CARACTERISTICS OF COPPER SINGLE CRYSTAL AS A NEUTRON MONOCHROMATOR

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A simple formula is given that permits the calculation of the neutron reflectivity from a single–crystal cut along different planes. The monochromatic features of copper single crystal are detailed in terms of the cutting plane, crystal mosaic spread and thickness within the wavelength band from 0.1 up to 0.4 nm. A computer program Cu-MONO written in FORTRAN-77, has been developed to carry out the required calculations. Calculation shows that, 5 mm thick Cu single-crystal cut along its (111) plane having 0.5° FWHM on mosaic spread are the optimum parameters when, it is used as an efficient neutron monochromator at wavelength 0.114 nm. Moreover the reflected 1st - order neutrons are almost free from the accompanying higher order ones at wavelengths up to 0.15 nm. The same features are of Cu (200) however, with less reflectivity than the later. While Cu (220) is preferred than the others at wavelengths up to 0.18 nm. The calculated integrated neutron intensity of the 1st -order reflection, when the monochromatic wavelength is longer than 0.25 nm, was found to be less than that of higher orders ones from a thermal reactor flux. However, from a cold reactor one, the intensity of the 1st order from Cu (111) within the wavelength band from 0.25 up to 0.4 nm is higher than the later. While, from Cu (200) is up to 0.36 nm, and from Cu (220) is only up to 0.3nm.

Keywords: Neutron Monochromator, Single-Crystal, Cu

INTRODUCTION

A range of wavelengths is selected from the incident incoming white neutron beam on a single-crystal monochromator according to Bragg’s law [1]. The intensity and width of the monochromatic beam depend on the crystal structure and its mosaicity. A larger mosaicity increases the produced beam intensity, but reduces the wavelength resolution [2]. Common materials used as monochromator crystals are pyrolytic graphite (PG) [3], silicon, copper, beryllium, iron, and haussler crystals for use in polarized neutron
scattering experiments. The choice of monochromator depends on the production of neutron wavelength band required for the experiment with the desired wavelength resolution. However, a beam of monochromatic neutrons, selected from the spectrum of a nuclear reactor diffracted by a monochromator crystal, will in general contaminate with higher-order components. Consequently the use of a filter is indispensable to eliminate higher order contaminations [4, 5].

Nowadays copper single crystal is commonly used as an efficient neutron monochromator, since it has high scattering cross-section [6]. Moreover, bulk copper crystals cut along different planes with a small mosaic spread are now available at a reasonable price. The neutron transmission through 10 mm thick copper single-crystal cut along both(111) and (200) planes with 0.2° FWHM on mosaic spread were measured at different take–off angles at ET-RR-1 reactor by Adib et al.[7]. Naguib and Adib [8] analyzed the experimental transmission data through Cu(111) using computer program ISCANF. They reported that 3 cm thick Cu(111) single crystal cooled to liquid nitrogen can be efficiently used as a neutron filter for removing fast neutrons of energies higher than 1 eV, while providing reasonable intensity of the transmitted thermal neutrons. Naguib and Adib[9] developed an improved computer ISCANF-1 program to investigate the effect of parasitic Bragg scattering on the neutron filtering characteristics of copper single crystal cut along (200) plane. However, Naguib and Adib [8,9] did not study the monochromatic features of copper single crystals having different cuts. Moreover the high scattering cross section value of copper permits higher neutron reflectivity, while it increases the effect of parasitic Bragg scattering on the neutron filtering characteristics. Moreover, copper absorption cross-section at thermal neutron energies is greater than 3.8 barns, thus decreasing the transmitted filtered neutrons. Consequently, copper single crystals are more preferred as a neutron monochromator than a neutron filter.

In the present work, a feasibility study of using Cu single- crystals cut along different planes as a neutron monochromator. The optimal monochromator parameters of crystal mosaic spread and thickness are discussed taking into consideration the ratio of higher order reflections to the first order one.

A computer program Cu-MONO has been developed to calculate the distribution of reflecting power \( P_{hk1}^\theta \), and neutron intensity \( I_{Ref} \) from the single-crystal as a function of wavelength, crystal orientation, mosaic spread, thickness and reactor moderating temperature. The Cu-MONO program is an adopted version of the computer code Mono-PG [10].

