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Windows for small-angle X-ray scattering cryostats

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To determine the suitability of commonly used windows for small-angle X-ray scattering, a range of materials, including Kapton, (aluminized) Mylar, beryllium, high-purity aluminium foil, mica and silicon nitride have been studied. At small wavevector transfers, Q, in the range 2×10^{-3} to 0.2 nm^{-1}, the scattering from Kapton, mica and beryllium is reasonably well described by power laws in Q with exponents of −3.25, −3.6 and −3.9, respectively. There are large variations in the scattering from mica, but a freshly cleaved natural mica window was by far the weakest scatterer. For applications where radiation in the infrared or visible range should be blocked, aluminized Mylar is the most suitable material. Both Mylar and Kapton can be used to make very simple demountable superfluid-tight windows using indium O-ring seals.

Keywords: SAXS; windows; parasitic scattering; cryostats.

1. Introduction

When performing small-angle X-ray scattering (SAXS) experiments on weakly scattering systems, it is important to minimize the background. An ideal SAXS set-up would be maintained entirely in vacuum, including the sample environment, and avoid all forms of windows. A complete in-vacuum design is not easily compatible with a cryostat, since in practice it is difficult to integrate a cryostat vacuum space with an X-ray beamline. Furthermore, some form of shielding against thermal radiation is required, and in many instances the sample cell has to be hermetically sealed. Hence, a cryogenic SAXS set-up, as shown in Fig. 1, requires a variety of windows. The presence of windows will lead to additional small-angle scattering, and it is important to have window materials that minimize parasitic scattering.

There have been a few papers reviewing the qualities of various window materials individually (Suzuki *et al.*, 1998; Bösecke & Diat, 1997) and a very good review of several typical windows by Ewen *et al.* (1981) as well as by Henderson (1995). However, in the last ten years the wide availability of synchrotron-based SAXS spectrometers has made measurements down to the ultra-small-angle X-ray scattering (USAXS) regime ($Q < 0.01$ nm^{-1}) routine. Consequently, there is a need for published data on the scattering from common window materials in the regime of very small wavevector transfer, Q. We have performed a survey of the SAXS from a selection of readily available window materials. The materials studied were Kapton, Mylar, aluminized Mylar, beryllium, aluminium, Si_3N_2 and mica. Details of the window materials are given in Table 1. All these materials have high mechanical strength and low atomic number, allowing good X-ray transmission and relatively little parasitic scattering.

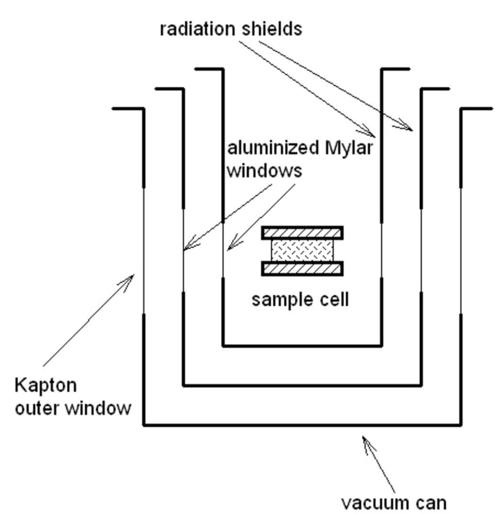

Figure 1
Typical arrangement of an experimental cell and tails of a cryostat intended for SAXS experiments. The outer windows have to withstand a pressure difference of 1 bar, the windows on the radiation shields block infra-red radiation while being transparent to X-rays, and the windows on the cell may have to be vacuum-, pressure- or even superfluid-tight.

Table 1
Window materials described in this paper.

Material	Thickness (μm)	Preparation	Source	Transmission at 8 keV
Kapton	13		Dupont (http://www.dupont.com/kapton)	0.99
	25		Dupont	0.98
	51		Dupont	0.96
Beryllium	254	IF-1 polished	Brush-Wellman (http://www.brushwellman.com)	0.95
	254	IF-1 unpolished	Brush-Wellman	0.95
Mylar	25	Clear	Dupont-Teijin (http://www.dupontteijinfilms.com)	0.98
	25	Aluminized (two-sided 1 μm)	Sunfilm (http://www.sunlightsupply.com)	0.94
Mica	12		Goodfellow (http://www.goodfellow.com)	0.88
	25	Freshly cleaved	Goodfellow	0.77
Al	10	99.999 pure, cold rolled	Goodfellow	0.88
Si_3N_4	1		Cornell University	0.99

