NEUTRON TRANSMISSION THROUGH BULK IMPERFECT SINGLE- CRYSTALS

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A formula is given that permits the calculation of the neutron transmission through bulk imperfect single crystals as a function of wavelength at both room and liquid nitrogen temperatures. The formula takes into consideration the crystal structure type and the orientation of its cutting plane w. r. t. the incident neutron beam direction. The reflecting power of the Bragg reflections from the allowed (hkl) planes, are calculated, assuming that the bulk single-crystal consists of thin layers where, the extensions inside each layer are negligible.

A computer code BISCF written in FORTRAN-77, has been developed to carry out the required calculations for cubic and hexagonal closed packing (HCP) crystal structure types. An overall agreement is indicated between the experimental neutron transmission data and the calculated ones. Such agreement justifies the applicability of the computer code.

Keywords: Cu , Zn & MgO imperfect single-crystals, neutron transmission.

INTRODUCTION

It was found by several authors [1-3], that the cross-section of single crystals at thermal neutron energies is very small at room temperature and could be reduced by further cooling the crystal. Accordingly, bulk single crystals could be used as thermal neutron filter which could pass thermal neutrons and exclude both fast neutrons and gamma rays [4]. However the parasitic Bragg scattering due to the reflections from crystal planes may limit its use as a thermal neutron filter [5].

The use of bulk imperfect single-crystals of various materials as filters for thermal neutron beams has long been known [6]. Several materials such as quartz
(SiO$_2$) [2], bismuth [6], silicon [7], lead [8], sapphire (Al$_2$O$_3$) [9-11], magnesium oxide (MgO) [12,13], magnesium fluoride (MgF$_2$) [14] have been suggested as most successful filter materials. At higher neutron energies, greater than about 1eV, the total neutron cross section $\sigma_t$ of each of the above mentioned materials is in the range of a few barns, but at neutron energies less than 0.1eV, where much of the coherent Bragg scattering is disallowed the effective cross-section for single-crystal specimens is reduced. That is also due to the decrease of the thermal diffuse scattering (TDS).

Naguib and Adib [15,16] reported a computer programs which allows calculating the total thermal cross-section of imperfect Cu & Zn-single crystals as a function of crystal constants: temperature and neutron energy, in the energy range between 3 meV to 10 eV. Recently [8,11,13], used different computer codes for calculations the neutron transmission through lead, Al$_2$O$_3$ and MgO. However, in these works, the effect of the primary and secondary extensions of the Bragg reflections inside bulk single crystals has not been considered. Therefore the present work deals with the development of a code for calculating the neutron transmission through cubic and hexagonal closed packing (HCP) crystal structure types. Moreover, the reflecting power of the Bragg reflections from the allowed (hkl) planes are calculated taking into account the primary and secondary extensions inside the bulk crystal.

**THEORETICAL TREATMENT**

The total cross-section determining the attenuation of neutrons by single-crystal is given by:

$$\sigma = \sigma_{abs} + \sigma_{tds} + \sigma_{Bragg}$$  \hspace{1cm} (1)

where $\sigma_{abs}$ the absorption cross-section due to nuclear capture processes, $\sigma_{tds}$ is the thermal diffuse scattering (TDS) and $\sigma_{Bragg}$ corresponds to elastic or Bragg scattering. As shown by Freund (1983) [3], the second contribution $\sigma_{tds}$ can be split in two parts, $\sigma_{mph}$ (multiple phonon) and $\sigma_{sph}$ (single phonon), depending on neutron energy. The single phonon scattering cross-section, concerns the energy range $E \ll k_B\theta_D$, where $k_B$ is Boltzmann’s constant and $\theta_D$ is the Debye temperature characteristic of the material. It is determined by phonon annihilation processes. While $\sigma_{mph}$ is predominant in the range $E \gg k_B T$.