THEORETICAL TREATMENT

The main parameters determining the quality of a single-crystal as a neutron monochromator with reasonable resolution are: crystal orientation, its mosaic spread,
the ratio of higher-order contaminations to the first one and the wavelength band of the reflected monochromatic neutrons [11].

It is well known that, the reflected neutrons from the \((hkl)\) planes satisfy the Bragg equation:

\[ n\lambda = 2d_{hkl}\sin\theta_{hkl} \]  

(1)

where, \(n\) is the order of reflection, \(\theta_{hkl}\) is the glancing angle to the \((hkl)\) plane.

The reflected power of the \((hkl)\) planes at glancing angle \(\theta\) within \(d\theta\) is given by [11].

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

(2)

where,

\[ d\theta = \frac{d\lambda}{2d\cos\theta} \]

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

(3)

and

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

(4)

in which \(\mu\) is the linear absorption coefficient, \(t_o\) is the crystal thickness, \(\gamma_o\) is the direction cosine of the incident neutron beam relative to the inward normal to the crystal face and \(=\).

The \(Q_{hkl}\) is crystal-graphic quantity given by:

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

(5)

where, \(F_{hkl}^2\) is the square of the structure factor of the unit cell for the \(hkl\) reflection and \(N\) is the number of nuclei per unit cell.

\(W(\theta)\) has a Gaussian distribution with standard deviation \(\eta\) on mosaic blocks of single crystal.

The integrated reflectivity \(R^\theta\) is given by:

\[ R^\theta = \int_{-\infty}^{+\infty} P_{hkl}^\theta d\theta \]  

(6)

While the reflected intensity \(I_{Ref}\) from crystals when a neutron reactor beam having Maxwellian distribution \(\Phi(\lambda)\) is given by:
\[ I_{\text{Ref}} = \Phi(\lambda) \ast P_{\text{hkl}}^\theta \]  \hspace{1cm} (7)

where \( \Phi(\lambda) \) having neutron gas temperature \( T \) is given by [12]:

\[ \Phi(\lambda) = \left( \text{constant} / \lambda^5 \right) \exp\left(-h^2 / 2mkT\lambda^2\right) \]  \hspace{1cm} (8)

From Eq. (8), for constant \( d\lambda \), the peak of curve \( \Phi(\lambda) \) occurs at a wavelength \( \lambda = h / \sqrt{5mkT} \). Thus for thermal reactor with \( T = 293 \text{k} \) and \( \lambda = 0.114 \text{ nm} \). While, for cold reactor with \( T = 20 \text{K} \) and \( \lambda = 0.43 \text{nm} \).

A computer code Cu-Mono has been developed to carry out the calculation of the reflecting power \( P_{\text{hkl}}^\theta \) as a function of neutron wavelength, mosaic spread, glancing angle to crystal orientation and crystal thickness. The code can also calculate the reflected intensity \( I_{\text{Ref}} \) of the neutron beam incident on the crystal from both thermal and cold reactor fluxes. The Cu-MONO code is an adopted version of the computer code MONO-PG written by Adib et.al. [10].

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

**Table 1. Physical parameters of Cu single crystal**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>System and space group of Cu</td>
<td>(F.C.C.) Fm3m</td>
</tr>
<tr>
<td>Atomic weight</td>
<td>63</td>
</tr>
<tr>
<td>Density ( gm/cm³)</td>
<td>8.93</td>
</tr>
<tr>
<td>Lattice parameters (nm)</td>
<td>( a_0 = 0.3615 )</td>
</tr>
<tr>
<td>No. of atoms / unit cell</td>
<td>4</td>
</tr>
<tr>
<td>No. of unit cells / m³</td>
<td>2.1167E+28</td>
</tr>
<tr>
<td>Atomic positions</td>
<td>((0,0,0),(1/2,0,1/2),(0,1/2,1/2),(1/2,10,1/2))</td>
</tr>
<tr>
<td>Coherent scattering lengths b (m)</td>
<td>0.771E-14</td>
</tr>
<tr>
<td>Total scattering cross section (barn)</td>
<td>8.03</td>
</tr>
<tr>
<td>Absorption cross section at 0.025 eV (barn)</td>
<td>3.78</td>
</tr>
</tbody>
</table>