A second consideration that is of special relevance to cryogenic windows is the need to prevent the transmission of infra-red radiation from room temperature into the sample environment. A window with an area of 1 cm^2 will radiate a power of 46 mW, while a 40 K window still radiates some 15 μW cm^{-2}. These heat loads can degrade the performance of the cryostat or produce temperature gradients in the sample. Thermal radiation can effectively be blocked by metal foils of sufficient thickness. In order for a window to transmit X-rays and minimize scattering, it is preferable to use a low-Z metal. Beryllium foils are usually thick enough not to transmit thermal radiation but, as shown below, show considerable low-angle scattering. Polymer films such as Kapton, Mylar, as well as mica, are transparent and require metallization. For electromagnetic radiation at frequency ν the skin depth in the metal is given by

$$\delta = 1/(\pi \mu_0 \sigma \nu)^{1/2}, \tag{1}$$

where $\mu_0 = 4\pi \times 10^{-7}$ H m^{-1} and σ is the conductivity of the metal. In the case of aluminium, $\sigma \simeq 2.5 \times 10^7$ S m^{-1}. For thermal radiation from a source at temperature T, the maximum in the spectrum is proportional to ν and the corresponding skin depths at 300 K and 40 K are 0.18 μm and 0.49 μm, respectively. Consequently, ‘super insulation’, used as multi-layer insulating material in cryogenic environments, which has a metallization in the range 0.05 μm to 0.1 μm, is entirely unsuitable for window applications. On the other hand, we found that aluminized Mylar intended as light reflector for indoor horticultural activities has a metallization of around 1 μm (on each side), sufficient to prevent transmission of thermal radiation through the cryostat windows. As an alternative one can use very thin high-purity (required to minimize parasitic X-ray scattering) aluminium foil. Supermarket-grade aluminium foil (Cub foods brand) was tested, but yielded substantially more scattering. This avoids scattering contributions from the polymer film, but the unsupported metal foils are quite fragile and difficult to work with.

2. Experimental set-up and materials

Measurements were made at two different beamlines at the Advanced Photon Source (APS). The UNICAT USAXS instrument (Jemian & Long, 1990; Long *et al.*, 1999) uses a Bonse–Hart geometry on an undulator beamline at the APS. Slits were adjusted to provide a 500 μm × 2000 μm beam incident on the sample and measurements were made at 10.6 keV. The beamline vacuum is terminated just after the incident slits with a beryllium window, so the X-ray beam is in air until the final detector. Parasitic small-angle scattering from air along the flight path is subtracted by making a second measurement without the sample in place. The minimum Q is determined by instrumental scattering and is typically $\sim$0.002 nm^{-1}. Since the direct beam can be measured on the same scale as the data, absolute X-ray intensities are obtained without reference to a secondary standard (Long *et al.*, 1991).

The X-ray scattering set-up at sector 8 of the APS employs a pinhole SAXS geometry. Measurements were made using a 20 μm × 20 μm beam size at an energy of 7.7 keV. Normalization of the SAXS data is based on previous calibration of the instrument against secondary standards calibrated at the UNICAT beamline. Data were recorded on a CCD area detector. The scattering from all the samples measured was found to be azimuthally symmetric and was reduced to a one-dimensional scattering pattern by circularly averaging. The X-ray path is enclosed entirely in vacuum up to 0.12 m in front of the CCD detector. Parasitic scattering was below detectable levels. Images were accumulated by averaging 50 1 s exposures.

Table 1 lists the different materials studied, their source and their preparation conditions. All are common window materials that have been used in similar forms and thickness at the APS. The thicknesses selected for the measured samples were typical of those used in situations where it was necessary to sustain a pressure differential of at least 1 bar over approximately a 2.5 cm-diameter window. The two exceptions are for the aluminium foil, which is not considered for use as a pressure window but rather as a thermal radiation shield, and Si_3N_4, which can only sustain a pressure difference of 1 bar over an area of approximately 1 cm^2. It is difficult to provide more definitive guidelines for appropriate window thicknesses since most windows are significantly deformed under pressure, and a theoretical treatment of the mechanical stress within a window deformed by more than its own thickness is quite complicated.

3. Results

The measured scattering data are shown in Fig. 2. The data from the two different set-ups agreed to within approximately

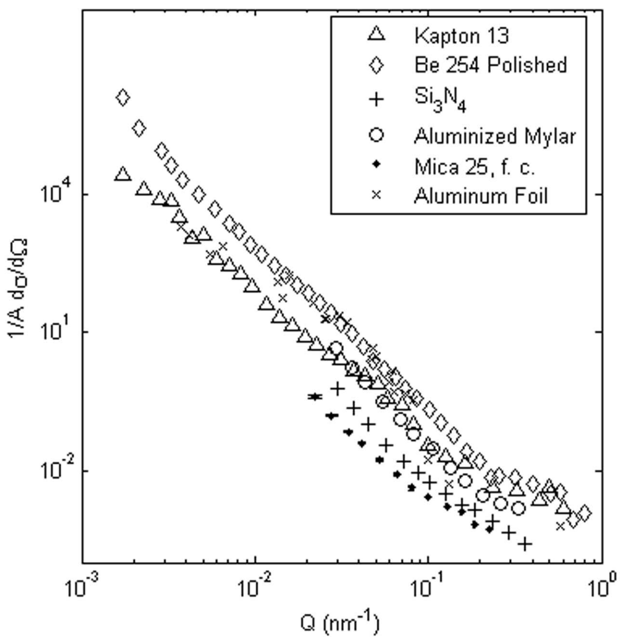

Figure 2
Comparison between the small-angle scattering from several window materials described in the text. Error bars are shown for the mica window. For all other windows, error bars are smaller than the size of the symbols.