The contribution of Bragg scattering $\sigma_{Bragg}$ to the total attenuation arises from coherent elastic scattering due to reflections from different (hkl) planes. In the case of single-crystal, the Bragg scattering cross-section is given by Naguib and Adib (1996) [15]:

\begin{align*}
M. Adib et al. & \quad 2
\end{align*}
\[ \sigma_{\text{Bragg}} = \frac{1}{N t_0} \ln \left( \prod_{hkl} \frac{1}{(1 - P_{hkl}^0)} \right) \]  

(2)

where, \( N \) is the number of atoms/cm\(^3\), \( t_0 \) is the thickness of the crystal in the beam direction, and \( P_{hkl}^0 \) is the reflecting power of the \((hkl)\) plane inclined by an angle \( \theta_{hkl} \) to the incident beam direction. In the case of imperfect crystal of finite absorption the reflecting power \( P_{hkl}^0 \) of the \((hkl)\) plane inclined by an angle \( \theta_{hkl} \) to the incident beam direction is given by Bacon (1975) [17]:

\[ P_{hkl}^0 d\theta = \frac{a \chi d\theta}{1 + a + (1 + 2a)^{1/2} \coth[A(1 + 2a)^{1/2}]} \]  

(3)

for the reflection method and;

\[ P_{hkl}^0 d\theta = \chi \sinh(Aa) e^{-A(1+a)} d\theta \]  

(4)

for the transmission method, where

\[ d\theta = d\lambda/2d_{hkl} \cos \theta \]

\[ A = \mu t_0/\gamma_o \]

\[ a = \frac{Q_{hkl} W(\theta)}{\mu} \]

\[ \mu \] is the linear absorption coefficient. \( W(\theta) \) has a Gaussian distribution with Standard deviation \( \eta \) on mosaic blocks of single crystal. The \( Q_{hkl} \) is crystal-graphic quantity given by Bacon (1975) [17]:

\[ Q_{hkl} = \lambda^3 N_c^2 F^2 / \sin 2\theta \]

As shown by Beacon (1975) [17], the integrated reflectively

\[ R^0 = \int_{-\infty}^{\infty} P_{hkl}^0 d\theta \] from imperfect crystal reaches saturation for bulk single crystal with thickness \( t_0 \). Such behavior is due to the primary and secondary extensions inside the filter. Therefore, when a bulk single-crystal is used as a neutron filter in transmission geometry, to take into account the effect of extensions within the crystal thickness \( t_0 \), Eq. (7) is calculated for a thickness \( t_s \) and Eqs. (5&6) is multiplied by a factor \( \chi = t_0/t_s \), where \( t_s \ll t_0 \).

The integrated reflection \( R^\lambda \) for a fixed crystal in white radiation beam is given by:

\[ R^\lambda = R^0 2d_{hkl} \cos \theta \]  

(6)

Since the reflecting power \( P_{hkl}^0 \) for an imperfect single crystal depends upon the
direction cosine of the diffracted beam $\gamma_{hkl}$ and the inclination angle of the $(hkl)$ plane to the crystal surface $\alpha_{hkl}$. Therefore in the present work we determine $\gamma_{hkl}$ and $\alpha_{hkl}$ for cubic and Hexagonal closed packing crystal structures.

1- Cubic structure

For cubic crystal structure with lattice constant $a_0$, the equation describing the cutting plane $(h_kl_c)$ which is parallel to the crystal surface can be given as:

$$\sqrt{h^2 + k^2 + l^2} Z = a_0$$

(7)

The interplanar distance $d_{hkl}$ is given by the relation

$$d_{hkl} = \frac{a_0}{\sqrt{h^2 + k^2 + l^2}}$$

(8)

If the angle between the neutron beam direction and the direction $(h_kl_c)$ is $\psi$, then $\gamma_0 = \cos \psi$, while, the direction cosine of the diffracted beam $\gamma_{hkl}$ from any $(hkl)$ plane can be expressed as:

$$\gamma_{hkl} = \frac{(hh_c + kk_c + ll_c) \cos \psi + l_c \left[ \frac{hh_c + kk_c}{\sqrt{h^2 + k^2}} - \frac{l \sqrt{h^2 + k^2}}{l_c} \right]}{\sqrt{h^2 + k^2 + l^2} \cdot \sqrt{h^2 + k^2 + l^2} \sin \psi}$$

