## Molybdenum/beryllium multilayer mirrors for normal incidence in the extreme ultraviolet

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> We report on a series of normal-incidence reflectance measurements at wavelengths just longer than the beryllium K-edge (11.1 nm) from molybdenum/beryllium multilayer mirrors. The highest peak reflectance was  $68.7 \pm 0.2\%$  at  $\lambda = 11.3$  nm obtained from a mirror with 70 bilayers ending in beryllium. To our knowledge, this is the highest normal-incidence reflectance that has been demonstrated in the 1–80-nm spectral range.

Key words: Beryllium, multilayer mirrors, extreme ultraviolet.

In the past few years, much attention has been devoted to the concept of extreme-ultraviolet (EUV) lithography as a possible route to very-large-scale integrated circuit manufacturing with feature sizes of 0.1 µm and smaller.1 The key item, without which this technology would be impossible, is high-reflectance multilayer mirrors for wavelengths near 10-15 nm. Some proposed systems use as many as eight normal-incidence reflections between the source and the resist-coated wafer,<sup>2</sup> and for this reason, high mirror reflectance is essential. Until now, Mo/Si mirrors have received the most attention, because this combination shows high theoretical reflectance  $(\sim 76\%)$  with 13.0-nm illumination. Intense activity has been devoted to efforts to achieve a reflectance close to this theoretical value and to demonstrate the thermal, radiation, and time stability of the multilayers. Measured reflectances of 63-65% (Ref. 3) are now achieved routinely with the Mo/Si system. On the other hand, multilayer mirrors that use Be as the spacer material with a low atomic number could theoretically achieve reflectances of 80% for wavelengths just longer than the Be K-edge at 11.1 nm. Mirrors with demonstrated reflectances of 70% or greater could have a significant effect on EUV lithog-

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raphy. For example, an increase in peak reflectance from 65% to 70% would correspond to a factor of  $(1.08)^8$ or an 80% net increase in photon throughput at the peak wavelength.

Wavelength considerations are of equal, if not greater, importance than reflectance considerations in EUV lithography. The Mo/Be multilayers have a peak reflectance near 11.4 nm compared with the 13.0–13.5-nm peak of the currently favored Mo/Si multilayers. This 16% increase in the photon energy leads to an increase in the penetration depth (1/e) in prototypical high-resolution resists, for example, poly(methyl methacrylate) from 0.24 to 0.34 µm, thus permitting a thicker resist to be used. This could lead to a higher-contrast latent image in surface imaging and near-surface imaging resists, better etch resistance, higher-sidewall angle, and lower defect density.4 In addition the thicker penetration depth opens up the possibility of new resist technologies, for example, bilayer schemes that employ new chemistry. In contrast with these potential advantages in resist development, the wavelength shift is not sufficiently significant to require a major redesign of current optical systems or to affect source considerations.

We report on a systematic investigation of the Mo/Be multilayer system, in particular a study of the peak reflectance R versus the number of bilayers Nand in various deposition conditions. We also report single observations from Nb/Be multilayers. We primarily targeted the 11.2–11.5-nm region, where theoretical reflectances are highest. The mirrors were fabricated at the Lawrence Livermore National Laboratory and tested at the Center for X-Ray Optics at Lawrence Berkeley Laboratory (CXRO/LBL).

Several groups have deposited multilayers with Be

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as the light or spacer element. The published study includes demonstrations in grazing incidence with nominally 1-keV x rays<sup>5-7</sup> and characterization of epitaxial single-crystal Be.<sup>8</sup> However, no group has reported tests of Be-containing multilayers at normal incidence in the wavelength regions of interest to EUV lithography.

In arriving at the choice of Be-containing multilayers for this investigation, we first surveyed candidate material pairs throughout the 3.0-30.0-nm region in 0.1-nm increments, looking at theoretical normalincidence reflectance with a fixed  $\Gamma = 0.4$ . We then repeated the calculation by letting  $\Gamma$  vary in 0.01 steps. Recall that  $\Gamma$  is the ratio of the thickness of the high atomic number to the entire bilayer thickness. We used the closed-form solution proposed by Rosenbluth<sup>9</sup> along with the optical constants provided by Henke et al. 10 We then selected interesting cases and confirmed the results using both a commercial multilayer code and one developed at CXRO/LBL using the formalism described by Underwood and Barbee. 11 The latter code allows for interfacial roughness, whereas both codes could model a compound formation at the interfaces. We found that, in the wavelength region just longer than 11.1 nm, Ru, Mo, Rh, and Nb in combination with Be could theoretically lead to mirrors with ~80% reflectance at normal incidence. Because so much research has been done with the Mo/Si system, we started with the analogous Mo/Be multilavers.