20%. The data are displayed as the differential scattering cross section per unit area, $(1/A)\,d\sigma/d\Omega$. This form does not take the thickness of the windows into effect, but makes no assumption as to whether the scattering originates from surface or bulk scattering. This distinction will be discussed, where appropriate, when each window material is described. The data can be converted to cross section per unit volume by using the window thicknesses given in Table 1.

For the Kapton (polyimide) windows the scattering intensity was found to depend linearly on the thickness of the window, for thicknesses between 13 µm and 51 µm, implying that the small-angle scattering is a bulk process. We also tested possible changes to the scattering owing to a pressure difference across the window, but found no significant change. The scattering from the Kapton windows (as well as those for some of the other windows) is well described by a simple power law in Q,

$$(1/V)\,d\sigma/d\Omega = CQ^{-\nu}, \quad (2)$$

where Q is in nm^{-1}. The parameters C and ν are shown in Table 2. These parameters are useful for evaluation of the window scattering, but it should be noted that, because of the different values of ν for the various window materials, a smaller value of C does not automatically imply less scattering.

Two different types of metal window materials were studied: pure aluminium and beryllium. Beryllium foils were purchased from Brush Wellman, in both polished and unpolished form. All foils were the highest purity available (IF-1) in order to minimize scattering from iron inclusions within the metal. When normalized by thickness, the polished foil shows less scattering than the unpolished foils. Systematic measurements as a function of thickness were not made for the metal foils, so that it is not known whether the majority of the scattering is due to surface or bulk scattering. The low-Q data for both is again well described by a power law in Q, but with an exponent significantly larger than that of Kapton. For $Q > 1 \times 10^{-2}$ nm^{-1}, the scattering from the aluminium foil resembles that of Kapton. However, for smaller Q values the scattering increases dramatically.

Table 2
Fitting parameters to equation (2).

For the purpose of comparison, the extrapolated value of $(1/V)\,d\sigma/d\Omega$ at Q = 0.01 nm^{-1} based on the power-law fits is shown.

Material	ν	C (cm^{-1})	$(1/V)\,d\sigma/d\Omega$ (cm^{-1}) at Q = 0.01 nm^{-1}
Kapton	3.3	1.8×10^{-2}	5.7×10^{4}
Beryllium, unpolished	3.9	9.8×10^{-4}	5.9×10^{4}
Beryllium, polished	3.9	4.1×10^{-4}	2.5×10^{4}
Mica, 12 µm	3.6	0.8×10^{-2}	1.3×10^{5}
Mica, 25 µm, freshly cleaved	3.5	2.2×10^{-4}	2.3×10^{3}

The scattering from double-sided aluminized Mylar (polyester) film with a thickness of 25 µm was also measured. The aluminized Mylar is useful for applications where thermal radiation or light need to be excluded. Interestingly, the aluminized Mylar actually produces less scattering than the uncoated films. We do not presently understand the origins of this observation; it may be due to a modification of the surface roughness in the aluminizing process or possibly to differences in the manufacturing process of the films themselves. The decrease in intensity with increasing Q is much stronger for Mylar than for Kapton. Over the somewhat limited Q range for which we have data for these windows, the scattering is not well described by a pure power law.

We also measured the scattering from two thin mica membranes made from natural mica mined in India. The scattering from mica is again well described by a power law over a wide range of wavevectors. However, the difference between the two samples is striking. The thinner (12 µm) membrane had been used for some indeterminate period of time as an X-ray window. It produced small-angle scattering comparable with that of the other window materials investigated. The second membrane (25 µm thick) was freshly cleaved just prior to the actual measurement and shows dramatically lower scattering. The origin of the apparent deterioration of the mica is currently unknown.

Measurements were also made on an ultra-thin Si_3N_4 membrane (1.0 µm). The membrane was manufactured at Cornell University, although commercial membranes are now available. Only one thickness of nitride was available, hence it was not possible to determine whether the scattering was of surface or bulk origin. Scattering from the nitride did not obey a power-law scattering form, and was quite weak compared with all the other materials except the mica.

