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Soft X-ray multilayer beam splitters

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A semitransparent Mo/Si multilayer beam splitter with a completely self-standing active area (10 × 10 mm) and a flatness of 1.1 nm (r.m.s.) was fabricated. The influence of the roughness of the membrane substrate on the reflectivity of a beam splitter was investigated for different materials and deposition schemes. Precise control of multilayer stress to give a slightly tensile state not only enables the fabrication of a large and flat reflection surface, but also makes it possible to etch away the supporting membrane and obtain a completely self-standing structure. The performance evaluation using synchrotron radiation revealed that the fabricated beam splitter works as a one-to-one beam splitter whose reflectivity and transmittance are both 27% (*s*-polarization, 45°, λ = 13.4 nm).

Keywords: soft X-rays; multilayers; semitransparent; self-standing beam splitter.

1. Introduction

At the beginning of the 1980s, the idea of a soft X-ray multilayer beam splitter was examined theoretically (Lee, 1982). In 1986 the first experiment using a soft X-ray multilayer beam splitter as an X-ray laser-cavity mirror was carried out at Lawrence Livermore National Laboratory (LLNL) (Hawryluk *et al.*, 1986). Beam splitters were deposited on a thin chemical-vapour-deposited SiN or BN membrane. Details of various beam splitters are listed in Table 1 (Stearns *et al.*, 1986; Ceglio, 1989).

Since 1988, studies on fabricating soft X-ray multilayer beam splitters have been undertaken at the Laboratoire pour l'Utilization du Rayonnement Electromagnétique of the Université Paris Sud and at the Laboratoire de Microstructures et de Microélectronique. At first, a W/C multilayer on a 2 µm-thick polypropylene foil was fabricated (Susini *et al.*, 1988). Subsequently, an Mo/C multilayer on a sputter-deposited SiC membrane was fabricated (Khan Malek *et al.*, 1989). Details of the structure and performance of several beam splitters are listed in Table 1. The stress of the films was effectively compensated by using an SiC/SiO$_2$ hybrid membrane.

Before this, intrinsic stress of the multilayers had not been positively controlled in order to preserve the multilayer properties. At the beginning of the 1990s, the use of a multilayer in the feasibility studies of extreme ultraviolet lithography (EUVL) raised new problems in that the intrinsic stress of the multilayer causes deformation of the mirror substrate, degrading imaging performance. Therefore, the origins of multilayer stress and ways in which it could be controlled were extensively investigated (Kola *et al.*, 1992; Nguyen, 1994; Windt *et al.*, 1995).

In addition, Kortright predicted that transmission multilayer filters could be used as phase retarders (Kortright & Underwood, 1990). Positive control of the multilayer stress and new capabilities of transmission multilayers opened a new era in the fabrication of multilayer beam splitters. As a result of collaboration between Lawrence Berkeley National Laboratories (LBNL) and Tohoku University, fully self-standing Mo/Si multilayer transmission phase retarders were fabricated by using the lift-off-and-remount method (Kortright *et al.*, 1992; Nomura *et al.*, 1992). The working range of the multilaer transmission filters was expanded to the C *K*-edge using Cr/C multilayers (DiFonzo *et al.*, 1994). At LBNL, the multilayer stress was controlled by changing the relative thickness of the multilayer (Γ) (Nguyen *et al.*, 1994). At LLNL, beam splitters for interferometric experiments (Da Silva *et al.*, 1995) were also fabricated. Performance details of these beam splitters are given in Table 1.

We have been studying the fabrication of beam splitters since 1993, and our goal is to fabricate a completely self-standing multilayer beam splitter and to obtain a high manufacturing yield. The main fabrication issues to be considered are the following: (i) for high manufacturing yield, membrane technologies are crucial; (ii) for preserving figure quality and mechanical strength, multilayer stress should be precisely controlled at a slightly tensile state; (iii) for high reflectivity, roughness of the initial supporting membranes should be minimized; (iv) for high transmittance, a process for removing the supporting membrane is crucial; and (v) for preserving multilayer properties, degradation during the fabrication process should be minimized.

In a previous paper (Tinone *et al.*, 1996), we clarified the relationship between the r.f. power applied to the magnetron source and the intrinsic stress of Mo and Si single and multilayered films. By using this relationship, we could very precisely control the stress of Mo/Si multilayer films at values ranging from nearly zero to a slightly tensile state without changing the other deposition parameters. This helped to minimize degradation of multilayer properties. We demonstrated the fabrication of 10 × 10 mm Mo/Si free-standing multilayers with flatnesses better than 5 nm (r.m.s.).