The mirrors were fabricated by dc magnetron sputtering in a cryopumped vacuum system. The sputtering targets and substrate face sideways to minimize debris. The sputtering targets for the Be deposition were obtained from Brush Wellman, Inc. The 3-in.-(7.6-cm-) diameter (100) silicon test wafer substrates were loaded into the system onto a rotating mandrel. As opposed to rotating over each source, the substrate was oscillated in front of the targets, so that each layer resulted from two passes. The target-to-substrate distance was 9.5 cm, the base pressure of the system was  $5 \times 10^{-8}$  Torr, and the nominal argon-sputtering pressure was 2.5 mTorr. Because Be dust is considered toxic, the face of the vacuum chamber was contained in a filtered enclosure.

The reflectance was measured at near-normal incidence (5 deg) with a laser-produced plasma source/reflectometer.<sup>12</sup> The measurements were performed with one pulse per data point with an incident intensity of  $\sim 10^5$  photons/pulse. The statistical uncertainty in the reflectance measurements is determined by the photon shot noise, and for a single measurement the relative rms uncertainty is  $\sim 0.7\%$ . The uncertainty in the measured peak reflectance is smaller than this because several points are averaged. Nonuniformity in the response of the detector may lead to an error in the measured reflectance if the direct and the reflected beams strike different spots on the detector. For this reason a silicon photodiode (International Radiation Detectors AXUV-100) with excellent uniformity (better than 0.1% at 11.3 nm) was used for these measurements. A Be filter was used to eliminate the effects of any higher harmonics or scattered light from the monochromator. The beam size at the sample was  $\sim\!1.0$  mm  $\times\,0.4$  mm.

It is possible that O could become incorporated into the Be during sputtering or that the surface could become oxidized after exposure to air. To investigate this possibility, we deposited a 0.5-µm Be film on a Be substrate for Rutherford backscattering analysis. The analysis showed a 2.9-nm equivalent thickness of BeO on the surface and a nominal 0.1-at.% level in the bulk. We assume that the multilayer mirrors ending in Be have the same amount of O on the surface as that of this test film. The O level in the bulk of this film is the same as in the target material, within experimental error. We found from modeling that the postulated BeO reduces ideal reflectance by  $\sim\!2\%$ , independent of the number of layers.

Figure 1 shows the peak reflectance versus number of bilayers. The circles represent the measured data, and the curve through the data is from a numerical model. The model parameters were d = 5.75 nm,  $\Gamma = 0.45$ , 0.68-nm rms interface roughness, and 3.0 nm of BeO on the surface. The measured reflectances do not increase as rapidly with the number of bilayers as the model suggests. This could possibly be due to the increasing value of interfacial roughness with increasing thickness. The data level off at N =70 with R = 67.5%. Also shown on this plot is a curve for ideal interfaces with no surface oxide. This curve asymptotically approaches R = 78.1%. We found that the wavelength of peak reflectance changed by 0.3 nm from the center to the edge of the wafer, corresponding to a 0.15-nm *d*-spacing variation.

After determining that N=70 gave the maximum reflectance, we adjusted  $\Gamma$  to  $0.36\pm0.02$ . The observed reflectance was  $68.7\pm0.2\%$ . This was the highest measured reflectance, and the reduced statistical uncertainty results from averaging more points at the wavelength of peak reflectance. We repeated

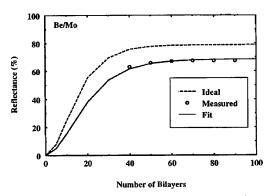


Fig. 1. Peak normal-incidence reflectance for the Mo/Be system as a function of the number of bilayers N. The dashed curve is calculated for multilayers with ideal interfaces, and the continuous curve results from a modeled interface roughness of  $\sigma=0.68$  nm, d=5.75 nm,  $\Gamma=0.45$ , and 3.0 mm of BeO on the surface. The points represent measured data.

this deposition onto a highly polished silicon wafer but found no improvement in reflectance. Several of the films N, which ended in Be, were remeasured over the four-month duration of the experiments, and it was determined that there was no loss in measured reflectance, within experimental error. The films were stored in Fluoroware containers in ambient air.

