1. Introduction

The use of long-wavelength neutrons at reactor and pulsed sources often requires a filter for the removal of unwanted epithermal neutrons. For instruments that only require neutrons having wavelength \(\lambda \geq 0.41\) nm, the polycristalline beryllium filter provides the best performance and is commonly used. For shorter wavelengths in the thermal neutron range, strong attenuation from Bragg scattering occurs in polycristalline materials since the Bragg condition may be satisfied over a continuous range of short wavelengths, whereas perfect single crystals eliminate the coherent Bragg scattering for all but a few narrow wavelength ranges that satisfy the Bragg condition for the particular crystal orientation. Other materials that can be produced economically as low-mosaic-spread and high-purity (>99.99%) single crystals are routinely used as filters for these thermal neutron applications (Holmyrd & Connor, 1969; Nieman et al., 1980; Freund, 1983). Single-crystal filters of sapphire (\(\text{Al}_2\text{O}_3\)) at room temperature (Mildner & Lamaze, 1998), or silicon (Freund et al., 1985) and magnesium oxide (Thiyagarajan et al., 1998) at cryogenic temperatures, are frequently used. Table 1 lists the cross sections for all the relevant elements involved, taken from Sears (1986, 1992).

Here we consider the potential of single-crystal magnesium fluoride. IR optical-quality single-crystal MgF\(_2\) is preferred to vitreous silica or single-crystal quartz for neutron optics experiments involving lenses (Eskildsen et al., 1998; Choi et al., 2000) because of its purity and greatly reduced microstructure. This results in high transmission with very little small-angle neutron scattering (SANS). To keep the absorption from impurities at less than 10% of that caused by Mg (for which the absorption cross section is only 0.063 b), the total impurity content must usually be less than 0.01 at%, with much lower levels for three highly absorbing elements (B < 2.6 \times 10^{-4}\%\), Cd < 8 \times 10^{-5}\% and Gd < 4 \times 10^{-6}\%). The absorption is low and the coherent scattering length is relatively high, with low incoherent scattering, so that MgF\(_2\) should also make a reasonable thermal neutron filter of epithermal neutrons.

2. Model calculations

Neglecting incoherent and Bragg scattering, the total macroscopic cross section \(\Sigma_{\text{tot}}\) as a function of temperature \(T\) can be separated into three attenuation processes: (1) temperature-independent absorption by the nuclei, (2) single-phonon scattering and (3) multiple-phonon scattering, i.e.

\[
\Sigma_{\text{tot}}(\lambda, T) = \Sigma_{\text{abs}}(\lambda) + \Sigma_{\text{mph}}(\lambda, T) + \Sigma_{\text{sph}}(\lambda, T). \tag{1}
\]

The absorption cross section is related to wavelength by

\[
\Sigma_{\text{abs}} = A_{\text{abs}} = \frac{N(\sigma_{\text{abs}}/\lambda_0)}{\lambda}, \tag{2}
\]

where \(N\) is the number of molecules per unit volume of material and \(\sigma_{\text{abs}}\) is the thermal absorption cross section per molecule at the standard tabulation wavelength \(\lambda_0 = 0.1798\) nm. For pure MgF\(_2\), \(N = 3.07\) b \(-1 \cdot m^{-1}\) and \(\sigma_{\text{abs}} = 0.082\) b at \(\lambda_0\), so that \(A_{\text{abs}} = 1.405\) m\(^{-1}\) nm\(^{-1}\).

Steyerl (1977) has given an expression for the calculation of the temperature-dependent single-phonon scattering as a function of temperature \(T\) as

\[
\Sigma_{\text{sph}}(\lambda, T) = A_{\text{sph}} = (3N\sigma_0/M)(2m_k\Theta_D/h^2)^{1/2}\lambda \times \sum_{n=0} B_n x^{n-1}/[n!(n + 2.5)], \tag{3}
\]