(9)

while, its inclination angle $\alpha_{hkl}$ to the crystal surface is given by :

$$\cos \alpha_{hkl} = \frac{(hh_c + kk_c + ll_c)}{\sqrt{h^2 + k^2 + l^2} \sqrt{h^2 + k^2 + l^2}}$$

(10)

If the cutting plane is $(00l_c)$, then $\gamma_{hkl}$ becomes

$$\gamma_{hkl} = \frac{l \cos \psi + k \sin \psi}{\sqrt{h^2 + k^2 + l^2}}$$

(11)

$$\cos \alpha_{hkl} = \frac{l}{\sqrt{h^2 + k^2 + l^2}}$$

(12)

2- Hexagonal closed packing structure

The equation of cutting plane $(h_kl_c)$ plane for HCP structure with lattice constants $a_o$ and $c_o$ is given by: $Z/d_c = 1$ and the equation of any $(hkl)$ plane is:
Debye temperature

A schematic diagram of flow chart of a computer code BISCF is given in Fig. (1)

\[ \text{transmissions are adjusted by the Gaussian resolution function (with standard deviation } \delta \lambda) \text{ of the experimental facility [18].} \]

A computer code Bulk Imperfect Single Crystal Filter (BISCF) written in FORTRAN-77, has been developed to carry out the required calculations for cubic and hexagonal closed packing crystal structure types. For comparison of the calculated neutron transmission versus wavelength with the experimental data, the calculated transmissions are adjusted by the Gaussian resolution function (with standard deviation \( \delta \lambda \)) of the experimental facility [18].

A schematic diagram of flow chart of a computer code BISCF is given in Fig. (1)

**COMPARISON WITH EXPERIMENT**

The main physical parameters of single crystals used for calculations are listed in Table 1.

**Table 1.** The main physical parameters of single crystals used for calculations.

<table>
<thead>
<tr>
<th>Element</th>
<th>Copper</th>
<th>Zinc</th>
<th>MgO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic weight</td>
<td>63</td>
<td>65.39</td>
<td>20.15</td>
</tr>
<tr>
<td>Crystal structure</td>
<td>F.C.C</td>
<td>H.C.P</td>
<td>F.C.C.</td>
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<tr>
<td>Lattice parameters</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>(a_0 = 0.3615 \text{ nm})</td>
<td>(a_0 = 0.2665 \text{ nm})</td>
<td>(a_0 = 0.4212 \text{ nm})</td>
</tr>
<tr>
<td></td>
<td>(c_0 = 0.4947 \text{ nm})</td>
<td>(c_0 = 0.4947 \text{ nm})</td>
<td>(c_0 = 0.4947 \text{ nm})</td>
</tr>
<tr>
<td>FWHM (degree)</td>
<td>0.5°</td>
<td>0.5°</td>
<td>0.5°</td>
</tr>
<tr>
<td>No. of unit cells/m^2</td>
<td>2.116E+28</td>
<td>3.286E+28</td>
<td>1.337E+28</td>
</tr>
<tr>
<td>Debye temperature (\theta_D)</td>
<td>362 K</td>
<td>327 K</td>
<td>938.2 K</td>
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### Atomic positions

<table>
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<tr>
<th></th>
<th>Mg: (0,0,0; 0,0.5,0.5; 0.5,0,0.5; 0.5,0.5,0)</th>
<th>O: (0.5,0.5,0.5; 0.0,0.5; 0,0.5,0.5)</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>0,0,0; ½, ½, 0; 0, ½</td>
<td>½, ½, ½; ½, ½, ½</td>
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### Coherent scattering length b (fm)

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<tbody>
<tr>
<td>b thigh</td>
<td>7.718</td>
<td>5.68</td>
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### Neutron capture cross-section at 0.025eV(barns)

<table>
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<tbody>
<tr>
<td>3.788</td>
<td>1.11</td>
<td>0.063</td>
</tr>
</tbody>
</table>

### Total scattering cross-section ($\sigma_{int}$)(barns)

<table>
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<th>O: 5.805</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.03</td>
<td>4.131</td>
<td>7.942</td>
</tr>
</tbody>
</table>

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**Figure 1.** A schematic diagram of flow chart of the computer code.
1- Cu single-crystals

