A number of samples of oriented UCAR graphite of various thicknesses and mosaic widths have been investigated for reflectivity of thermal neutrons. The observed values agree rather well with those calculated on the assumption of ideally imperfect samples. The peak reflectivity and integrated reflectivity were found to be superior to other monochromators tested by a factor of about 2-10. For 4 Å neutrons peak reflectivities above 0.9 can easily be attained. An important feature is the absence of double Bragg scattering in graphite, leading to a considerable reduction of the intensity of extraneous neutrons in the primary beam.

1. Introduction

Oriented graphite, also known as pyrolytic graphite, has highly preferred orientation of (00l)-planes similarly to a single crystal. All the other (hkl)-planes, however, are aligned at random, and give rise to powder peaks in a diffraction diagram, similarly to a polycrystalline specimen. Oriented graphite of a specified quality is now commercially available from Union Carbide Corporation. The specification given is the degree of alignment of the c-axes, as measured by the width \( \beta \) of the rocking curve of an (00l) reflection.

Highly oriented graphite, having \( \beta < 1^\circ \), has been reported by Sparks to be a very efficient monochromator for X-rays. Bergsma and van Dijk have investigated the properties of oriented graphite, having \( \beta \sim 5^\circ \), as a second order neutron filter, and Loopstra has demonstrated the use of such a filter in neutron diffractometry with 2.6 Å neutrons.

W. C. Koehler made the first preliminary test of oriented graphite as a neutron monochromator. We have performed a systematic test of the same samples.

2. Reflectivity formulas

All of the samples were investigated in the symmetric reflection geometry. The appropriate formulas have been given by Bacon and Lowde and are reproduced below.

The mosaic distribution of a crystal is specified by the parameter \( \eta \) which is related to \( \beta \), the full width at half maximum of the rocking curve, by

\[
\eta = 0.424 \beta. \tag{1}
\]

If the mosaic distribution is given by

\[
W(\Delta) = \frac{1}{\eta \sqrt{(2\pi)}} \exp(-\Delta^2/2\eta^2), \tag{2}
\]

the maximum reflectivity at \( \Delta = 0 \) is for an ideally imperfect, nonabsorbing crystal given by

\[
R_{\text{max}} = \frac{1}{\sqrt{(2\pi)}} \frac{Qt_0}{\eta \sin \theta} \left[ 1 + \frac{1}{\sqrt{(2\pi)}} \frac{Qt_0}{\eta \sin \theta} \right]. \tag{3}
\]

\( \theta \) is the neutron glancing angle, \( t_0 \) is the crystal thickness in the direction of the scattering vector, and the crystallographic quantity \( Q \) is defined by

\[
Q = \lambda^3 N_e F^2 / \sin 2\theta, \tag{4}
\]

in which \( \lambda \) is the neutron wavelength, \( N_e \) is the number of unit cells per cm\(^3\), and \( F \) is the structure factor. When \( \lambda \) is given in Ångstrom, the value for graphite is

\[
Q = (\lambda^3 / \sin 2\theta) 5.65 \times 10^{-3}. \tag{5}
\]

While \( R_{\text{max}} \) is the peak reflectivity in a monochromatic beam, \( R^a \) is the integrated reflectivity for a mosaic crystal that is rotated in the same beam. It has the dimension of an angle and gives the angular region over which a crystal is totally reflecting. For large values of \( R^a \), when secondary extinction is no longer negligible, the reflectivity is expected to be reproduced within five percent by

\[
R^a = 0.96(\eta Q t_0 / \sin \theta)^{\frac{3}{2}}. \tag{6}
\]

A stationary monochromator crystal in a white neutron beam is totally reflecting over a wavelength range given by

\[
R^l = R^a \cot \theta. \tag{7}
\]

All of the reflectivity formulas above are valid for ideally imperfect, nonabsorbing crystals. Graphite belongs to this category when \( \lambda > 3 \) Å. At shorter wavelengths thermal diffuse scattering and disordered Bragg scattering give an attenuation of the transmitted beam that is similar to absorption. An inspection of the curves given by Bacon reveals that the attenuation can be approximately taken care of by including a multiplying attenuation factor in formula (6). For the maximum reflectivity given by (3) we expect a negligible attenuation factor for graphite, the penetration depth of the beam being so small.
3. Experimental

The UCAR graphite samples tested are in the form of thin plates (0.02–0.37 cm) and have different shapes: rectangular, circular or circular segment. The maximum area of any of the samples was about 75 cm². The face having the largest area is always a basal plane. The dimension of the hexagonal unit cell of graphite is given by c=6.71 Å and a=2.46 Å.