valid for \(x \leq 6\), where \(x = \Theta_D/T\). [Note that for \(x > 6\) the summation in equation (3) is replaced by \(3.3x^{3/2}\).] Here \(\Theta_D\) is the Debye temperature and \(T\) is the physical temperature of the material in Kelvin, \(k\) is the Boltzmann constant, \(m_k\) is the neutron mass, \(h\) is Planck’s constant, and \(B_n\) are the Bernoulli numbers. \(M\) is the bound-atom cross-section-weighted harmonic mean atomic mass number. For MgF\(_2\), \(M\) is calculated using the expression \(M^{-1} = (\sigma_{Mg}/M_{Mg} + 2\sigma_F/M_F)/(\sigma_{Mg} + 2\sigma_F)\), where \(M_{Mg}\) and \(M_F\) are the atomic mass numbers, and \(\sigma_{Mg}\) and \(\sigma_F\) are the bound-atom cross sections for magnesium.
and fluorine, respectively. Hence, $M = 20.41$ and the bound-atom cross section $\sigma_b = \sigma_{\text{Mg}} + 2\sigma_T = 11.75 (5)$ b. The recommended Debye temperature for MgF$_2$ is 610 K (Kim, 2005), calculated from room-temperature elastic stiffness coefficients, in approximate agreement with the value of 626 K (Jones, 1977) deduced from acoustic measurements. [Note that Freund (1983) has given a value of $\Theta_D = 440$ K, without reference]. This results in single-phonon scattering of 3.66 m$^{-1}$ at 293 K and 0.100 m$^{-1}$ at 77 K. Equation (3) shows that the single-phonon cross section increases as $T^{3/2}$ at low temperatures and increases linearly with $T$ above room temperature.

The multiple-phonon scattering that dominates at short wavelengths has been modeled as (Freund, 1983)

$$\Sigma_{\text{muph}}(\lambda, T) = \Sigma_{\text{tot}}[1 - \exp\left(-\left(B_0 + B_T\right)\sin^2(\theta/2)/\lambda^2\right)],$$

where $\theta$ is the scattering angle. The average quantity $\langle\sin^2(\theta/2)\rangle = 1/2 - 1/(3\theta_D^2)$, which for MgF$_2$ is 0.48. The free-atom macroscopic scattering cross section $\Sigma_{\text{tot}} = N[M_{\text{Mg}}^2/(M_{\text{Mg}} + 1)^2]\sigma_{\text{Mg}} + 2M_{\text{F}}^2/(M_{\text{F}} + 1)^2\sigma_{\text{F}}] = 32.8 (2)$ m$^{-1}$. The temperature-independent quantity $B_0 = 3h^2/(2k\Theta_D^2M_{\text{Mg}})$, where $m_p$ is the proton rest mass, corresponds to zero-point motion and is numerically equal to $2.29 \times 10^{-3}$ nm$^2$. The temperature-dependent term is given by

$$B_T = (4B_0 T^2/\Theta_D^2) + \Theta_D/T \sqrt{\int_0^{\lambda} z J_z \exp(-z) - 1}.$$  

For $\Theta_D < 2\pi$, $B_T = 6h^2/(M_{\text{Mg}}\theta_D)k(T)/(k\Theta_D^2n)\sum_{n=0}B_n(\Theta_D/T)^n/(n + 1)!$ and is equal to 2.62 $\times 10^{-3}$ nm$^2$ at 293 K. At 77 K, $B_T = 0.24 \times 10^{-3}$ nm$^2$. The Debye–Waller factor increases as $T^2$ at low temperatures and increases linearly with $T$ above room temperature. Note that Scharenberg (1972) has given a slightly different form for the multiple-phonon cross section:

$$\Sigma_{\text{muph}} = \Sigma_{\text{tot}} \left[1 - \frac{2B}{2B} \left(1 - \exp\left(-\frac{2B}{\lambda^2}\right)\right)\right].$$

where the Debye–Waller factor is $B = B_0 + B_T$. Both this form and equation (4) reduce to the same expression to first order.

### 3. Transmission Measurements

Neutron transmission measurements have been performed with both monochromated beams and pulsed white beams using time-of-flight spectrometry. The measurements used different combinations of six right-angle prisms of optical-quality MgF$_2$ with sides of 25.4 mm and a width of 25.4 mm purchased from ISP Optics of Irvington, NY. The prisms were initially procured to compensate for chromatic aberrations caused by gravity in SANS measurements (Forgan & Cubitt, 1998). The crystal purity is unknown. The MgF$_2$ crystal has the rutile (TiO$_2$) structure, which exhibits tetragonal symmetry with $a_0 = 0.4623$, $c_0 = 0.3052$ nm. No special crystal orientation is specified for the prisms. For our purposes the orientation is unimportant because the single crystals have such narrow mosaic spread. Consequently the Bragg dips are very narrow in wavelength range.

All transmission measurements were taken with and without prisms, and in a sample container when cooled. The prisms were stacked in various arrangements, an example of which is shown in Fig. 1, to produce a total length of 107.8 mm. The transmission $T(\lambda)$ is determined from the ratio of the unscattered beam intensity with and without prisms, normalized by a beam monitor. The wavelength-dependent total macroscopic scattering cross section $\Sigma_{\text{tot}}(\lambda)$ for the filter is determined from $T(\lambda) = \exp[-\Sigma_{\text{tot}}(\lambda)/\lambda]$, where $\lambda$ is the length of the filter in the beam.

