Tri-material multilayer coatings with high reflectivity and wide bandwidth for 25 to 50 nm extreme ultraviolet light

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Abstract: Magnesium/silicon carbide (Mg/SiC) multilayers have been fabricated with normal incidence reflectivity in the vicinity of 40% to 50% for wavelengths in the 25 to 50 nm wavelength range. However many applications, for example solar telescopes [1] and ultrafast studies using high harmonic generation sources [2], desire larger bandwidths than provided by high reflectivity Mg/SiC multilayers. We investigate introducing a third material, Scandium, to create a tri-material Mg/Sc/SiC multilayer allowing an increase the bandwidth while maintaining high reflectivity.

OSIC Codes: (260.7200) Ultraviolet, extreme; (310.4165) Multilayer design.

References and Links

1. Regina Soufli, David L. Windt, Jeff C. Robinson, Sherry L. Baker, Eberhard Spiller, Franklin J. Dollar, Andrew L. Aquila, Eric M. Gullikson, Benjawan Kjornrattanawanich, John F. Seely, Leon Golub, "Development and testing of EUV multilayer coatings for the Atmospheric Imaging Assembly instrument aboard the Solar Dynamics Observatory," Proc. of SPIE Vol. 5901 59010M-1 (2005)
2. N. Dudovich, O. Smirnova, J. Levesque, Y. Mairesse, M. Yu. Ivanov, D. M. Villeneuve, and P. B. Corkum. Measuring and controlling the birth of attosecond XUV pulses. Nature Physics, 2:781, 2006.
3. Hisataka Takenaka, Satoshi Ichimaru, Tadayuki Ohchi, E.M. Gullikson, "Soft-X-ray reflectivity and heat resistance of SiC/Mg multilayer," Journal of Electron Spectroscopy and Related Phenomena 144–147, 1047–1049 (2005)
4. Igor V. Kozhevnikov, Inna N. Bukreeva, and Eric Ziegler, "Design of X-ray supermirrors," Nuc. Inst. & Methods in Physics Research A 460, 424-443 (2001)
5. A. L. Aquila, F. Salmassi, F. Dollar, Y. Liu, and E. Gullikson, "Developments in realistic design for aperiodic Mo/Si multilayer mirrors," Optics Express 14 10073-10078 (2006)
6. Andrew Aquila, Farhad Salmassi, and Eric Gullikson, "Metrologies for the phase characterization of attosecond extreme ultraviolet optics," Opt. Lett. 33, 455-457 (2008)
7. Juan I. Larruquert, "Reflectance enhancement with subquarterwave multilayers of highly absorbing materials," J. Opt. Soc. Am. A 18, 1406–1414 (2001).
8. Juan I. Larruquert, "General theory of sub-quarterwave multilayers with highly absorbing materials," J. Opt. Soc. Am. A 18, 2617-2627 (2001)
9. Julien Gautier, Franck Delmotte, Marc Roulliay, Françoise Bridou, Marie-Françoise Ravet, and Arnaud Jérome, "Study of normal incidence of three-component multilayer mirrors in the range 20-40 nm," Applied Optics 44, 384- 390 (2005)
10. P. Boher, L, Hennet, P. Houdy, "Three materials soft X-ray mirrors: theory and application," SPIE 1345, 198-212 (1990)
11. Yu. A. Uspenskii, V. E. Levashov, A. V. Vinogradov, A. I. Fedorenko, V. V. Kondratenko, Yu. P. Pershin, E. N. Zubarev, and V. Yu. Fedotov, "High-reflectivity multilayer mirrors for a vacuum-ultraviolet interval of 35–50 nm," Optics Letters 23, 771-773 (1998)
12. E. M. Gullikson, S. Mrowka, and B. B. Kaufmann, "Recent developments in EUV reflectometry at the Advanced Light Source," in Emerging Lithographic Technologies V, E. A. Dobisz ed., Proc. SPIE 4343, 363 (2001).
13. B.L. Henke, E.M. Gullikson, J.C. Davis, "X-ray interactions: photoabsorption, scattering, transmission, and reflection at E=50-30000 eV, Z=1-92", Atomic Data and Nuclear Data Tables 54, 181-342 (1993).
14. E. M. Gullikson, "X-ray interactions with matter" http://www-cxro.lbl.gov/optical_constants.
15. Yu. A. Uspenskii, J. F. Seely, N. L. Popov, A. V. Vinogradov, Yu. P. Pershin,and V. V. Kondratenko. Efficient method for the determination of extreme ultraviolet optical constants in reactive materials: application to scandium and titanium. Journal of the Optical Society of America A, 21(2):298, 2004.

