such crystal will be distorted. Moreover, the wavelength distribution of the transmitted neutrons due to reflection from the crystal surface at different ψ may be overlaped with reflections from other (hkl) planes. Thus the straight line relation will not be valid.

The present work deals with a mosaic spread determination method using the shape fit analysis of the wavelength distribution of the transmitted neutrons when the crystal is set at Bragg reflection from a certain (hkl) plane, with the measured one.

Principal of the method: The neutron transmission through a slab of single crystal is given by

$$T = e^{-Nt_0\sigma} \tag{1}$$

where N is the number of atoms per cubic centimeter, t_0 is the effective thickness of the crystal in cm and σ is the total cross-section which is given by[10].

$$\sigma = \sigma_{abs} + \sigma_{tds} + \sigma_{Bragg} \tag{2}$$

The first contribution σ_{abs} for most of the elements obeys the 1/v law where v is the neutron velocity.

The second contribution σ_{tds} is the sum of single and multi-phonon-scattering cross-section and can be calculated as given by Freund[10] from the crystal physical parameters. Following Naguib and Adib[11], the Bragg scattering cross-section σ_{Bragg} by a single crystal due to reflection from a certain (hkl) plane is given by:

$$\sigma_{Bragg} = -\tfrac{1}{Nt_o}\ln(1 - P^{\theta}_{hkl}) \qquad (3)$$

where P^{θ}_{hkl} is the reflecting power of the (hkl) plane inclined by an angle θ_{hkl} to the neutron beam direction and is given by Bacon[7].

$$P^{\theta}_{hkl} = \frac{a}{1 + a(1+2a)^{1/2}\cot[b(1+2a)^{1/2}]} \qquad (4)$$

where:

$$b = {}^{\mu t_o}/_{\cos\theta_{hkl}}$$
$$a = {}^{Q_{hkl}W(\Delta)}/_{\mu}$$

in which μ is the linear absorption coefficient and Q_{hkl} is the well known crystallographic quantity.

W(Δ) is the distribution of the crystal's mosaic blocks and defind so that W(Δ)dΔ is the fraction of mosaic blocks which have their normals between the angles Δ and Δ+ dΔ from the crystal surface. There is good evidence that W(Δ) has a Gaussian form and can be written[7] as:

$$W(\Delta) = \tfrac{1}{\eta\sqrt{(2\pi)}}e^{-\Delta^2/2\eta^2} \qquad (5)$$

where, η is the standard deviation of the mosaic blocks,

Let the angle between the neutron beam direction and the direction of the cutting plane $[h_c,k_c,l_c]$ (i.e.perpendicular to the crystal suface) be ψ then following Naguib and Adib[12] the direction cosine of the diffracted beam can be given as:

$$\cos\theta_{hkl} = \frac{(hh_c + kk_c + ll_c)\cos\psi + l_c\begin{pmatrix}\dfrac{hh_c+kk_c}{\sqrt{h_c^2+k_c^2}}\\ -\dfrac{l\sqrt{h_c^2+k_c^2}}{l_c}\end{pmatrix}\sin\psi}{\sqrt{h_c^2+k_c^2+l_c^2}.\sqrt{h^2+k^2+l^2}} \qquad (6)$$

For face centered cubic structure (fcc) with lattice parameter a_o.
and

$$\cos\theta_{hkl} = d_c d_{hkl}\left\{\begin{matrix}\left[\begin{matrix}\dfrac{4}{3a_o^2}\left\{hh_c + kk_c + \begin{pmatrix}hk_c\\ +kh_c\end{pmatrix}/2\right\}\\ +ll_c/c_o^2\end{matrix}\right]\cos\psi + \\ +\dfrac{2l}{\sqrt{3}a_oc_o}\left[M_c\begin{pmatrix}hh_c + kk_c + \begin{pmatrix}kh_c\\ +hk_c\end{pmatrix}\\ /2 - \dfrac{1}{M_c}\end{pmatrix}\right]\sin\psi\end{matrix}\right\} \qquad (7)$$

Where:

$$d_c = \frac{1}{\sqrt{\dfrac{4}{3a_o^2}\left(h_c^2 + k_c^2 + h_ck_c\right) + \dfrac{l_c^2}{c_o^2}}}$$

and

$$M_c = \frac{1}{\sqrt{h_c^2 + k_c^2 + h_ck_c}}$$

For hexagonal closed packing structure (hcp) with lattice constants a_o and c_o

CRYSHAPE program was developed for the use with personal computers to calculate the neutron transmission for single crystal set at an angle ψ at a which single Bragg reflection from (hkl) plane took place. The code can also provide the fitting of the calculated transmission with the experimental data and defining the crystal mosaic spread value.