In this paper we describe the solution for the remaining issue, that is, the evaluation of the influence of the supporting membrane material and deposition processes on the reflectivity of the multilayers, and optimization of the removal process of the supporting membrane. As a result of this study, we were able to fabricate large-area self-standing multilayer beam splitters with high reflectivities, transmittances and manufacturing yields.

2. Influence of the surface roughness of supporting membranes

Multilayer beam splitters were fabricated by a multistep process utilizing the membrane technology developed for X-ray mask fabrication. The process sequence has been described in detail previously (Haga *et al.*, 1996). We have used two different materials and deposition methods for making the supporting membrane: low-pressure chemical-vapour-deposited (LPCVD) SiN and electron-cyclotron-resonance-plasma chemical-vapour-deposited (ECR-plasma CVD) SiC (Shimada *et al.*, 1995). We compared three different surface finishes of the supporting membrane: as-deposited, back-surface (proposed by the LLNL group) and polished. In summary, we prepared four kinds of self-supporting membranes with a 10 × 10 mm open area: (*a*) as-deposited SiN, (*b*) back-surface SiN, (*c*) polished SiN, (*d*) as-deposited ECR-plasma CVD SiC and, as a reference, (*e*) a polished Si wafer substrate. All the membranes were about 200 nm thick and their stress was controlled to about 20 MPa in the tensile state.

Table 1
Soft X-ray beam splitters.

Reference	Multilayer	Membrane	Size	Peak reflectivity (%)	Transmittance (%)	Remarks (λ = wavelength, θ = angle from normal incidence)
Stearns *et al.* (1986)	Mo/Si (26 pairs)	Si_3N_4 (30 nm)	<5 × 15 mm	~20	~4	$\lambda = 20.8$ nm, $\theta = 0.5^\circ$
Ceglio (1989)	Mo/Si (13 pairs)	Si_3N_4 (30 nm)	10–20 mm^2	~13.4	~45	$\lambda \simeq 13$ nm, $\theta = 0.5^\circ$
Susini *et al.* (1988)	W/C (15 pairs)	Polypropylene (2 μm)	25 × 12 mm	0.3	0.03	$\lambda = 1.24$ nm, $\theta = 79.05^\circ$
Khan Malek *et al.* (1989)	Mo/C (35 pairs)	SiC (300 nm)	10 × 10 mm	6	0.45	$\lambda = 1.33$ nm, $\theta = 78.5^\circ$
Nomura *et al.* (1992)	Mo/Si (40–80 pairs)	Self-standing	8 mm diameter	81	7	$\lambda = 12.8$ nm, $\theta = 45^\circ$
Nguyen *et al.* (1994)	Mo/Si (6 pairs)	SiN (150 nm)	2.5 × 2.5 mm	~15	~10	$\lambda = 13.6$ nm, $\theta = 45^\circ$
Da Silva *et al.* (1995)	Mo/Si (8–12 pairs)	SiN (100 nm)	12 × 12 mm	20	15	$\lambda = 15.5$ nm

Mo/Si multilayers were deposited on these different membranes using an r.f. magnetron sputter system, described in detail previously (Tinone *et al.*, 1996). The same deposition conditions were used on the different membranes, which were optimized to control the stress of Mo/Si multilayers (N = 20.5 pairs, d = 10 nm, Γ = 0.36) to about 50 MPa in the tensile state. The differences between the membranes did not interfere in the figure quality of the beam splitters and better than 5 nm (r.m.s.) flatness was obtained.