Introduction

Highly reflective multilayer optics in the extreme ultraviolet (EUV) have enabled many areas of scientific study. Specifically, in the wavelength range of 25 to 50 nm multilayers are used as focusing and imaging elements for research in high harmonic femtosecond chemistry and solar astrophysics imaging such as the He II 30.4nm line. Currently the highest published results on reflectivity in this region are obtained from Magnesium /Silicon Carbide (Mg/SiC) multilayers and are around 40% to 50% [3].

The relatively high reflectivity of these mirrors comes with a drawback of reduced bandwidth. In femtosecond/attosecond applications a large bandwidth is very important to maintain short pulses. In many applications integrated reflectivity is more desirable as a merit function for mirror performance than peak reflectivity. For example the odd harmonics of a Ti-sapphire high harmonic generator are separated by 2 nm in the vicinity of the 27th harmonic (~30 nm). The typical bandwidth of an optically thick (highest obtainable reflectivity) Mg/SiC multilayer is 2.9 nm limiting their use to a single harmonic.

Several methods have been implemented to increase the bandwidth of multilayer coatings. The simplest method to increase bandwidth is to use fewer periods in the multilayer stack. Multilayers are artificial Bragg crystals; they use temporal coherence to obtain high reflectivity. In other words each reflective interface adds coherently with the one next to it. For an optically thin stack the temporal/longitude coherence length is proportional to the thickness of the multilayer:

$$L_{coh} \equiv \frac{\lambda^2}{2\Delta\lambda} \approx d \cdot N \Rightarrow \Delta\lambda \propto N^{-1} \qquad (1)$$

Where L_{coh} is the coherence length, λ is the center wavelength, $\Delta\lambda$ is the bandwidth, d is the optical period (d-spacing) of the multilayer, and N is the number of periods of thickness d. However reducing the number of layers also decreases the peak reflectivity, as the peak reflectivity scales as N^2.

The second method implemented is to make the multilayer aperiodic [4,5]. This method also trades off peak reflectivity for larger bandwidth, however the integrated reflectivity is often higher than it is by just reducing the number of layers. However the aperiodic structure can introduce non-uniform phase response and affect the temporal structure of the reflected radiation. Although this provides a mechanism for pulse compression [6], in general the use of aperiodic optics with high harmonic sources in femtosecond dynamic studies requires full characterization of the source, as well as the optics, to insure that the femtosecond pulses are not inadvertently broadened.

An alternative solution that has been used at longer wavelength (>50 nm) [7,8] is to use more than two materials in each repeating period of the stack. This allows for higher reflectivity per period in the multilayer stack. Thus higher reflectivity is obtained in fewer periods, therefore increasing the bandwidth. Three material multilayers have also been proposed and measured for use in both the extreme ultraviolet and the soft x-ray regime of the spectrum [9,10]. At 25-50 nm, previous research using tri-material multilayers did not demonstrate improvement over standard bilayer Mg/SiC due to the materials chosen in the study [9]. Here, we report the design and fabrication of a tri-material multilayer that shows good improvements in both the reflectivity and bandwidth.

Material selection, optimization, and fabrication

The three materials chosen for improving multilayer bandwidth and maintaining high reflectivity in the 25 to 50 nm range were Mg, SiC, and Sc. Mg was chosen as the low Z element, because the L3 absorption edge is located at 25.0 nm. This makes Mg optically transparent, and it is typically used as a spacer for multilayers in this wavelength range. SiC is usually chosen as the high Z compound (absorber) in Mg based multilayer due to its low