Figure 2 shows the reflectance measured from the multilayer referred to above, along with simulations. Note that the d spacing that best simulates the measurements is 5.72 nm, which, with no refractive correction, would show a peak at 11.44 nm. The observed peak at 11.3 nm indicates the necessity of accounting for a refractive shift in the wavelength corresponding to peak reflectance. We find excellent agreement between the measured and simulated data.

Several other depositions to determine the effect of certain processing conditions on observed reflectance were tried, for example, a Be/Mo multilayer with N=40,  $\Gamma=0.45$  that ended in Mo. Its reflectance was  $58.0\pm0.3\%$ , compared with  $63.0\pm0.3\%$  for the multilayer processed in identical conditions that ended with Be. We did not test for a time dependence of the reflectance of this Mo-ending multilayer. Such a phenomenon has been seen in Mo/Si multilayers ending in Mo.  $^{13}$ 

We deposited a multilayer at a reduced sputtering pressure of 1.75 mTorr. We then adjusted the power supplies to give a multilayer spacing of approximately d=5.7 nm and  $\Gamma=0.45$  to resemble the depositions shown in Fig. 1. For N=40 the reflectance was  $R=63.0\pm0.3\%$ . Although the reflectance did not increase, much better uniformity over the 7.6-cm wafer was achieved, with the wavelength of peak reflectance being within 0.1 nm.

Two multilayers were deposited at 2.5 mTorr with Nb instead of Mo. With  $N=50,~\Gamma=0.38,~$  we observed  $R=57.5\pm0.3\%$  for the mirror ending in Be and  $R=54.0\pm0.3\%$  for the mirror ending in Nb. We did not continue with the Nb-based multilayers, because for equivalent conditions it appeared that the Mo/Be mirrors performed better than the Nb/Be mirrors.

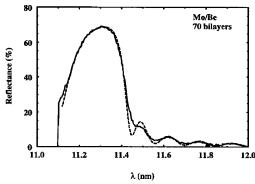


Fig. 2. Reflectance of a Mo/Be multilayer,  $\Gamma=0.36$ , N=70, ending in Be. The measured peak reflectance is  $68.7\pm0.2\%$  at 11.3 nm (continuous curve). The dashed curve represents the modeled curve; the following parameters are used: d=5.72 nm,  $\Gamma=0.36$ ,  $\sigma=0.63$  nm, N=70, 3.0 nm BeO on the surface.

Table 1. Comparison of Integrated Reflectance for the Mo/Si System and the Mo/Be System for an Eight-Bounce System<sup>a</sup>

Parameter	Mo/Si	${f Mo/Be}$
N	50	70
d $(nm)$	6.6	5.75
Γ	0.36	0.36
$\sigma$ (nm)	0.65	0.65
$R_{ m peak}$	0.031	0.048
$R_{ m integ}$	0.044	0.058
$\Delta E (eV)$	1.3	1.2
$\Delta E/E$	0.0135	0.0110

<sup>&</sup>lt;sup>a</sup>Modeling parameters are shown along with the results.

A modeling experiment was conducted to show how the integrated reflectance  $R_{\rm integ}$  compares with peak reflectance R between the Mo/Be and Mo/Si systems. The results are for an eight-bounce system. The modeling parameters and the results are shown in Table 1. After eight reflections the peak reflectance of Mo/Be is 55% greater than that of Mo/Si. The integrated reflectance increases by only 32% because of the narrower bandwidth of the Mo/Be. Thus the throughput gain in a lithography system resulting from the increased peak reflectance of Mo/Be depends on the relative bandwidths of the source and multilayers. Also, the smaller bandwidth of the Mo/Be system will impose greater constraints on the control of the d spacing for the eight reflection surfaces.  $^{14}$ 

We observed a reflectance in near normal incidence of  $68.7 \pm 0.2\%$  at 11.3 nm for a Mo/Be multilayer with 70 periods,  $\Gamma = 0.36$ , ending in Be. We believe that this is the highest normal incidence reflectance that has been attained for any material in the spectral range of 1–80 nm. For wavelengths longer than  $\sim$ 80 nm clean aluminum has a very high reflectance, and for wavelengths of less than  $\sim 1$  nm there are perfect crystals that have peak reflectances approaching unity. This new multilayer combination with its higher reflectance and shorter peak wavelength may offer significant advantages over the Mo/Si combination for EUV projection lithography. More experiments are clearly necessary to optimize the deposition conditions to minimize interfacial roughness and optimize peak reflectance.