The (00l) planes with l=2n all give rise to Bragg reflections, the highest reflectivity being that of (002). The usefulness of this reflection for monochromating a neutron beam is qualitatively demonstrated by fig. 1. The whole Maxwellian neutron spectrum was in this case incident on a graphite sample, which was mounted with θ=11°, and the transmitted spectrum was measured by the time-of-flight technique. The strong dip around λ=1.28 Å is due to the (002) reflection and a maximum reflectivity in excess of 70 percent is apparent.

Most of the reflectivity measurements were performed on (004), for three reasons. Firstly, with this reflection, an almost parallel arrangement could be obtained at a double axis spectrometer with Be(002) as a monochromator. The rocking curve then gives an almost direct figure for the β parameter of the sample, the monochromator having a much narrower mosaic. Secondly, the reflectivity is low enough for the wavelength used that the curves can be treated as Gaussian. Thirdly, the Bragg angle θ is higher than for (002) and it is easier to make sure that all of the incident beam is hitting the sample.

The measurements were performed at λ=1.20 Å. The incident beam had to pass an entrance slit of 5 mm diameter before reaching the sample. The in-pile collimation is ~ 1°. In front of the detector a Soller collimator was mounted, the dividing slits define a divergence angle of 0.8°. When measuring the reflectivity the counter was left stationary and the crystal was rotated through the Bragg position. In this way the reflectivity is always referred to the same interval of neutron wavelength. For the samples having the narrowest mosaic spread, the integrated intensity observed when rotating the crystal and counter coupled with angular speeds in a 1:2 ratio actually differ only little from the intensity measured in the way described above. There is a pronounced difference when the crystal has a wide mosaic, however.

The reflectivities were put on an absolute scale by measuring the maximum intensity of the direct beam with the same collimation as above.

4. Results

The results obtained for $R_{\text{max}}$ and $R^\theta$ are given in table 1. It is difficult to quote any accuracy on the measurements. Extinction effects tend to make the observed β- (or η-) values too large, and geometrical
Table 1
Calculated and observed reflecting properties of graphite (004) at $\lambda = 1.20$ Å (for symbols, see text).

<table>
<thead>
<tr>
<th>Sample shape and dimension (mm)</th>
<th>$\beta$ (degrees)</th>
<th>$R_{\text{max}}$ calc. from eq. (3)</th>
<th>$R_{\text{max}}$ obs.</th>
<th>$R_{0} \times 10^{3}$ calc. from eq. (6)</th>
<th>$R_{0} \times 10^{3}$ obs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Phi = 95$ Segment $h = 70$</td>
<td>1.73</td>
<td>0.28</td>
<td>0.22</td>
<td>12.4</td>
<td>8.1</td>
</tr>
<tr>
<td>$t_0 = 3.3$ Rectangle $30 \times 74 \times 2.2$</td>
<td>1.36</td>
<td>0.25</td>
<td>0.25</td>
<td>8.9</td>
<td>7.0</td>
</tr>
<tr>
<td>Rectangle $30 \times 74 \times 2.1$</td>
<td>1.06</td>
<td>0.30</td>
<td>0.32</td>
<td>7.8</td>
<td>5.8</td>
</tr>
<tr>
<td>Rectangle $33 \times 54 \times 0.8$</td>
<td>0.83</td>
<td>0.34</td>
<td>0.32</td>
<td>6.6</td>
<td>5.4</td>
</tr>
<tr>
<td>Rectangle $28 \times 32 \times 2.5$</td>
<td>1.4</td>
<td>0.28</td>
<td>0.32</td>
<td>9.5</td>
<td>9.1</td>
</tr>
<tr>
<td>Circle $\Phi = 98$, $t_0 = 3.7$</td>
<td>1.13</td>
<td>0.41</td>
<td>0.42</td>
<td>10.5</td>
<td>9.7</td>
</tr>
<tr>
<td>Rectangle $36 \times 79 \times 0.8$</td>
<td>3.0</td>
<td>0.014</td>
<td>0.02</td>
<td>0.4</td>
<td>1.2</td>
</tr>
</tbody>
</table>

Errors (reflected beam not hitting counter etc.) would give $R_0$ too low.