The neutron transmission through 10mm Cu(002) single crystal at $\psi = 13.4^\circ$ as a function of wavelength $\lambda$ is calculated using the BISCF code assuming different values of $t_s$. The calculated transmission values are compared with experimental ones measured by Naguib & Adib, (1998)[16]. While through 10mm Cu (111) single crystal at $\psi = 30^\circ$ is compared with the experimental ones measured by Naguib & Adib (1996)[15]. The result of calculation is displayed in Fig. (2 a&b), respectively. Fig. (2) shows that the calculated neutron transmission excluding the range of the Bragg dips are in reasonable agreement with experimental ones. Moreover, with decreasing $t_s$ the shape and dip area becomes closer to the experimental data.

To show the effect of single crystal layer thickness $t_s$ on the integrated reflectivity $R^\lambda$ from Cu (002), $R^\lambda (t_s)$ is calculated for both (111) plane (at $\lambda_{111}=0.29$ nm) and (002) plane (at $\lambda_{002}=0.35$ nm). The result of calculation is displayed in Fig. (3a&b), respectively. While for Cu (111) single crystal set at $\psi = 30^\circ$ for reflection from (111) plane (at $\lambda_{111}=0.361$ nm), is displayed in Fig. (3c).

![Figure 2](image)

Figure 2. Neutron transmission through Cu single-crystal at different layer thickness $t_s$.

Figure (3) shows that, the integrated reflectivity $R^\lambda$ is decreasing with increasing the value of layer thickness $t_s$. However, it is almost constant for layer thickness less than 60 $\mu$m. The calculated $R^\lambda_{111}$ for Cu (002) at $\psi = 13.4^\circ$ was found to be in agreement with experimental value reported by Naguib and Adib (1998)[16] for layer thickness $t_s$ ranging from 10 - 60 $\mu$m. The same range of layer thickness was found for $R^\lambda_{002}$. While, from Fig.(3c), the agreement between the calculated $R^\lambda_{111}$ from Cu (111) and the experimental one was found for layer thicknesses $t_s$ from 20-50 $\mu$m.
2- MgO single-crystal

The experimental neutron transmission data through 100 mm thick ($\eta = 0.5^0$) (FCC) MgO(100) single crystal at $\psi=0^0$ at both R.T. & L.N. temperature reported by Thiyagarajan et al., (1998) [12], are displayed in Fig.(4) as closed circles. For comparison, the calculated transmission data for various values of $t_s$ are also displayed in Fig. (4) as solid lines.

Figure 3. $R^2$ versus layer thickness $t_s$ for Cu single crystals.

Figure 4. Neutron transmission through MgO single -crystal at different layer thickness $t_s$. 
Figure 5. $R^4$ versus layer thickness $t_s$ for MgO single crystal.

The integrated neutron reflectively $R^4_{420}$ (at $\lambda_{420} =0.16$ nm) and $R^4_{220}$ (at $\lambda_{220} =0.219$ nm) from MgO were calculated under the same conditions for versus layer thickness $t_s$ and displayed in Fig. (5).

Figure (5) also shows that the reflecting power $R^4$ is constant within accuracy 5% for layer thickness $t_s$ from 10 $\mu$m up to 500 $\mu$m. Such wide range of $t_s$ than for Cu is due to the fact that absorption cross-section of MgO is much less than for copper.

3- Zn single-crystal

The neutron transmission through 22mm thick ($\eta=0.5^\circ$) Zn (HCP) single crystal set at glancing angles $\Psi=18^\circ$ and $\Psi=32^\circ$ cut along (002) plane were calculated using the computer code as a function of wavelength $\lambda$ at different values of $t_s$. The results were displayed in Fig. (6a&b) respectively as solid lines and compared with experimental ones measured by Adib et al. (1988) [19] at the same conditions. To show the effect of single crystal layer thickness on $R^4$ from Zn (002) with ($\eta=0.5^\circ$) set at $\psi=18^\circ$ and $\psi=32^\circ$, $R^0$ was calculated versus layer thickness $t_s$. The results of calculation are displayed respectively in Fig. (7a&b).
Figure 6. Neutron transmission through Zn single crystal at different layer thickness $t_s$.