For the fit of the calculated wavelength distribution of the transmitted neutrons with the experimental one, the program takes into consideration the effects of both neutron wavelength resolution and incident neutron beam divergence. As reported by Naguib and Adib[11], they assumed that these resolutions for simplicity have a rectangular shape with wavelength spread Δλ.

However, it was shown that the time spread and consequently the wavelength spread Δλ in the TOF spectrometry is the convolution of several independent uncertainties (burst time, time channel width, neutron detector radius, etc.[12]). They reported that the resolution function can be considered to have a Gaussian distributution where the FWHM of the wavelength spread Δλ of the distribution can be given as:

$$\Delta\lambda = \frac{26.9}{L}\sqrt{\frac{2h^2}{\omega^2r^2} + \Delta^2 + \left(\frac{2\beta}{\omega}\right)^2}\ \ldots\ldots\ldots\ldots\ldots\ldots\text{nm} \qquad (8)$$

Where, r (in m) is the shopper radius having slit width h (in m) and angular velocity ω (in $\sec^{-1}$), Δ (in sec) is the channel time width of the used time analyzer, L (in m) is the flight path, β is the FWHM of the angular divergence of the experimental arrangement.

Therefore the effect of both wavelength resolution and beam divergence in CRYSHAPE program are calculated[7].

Neutron measurements: The neutron transmission measurements were carried out for lead and zinc single crystals in the neutron wavelength range from 0.04-0.52 nm using both TOF and fixed-angle scattering spectrometers, installed in front of the ER-RR-1 reactor horizontal channels. The wavelength spread of the poly-energetic burst of the TOF spectrometer was 0.006 nm.

The lead single crystal cut along (311) plane used has a parallelepiped shape with dimensions 6x17.6x1.1 cm^3. The 6x17.6 cm^2 face was parallel to its cutting plane.The zinc single crystal cut along (002) was a segment of a cylindrical shape 8 cm in diameter. The (002) plane was parallel to the face of the cylindical segment with cross-sectional area 6x16 cm^2.

The measured neutron transmission data of lead and zinc versus neutron wavelenght at different Ψ, reported by Adib *et al.*[13] are displayed in Fig 1and 2 respectively.

From the lead experimental transmission data, the dips at neutron wavelengths 0.4484, 0.3884 and 0.5332 nm due to reflections from (200) plane at Ψ = 0, (111) at Ψ =31° and (1$\bar{1}$) at Ψ = 42° respectively were selected for shape fit analysis. While reflections from (002) plane at Ψ = 18°, 32° and 51° were selected for zinc.

RESULTS AND DISCUSSIONS

The formula given by Eq. 1 was calculated versus neutron wavelength and fitted to experimental data for the selected reflections from (hkl) planes of lead and zinc crystals. The main physical parameters required in these calclations are listed in Table 1.

Lead: The lead coherent scattering cross-section due to neutron reflection from the (200) plane versus neutron wavelength, when the neutron beam incident perpendicular to the crystal surface i.e. at Ψ = 0° were calculated using Eq. 3 assuming different values of crystal mosaic spread. The result of calculation is displayed in Fig. 3a. Similar calculations were carried out for neutron reflection from (111) plane at Ψ = 31° and (11$\bar{1}$) at Ψ = 42°

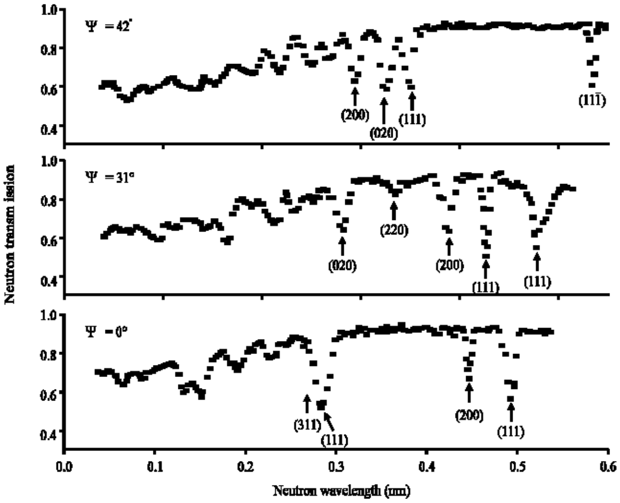

Fig. 1: The measured neutron transmission through lead single crystal[13]