Fig. 2 gives a comparison between calculated and observed values for the peak reflectivity $R_{\text{max}}$. The agreement is rather good even when absorption effects are neglected. The lower abscissa applies to a sample having $\beta = 1.13^\circ$ and $t_0 = 0.37$ cm. One data point at $\lambda = 1.86$ Å for (002) is included. According to the calculated curve one expects peak reflectivities higher than 90 percent for this crystal. This expectation was verified through transmission measurements similar to that in fig. 1, but with larger values of $\theta$.

Fig. 3 gives a comparison between calculated and observed values for $R_0$. The data do on the average fall some 20 percent below the calculated curve for zero absorption, this suggests that an empirical attenuation factor of 0.8 should be included in (6), as discussed above, when working at $\lambda = 1.20$ Å.

5. Comparison with other neutron monochromators

Some of the monochromator crystals at our laboratory have earlier been investigated at $\lambda = 1.27$ Å. The values obtained are thought to be representative for the kind of material they are made of. Using eqs. (3) and (6) above, and including an attenuation factor of 0.8 in the latter, we may, with an accuracy of ~10 percent, calculate the reflectivity of graphite at the same wavelength. The sample having $\beta = 1.13^\circ$ and $t_0 = 0.37$ cm was chosen for this comparison, and numbers are given in table 2. The comparison includes all of the quantities $R_0$, $R_0/\beta$, $R^4$ and $R_{\text{max}}$. The relevant figures to compare depend on the specific purpose. $R_0/\beta$ and $R_{\text{max}}$ are relevant when the incident beam is monochromatic, e.g. when the crystal is used as an energy analyzer in triple axis spectrometry.

It may at first seem puzzling why graphite performs so much better than other monochromators. Beryllium e.g. is expected to perform equally well on account of theoretical values of $Q$ and of the absorption

![Fig. 2. Calculated and observed value of $R_{\text{max}}$ at $\lambda = 1.20$ Å. The lower abscissa applies to (002) of a sample having parameters $t_0 = 0.37$ cm and $\beta = 1.13^\circ$. One data point for this sample at $\lambda = 1.86$ Å is included.](image1)

![Fig. 3. $R_0$ for a rotated sample at $\lambda = 1.20$ Å, (004) reflections. The upper curve is calculated for an ideally imperfect, non-absorbing crystal. For the lower curve an empirical attenuation factor of 0.8 is included.](image2)
Table 2
The performance at $\lambda = 1.27$ Å of different monochromators available at Kjeller.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Reflection</th>
<th>$\beta$ (min)</th>
<th>$R_\theta$ (rad)</th>
<th>$R_\theta/\beta$</th>
<th>$R_\lambda \times 10^3$ (Å)</th>
<th>$R_{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be</td>
<td>002</td>
<td>22</td>
<td>11</td>
<td>3.2</td>
<td>0.5</td>
<td>1.1</td>
</tr>
<tr>
<td>Cu</td>
<td>111</td>
<td>22</td>
<td>4.7</td>
<td>1.4</td>
<td>0.19</td>
<td>0.53</td>
</tr>
<tr>
<td>Zn</td>
<td>002</td>
<td>34</td>
<td>13.6</td>
<td>4.0</td>
<td>0.39</td>
<td>1.9</td>
</tr>
<tr>
<td>Ge</td>
<td>111</td>
<td>18</td>
<td>4.8</td>
<td>1.4</td>
<td>0.27</td>
<td>0.9</td>
</tr>
<tr>
<td>Graphite</td>
<td>002</td>
<td>68</td>
<td>58</td>
<td>17</td>
<td>0.86</td>
<td>8.7</td>
</tr>
</tbody>
</table>

coefficient. There are probably two reasons for the difference in performance, however. Firstly the dislocation patterns may be different, leading to more primary extinction in other crystals than in graphite. The absence of primary extinction makes graphite behave so closely to the ideally imperfect case with secondary extinction as the prevailing attenuation process.