**MONOCHROMATIC FEATURES OF Cu SINGLE-CRYSTAL**

From Eq.(2) the highest reflectivity is expected from the surface of a Cu single-crystal cut along \((hkl)\) at glancing angle \( \theta \) whose inter-planer distance \( d_{hkl} \) is the
longest and its structure factor \( F_{hkl}^2 \) has the biggest value among the other planes. From the crystal structure of Cu the (111), (200) and (220) cutting planes satisfies these requirements. Therefore the distribution of the reflected neutrons \( P_{hkl}^\theta \) at \( \lambda =0.114 \text{ nm} \) from Cu single-crystals cut along (111), (200) and (220) as a function of mosaic spread were calculated at the same \( d\theta \) and effective thickness \( t_{\text{eff}} \) \((t_{\text{eff}} = t_0 / \cos \theta \) ). The input parameters used for calculations of the reflecting power from different cuts of Cu single crystals are given in Table 2. The result of calculation are displayed in Fig. 1 a, b and c respectively.

**Table 2.** Input parameters for calculation the reflecting power \( P_{hkl}^\theta \)

<table>
<thead>
<tr>
<th>Cutting plane (hkl)</th>
<th>Glancing angle ( \theta )°</th>
<th>Thickness ( t_0 ) (mm)</th>
<th>( \lambda ) ( \text{min} ) (nm)</th>
<th>( \lambda ) ( \text{max} ) (nm)</th>
<th>( \Delta \lambda ) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 111</td>
<td>15.85</td>
<td>5.00</td>
<td>0.001</td>
<td>0.45</td>
<td>0.10</td>
</tr>
<tr>
<td>2 200</td>
<td>18.38</td>
<td>5.50</td>
<td>0.001</td>
<td>0.45</td>
<td>0.086</td>
</tr>
<tr>
<td>3 220</td>
<td>26.49</td>
<td>8.20</td>
<td>0.001</td>
<td>0.45</td>
<td>0.057</td>
</tr>
</tbody>
</table>

Fig.1 shows that the reflective power from Cu (111) at \( \lambda =0.114 \text{ nm} \) is more than that of (200) and (220). Such result gives the opportunity of using Cu(111) as a neutron monochromator than the others. However the distribution of its reflective power is more broadened than the others.

The wavelength resolution \( \Delta \lambda / \lambda \) as a function of mosaic spread was determined as a ratio of the FWHM of the distribution \( P_{hkl}^\theta \) to the wavelength \( \lambda_{hkl} \). The values of peak reflecting power and \( \Delta \lambda / \lambda \) for versus \( \eta \) for Cu(111), Cu(200) and Cu(220) are displayed in Fig. (2) a, b and c respectively. Fig. (2) shows that \( \Delta \lambda / \lambda \) is increasing with the increase of the mosaic spread \( \eta \). This is consistent of the fact that low quality crystal (i.e., high value of \( \eta \) ) the distribution of the reflected neutrons is broadened consequently the resolution becomes worst.
Figure 1. Reflecting power $P^{\theta}_{hkl}$ at various $\eta$. 
Figure 2. Peak reflecting power $P^{\theta}_{hkl}$ and wavelength resolution $\Delta \lambda / \lambda \%$ versus $\eta$.

Figure (2) shows also that $P^{\text{max}}$ reaches a maximum value at FWHM on mosaic spread $\eta = 0.4^0$. Such behavior may be due to the fact that the assumed incident neutron beam divergence ($0.4^0$) is comparable with the mosaic spread value. Moreover, Cu(220) with $0.3^0$FWHM on mosaic spread $\eta$ is in some cases may be more preferred than the others when it used as a monochromator since the glancing angle ($\Theta \approx 26.0^0$) is more suitable to carry out the neutron diffraction experiment. The integrated reflecting power $R^{\theta}(n)$ of the monochromatic neutrons of 1$^{\text{st}}$ order reflection from Cu(111),Cu(200) and Cu(220) single-crystal having the same mosaic spread ($\eta = 0.4^0$) along with the higher order contaminations accompanying the 1$^{\text{st}}$-one are calculated versus crystal thickness are displayed in Fig. (3)a,b&c, respectively.
From Fig.(3) one can see that 5mm thick is the optimum thickness of Cu(111) single-crystal as a neutron monochromator. However Cu(220), in some cases, is more promising when it used as a neutron monochromator since the contaminations of higher orders are less. These contaminations are true when the incident neutron beam distribution is constant. However the incident neutron beam distribution from steady state reactor obeys Maxwellian distribution with neutron gas temperature $T$. Therefore the reflected neutron intensities from Cu single crystals cut along (111),(200) and (220) were calculated at same selected reflected monochromatic neutrons of 0.114 nm, 0.34 nm from thermal reactor flux. The result is displayed in Fig.(4)a & b respectively.
While, Fig.(4c) displays the reflected monochromatic neutrons at 0.35 nm from cold reactor flux.

