Normal-incidence silicon–gadolinium multilayers for imaging at 63 nm wavelength

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Si/Gd multilayers designed as narrowband reflective coatings near 63 nm were developed. The highest peak reflectance of 26.2% at a 5° incident angle was obtained at 62 nm, and the spectral bandwidth was 7.3 nm FWHM. The fits for x-ray and extreme ultraviolet reflectance data of Si/Gd multilayers indicate the possibility of silicide formation at the Si–Gd interfaces. B_4C, W, and SiN were deposited as interface barrier layers to improve the reflectance of Si/Gd multilayers. More than an 8% increase in reflectance was observed from the interface-engineered Si/W/Gd and Si/B_4C/Gd multilayers. © 2008 Optical Society of America

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Narrow bandpass solar imaging in the extreme ultraviolet (EUV) using telescopes with multilayer coatings, e.g., the SOHO/EIT [1] and TRACE [2], has led to increased understanding of solar phenomena that affect space weather and global climate change. Existing multilayer coatings, e.g., the widely used Mo/Si coating and others, have made possible the observation of such low-temperature spectral lines as He II (8×10^4 K) at 30.4 nm wavelength and such high-temperature Fe lines as Fe XV (2×10^6 K) at 28.4 nm and Fe XVI (3×10^6 K) at 33.5 nm. However, it is difficult to observe intermediate-temperature spectral lines in the well-studied 17–34 nm solar spectral range, because these lines are not adequately isolated and any recordable spectral images are most likely contaminated by higher- or lower-temperature lines. Thus it is desirable to develop narrowband multilayers for other EUV wavelengths where the intermediate-temperature lines are well isolated. For example, narrowband imaging of the O V line at 62.97 nm (2.4×10^5 K) would fill the gap between the low-temperature He II line and the high-temperature Fe lines. The need for multilayers operating near 63 nm motivated this Letter.

A multilayer coating comprises a stack of two alternating materials, termed spacer and scatterer, with high optical contrast, low absorption, and a combined thickness (period d) about half the wavelength of interest when operating at normal incidence. Coating the telescope mirrors with suitable multilayers increases the EUV reflectance and enables high-cadence normal-incidence solar imaging observations. Multilayers such as Mo/Si, Mo_2C/Si, Mo/Y, and SiC/Mg have been selected as coatings for solar EUV imaging telescopes [1–3]. These multilayers possess high normal-incidence reflectance, good thermal and long-term stability, and narrow spectral bandwidth and together provide excellent wavelength coverage from approximately 8–35 nm.

To develop multilayers for imaging the longer EUV wavelengths >35 nm, in particular the O V 62.97 nm solar emission, suitably transmissive materials other than Si, which is a great spacer for the shorter wavelength range, must be identified, and this requires a good knowledge of the optical constants of the candidate materials. The optical constants of most materials at >35 nm are not accurately known due to high absorption as well as oxidation/contamination of pure materials that can significantly affect the experimental measurements. We initiated an experimental program to determine the optical constants of materials for long-wavelength EUV multilayers and identified the rare earth elements with 5d or 4f open atomic shells as candidate spacer materials [4–6]. Tb-based multilayers, e.g., Si/Tb and SiC/Tb, had comparable peak reflectances of ~23% around 63 nm, but both material pairs had substantial interface diffusion as indicated by transmission electron microscopy (TEM) images, and the maximum achievable reflectances were much lower than the theoretical predictions.

Here we report the development of Si/Gd multilayers for high reflectance and narrow bandpass imaging at 63 nm wavelength. The multilayers were fabricated using a dc magnetron sputtering system at Reflective X-ray Optics LLC. The coating system utilizes S-Gun cathodes operating in a constant-power mode. The base pressure of the coating chamber was 2×10^{-7} Torr or lower prior to deposition. Ar was used as a process gas with the pressure maintained at 2.0 mTorr during deposition. Each multilayer film was grown on a 16 mm × 16 mm Si (100) substrate. The number of bilayers N deposited for each multilayer was 10, since adding more bilayers did not improve the reflectance. Each deposition began with Gd and finished with Si, to allow formation of a silicon dioxide top layer that protected the layers underneath from oxidation and contamination.

The multilayer structure was predetermined by grazing incidence x-ray reflectance (XRR), using an x-ray diffractometer with a rotating Cu anode source. The thickness of each individual layer and the roughness/diffusion (σ) at the surface and interfaces were determined by fitting [7,8] to the measured XRR data using (1) published x-ray optical constants [9] assuming bulk densities in all layers and (2) the standard two-layer model in which each multilayer period was composed of pure Gd and Si layers and the last Si layer was partially oxidized and formed SiO_2.