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## References

- N. M. Ceglio, A. M. Hawryluk, D. G. Stearns, D. P. Gaines, R. S. Rosen and S. P. Vernon, "Soft x-ray projection lithography," J. Vac. Sci. Technol. B8, 1325–1328 (1990).
- D. P. Gaines, G. E. Sommargren, S. P. Vernon, and R. E. English, "X-ray characterization of a three-element condenser system for SXPL," in Soft X-ray Projection Lithography, Vol.

- 18 of OSA Proceedings (Optical Society of America, Washington, D.C., 1993), p. 66–69.
- 3. Unpublished survey results from T. W. Barbee, "Multilayer survey results and discussion," in *Physics of X-Ray Multilayer Structures*, 1994 OSA Technical Digest Series, Vol. 6 (Optical Society of America, Washington, D.C., 1994), p. ix, provided by Dan Stearns, Lawrence Livermore National Laboratory.
- O. R. Wood II, J. E. Bjorkholm, L. Fetter, M. D. Himel, D. M. Tennant, A. A. MacDowell, B. LaFontaine, J. E. Griffith, G. N. Taylor, W. K. Waskiewicz, D. L. Windt, J. B. Kortright, E. M. Gullikson, and K. Nguyen, "Wavelength dependence of the resist sidewall angle in extreme ultraviolet lithography," J. Vac. Sci. Technol. B12, 3841–3845 (1994).
- Y. Utsumi, H. Kyuragi, T. Urisu, and H. Maezawa, "Tungstenberyllium multilayer mirrors for soft x rays," Appl. Opt. 27, 3933–3936 (1988).
- O. Renner, M. Kopecky, E. Krousky, F. Schafers, B. R. Muller and N. I. Chkhalo, "Properties of laser-sputtered Ti/Be multilayers," Rev. Sci. Instrum. 63, 1478–1481 (1992).
- Y. Utsumi, H. Kyuragi, and T. Urisu, "Spatial period division with synchrotron radiation bandwidth control by W/Be multilayer mirror," J. Vac. Sci. Technol. B8, 436–438 (1990).
- 8. J. A. Ruffner, J. M. Slaughter, J. Eickmann, and C. M. Falco,

- "Epitaxial growth and surface structure of (0001) Be on (111) Si,"Appl. Phys. Lett. **64**, 31–33 (1994).
- A. E. Rosenbluth, "Computer search for layer materials that maximize the reflectivity of x-ray multilayers," Rev. Phys. Appl. 23, 1599–1621 (1988).
- 10. B. L. Henke, E. M. Gullikson, and J. C. Davis, "X-ray interactions, photoabsorption, scattering, transmission, and reflection, E=50-30,000 eV, Z=1-92," At. Data Nucl. Data Tables **54**, 181–343 (1993).
- J. H. Underwood and T. W. Barbee, Jr., "Layered synthetic microstructures as Bragg diffractors for x rays and extreme ultraviolet: theory and predicted performance," Appl. Opt. 20, 3027–3034 (1981)
- E. M. Gullikson, J. H. Underwood, P. C. Batson, and V. Nikitin, "A soft x-ray/EUV reflectometer based on a laser produced plasma source," J. X-Ray Sci. Technol. 3, 283–299 (1992).
- J. H. Underwood, E. M. Gullikson, and K. Nguyen, "Tarnishing of Mo/Si multilayer x-ray mirrors," Appl. Opt. 32, 6985
  6990 (1993).
- J. B. Kortright, E. M. Gullikson, and P. E. Denham, "Masked deposition techniques for achieving multilayer period variations required for short-wavelength (68-Å) soft x-ray imaging optics," Appl. Opt. 32, 6961–6968 (1993).