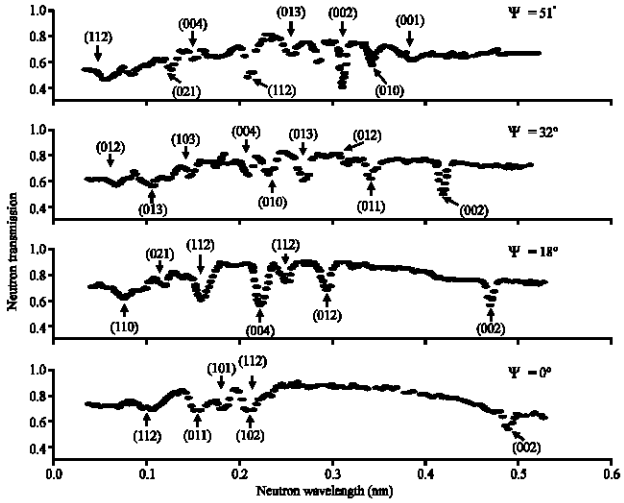

Fig. 2: The measured neutron transmission through zinc single crystal[13]

Table 1: Physical parameters of lead and zinc

	Lead	Zinc
Atomic weight	207.21	65.39
Crystal structure	FCC	HCP
Lattice constant	a_o = 0.495 nm	a_o=0.2665 nm c_o=0.4947 nm
Number of atoms per unit cell	4	2
Atomic positions	000, 1/2 1/2 0, 1/2 0 1/2, 0 1/2 1/2	1/3 2/3 1/4, 2/3 1/3 3/4
Number of unit cells cm^{-3}	0.8245 E+22	3.28 E+22
Debye temperature	200 K	327 K
Neutron capture cross-section at 0.025 eV	0.017 barn	1.11 barn
Total scattering cross-section	11.106 barns	4.131 barn
Coherent scattering amplitude	9.3 fm	5.68 fm

and displayed in Fig. 3b and 3c, respectively. As expected the distribution of the Bragg coherent scattering cross-section is broadening with the increase of the mosaic spread value and the coherent scattering cross-section reaches a maximum value at neutron wavelength satisfying the Bragg condition $\lambda = 2d_{hkl} \sin \theta_{hkl}$.

The neutron transmission through 1.1 cm lead single crystal cut along (311) plane was calculatd as a function of neutron wavelength with steps of 0.001 nm for the case when the neutrons incident perpendicular to the crystal surface assuming different mosaic spread values η. The CRYSHAPE code convoluted the transmission values with the Gaussian distribution of the resolution function of the TOF spectrometer having FWHM of wavelength spread 0.006 nm. The sum of $\chi^2(\eta)$ were calculated versus mosaic spread as:

$$\chi^2(\eta) = \sum_i^n \left(T_{cal}(i) - T_{exp}(i)\right)^2$$ where, T_{cal} (i) and T_{exp} (i)

are the calculated and experimentally measured trasmission respectively at the same neutron wavelength λ. The result of calculation of χ^2 versus crystal mosaic spread is displayed in Fig. 4a. Similar calculation of χ^2 were carried out at $\Psi = 31°$ and $\Psi = 42°$ and displayed in Fig. 4b and 4c, respectively. Figure 4 shows that for the three cases χ^2 reaches a minimum value at $\eta = 2.8 \pm 0.4$ m radian. The obtained lead mosaic spread was found to be in reasonable agreement with the value 2.9 m radian. (10 min of arc) given by the manufacturer (The Polish Academy of Science, Institute of Nuclear Research)[13].

The calculated neutron transmission through lead single crystal at minimum χ^2 are displayed in Fig. 5 for the three cases. For comparison the experimental transmission data are also displayed in the same figure. It is evident that the calculated transmission values fit well the experimental ones.

