Normal-incidence reflectance of optimized W/B_4C x-ray multilayers in the range 1.4 nm < λ < 2.4 nm

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Received July 15, 2002

We have fabricated W/B_4C multilayers having periods in the range $d = 0.8-1.2$ nm and measured their soft-x-ray performance near normal incidence in the wavelength range $1.4 < \lambda < 2.4$ nm. By adjusting the fractional layer thickness of W we have produced structures having interface widths $\sigma \sim 0.29$ nm (i.e., as determined from normal-incidence reflectometry), thus having optimal soft-x-ray performance. We describe our results and discuss their implications, particularly with regard to the development of short-wavelength normal-incidence x-ray optics. © 2002 Optical Society of America

OCIS codes: 110.7440, 230.1480, 230.4170, 240.0310, 310.1620, 340.7470.

Nanometer-scale multilayer coatings optimized for normal-incidence reflectance at soft-x-ray wavelengths have enabled the development in recent years of breakthrough instrumentation in a variety of technological and scientific disciplines, including photolithography, plasma physics, synchrotron radiation, and space astronomy. Of particular interest here, normal-incidence x-ray multilayers operating over the wavelength range $\lambda \sim 17-30$ nm have been used to construct high-resolution telescopes and spectrographs for solar physics, which have been implemented in a number of major satellite missions. The motivation for the present study is to develop normal-incidence multilayer x-ray instrumentation that works at even shorter wavelengths, in particular in the range $\lambda \sim 1.5-3$ nm, in order to observe the important emission lines from O VII, O VIII, and Fe XVII present in many solar and nonsolar astrophysical plasmas.

X-ray multilayer structures are composed of alternating layers of optically dissimilar materials arranged so that the small x-ray reflections at each interface add coherently. Periodic x-ray multilayers designed to reflect near normal incidence operate over a narrow range of wavelengths: The condition for constructive interference is given by Bragg's law, $n\lambda = 2d \cos\theta$, where n is the Bragg order, d is the multilayer period, θ is the incidence angle, and λ is the x-ray wavelength. The peak reflectance increases (and the reflectance bandwidth decreases) with the number of bilayers, although the maximum useful number of bilayers is limited by the absorption of the materials; by adjusting the multilayer period, one can tune the wavelength response of the coating, as desired. Normal-incidence operation below $\lambda \sim 3$ nm, for example, thus requires multilayers containing hundreds of bilayers with periods below $d \sim 1.5$ nm.

The peak reflectance attainable from a periodic x-ray multilayer depends in practice on the effective reflection coefficient at each interface, determined by the optical constants of the materials that make up the multilayer and by the interface width σ that characterizes the degree of interface perfection; both interfacial roughness and interfacial diffuseness contribute to the interface width and thus reduce the effective reflection coefficient at each interface.[1] The loss in reflectance due to interface imperfections can be modeled simply as $\tilde{w}(s) = \exp(-s^2\sigma^2/2)$ (i.e., assuming an error-function interface profile), where $s = 4\pi \cos\theta/\lambda$; the effective reflection coefficient at each interface in the multilayer stack is computed as $r' = r \times \tilde{w}(s)$, where r is the Fresnel reflection coefficient[2] for the case of a perfectly smooth and sharp interface. Clearly the loss in reflectance near normal incidence owing to interface imperfections can become enormous at short wavelengths, where the argument of $\tilde{w}(s)$ becomes large. The challenge to develop x-ray multilayers that operate efficiently below $\lambda \sim 3$ nm is thus to find (stable) material pairs that have both large optical contrast and that can be used to grow structures containing the requisite number of bilayers while maintaining maximally smooth and sharp interfaces (i.e., having $\sigma < \sim 0.3$ nm) for periods below $d \sim 1.5$ nm.

The W/B_4C system has already been identified as a promising candidate for ultrashort-period x-ray multilayer structures[3–5]; in particular, Walton[6] produced W/B_4C structures containing $N = 100$ bilayers with periods as small as $d = 0.47$ nm. He measured the reflectance at $\theta = 45°$ and $\theta = 5°$ for sputtered films with $d \sim 1.0-3.2$ nm (and fractional W layer thicknesses $\Gamma = d_w/d = 0.28-0.4$); his results are consistent with interface widths of $\sigma \sim 0.35$ nm (i.e., in the framework of the modified Fresnel coefficient formalism described above, and assuming pure W and B_4C layers having bulk densities.) He observed by electron microscopy an expansion of the W-rich layers by 60–80% from their nominal values, consistent with intermixing of the two materials during growth, and discontinuous W-rich layers for periods below $d \sim 1.5$ nm.

