

Mo/Si aperiodic multilayer broadband reflective mirror for 12.5–28.5-nm wavelength range

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Aperiodic molybdenum/silicon (Mo/Si) multilayer designed as a broadband reflective mirror with mean reflectivity of 10% over a wide wavelength range of 12.5–28.5 nm at incidence angle of 5° is developed using a numerical optimized method. The multilayer is prepared using direct current magnetron sputtering technology. The reflectivity is measured using synchrotron radiation. The measured mean reflectivity is 7.0% in the design wavelength range of 12.5–28.5 nm. This multilayer broadband reflective mirror can be used in extreme ultraviolet measurements and will greatly simplify the experimental arrangements.

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In the extreme ultraviolet (EUV) region, the real parts of the refractive indices of all materials are very close to unity. This, along with high absorption, makes the reflectivity of a single-layer film mirror extremely at near-normal incidence of only 10^{-4} based on the Fresnel formula^[1]. Multilayer film mirror^[1,2] composed of an absorber and a spacer layer can improve reflectivity in the EUV region, which essentially indicates the enhancement of multiple-beam interference. For example, the highest near-normal incidence reflectivity of 70% at 13.5 nm has been successfully obtained using molybdenum/silicon (Mo/Si) multilayer under the motivation of EUV lithographic in the integrated circuit industry. However, this interference structure of multilayer causes the high reflectivity to be effective only over a very narrow wavelength range. For instance, the bandwidth (full-width of half-maximum, FWHM) of traditional periodic Mo/Si multilayer is only 0.9 nm at 13.5 nm. Such narrow bandpass property may be disadvantageous for some studies, such as microscopy, astronomical telescope, and spectroscopy, in which both high reflectivity and broadband performance are desired and broadband multilayer mirrors are required. In order to increase the bandpass, graded-multilayer structures called “supermirror” were proposed for neutron optics and then developed for X-ray astrophysics and synchrotron radiation applications^[3–5]. Theoretically, supermirror is a type of aperiodic multilayer, in which the layer thickness changes along the depth of the multilayer stack to satisfy the Bragg condition in wide wavelength or angle range to get flat and high reflectivity. Wang *et al.* successfully further developed this kind of aperiodic multilayer as EUV broadband polarization elements, including reflective analyzer and transmission phase retarder^[6–11]. The use of a wide spectral range multilayer would greatly simplify experiments because the optical system would have a fixed shape and incident angle. In this letter, the design, fabrication, and measurement of broadband Mo/Si aperiodic

multilayer reflective mirror working in the 12.5–28.5-nm wavelength range at incident angle of 5° are described.

The design of a broadband multilayer requires the determination of materials combination and layer thickness distribution. The best materials combinations are those that form smooth and abrupt interfaces with high optical contrast and low absorption^[1,12]. Mo/Si combination is suitable for wavelengths longer than 12.4 nm because of the Si L-shell absorption edge. Mo/Si multilayer has been widely used in EUV lithograph and astronomy physics for its high stability and fairly high reflectivity.

The layer thickness distribution is a solution of the inverse problem of the multilayer reflectivity, which is difficult to calculate. There are three algorithms used in our group: simulated annealing, random search, and local optimization. The ideal solution is a global optimization, which will prevent convergence to local minima, rather than to the global minimum. However, the global method will result in lengthy calculations, and it is difficult to obtain ideal layer stack suitable for deposition (layer thickness changes smoothly and is not too thin to deposit). In this letter, the design method is a combination of analytical calculation with numerical optimization. The main ideas of this method are as follows: firstly, based on simplified analytic formula, approximate solution of multilayer structures with reflectivity profiles was deduced, which oscillated strongly over the design wavelength range. Secondly, the merit function was minimized using the standard Levenberg-Marquardt algorithm. As a result, the risk of local minimization of the merit function disappears because the analytical depth distribution is very close to that providing a sufficiently deep minimum of the merit function. Finally, the layer distribution can be achieved by minimizing the merit function (MF):

$$\text{MF} = \frac{1}{m} \sum_{i=1}^m [R(\lambda_i) - R_o]^2, \quad (1)$$

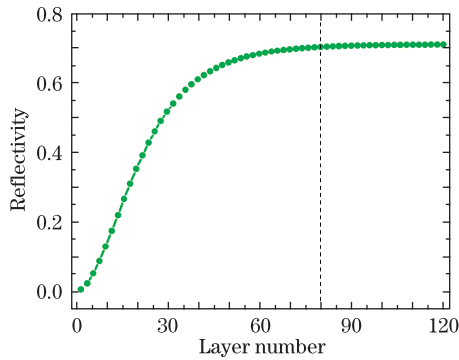


Fig. 1. Layer number dependence of calculated reflectivities of Mo/Si multilayers for use at wavelength of 14.5 nm at incident angle of 5.0° .