Zinc: It was found that the dips in the measured transmission curve through Zn single crystal due to reflection from (002) plane at angles Ψ = 18°, 32° and 51° are suitable for carring out the fit shape analysis. Since these dips are single and symetric. The observed dip

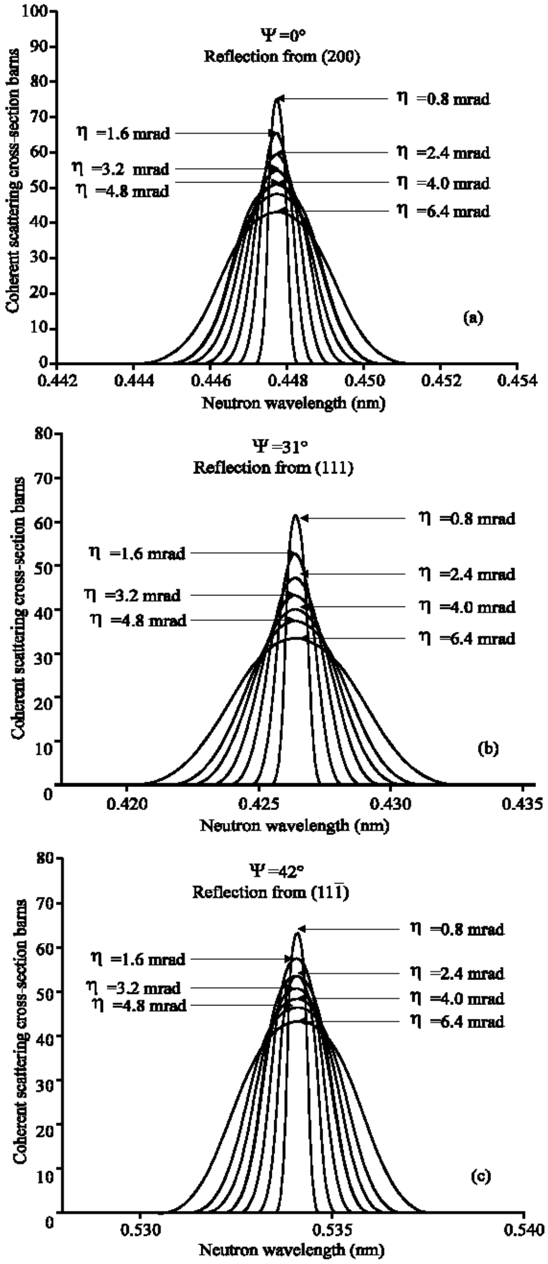

Fig. 3: Coherent scattering cross-section of lead at different mosaic spread values