0146-9592/02/242212-03$15.00/0

Following this work by Walton, here we have attempted to further optimize the reflectance of W/B_4C multilayers by producing films having $N = 300$ periods and even smaller interface widths attained by adjusting Γ. The films were deposited onto polished Si (100) wafer segments (1 cm × 1 cm), using a magnetron sputtering system that has been described previously.[7] The background pressure in the vacuum chamber prior to deposition was less than 5×10^{-7} Torr in all cases, and the sputter gas (argon) pressure was maintained at 1.50 ± 0.01 mTorr during growth. The effective deposition rate was found to be ~0.071 nm/s for both W and B_4C, deposited using 100 and 500 W of dc power, respectively. These rates were determined as the layer thicknesses (measured by using x-ray reflectometry, described below) divided by the exposure times of the substrate to the magnetron cathodes, for multilayers having periods in the range $d \sim 1-10$ nm, and were found to be linear to within ±1% over this entire range of periods. Individual W and B_4C layer thicknesses were adjusted in subsequent multilayers, using the measured rates by controlling the exposure times.

We first prepared a series of multilayers having $d \sim 1.2$ nm, $N = 120$, and Γ in the range 0.16–0.4. We measured the grazing-incidence x-ray reflectance (XRR) of these films, using a four-circle x-ray diffractometer having a sealed-tube Cu source and a Ge (111) monochromator tuned to the Cu $K\alpha$ line ($\lambda = 0.154$ nm). The angular resolution of this system is $\delta\theta \sim 0.015°$. Fits to the XRR data[8] were used to infer apparent interface widths σ. The results are presented in Fig. 1: The interface widths decrease monotonically from a maximum value of $\sigma = 0.38$ nm for $\Gamma = 0.4$ to $\sigma = 0.255$ nm for $\Gamma = 0.16$.[9] Also shown in Fig. 1 are the peak normal-incidence soft-x-ray reflectance values expected for these films: The theoretical reflectance increases with decreasing σ, in general, as discussed above, but also decreases with decreasing Γ, due to the effect of this parameter on the relative phases of the waves reflected from the W-on-B_4C versus the B_4C-on-W interfaces. Consequently, the theoretical reflectance curve shown in Fig. 1 exhibits a maximum, near $\Gamma = 0.21$.

Based on the results of Fig. 1, we prepared another series of multilayers, in this case with $\Gamma = 0.21$ and $N = 300$ and with periods in the range $d = 0.7-1.2$ nm, thus optimized for normal-incidence reflectance in the range $\lambda \sim 1.4-2.4$ nm. The XRR curves obtained at $\lambda = 0.154$ nm are shown in Fig. 2. The interface widths determined from the fits to these data are $\sigma = 0.29$ nm in all cases, except for the $d = 0.7$ nm film, for which we find $\sigma = 0.30$ nm. XRR data for these films were obtained again four months after deposition; the films had been stored in air during this time, and no significant changes were found, again with the exception of the $d = 0.7$ nm film, for which no Bragg peak was observed and which had visibly crazed.

The normal-incidence soft-x-ray reflectance of the films shown in Fig. 2 was measured using synchrotron radiation at the Calibration and Standards Beamline 6.3.2 synchrotron at the Advanced Light Source.[10] The measured reflectance curves ($\theta = 2.5°$) are shown in Fig. 3, along with the fits to these curves. The peak reflectance ranges from 1.3% at $\lambda = 2.4$ nm ($d = 1.2$ nm) to 0.08% at $\lambda = 1.6$ nm ($d = 0.8$ nm); peak reflectance of 0.08% was also measured at $\lambda = 1.4$ nm, with $\theta = 15°$ and $\theta = 30°$, in the case of the $d = 0.8$ nm sample, also shown in Fig. 3. (Note that the measured peak reflectance does not vary significantly with incidence angle over this range of angles because the synchrotron beam is ~95% *s* polarized.) Again in the context of the modified Fresnel coefficient formalism, assuming error-function interface profiles and pure W and B_4C layers having bulk densities, we infer interface widths $\sigma = 0.29$ nm in all cases. The measured bandwidths for these multilayers were found to range from $\Delta\lambda \sim 0.01$ nm ($d = 1.2$ nm) to $\Delta\lambda \sim 0.005$ nm ($d = 0.8$ nm) and agree well with the theoretical values, indicating good stability of the deposition rates during growth.