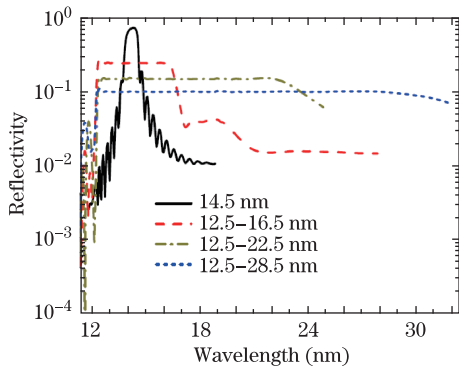


Fig. 2. Calculated reflectivities of broadband Mo/Si aperiodic multilayer mirrors designed for the wavelength ranges of 12.5–16.5, 12.5–22.5, and 12.5–28.5 nm.

where the summation is over a selection of discrete wavelengths with the number of m in the desired range; R_0 is the desired reflectivity; $R(\lambda_i)$ is the reflectivity of the multilayer at the i th wavelength. Only randomly selected layer thickness changes that decreased MF are retained, leading to an optimized layer thickness distribution that provides a minimum value of MF. At each stage in the optimization, the reflectivities are calculated using the Fresnel formula and tabulated optical constants^[1,13].

We used the initial layer distribution produced by method proposed in Ref. [14], which is used to design X-ray supermirror. We extended this method to successfully design broadband high-reflective and polarization aperiodic multilayer in EUV^[6–11]. The most important modification is the initial layer distribution produced; the MF is optimized by the simplex algorithms at a reasonable computation time. The most advantageous is the optimization, which is very effective at only several minutes using a standard personal microcomputer (PC)^[15]. In this letter, the design incidence angle was 5° , and the layer number of multilayer was 80 (40 bi-layers). The layer number at which the reflectivity near the short wavelength reached saturation to ensure high reflectivity within the entire design wavelength range was chosen, as shown in Fig. 1. The layer thickness distribution was used as the independent variable values (values: 80) during the recursive optimization.

Figure 2 shows some optimized results for wavelength ranges of 12.5–16.5, 12.5–22.5, and 12.5–28.5 nm, with the desired reflectivities (R_0) at 25%, 15%, and 10%, respectively. The periodic multilayer for 14.5-nm wave-

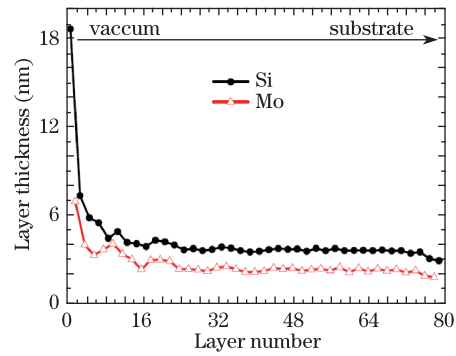


Fig. 3. Layer thickness distribution of the aperiodic Mo/Si multilayer designed for 12.5–28.5-nm wavelength range.

length is shown for comparison with the aperiodic broadband multilayers. It can be seen that the reflectivity profiles are all reasonably flat in the design wavelength ranges.

For the design wavelength range of 12.5–28.5 nm, the bandwidth is 16.0 nm, which covers most of the wavelength range where Mo/Si can be used. The working wavelength was extended from Si-L absorption edge (12.4 nm) to Mg-L edge (25.1 nm), making this kind of broadband mirror convenient in the experiments. Even though the desired reflectivity is only 10%, this low reflectivity is enough for synchrotron radiation applications because of its high photon flux. Hence, this broadband multilayer was selected to be further prepared and measured in this letter. Figure 2 shows the layer thickness distribution in the multilayer stack. The layer thickness of Mo varies between 1.8 and 7.0 nm, whereas that of Si varies between 2.9 and 18.6 nm. The designed Mo/Si multilayer mirror was deposited using an ultrahigh vacuum direct current magnetron sputtering system^[16] onto 20×30 (mm) polished silicon substrates. The base pressure of the vacuum chamber was 4×10^{-4} Pa. The working pressure of Ar (purity of 99.9999%) was 0.1 Pa during deposition. The power applied to Mo (purity of 99.95%) and Si (purity of 99.999%) was kept constant at 20 and 35 W, respectively. For layer thickness measurement and quality control, the deposited multilayer was measured using a grazing angle X-ray diffractometer.