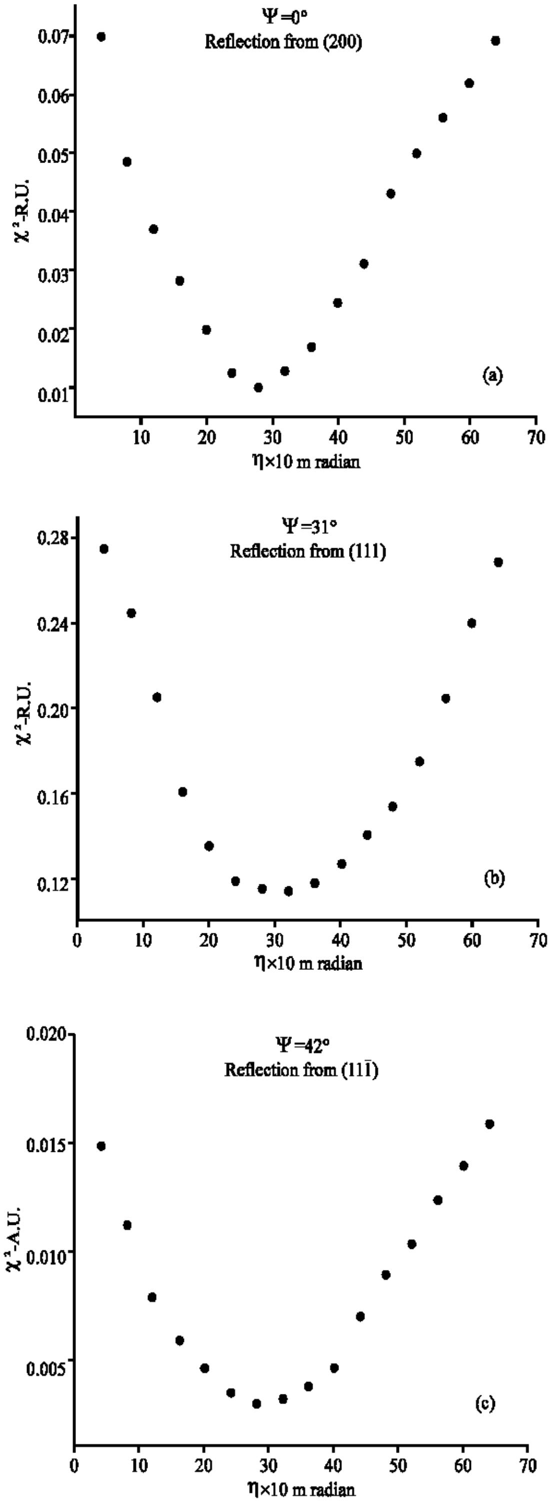

Fig. 4: χ^2 at different mosaic spread values of Pb

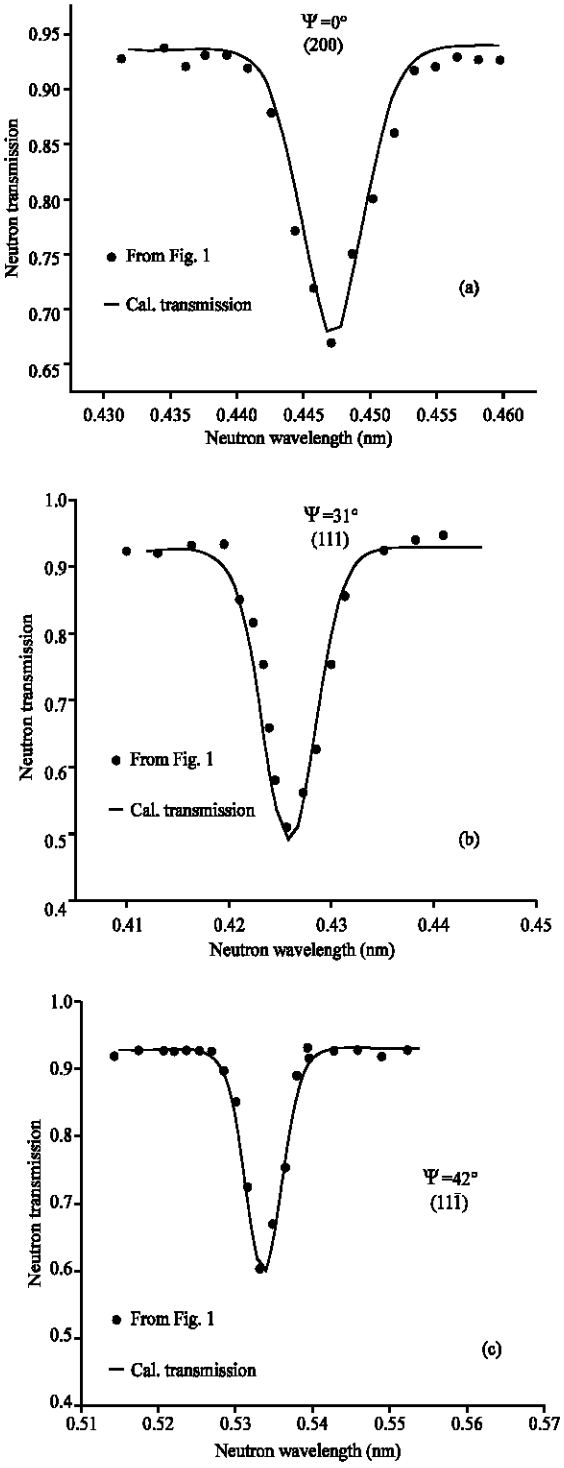

Fig. 5: The calculated neutron transmission through Pb single crystal at minimum χ^2

arround 0.47 nm when the zinc single crystal was set at Ψ = 18° was found to satisfy Bragg equation: $\lambda = 2d_{002} \cos 18$