Figure 7. $R^k$ Versus layer thickness $t_s$ for Zn single-crystal.

Figure (7) shows that reflecting power $R^k$ is constant within accuracy 5 % for layer thickness from 10 µm up to 60 µm. The calculated reflecting power $R^k_{002}$ for Zn (002) at cut-off angles $\psi=18^0$ and $\psi=32^0$ are 0.00148 and 0.00091 nm respectively. These values are in agreement with measured one at the same conditions. The main features of reflecting power $R^k$ from different single-crystals are listed in Table 2.
Table 2: The features of reflecting power $R^d$ form different single-crystals

<table>
<thead>
<tr>
<th>Single-crystal</th>
<th>$h,k,l$</th>
<th>$\Psi$ Degree</th>
<th>$t_0$ (mm)</th>
<th>T (K)</th>
<th>Reflection Plane $hkl$</th>
<th>$\lambda_{hkl}$ (nm)</th>
<th>$R^d \times 10^4$ (nm) (Experimental)</th>
<th>Range of layer thickness ($t_\circ$)-μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Copper</td>
<td>002</td>
<td>13.4</td>
<td>10</td>
<td>R.T</td>
<td>111</td>
<td>0.350</td>
<td>6.80 ± 0.34</td>
<td>10-60</td>
</tr>
<tr>
<td>Copper</td>
<td>002</td>
<td>13.4</td>
<td>10</td>
<td>R.T</td>
<td>002</td>
<td>0.290</td>
<td>9.70 ± 0.50</td>
<td>10-60</td>
</tr>
<tr>
<td>Copper</td>
<td>111</td>
<td>30</td>
<td>10</td>
<td>R.T</td>
<td>111</td>
<td>0.361</td>
<td>12.50 ± 0.62</td>
<td>10-50</td>
</tr>
<tr>
<td>Zinc</td>
<td>002</td>
<td>18</td>
<td>22</td>
<td>R.T</td>
<td>002</td>
<td>0.470</td>
<td>14.40 ± 0.72</td>
<td>40-70</td>
</tr>
<tr>
<td>Zinc</td>
<td>002</td>
<td>32</td>
<td>22</td>
<td>R.T</td>
<td>002</td>
<td>0.419</td>
<td>8.95 ± 0.44</td>
<td>40-70</td>
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<tr>
<td>MgO</td>
<td>100</td>
<td>Zero</td>
<td>100</td>
<td>R.T</td>
<td>420</td>
<td>0.160</td>
<td>25.0 ± 1.3</td>
<td>10-500</td>
</tr>
<tr>
<td>MgO</td>
<td>100</td>
<td>Zero</td>
<td>100</td>
<td>R.T</td>
<td>220</td>
<td>0.219</td>
<td>30.3 ± 1.5</td>
<td>10-500</td>
</tr>
<tr>
<td>MgO</td>
<td>100</td>
<td>Zero</td>
<td>100</td>
<td>L.N</td>
<td>420</td>
<td>0.160</td>
<td>26.5 ± 1.3</td>
<td>10-550</td>
</tr>
<tr>
<td>MgO</td>
<td>100</td>
<td>Zero</td>
<td>100</td>
<td>L.N</td>
<td>220</td>
<td>0.219</td>
<td>30.4 ± 1.5</td>
<td>10-550</td>
</tr>
</tbody>
</table>

CONCLUSION

Use has been made of the simple additive formula determining the reflectivity and attenuation of neutrons by a single-crystal, together with the BISCF code, which have been developed and presented in this manuscript.

The obtained agreement between the calculated neutron transmission through bulk cubic and hexagonal closed packing (HCP) bulk single-crystals and experimental data using the computer code justifies its applicability.

Moreover, the reflecting power of the Bragg reflections from the allowed $hkl$ planes are calculated by assuming that the bulk single-crystal consists of thin layers such that the extensions inside the layer are negligible. Such method allows calculating the contribution of the Bragg scattering to neutron transmission of imperfect single crystal which are found to be in reasonable agreement with the experimental data.

REFERENCES