After preparation, the reflectivity of broadband Mo/Si multilayer mirror was measured using a reflectometer on beam line U27 at the National Synchrotron Radiation Laboratory (NSRL) in Hefei, China. The monochromator grating (600 lines/mm) with spectral resolution $\lambda/\Delta\lambda$ above 192 was used in the measurement. The silicon filter was inserted into the beam path to improve the spectral purity. Reflectivity was measured using wavelength scanning mode at the fixed incident angle of 5° . The measured mean reflectivity is 7.0% in the design wavelength range of 12.5–28.5 nm. This multilayer broadband reflective mirror can be used in EUV measurements and will greatly simplify experimental arrangements. The results are summarized and compared with the design values, as shown in Table 1 and Figs. 4 and 5. The measured curve shows that an almost flat reflectivity was obtained over the broadband wavelength range of 12.5–25.8 nm. The measured results were fitted by using the Stearns model^[17] and optical constant form center for X-ray optics (CXRO) to estimate the influence of interface

Table 1. Design Parameters, Calculated, and Measured Data of Mo/Si Multilayer Mirror for the Spectral Bandwidth of 12.5–28.5 nm Shown in Figs. 4 and 5 (Error is the Standard Deviation)

Wavelength Range (nm)	Spectral Bandwidth (nm)	Layer Number	R_{mean} (%) (Calculated)	R_{mean} (%) (Measured)	Interface Roughness/Diffusion (nm)
12.5–28.5	16.0	80	10.0 ± 0.1	7.0 ± 1.3	0.8

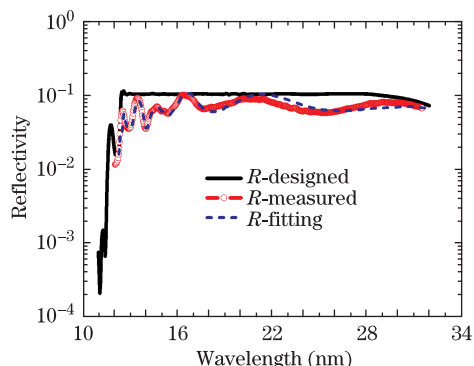
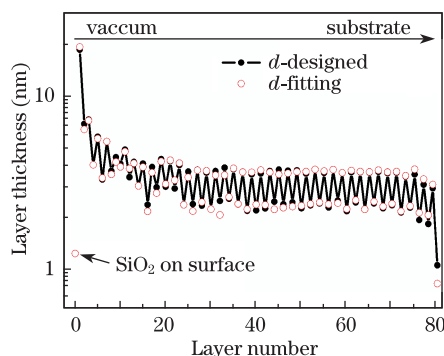
Fig. 4. Measured, designed, and fitting reflectivity curves of Mo/Si broadband multilayer mirror at incident angle of 5° .

Fig. 5. Layer thickness distribution of the designed and fitted results of aperiodic Mo/Si multilayer designed for 12.5–28.5-nm wavelength range.

imperfection, roughness, and diffusion. In this model, each interface is assumed to be described by a surface having statistically random roughness with a well-behaved power spectrum. The model also accounts for arbitrary correlation of the roughness between different interfaces. Because the surface of the top Si layer was partially oxidized into SiO_2 ^[19], the fitting multilayer structure model was modified into $\text{SiO}_2/[\text{Si}/\text{Mo}]_{40}/\text{substrate}$, and the layer thicknesses of Mo and Si are set as independent parameters during the fitting. Figure 4 shows the fitted reflectivity curve. Figure 5 shows the fitted layer thickness, including surface-oxidized layer, with consideration for design. The fitted results indicate that the first layer Si was oxidized partially to SiO_2 with thickness of 1.25 nm. The deposited layers coincide with the design, and the roughness/diffusion between the Mo and Si layers was approximately constant at 0.8 nm. This value is agreement with the report in Refs. [18–22].

In conclusion, the broadband reflective mirrors based on aperiodic Mo/Si multilayer structure with broad spectral bandwidth in the EUV region are described. These aperiodic multilayers are designed using a combined an-

alytical and numerical method. In the wavelength range of 12.5–28.5 nm, the design reflectivity curve of Mo/Si aperiodic multilayer is reasonably flat, and the mean reflectivity is 10% at the incident angle of 5° . The aperiodic Mo/Si multilayer mirror is deposited by a high-vacuum magnetron sputtering system. The reflectivity is measured by synchrotron radiation. In the desired wavelength range of 12.5–28.5 nm, the measured mean reflectivity is 7.0%, which is available for synchrotron radiation application. This kind of broadband multilayer mirror will greatly simplify experimental arrangements and is convenient since all the measurement setups in the EUV should be put into vacuum chamber. Furthermore, the design and preparation methods described in this letter are generally applicable for other material pairs and wavelength ranges in EUV applications.