The neutron transmissions as a function of neutron wavelength arround the dip at $\lambda = 0.47$ nm and assuming constant wavelength spread of the incident neutron beam

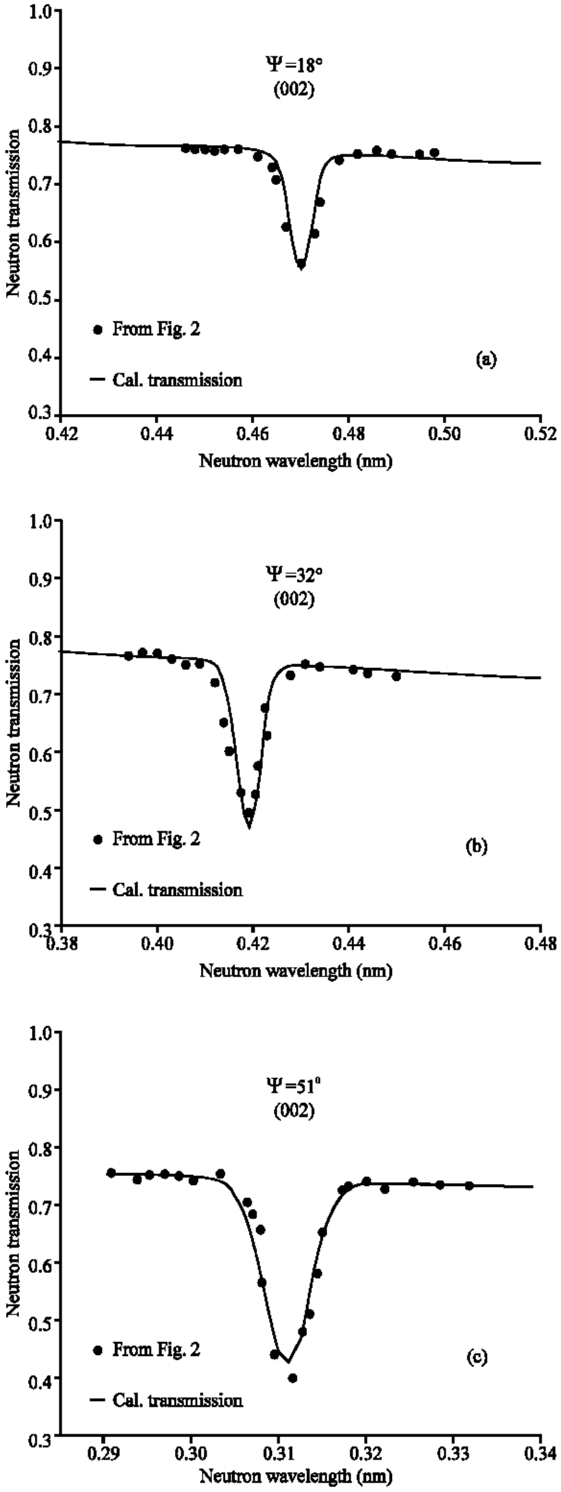

Fig. 6: The calculated neutron transmission through Zn single crystal at minimum χ^2

were calculated at different crystal mosaic spread values η. The χ^2 were deduced at each in similar way as for lead.

The result of calculation shows that χ^2 has a minimum value at η = 2.4 m radian. Figure 6a displays the calculated neutron transmission at minimum χ^2 as solid line along with the measured data as dots. The figure shows that the calculated values fits the experimental ones.

Similar calculations were carried out at Ψ = 32° and 51° and the calculated neutron transmission data at minimum χ^2 along with the measured ones are also displayed in Fig. 6b and c, respectively.

The least square fit for the three cases shows that the mosaic spread of zinc was found to be (2.4±0.4) m radian. The obtained zinc mosaic spread was found to be in reasonable agreement with the value 10 min of arc given by the manufacturer (the Polish Academy of Science, Institute of Nuclear Research[13]).

CONCLUSIONS

The developed method permits the determination of the crystal mosaic spread within accuracy of 1.2 min of arc, which is sufficient when the crystal is used as neutron monochromator and filter. The accuracy could be improved by using high resolution TOF spectrometer. Moreover, the method can be applied directly on the site at the reactor channel.

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