The EUV reflectance was measured at a 5° normal-incidence angle using the reflectometer at the NSLS Beamline X24C at Brookhaven National Laboratory. A monochromatic beam with energy extending from 11 to 465 eV and spectral resolution up to 600 was generated by the dual-element monochromator utilizing an Au mirror and a 600 line/mm grating. The multilayer fitting parameters such as d, Γ (commonly defined as scatterer thickness divided by d), and σ determined from EUV and XRR measurements were compared. Such a comparison is extremely useful to truly identify the multilayer structure and evaluate the accuracy of the optical constants used in the multilayer reflectance simulations.

For typical Si/Gd multilayers, preliminary experiments showed that samples with higher Γ had broader spectral bandwidth. The Γ values determined from XRR fits were in the range of 0.34–0.64. Our record high reflectance of 26.2% at 62 nm was achieved from a Si/Gd multilayer with the fitted Γ value of 0.34, which also resulted in the smallest spectral bandpass (FWHM) of 7.34 nm. Figure 1(a) shows the measured XRR of this particular sample and the best XRR fit. The detailed structure of this multilayer is $d \sim 33.18$ nm, $\Gamma \sim 0.34$, $N=10$, and $d_{SiO_2} \sim 2.5$ nm. The value of σ was assumed to be equal at the surface and all interfaces and was found to be ~ 0.34 nm. The d value obtained from the XRR fit was in excellent agreement with the value calculated based on the modified Bragg's equation knowing the diffraction orders and the diffracted peak positions.

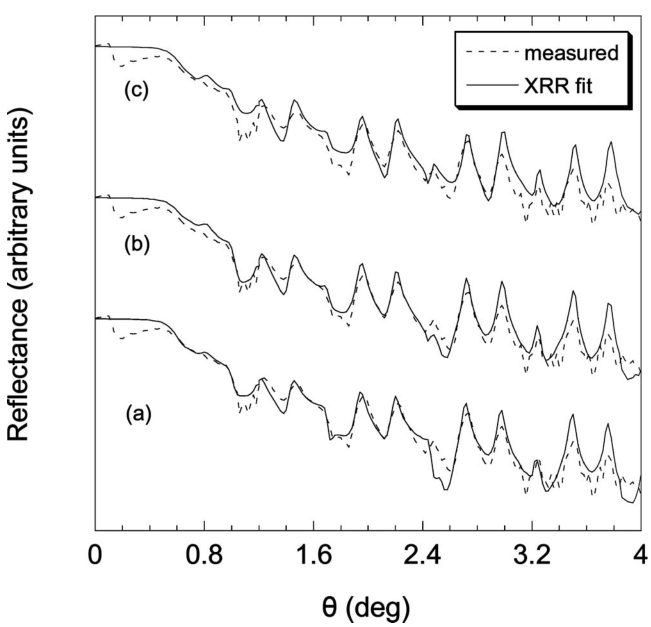

Fig. 1. Measured XRR reflectance of a Si/Gd multilayer and the XRR fits using (a) two-layer model, (b) and (c) four-layer model with Gd_5Si_3 and $GdSi_2$ formation at the interfaces, respectively.

The EUV reflectance of the same multilayer was fitted using newly determined Gd [6] optical constants and available Si and SiO_2 optical constants from various sources [7]. As shown in Fig. 2, the measured EUV reflectance was compared with the values calculated using the XRR and EUV fit parameters. The multilayer structure determined from the best EUV fit is $d \sim 32.41$ nm, $\Gamma \sim 0.64$, $N=10$, and $d_{SiO_2} \sim 2.5$ nm. The σ value was predicted to be as high as 2.0 nm at all interfaces. Since there are considerable discrepancies between EUV and XRR fit parameters, this suggests that the two-layer model may not be the best representation of the Si/Gd multilayer structure. To demonstrate this, we fitted the XRR reflectance using the four-layer model, in which each multilayer period was composed of pure Gd, Gd silicide, pure Si, and Gd silicide. Again, the last layer of Si was assumed to be partially oxidized. Among four well-known Gd silicides, which are GdSi, Gd_3Si_5, Gd_5Si_3, and $GdSi_2$, only Gd_5Si_3 and $GdSi_2$ have enthalpy of formation reported in literature [10], and only these two could provide reasonably good fits to the XRR data as shown in Figs. 1(b) and 1(c), respectively. Assuming Gd_5Si_3 formation, the fit parameters were $d_{Gd} \sim 13.09$ nm, $d_{\text{Si-on-Gd}} \sim 6.98$ nm, $d_{Si} \sim 11.0$ nm, $d_{\text{Gd-on-Si}} \sim 2.02$ nm, $d_{SiO_2} \sim 2.5$ nm, and $\sigma \sim 0.37$ nm. For the case of $GdSi_2$ formation, the fit parameters were $d_{Gd} \sim 17.02$ nm, $d_{\text{Si-on-Gd}} \sim 3.16$ nm, $d_{Si} \sim 11.76$ nm, $d_{\text{Gd-on-Si}} \sim 2.18$ nm, $d_{SiO_2} \sim 2.5$ nm, and $\sigma \sim 0.37$ nm. The fits for the silicide thicknesses indicate asymmetric silicide formation where Si-on-Gd was thicker than Gd-on-Si in both cases. Asymmetric interfaces have also been observed in the TEM images of Mo/Si and Si/Tb multilayers. Because good XRR fits can be obtained with and without silicide formation using completely different layer structures, this indicates the need for similar modeling of EUV data. Good agreement between the XRR and EUV fit parameters for the same layer model would