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References

1. E. Spiller, *Soft X-ray optics* (SPIE Press, Bellingham, 1994).
2. C. Montcalm, R. F. Grabner, R. M. Hudyma, M. A. Schmidt, E. Spiller, C. C. Walton, M. Wedowski, and J. A. Folta, *Appl. Opt.* **41**, 3262 (2002).
3. F. Mezei, *Proc. SPIE* **1738**, 107 (1992).
4. K. D. Joensen, F. E. Christensen, H. W. Schnopper, P. Gorenstein, J. Susini, P. Høghøj, R. Hustache, J. Wood, and K. Parker, *Proc. SPIE* **1736**, 239 (1992).
5. P. Høghøj, E. Ziegler, J. Susini, A. K. Freund, K. D. Joensen, P. Gorenstein, and J. L. Wood, *Nucl. Instr. Meth. B* **132**, 528 (1997).
6. Z. Wang, H. Wang, J. Zhu, F. Wang, Z. Gu, L. Chen, A. Michette, A. Powell, S. Pfauntsch, and F. Schafers, *J. Appl. Phys.* **99**, 056108 (2009).
7. Z. S. Wang, H. C. Wang, J. T. Zhu, F. L. Wang, Z. X. Gu, L. Y. Chen, A. G. Michette, A. K. Powell, S. J. Pfauntsch, and F. Schäfers, *Opt. Express* **14**, 2533 (2006).
8. H. Wang, J. Zhu, Z. Wang, Z. Zhang, S. Zhang, W. Wu, L. Chen, A. Michette, A. Powell, and S. Pfauntsch, *Thin Solid Films* **515**, 2523 (2006).
9. Z. Wang, H. Wang, J. Zhu, Y. Xu, S. M. Zhang, C. Li, F. Wang, Z. Zhang, Y. Wu, X. Cheng, L. Chen, A. G. Michette, S. J. Pfauntsch, A. K. Powell, F. Schafers, A. Gaupp, and M. MacDonald, *Appl. Phys. Lett.* **89**, 24120 (2006).

10. Z. Wang, H. Wang, J. Zhu, Z. Zhang, Y. Xu, S. Zhang, W. Wu, F. Wang, B. Wang, L. Liu, L. Chen, A. G. Michette, S. J. Pfauntsch, A. K. Powell, F. Schafers, A. Gaupp, and M. MacDonald, *Appl. Phys. Lett.* **90**, 031901 (2007).
11. Z. S. Wang, H. C. Wang, J. T. Zhu, Z. Zhang, F. L. Wang, Y. Xu, S. M. Zhang, W. J. Wu, L. Y. Chen, A. G. Michette, S. J. Pfauntsch, A. K. Powell, F. Schafers, A. Gaupp, M. Q. Cui, L. J. Sun, and M. MacDonald, *Appl. Phys. Lett.* **90**, 081910 (2007).
12. Z. Wang, S. Zhang, W. Wu, J. Zhu, H. Wang, C. Li, Y. Xu, F. Wang, Z. Zhang, and L. Chen, *Chin. Opt. Lett.* **4**, 611 (2006).
13. P. Naulleau "The center for X-ray optics" <http://www-cxro.lbl.gov/> (May 28, 2010).
14. I. V. Kozhevnikov, I. N. Bukreeva, and E. Ziegler, *Nucl. Instrum. Methods Phys. Res. A* **460**, 424 (2001).
15. Z. Zhang, Z. Wang, F. Wang, W. Wu, H. Wang, S. Qin, and L. Chen, *Chin. Opt. Lett.* **3**, 422 (2005).
16. F. Wang, Z. Wang, J. Zhu, Z. Zhang, W. Wu, S. Zhang, and L. Chen, *Chin. Opt. Lett.* **4**, 550 (2006).
17. D. G. Stearns, *J. Appl. Phys.* **71**, 4286 (1992).
18. H. Maury, P. Jonnard, J.-M. André, J. Gautier, F. Bridou, F. Delmotte, and M.-F. Ravet, *Surf. Sci.* **601**, 2315 (2007).
19. J. H. Underwood, E. M. Gullikson, and K. Nguyen, *Appl. Opt.* **32**, 6985 (1993).
20. K. Le Guen, H. Maury, J.-M. André, H. Wang, J. Zhu, Z. Wang, and P. Jonnard, *Appl. Surf. Sci.* **253**, 8443 (2007).
21. M. Dai, Z. Zhang, J. Zhu, X. Wang, J. Xu, X. Fu, L. Bai, Q. Huang, Z. Wang, and L. Chen, *Chin. Opt. Lett.* **7**, 738 (2009).
22. J. Zhu, H. Li, X. Wang, Q. Huang, Z. Wang, Y. Li, H. Li, D. Wang, J. Zhao, and W. Lu, *Chin. Opt. Lett.* **8**, 167 (2010).