The W-rich layer expansion observed by electron microscopy by Walton has almost certainly occurred in the present films, and this expansion results in an

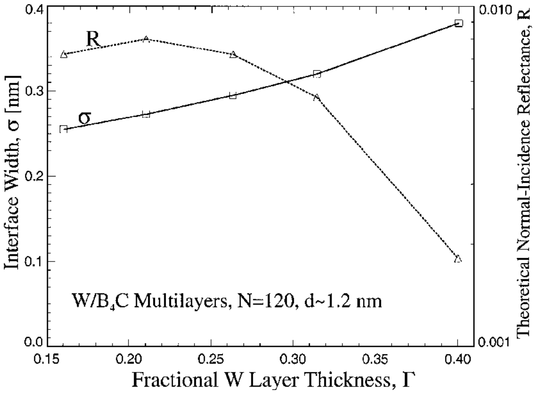

Fig. 1. Interface widths σ determined for grazing-incidence XRR measurements made on W/B_4C multilayers as a function of the fractional W layer thickness Γ. Also shown are the theoretical peak normal-incidence soft-x-ray reflectance values R for these same films, based on the measured σ values.

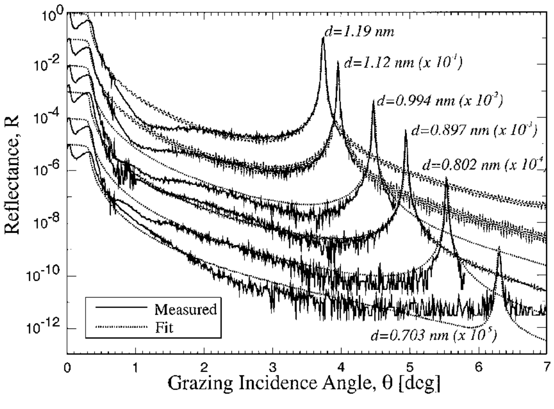

Fig. 2. Grazing-incidence XRR measurements ($\lambda = 0.154$ nm) for periodic W/B_4C multilayers having $N = 300$ bilayers, $\Gamma = 0.21$, with periods in the range $d = 1.2-0.7$ nm, as indicated.

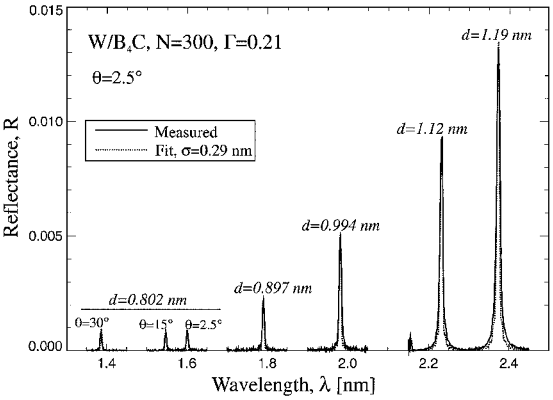

Fig. 3. Normal-incidence soft-x-ray reflectance measurements of the films shown in Fig. 2, made using synchrotron radiation. The measurements were all made at $\theta = 2.5°$ incidence, except for the two curves peaked near $\lambda = 1.55$ nm and $\lambda = 1.39$ nm, which were made at $\theta = 15°$ and $\theta = 30°$, respectively. Fits to these data indicate interface widths $\sigma = 0.29$ nm in all cases.