The first set of measurements were made using the NG3 30 m SANS instrument (Glinka et al., 1998) at the NCNR (NIST Center for Neutron Research) at room temperature, $T = 293$ (2) K, over the wavelength range $0.5 < \lambda < 2.0$ nm. The helical velocity selector produces a triangular-shaped distribution with a full width at half-maximum $\Delta\lambda/\lambda = 15\%$. A further measurement was made on the perfect crystal diffractometer (Barker et al., 2005) with $\lambda = 0.24$ nm and $\Delta\lambda/\lambda = 6\%$.

Transmission measurements using the same six prisms, but now placed within a cryostat cooled with liquid nitrogen and with aluminum windows, were taken on an 8 m SANS instrument at the NCNR with $\Delta\lambda/\lambda = 15\%$, at both room temperature and 77 K. For these measurements the incident beam direction was normal to the sides of constant thickness of the six prisms for a total length of 152.4 mm. There was no special gridding needed on the sides of the prisms.

![Figure 1](image_url)
Additional room-temperature transmission measurements were performed on the time-of-flight small-angle neutron diffractometer (SAND; Thiyagarajan et al., 1997) at the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory, in order to extend the wavelength range into the thermal neutron region with \( \lambda < 0.5 \) nm and particularly into the region of minimum attenuation. Details of such measurements are given by Mildner & Lamaze (1998). The incident beam was normal to the sides of constant thickness for five prisms with a total length of 127.0 mm, such that the transmission was between 0.4 and 0.7 for the wavelength range \( 0.15 < \lambda < 1.2 \) nm, but significantly less than 0.4 for \( \lambda < 0.15 \) nm. The wavelength resolution after rebinning of data was \( \Delta \lambda / \lambda \leq 5\% \).

Further transmission measurements were made on the time-of-flight Disk Chopper Spectrometer (DCS; Copley & Cook, 2003) at the NCNR to extend the \( T = 77 \) K measurements into the thermal neutron region range \( 0.1 < \lambda < 1.5 \) nm, with \( \Delta \lambda / \lambda = 0.03 \) nm after rebinning of data. The sample thickness was 107.8 mm, and a close cycle refrigerator was used to control the temperature.

Finally, transmission measurements were made on the NG7 30 m SANS instrument at the NCNR using four prisms stacked to produce a 50.8 mm length in the beam over the wavelength range \( 0.6 \leq \lambda \leq 2.0 \) nm, with \( \Delta \lambda / \lambda = 15\% \). Transmissions were measured at three separate temperatures of 293, 373 and 523 K.

Fig. 2 shows the attenuation results as a function of wavelength at both room temperature and 77 K. Dips in the transmission caused by Bragg reflections were not observed in MgF\(_2\), indicating that the crystalline mosaic spread is sufficiently small. The linear dependence at long wavelengths accounts for both the absorption (independent of temperature) and the single-phonon scattering, which is temperature dependent and may be reduced by cryogenic means. Linear fits to the data having \( \lambda \geq 0.8 \) nm indicate an attenuation of 6.32 m\(^{-1}\) nm\(^{-1}\) at room temperature and 1.49 m\(^{-1}\) nm\(^{-1}\) at 77 K. The data indicate that most of the phonon scattering present in the 293 K data has been eliminated at 77 K.

4. Analysis

The data were fitted to a curve of the form

\[
\Sigma_{\text{tot}}(\lambda, T) = A \lambda + \Sigma_{\text{sp}} \left[ 1 - \exp\left( -\frac{B}{\lambda^3} + D/\lambda^6 \right) \right],
\]

which accounts for both the linear attenuation at long wavelengths for absorption and single-phonon scattering, and the multiple-phonon scattering at short wavelengths. The additional term \( D \) was originally introduced by Carpenter et al. (1989) to account for anharmonic effects in the Debye–Waller factor to improve the quality of the fit.