Since the reflected neutron monochromatic intensities and its higher orders contaminations from Cu single-crystal was found to depend upon both the reactor moderating temperature and the value of the selected monochromatic wavelength, therefore the integrated intensity of 1\textsuperscript{st} order monochromatic neutrons from 5mm thick Cu(111),Cu(200) and Cu(220) single crystals along with the accompanying higher orders ones were calculated as a function of the neutron wavelength. The result of calculation is displayed in Fig. (5)a, b & c respectively for incident thermal reactor neutron beam, while Fig. (6), for cold reactor one.

**Figure 4.** Reflected neutron intensities at various selected monochromatic wavelengths.
Figure 5. The integrated intensity of monochromatic neutrons and its higher orders from thermal reactor flux versus wavelength $\lambda$. 
Fig. (5) shows that Cu (111) is the best choice as a neutron monochromator without need for a filter for selected monochromatic neutrons from a thermal reactor flux up to $\lambda = 0.15$ nm. The same features are of Cu(200) however, with less integrated reflectivity than the later. While, Cu (220) is preferred than the others cuts at wavelengths up to 0.18 nm.

One can see from Fig.(6) that if a cold reactor is available, the integrated intensity of the 1st order from Cu (111) is higher than from other cuts within the wavelength band from 0.25 up to 0.4 nm. While, from Cu(200) is up to 0.36 nm, and from Cu(220) is only up to 0.3nm.

**Figure 6.** The integrated intensity of monochromatic neutrons and its higher orders from cold reactor flux versus wavelength $\lambda$. 

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CONCLUSION

The developed Cu Mono-code, used to calculate the reflectivity from single-crystals was found to be sufficient for determining its neutron monochromatic characteristics.

It was shown, that 5 mm thick Cu single-crystal cut along its (111) plane having 0.5° FWHM on mosaic spread is the best choice as a neutron monochromator without need for a filter for selected monochromatic neutrons from a thermal reactor flux up to $\lambda = 0.15$ nm. The same features are of Cu(200) however, with less integrated reflectivity than the later. While, Cu (220) is preferred than the others cuts at wavelengths up to 0.18 nm. If a cold reactor is available, the use of the Cu (111) single crystal as a neutron monochromator free from higher order contaminations at wavelengths $\lambda$ up to 0.4 nm is more appreciated than the other cuts.

REFERENCES

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في هذا البحث تم دراسة انعكاس النبوترونات من مستويات قطع مختلفة للبللورة النحاس الاحادية واستنتاج انسب البارامترات لهذه البللورة من حيث مستوى القطع - التوزيع الموجي - سمك البللورة وذلك في مدى الأطوال الموجية من 0.1 إلى 0.4 نانومتر. وتم توضيح برنامج Cu-MONO الموجي علاوة على ذلك. أظهرت الحسابات ان بللورة النحاس قطعت على المستوى 111 هي ذات سمك 0.5 mm و التوزيع الموجي 0.15 nm عند الطول الموجي 0.114 nm. وتم الحصول على نفس النتائج بالنسبة لمستوى القطع 200 ولكن مع انعكاس أعلى. بينما عند مستوى القطع 220 يكون أفضل من المستوى الأخيرين عند الطول الموجي 0.18 nm. وحسب تكامل كثافة نبوترونات الرتب الأولى عند الطول الموجي 0.25 nm بالنسبة لسيل نبوترونات المفاعل الحراري وجد انها أقل منها انعكاس نبوترونات الرتب الأعلى بينما بالنسبة لسيل نبوترونات المفاعل ذو الفيض البارد فا: تكامل كثافة الرتب الأولى من بللورة النحاس المقطوعة على المستوى 111 من الطول الموجي 0.25 nm حتى 0.4 nm أعلى مما بعده. بينما مستوى القطع 200 عند الطول الموجي 0.36 nm ومستوى القطع 220 حتى الطول الموجي 0.3 nm.