Secondly, and probably more important, is the absence of double Bragg scattering in graphite, due to its polycrystalline character.

Double Bragg scattering contributes strongly to the background scattering when a single crystal is exposed to a white neutron beam. The extraneous intensity from double processes is particularly strong for thick crystals.

Fig. 4. Comparison of neutron spectra reflected from zinc and graphite monochromators at $\theta = 25^\circ$. Notice the different signal-to-background ratios.
having large unit cells, and for short neutron wave-
lenghths\(^8\)). On this account the zinc monochromator
listed in table 2 is expected to have a serious con-
taminant component in the primary beam. The different
qualities of graphite and zinc in this respect are clearly
demonstrated in fig. 4. This plot gives the neutron
spectra scattered for \(\theta = 25^\circ\), as measured by time-of-
flight technique with a Fermi-type chopper. The peak-
to-background ratio is about a factor 200 higher for
graphite than for zinc.

6. Application

Since the first results of the present investigations
were communicated\(^9\), oriented graphite samples have
been used as monochromators and analyzers in neutron
spectrometry.

At our laboratory we have combined a graphite
monochromator with a semi-monochromating chopper
in an instrument for inelastic scattering\(^10; 11\). The
chopper serves two purposes. It eliminates very effec-
tively the Bragg components arising from higher order
reflections of (002), as seen from fig. 5, and it forms
part of a time-of-flight analyzing unit. By working at
long wavelengths it is possible to take advantage of the
large mosaic spread, using collimation \(\sim 1^\circ\), without
making the resolution intolerably poor. With this setup
it has proved possible to measure the spin wave
dispersion relation in antiferromagnetic MnS\(_2\) for a
sample volume of \(0.35 \text{ cm}^3\)\(^12\)) at a 2MW reactor.

In triple axis spectrometry one may use graphite both
as a monochromator and as an analyzer. This combina-
tion is now being used at Brookhaven National
Laboratory, and the expected intensity gain of \(\sim 10\)
over Ge has been verified\(^13\).

At the monochromator position only the (002) reflection
may be used without complication due to extraneous
wavelength components. It would be desirable
to be able to work at (004) and possibly also at (006) in
order to produce short wavelength neutrons with a
reasonably high take-off angle. This could be achieved
with a double-rotor setup.

The oriented graphite tested here has some advantage
over less well oriented graphite as a second order
neutron filter. The additional transmission dip due to
Bragg scattering, as seen from fig. 1, may be used to
reduce the thickness of the filter and thus to increase
the intensity ratio of first to second order neutrons.

7. Conclusions

Oriented graphite having mosaic spread of 1–2\(^\circ\), as
measured by fwhm of the rocking curve, has been
measured to possess very valuable properties as a
neutron monochromator. It is found that absolute
values of the maximum and integrated reflectivities
may be calculated with confidence from the standard
formulas for an ideally imperfect crystal. For the
integrated reflectivity an empirical attenuation factor
of 0.8 had to be introduced, but this is possibly partly
due to systematic errors in our measurements.

Recently highly oriented graphite having a mosaic \(\beta\)
as low as 0.3\(^\circ\) has become available\(^1\)). One can not
exclude the possibility that such samples may contain

Fig. 5. The neutron spectrum reflected from graphite at \(\theta = 37^\circ\), when transmitted through a semi-monochromating chopper.
a different dislocation pattern than the less well oriented ones, and consequently be more susceptible to primary extinction effects. We have had the possibility to test only one sample of the highly oriented type, having a nominal mosaic width of 0.35°. The measured values of $R^{max}$ and $R^0$ fall about 15 and 30 percent, respectively, below the curves of figs. 2 and 3. Although it is not possible to draw any conclusion from this single measurement, it suggests that a systematic study of several samples might be useful. The highly oriented samples are in any case superior to other known monochromator crystals, however. The sample tested by us had a peak reflectivity of 0.55 for 1.22 Å neutrons, and the results from Brookhaven\textsuperscript{13)}, mentioned above, were actually obtained with samples of the highly oriented quality.

We are very much indebted to Dr. W. C. Koehler for calling our attention to graphite monochromators, and to the Union Carbide Corporation for the loan of the samples.

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