interfacial diffuseness contribution to σ. But, in addition, some interfacial roughness is likely present as well, and the variation of the roughness contribution to σ can explain the observed dependence of interface width on d and Γ. Specifically, this dependence can plausibly be related to the discontinuous W-layer growth observed by Walton for multilayers having periods below $d = 1.5$ nm, with $\Gamma = 0.32$. The nominal W layer thicknesses are below the expected threshold value for continuous W layer growth (i.e., 0.5 nm) in all the films studied here. The extent to which the surface roughness of the discontinuous W layer propagates to the next W-on-B$_4$C interface should depend on the thickness of the overlying B$_4$C layer, which can act to smooth this roughness. This mechanism can explain the apparent reduction in σ with decreasing Γ (Fig. 1) as well as the constancy of σ with decreasing d and constant Γ (Figs. 2 and 3.) Alternatively, the dependence of σ on d and Γ could be related to stress: In principle the interface roughness (and thus the interface area) can vary in order that the interfacial stresses in these structures equilibrate with the layer stresses, which are, in general, layer-thickness dependent. Future investigations using both specular and diffuse x-ray scattering could be used to test these hypotheses.

We can identify three possibilities for further improvements in these multilayers. First, increasing the number of periods will almost certainly result in increased reflectance. For example, increasing the number of bilayers from $N = 300$ to $N = 600$ (while maintaining $\sigma = 0.29$ nm) is expected to increase the peak reflectance by a factor of $\sim 1.5-2.1$ for structures having $d = 1.2-0.8$ nm. (The maximum useful number of bilayers in these films is essentially $N = 600$.) Second, our assumption of pure W and B$_4$C layers having bulk densities is inaccurate at some level of detail. More-accurate modeling of the microstructure (particularly the composition and density of the expanded W-rich layers) therefore may lead to further refinements in the optimal value of Γ, which may depend on period. Finally, the application of a negative substrate bias during sputter growth might result in smaller interface widths in these films, as was found in recently developed Cr/Sc multilayers designed for somewhat longer wavelengths than those considered here,[11] because enhanced bombardment of the growing film by Ar$^+$ ions can effectively increase the adatom's surface mobility, thus resulting in reduced interfacial roughness. We plan to explore all these possibilities in future investigations.

The results shown in Fig. 3 represent, to our knowledge, the smallest interface widths and the highest normal-incidence multilayer reflectance values reported for x-ray multilayers having periods smaller than $d \sim 2$ nm. Although the peak reflectance values are relatively small, these results indicate nevertheless that normal-incidence soft-x-ray instrumentation, particularly normal-incidence astronomical telescopes, as well as x-ray microscopes, spectrographs, collection optics, etc. for a variety of other scientific and technological applications are now realizable. Further improvements in the performance of these and perhaps other ultrashort-period x-ray multilayer structures (e.g., that comprise materials with even greater optical contrast, lower absorption, and (or) having smaller interface widths), would result in even more practical implementations of this technology.

The authors kindly thank Andy Aquila for assistance with the synchrotron measurements. This work was funded, in part, by grants from NASA. D. L. Windt's e-mail address is windt@astro.columbia.edu.

References

1. D. G. Stearns, J. Appl. Phys. **65,** 491 (1989).
2. J. D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975), pp. 281–282.
3. A. F. Jankowski, L. R. Schrawyer, and M. A. Wall, J. Appl. Phys. **68,** 5162 (1990).
4. J. F. Seely, G. Gutman, and J. Wood, Appl. Opt. **32,** 3541 (1993).
5. G. Gutman, J. X-Ray Sci. Technol. **4,** 142 (1994).
6. C. C. Walton, "Ultra-short-period W/B$_4$C multilayers for x-ray optics—microstructure limits on reflectivity," Ph.D. dissertation University of California, Berkeley, Berkeley, Calif., (1997).
7. D. L. Windt and W. K. Waskiewicz, J. Vac. Sci. Technol. B **12,** 3826 (1994).
8. D. L. Windt, Computers Phys. **12,** 360 (1998).
9. The validity of relating the interface width parameter σ (used in the modified Fresnel formalism to describe the x-ray reflectance data) to a precise physical description of the interface remains to be determined comprehensively, particularly when σ is comparable to the layer thickness. Nevertheless, even in multilayers that locally have large interface widths or discontinuous layers, we find σ to be a useful parameter with which to quantify the interface width and to compare multilayers and multilayer systems.
10. J. H. Underwood, E. M. Gullikson, M. Koike, P. J. Batson, P. E. Denham, K. D. Franck, R. E. Tackaberry, and W. F. Steele, Rev. Sci. Instrum. **67,** 3372 (1996).
11. F. Eriksson, G. A. Johansson, H. M. Hertz, and J. Birch, Proc. SPIE **4506,** 14 (2001).