The scatterings from the materials described are shown together in Fig. 2. Freshly cleaved mica produces by far the smallest amount of parasitic scattering. However, there are concerns about its long-term behavior. Of the other materials investigated here, Kapton appears to be the best window material in the USAXS regime ($Q < 0.01$ nm^{-1}). In the SAXS regime ($Q > 0.01$ nm^{-1}), Si_3N_4 is optimal. Beryllium appears to be the worst window material in either range. Aluminized Mylar is close to Kapton in both ranges. The convenience of being able to obtain aluminized Mylar films makes this a good compromise choice

In previous measurements, Henderson (1995) reached a very different conclusion regarding the suitability of various windows, finding, in particular, that beryllium was the best choice of window. The difference between that study and our own is likely due to the different ranges of wavevector studied. Henderson's work measured parasitic window scattering over the range $0.05 < Q < 2$ nm^{-1}, whereas the present study examined the range $0.002 < Q < 2$ nm^{-1}. Since beryllium was found to have the strongest power-law decay, its scattering increases more rapidly at small Q than the other window materials studied, which would make it appear to be a better material when only examined at the upper end of the wavevector range. Furthermore, based on Henderson's work, both Kapton and mica appear to have substantially more scattering at intermediate wavevectors $Q > 0.5$ nm^{-1}, which may make beryllium a more suitable choice for work in the wide-angle X-ray scattering regime.

4. Mounting of the windows

All window materials mentioned in this paper (with the possible exception of mica) can be sealed at room temperature using a simple O-ring arrangement. In cryostats that have a single vacuum space, the windows intended to block thermal radiation do not have to be sealed and, as long as some care is taken when evacuating the cryostat, will not have to withstand a significant pressure difference. In many experiments, especially those involving liquid (superfluid) helium, the windows on the cell have to be hermetically sealed. Over the years a number of, sometimes fairly involved, methods for mounting polymer windows have been suggested (Silvera, 1970; Adams, 1988). We find that in many instances gluing the windows to the cell body using a suitable epoxy provides a perfectly adequate seal. We tested windows of Kapton, Mylar and aluminized Mylar sealed to a copper body with either low- or medium-viscosity epoxies (Trabond 2115, 250 cP and FDA2, 14000 cP, both made by Tra-con (http://www.tra-con.com). All combinations of film and epoxy survived rapid cooling to 77 K, and, on pressurizing, failed through the rupture of the film while leaving the epoxy joint intact. We repeatedly cooled epoxy-sealed Kapton windows from room temperature to below 1 K.

A demountable seal between a polymer film and metal cell body can easily be achieved using an indium O-ring. Fig. 3 shows a typical arrangement. Indium wire with a diameter in the range 0.5–1.0 mm can be used, positioned around a similar size step on the cell body. A clamping ring squeezes down on to the film and indium O-ring. A small clearance between the inner edge of the clamping ring and the outer edge of the step is desirable to avoid cutting the film. Both the epoxy and the indium mounting methods have proven reliable on repeated cooling to below 0.5 K in experiments involving superfluid ^{4}He.

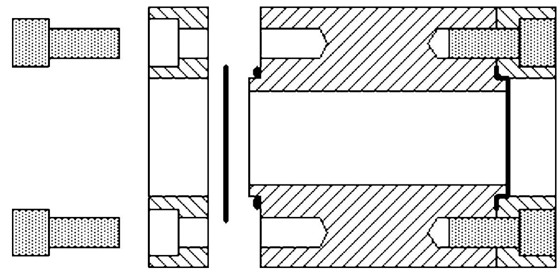

Figure 3
Kapton and Mylar films can be sealed to a cell body using an indium gasket, thus forming an easily demountable window.

5. Conclusions

In conclusion, we have characterized the small-angle X-ray scattering from a variety of window materials suitable for cryogenic applications. With the exception of mica, the volume-normalized scattering power of the materials investigated does not differ greatly. Consequently, materials that can be used in the form of thin foils or membranes clearly have an advantage. The optimal window material depends on the range of wavevectors of interest. For general-purpose SAXS work, aluminized Mylar is an inexpensive solution that gives low parasitic scattering over a wide range of wavevectors and also effectively blocks infrared and visible light. In the USAXS Q range, thin Kapton windows produce somewhat less scattering. For SAXS experiments requiring extremely low backgrounds, nitride windows, which are available with thicknesses of as little a 1 µm, can be used and, in cases where mechanical properties are less of a concern, freshly cleaved mica does an excellent job. Finally, we note that the most commonly used X-ray window material in commercial cryostats, beryllium, is not an optimal choice, largely because this brittle material is invariably used in the form of rather thick foils or shells. For Kapton, beryllium and mica the scattering could be described by a simple power law over a large range of Q. The power-law exponents vary from material to material, and we do not have a theory as to the origin of these exponents.

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