Fig. 3 shows the total cross sections determined from all the measurements normalized by the wavelength \( \lambda \). The long-wavelength-independent asymptotic values of \( A \) appear as horizontal lines in the figure. From these data we obtain \( A = 6.32 (13) \) m\(^{-1}\) nm\(^{-1}\) at \( T = 293 \) K, and \( A = 1.49 (7) \) m\(^{-1}\) nm\(^{-1}\) at \( T = 77 \) K. Using only data for \( \lambda \geq 0.8 \) nm obtained at four different temperatures, we show the experimental values for \( A \) in Fig. 4 as symbols, together with the calculated values \( A = A_{\text{abs}} + A_{\text{sp}} \) obtained using equations (2) and (3) with \( \Theta_D = 610 \) K as a dashed line. The observed values for \( A \) at the higher temperatures considerably exceed the calculation (\( A = 5.06 \) m\(^{-1}\) nm\(^{-1}\) at \( T = 293 \) K). Better agreement between the

---

**Figure 2**
The attenuation coefficients for single-crystal MgF\(_2\) measured by time-of-flight at ambient temperature \( T = 293 \) K using the SAND instrument at IPNS (squares), and at \( T = 77 \) K using the DCS instrument at the NCNR (circles). The straight lines are fits to data having \( \lambda \geq 0.8 \) nm according to equation (7). The error bars represent \( \pm 1\sigma \).

**Figure 3**
The total cross section \( \Sigma_{\text{tot}} \) normalized by wavelength \( \lambda \). Open symbols are from time-of-flight measurements and closed symbols are from monochromatic beam measurements. The horizontal lines are average values obtained for data having \( \lambda \geq 0.8 \) nm. The three separate lines at \( T = 293 \) K were obtained by fitting data from each measurement separately.
![Image](https://via.placeholder.com/150)

**Table 2**

A compilation of material parameters for several different filter materials.

The order of materials is arranged from best to worst figure-of-merit ratio \( R \) at \( T = 0 \) K according to equation (7). References for Debye temperatures are given in the text.

<table>
<thead>
<tr>
<th>Material</th>
<th>( N ) (b(^{-1}) m(^{-3}))</th>
<th>( M ) (u)</th>
<th>( \lambda_{\text{abs}} ) (m(^{-1}) nm(^{-1}))</th>
<th>( \Theta_D ) (K)</th>
<th>( \Sigma_{\text{sph}} ) (m(^{-1}))</th>
<th>( \lambda_{\text{min}} ) (nm)</th>
<th>( R(\lambda_{\text{min}}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be</td>
<td>12.35 (1)</td>
<td>9.0122 (1)</td>
<td>0.52 (5)</td>
<td>1481 (16)</td>
<td>76.3 (2)</td>
<td>0.68 (2)</td>
<td>0.0070 (5)</td>
</tr>
<tr>
<td>MgO</td>
<td>5.357 (2)</td>
<td>19.04 (1)</td>
<td>1.88 (9)</td>
<td>938</td>
<td>38.4 (2)</td>
<td>0.32 (1)</td>
<td>0.023 (1)</td>
</tr>
<tr>
<td>MgF(_2)</td>
<td>3.071 (3)</td>
<td>20.405 (5)</td>
<td>1.40 (7)</td>
<td>610</td>
<td>32.8 (2)</td>
<td>0.38 (1)</td>
<td>0.024 (1)</td>
</tr>
<tr>
<td>SiO(_2)</td>
<td>2.66 (1)</td>
<td>17.538 (3)</td>
<td>2.54 (4)</td>
<td>580</td>
<td>25.31 (5)</td>
<td>0.304 (2)</td>
<td>0.0456 (7)</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>2.345 (6)</td>
<td>17.351 (2)</td>
<td>6.05 (8)</td>
<td>1032</td>
<td>32.99 (5)</td>
<td>0.206 (2)</td>
<td>0.0565 (6)</td>
</tr>
<tr>
<td>Si</td>
<td>4.9939 (2)</td>
<td>28.086 (1)</td>
<td>4.76 (8)</td>
<td>543</td>
<td>10.10 (4)</td>
<td>0.158 (1)</td>
<td>0.112 (1)</td>
</tr>
</tbody>
</table>

![Figure 4](https://via.placeholder.com/150)

**Figure 4**
The mean value for the measured total cross section normalized by wavelength for all data having \( \lambda \geq 0.8 \) nm obtained at four different temperatures. The curves show the sum of the single-phonon and the absorption cross section \( (\Sigma_{\text{sph}} + \Sigma_{\text{abs}})/\lambda = A_{\text{abs}} + A_{\text{sph}} \), calculated assuming a Debye temperature, \( \Theta_D \), of 610 K for the dashed curve and 520 K for the solid curve.

![Figure 5](https://via.placeholder.com/150)

**Figure 5**
The attenuation coefficient for single-crystal Mg\(_2\)F\(_2\), measured by time-of-flight at ambient temperature \( T = 293 \) K at IPNS (squares), and at \( T = 77 \) K using the DCS instrument at the NCNR (circles). The solid curves use fits to determine \( A, B \) and \( D \) as shown in Table 3. The dashed curves assume \( D = 0 \), with \( A \) estimated from equations (2) and (3) and \( B \) estimated according to equation (5) with \( \Theta_D = 520 \) K.
merit. Since the atomic fraction of magnesium is a third less in MgF\textsubscript{2} than in MgO, the absorption per molecule will be a third lower. However, this is largely compensated by MgF\textsubscript{2} having a Debye temperature approximately a third lower than MgO.