The performance of the fabricated beam splitters was evaluated using a soft X-ray reflectometer at the NTT synchrotron radiation facility (Super-ALIS, SBL-8). The reflectivities for *s*-polarized radiation at an incident angle of 45° are summarized in Table 2. The difference in reflectivities can be explained by the roughness of the multilayer, which is mainly caused by the surface roughness of the membrane. Using an atomic force microscope (AFM), we measured the initial surface of the membranes before multilayer deposition (Fig. 1). The measured reflectivities were evaluated using Debye–Waller factors (DWF). Table 2 summarizes the measured roughnesses and estimated DWF for each of the samples studied. It can be clearly seen that polished SiN (Fig. 1*c*), as-deposited ECR-plasma CVD SiC (Fig. 1*d*), and the polished Si wafer substrate (Fig. 1*e*) are very smooth. As-deposited SiN (Fig. 1*a*) surfaces are filled with square structures, grains of polycrystal, approximately 400 nm across. These structures are smoothed out by polishing (Fig. 1*c*), but the act of polishing can also damage the surface by scratching it (Fig. 1*f*). Depending on the position of measurement, the roughness of polished SiN surfaces can be as small as 1.1 nm or as large as 3.3 nm in areas where scratches remain. Using the back-surface of the membrane had some effect, but there were still grains that reduced the smoothness of the surface (Fig. 1*b*). In conclusion, we verified the importance of the quality of the surface of the supporting membrane for the reflectivity of the beam splitter. The highest reflectivity was obtained using an ECR-plasma CVD SiC membrane. It is smooth as deposited, whereas the roughness of as-deposited LPCVD SiN membranes restricts their use as substrates for multilayer beam splitters. They need to be polished before multilayer deposition if similar results are to be obtained. Therefore, ECR-plasma CVD SiC membranes are the best choice from the manufacturing point of view.

Table 2
Influence of the membrane on the multilayer reflectivity.

Membrane material and surface finish	Reflectivity (%)	DWF σ (nm)	Measured roughness (r.m.s.) (nm)
(*a*) As-deposited LPCVD-SiN	2.5	2.77	3.19
(*b*) Back surface of LPCVD-SiN	22.0	1.80	1.41
(*c*) Polished LPCVD-SiN	38.0	1.55	1.24
(*d*) As-deposited ECR-plasma CVD-SiC	40.0	1.47	1.03
(*e*) Polished Si substrate (reference)	49.0	1.22	0.86

3. Membrane removal process

The last fabrication issue is the complete removal of the supporting membrane in order to achieve high transmittance. Supporting membranes were removed by reactive ion etching (RIE). In this removal process, the damage to the multilayer properties needs to be considered. The first problem was the radiation from plasma, which increased the temperature of the multilayer and annealed it. This caused the stress of the multilayers to change, degrading the figure quality, or demolishing the multilayer structure. In the worst case, the rapid thermal expansion of the multilayer films caused them to break. Therefore, we reduced the initial membrane thickness to 200 nm in order to make the total etching time as short as possible, while maintaining manufacturing yields. Freon (CF_4, C_2F_6) was used as the etching gas (Matsuo, 1980). To improve the etching rate for membrane material under low power density, oxygen gas was mixed with the Freon. Typical etching rates for SiN and SiC were 34 and 18 nm min^{-1}, respectively, when the r.f. power density was 0.13 W cm^{-2}

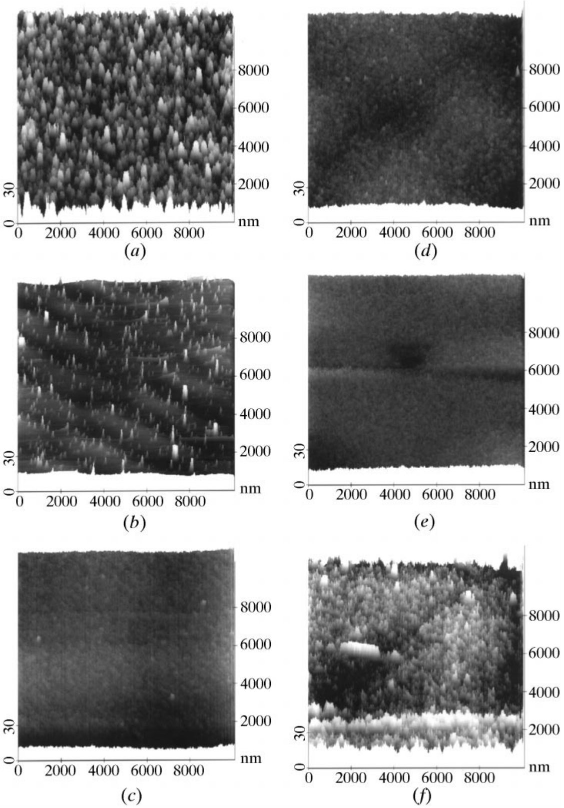

Figure 1
AFM surface profiles of different supporting membranes. (*a*) As-deposited SiN, (*b*) back-surface of SiN, (*c*) polished SiN, (*d*) as-deposited ECR-plasma CVD SiC, (*e*) polished Si wafer substrate, and (*f*) another portion of polished SiN (showing scratches).