interdiffusion and roughness. In this work the authors chose Scandium to be a second high Z material. Sc was chosen because of its M2,3 absorption edge at 43.8 nm makes the real part of its index of refraction greater than one and it is also used in multilayers between 35 and 50 nm [11]. It is expected that the reflection from the Sc/SiC interface is strong, since the Fresnel reflectivity scales with the difference in the index of refraction. The optical constants [13, 14, 15] of the 3 materials are shown in Fig. 1.
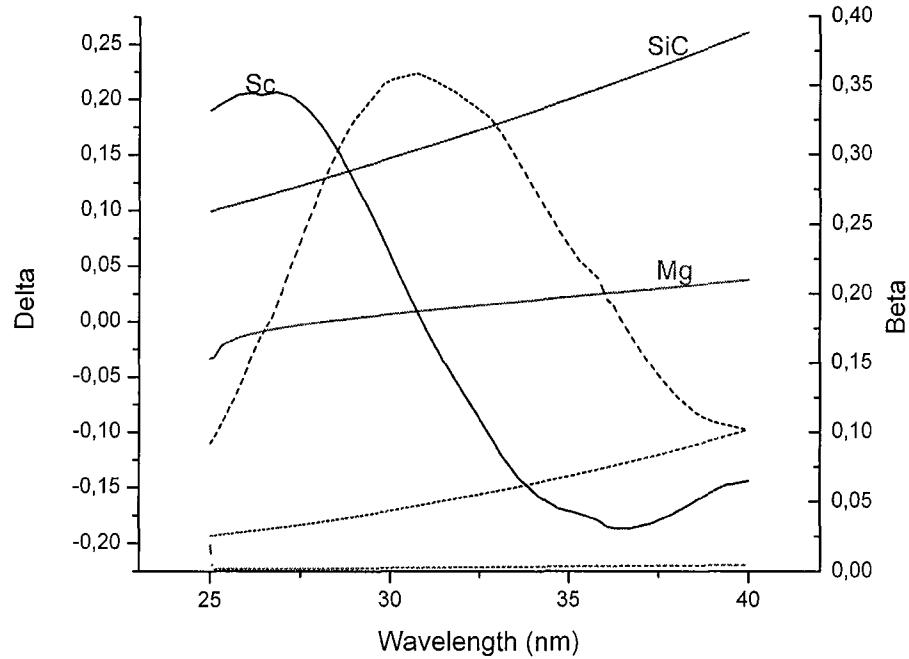

Fig. 1. This graph shows the optical constants for Mg, Sc, and SiC. The solid line represents delta while the doted lines represent beta. The index of refraction uses the standard convention: $n = 1-\delta+i\beta$.

Four samples were made for this experiment: 2 Mg/SiC multilayers and 2 Mg/Sc/SiC. One set (1 Mg/SiC and 1 Mg/Sc/SiC) was designed for a peak reflectivity at 37 nm near the optimal location for the optical constants. The optimal is located by maximizing the difference in the real parts of the index of refraction is large, and minimizing the complex or absorptive part for both materials. A larger difference in the real parts increases reflectivity at each interface, and the lower absorption allows for more layer also increasing reflectivity. The other set was designed for a reflectivity peak at 28 nm to be away from the optimal difference in optical constants. For the 28 nm samples we used non-optically thick stacks for both samples. A brute force reflectivity simulation was performed to optimize the material thickness of each sample by simulating the peak reflectivity, at a chosen wavelength, as a function of the thickness of the three materials. The brute force multilayers simulation was preformed on optically thick stacks and for thickness of the 3 materials ranging from 0 nm to half of the chosen wavelength in 0.1 nm steps for all combinations of thicknesses. The thickness parameters that created the global maximum reflectivity were chosen. As predicted by the theory [8] the order of the materials is critical when three or more materials are used.

For the 37 nm samples, the optimal parameters for the Mg/Sc/SiC were: d = 19 nm, $\gamma 1$ = 0.2 (SiC thickness / d), $\gamma 2$ = 0.13 (Sc thickness / d), N = 30 tri-layers. The Mg/SiC sample d = 19.5 nm, $\gamma 1$ = 0.33, N = 35 bi-layers. For the 28 nm samples, the optimal parameters for the

Mg/Sc/SiC were: d = 15.4 nm, $\gamma 1$ = .2, $\gamma 2$ = .07, N = 30 tri-layers. The Mg/SiC sample d = 14.0 nm, $\gamma 1$ = .3, N = 40 bi-layers.

The samples were fabricated on polished silicon wafers using magnetron sputtering. DC sputtering was used for Mg and Sc targets at 100 W. While RF sputtering was used SiC, at 275 W. The samples were fabricated under a 1.0 mTorr argon pressure during the deposition. The top layer of each sample is SiC to prevent oxidation.