The Debye temperature \( \Theta_D \) for a given material is often found to differ somewhat depending on the specific measurement and temperature. The values of \( \Theta_D \) for the materials in Table 2 are those obtained from Ahlers (1966) for Be, Striefler & Barsch (1975) for SiO\textsubscript{2} and Batterman & Chipman (1962) for Si, whereas the values for Al\textsubscript{2}O\textsubscript{3} and MgO are those used by Mildner & Lamaze (1998) and Thiyagarajan et al. (1998). Freund (1983) has also given values for Debye temperature for many filter materials, typically lower than the above reference values.

Fig. 6 shows the fitted curves of the figure-of-merit \( R \) of four different filter materials (Si, Al\textsubscript{2}O\textsubscript{3}, MgO and MgF\textsubscript{2}) as a function of wavelength. Table 3 gives the fit parameters used for these curves; the values for MgF\textsubscript{2} are from this article, while those for Al\textsubscript{2}O\textsubscript{3} are from Mildner & Lamaze (1998), those for MgO are from Thiyagarajan et al. (1998) and those for Si are from Freund et al. (1985). High-quality single-crystal silicon is the cheapest material but gives the poorest performance owing to the much higher atomic absorption cross section of Si. The next cheapest material to obtain in suitable quality is sapphire, Al\textsubscript{2}O\textsubscript{3}. The rather high Debye temperature allows this material to be used at room temperature with performance nearly twice as good as that of cooled silicon. MgO and MgF\textsubscript{2} when cooled both show much better performance than sapphire. Both materials cost more than sapphire but are readily available in suitable quality. MgO single crystals are often used as substrates for growing thin films. High-quality MgF\textsubscript{2} is used in refractive optics with infrared radia-

### Table 3

<table>
<thead>
<tr>
<th>Material</th>
<th>( T ) (K)</th>
<th>( A ) (m\textsuperscript{-1} nm\textsuperscript{3})</th>
<th>( \Sigma_0 ) (m\textsuperscript{-1})</th>
<th>( B ) (10\textsuperscript{-3} nm\textsuperscript{3})</th>
<th>( D ) (10\textsuperscript{-3} nm\textsuperscript{3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>77</td>
<td>4.83</td>
<td>10.10 (4)</td>
<td>2.11</td>
<td>0</td>
</tr>
<tr>
<td>Al\textsubscript{2}O\textsubscript{3}</td>
<td>300</td>
<td>8.11 (6)</td>
<td>21.2 (4)</td>
<td>1.61 (8)</td>
<td>1.29 (10)</td>
</tr>
<tr>
<td>MgO</td>
<td>77</td>
<td>2.62 (24)</td>
<td>38.4 (2)</td>
<td>1.3 (1)</td>
<td>0</td>
</tr>
<tr>
<td>MgF\textsubscript{2}</td>
<td>77</td>
<td>1.49 (7)</td>
<td>32.8 (2)</td>
<td>0.35 (6)</td>
<td>2.85 (15)</td>
</tr>
<tr>
<td>MgF\textsubscript{2}</td>
<td>293</td>
<td>6.32 (13)</td>
<td>32.8 (2)</td>
<td>3.68 (10)</td>
<td>0.72 (6)</td>
</tr>
</tbody>
</table>

Data were allowed to differ from the calculated values shown in Table 2.

We thank Denis Wozniak for his assistance in the SAND experiments at IPNS funded by the US Department of Energy, BES, under contract No. DE-AC02-06CH11357 to UChicago.

Figure 6

The relative figure-of-merit \( R = \Sigma_0 \lambda / \Sigma_0 = 3, \) as a function of wavelength for four single-crystals filters. The parameters used in the fits are shown in Table 3; the values for MgF\textsubscript{2} are from this article, while those for Al\textsubscript{2}O\textsubscript{3} are from Mildner & Lamaze (1998), those for MgO are from Thiyagarajan et al. (1998) and those for Si are from Freund et al. (1985).
Argonne, LLC, and Juscelino Leao and John Copley on DCS at the NCNR for aid in setting up the close cycle refrigerator and data analysis. The elements of this work utilizing the DCS, PCD and NG3 30 m SANS instruments at the NCNR are supported in part by the National Science Foundation under agreement No. DMR-0454672.

References