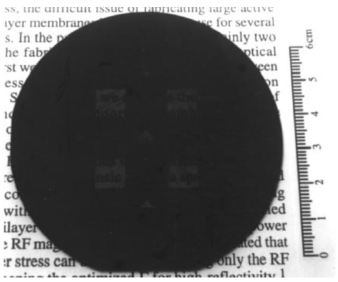

Figure 2
Photograph of the beam splitter.

and the working pressure was 6 Pa. Even under these etching conditions, the temperature of the multilayer films increased and figure quality was degraded. We therefore used an intermittent etching sequence, in which the etching process was divided into short periods of 2 min. This helped to reduce the temperature rise over 10 min intervals. Measuring the surface figure revealed no significant change in surface quality.

Another problem was multilayer damage due to the etching process itself. Etching selectivity of the Mo/Si multilayer against the membrane materials was less than 0.5, therefore over-etching will cause serious damage to multilayers. Moreover, reflectivity measurements revealed that some of the layer pairs were lost during etching, even though the membrane still remained. This indicated that the etching damage was induced not only by the over-etching but also by the fluorine radicals that attacked the front surface of the multilayer films. We used an Ru layer on both sides of the Mo/Si multilayer for preventing damage from both over-etching and fluorine radicals. At around 13 nm, Ru has nearly the same optical properties as Mo, and its etching selectivity against the membrane materials is more than 20. The thickness of each Ru layer was made the same as that of each Mo layer and the Ru layer replaced the Mo layer to preserve the multilayer structure. Because of the Ru etch-protection layer, the supporting membrane was completely removed without any degradation of the properties of the multilayer.

4. Fabrication and evaluation of a self-standing beam splitter

Self-standing Mo/Si multilayer beam splitters were fabricated by depositing beam-splitter layers on an initial supporting membrane of ECR-plasma CVD SiC and then removing that membrane by RIE. To obtain a one-to-one beam splitter, beam splitters with 5–10 layer pairs were tested. The measured flatness of these beam splitters was better than 5 nm (r.m.s.) in all cases, and the best flatness was 1.1 nm (r.m.s.) in the central 7 × 7 mm active area. A photograph of a 7.6 cm wafer with four 10 × 10 mm beam splitters is shown in Fig. 2. Fig. 3 shows the reflectivity and transmission curves of an eight-pair Mo/Si multilayer beam splitter. The same reflectivity and transmittance rates, 27%, were obtained at a wavelength of 13.4 nm. Thus the fabricated beam splitter worked as a one-to-one beam splitter at an incident angle of 45°.

5. Conclusions

In summary, we were able to fabricate large-area self-standing multilayer beam splitters with high reflectivities, transmittances and manufacturing yields. Investigation of the supporting membrane materials and deposition processes clarified their influences on the reflectivity of the multilayers and indicated that ECR-plasma CVD SiC is the best material to use as the initial supporting membrane. To remove this membrane, we used an intermittent etching sequence and an Ru etch-protection layer to prevent degradation of the multilayer properties. The performance of the beam splitter is very promising and indicates the likelihood of their use as optical components for soft X-ray interferometry, laser cavity, holography and in other new applications of soft X-ray optics.

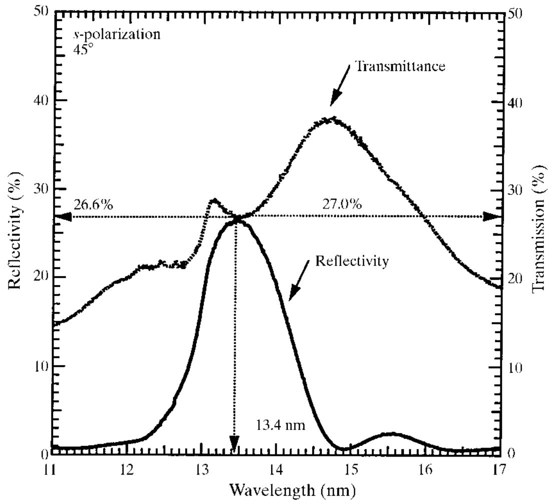

Figure 3
Measured reflectivity and transmittance of the beam splitter.

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