Reflectivity Measurements

The samples were measured at the Advanced Light Source beamline 6.3.2. The beamline is designed for EUV optical metrology and reflectivity measurements [12]. The beamline has high spectral purity, and a spectral resolving power ($\Delta\lambda/\lambda$) of up to 7000, a wavelength accuracy of 2×10^{-3} nm, and a reflectivity accuracy of 0.1% (absolute). The samples were measured at 5 degrees from normal incidence. The reflectivity for the 37 nm samples is shown in Fig. 2, while the reflectivity for the 28 nm samples is shown in Fig. 3.
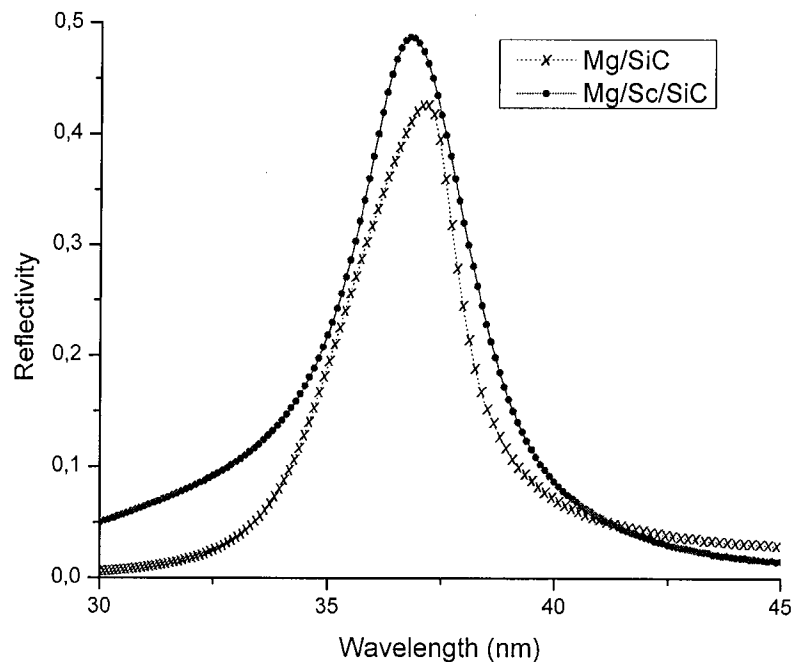

Fig. 2. The reflectivity of optimized, optically thick Mg/SiC and Mg/Sc/SiC multilayers is shown for the samples optimize for 37 nm. The tri-layer mulitlayer has a FWHM bandwidth of 3.2 nm and a peak reflectivity of 48.7% at 36.8 nm. While the bi-layer Mg/SiC has a FWHM bandwidth of 3 nm and a peak reflectivity of 42.5% at 37.1 nm.
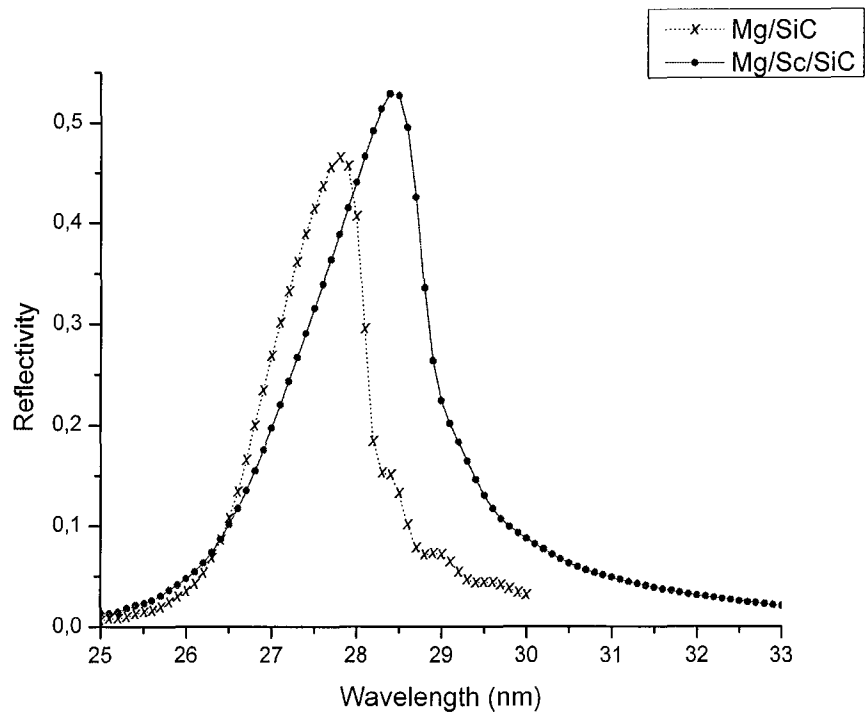
Fig. 3. The reflectivity of optimized, 30 period Mg/SiC and Mg/Sc/SiC multilayers is shown for the samples optimized for 28 nm. The tri-laer mulitlayer has a FWHM bandwidth of 1.6 nm and a peak reflectivity of 52.8% at 28.4 nm. While the bi-layer Mg/SiC has a FWHM bandwidth of 1.2 nm and a peak reflectivity of 46.5% at 27.8 nm.

Results & Conclusion

As shown in figures 2 & 3 the reflectivity of the tri-material multilayer is comparable to the Mg/SiC multilayers. In figure 2 the reflectivity of the optimized tri-material stack is even higher then the optimized bi-material stack. Because the tri-material stack uses fewer periods the intergraded reflectivity & bandwidth are larger.

We have optimized tri-material multilayers for use in the range of 25 to 40 nm EUV light. The tri-materials allows for larger bandwidths without sacrificing peak reflectivity.