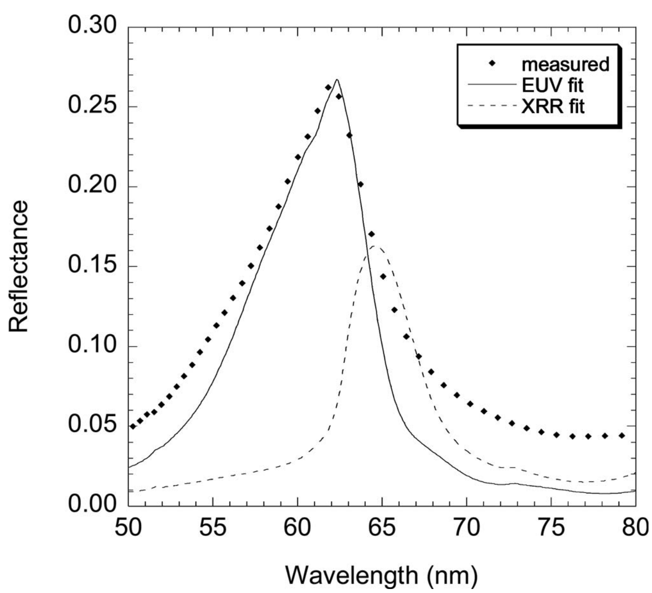

Fig. 2. Measured EUV reflectance of a Si/Gd multilayer compared with the reflectances calculated using the best EUV and XRR fit parameters.

be the best indication of the real multilayer structure.

Under the independent atomic approximation, the optical constants of Gd silicide at Cu K_{α} x-ray energy may be calculated, given that the optical constants of Si and Gd and the composition and density of Gd silicide are known. But in the vicinity of the absorption thresholds of Gd at 63 nm, such an approximation is invalid, and optical constants of Gd silicide can be determined only from experiments. To obtain a more meaningful model of the EUV data, this requires significant efforts in determining the chemical composition/density of Gd silicide that actually formed at Gd–Si interfaces and its optical constants at EUV wavelength. Until this is accomplished, the multilayer parameters based on the XRR fits still provide useful guidance for reflectance optimization, even though that may not be the best representation of the actual layer structure.

To improve the reflectance of Si/Gd multilayers, B_4C barrier layers were deposited at the Si–Gd interfaces with the thickness varying from 0.1 to 0.5 nm. It can be visualized that the peak position of a multilayer may be altered by changing either its period or Γ value. To understand the change of EUV reflectances as a function of barrier layer thicknesses, the value Γ redefined here as $d_{\mathrm{Si}}/(d_{\mathrm{Gd}}+d_{\mathrm{Si}})$ and the final period $d=(d_{\mathrm{B_4C}}+d_{\mathrm{Gd}}+d_{\mathrm{B_4C}}+d_{\mathrm{Si}})$ must be kept constant to maintain the same peak wavelengths in all samples. No drastic change in EUV reflectances was observed among the Si/B_4C/Gd multilayers with 0.1–0.5 nm B_4C thicknesses.

Thicker B_4C, W, and SiN barrier layers up to 2.0 nm were tested. The SiN alloy was prepared by leaking N_2 gas at a controllable rate during the sputter deposition from the Si target. The reflectance as a function of B_4C-, W-, and SiN-barrier layer thicknesses are summarized in Fig. 3. The peak reflectance of the Si/Gd multilayer with no barrier layers was 21.9% at 62.2 nm, much lower than the record high reflectance sample (26.2%) as a result of unoptimized Γ. Reflectance improvement was observed only when using B_4C or W as barrier layers. The percent increases relative to the original reflectance values were ~11% for 2.0 nm thick W and ~8% for 2.0 nm thick B_4C.The gradual decrease in reflectance with increasing SiN thickness is clearly a result of absorption in the SiN layer. Further tests to reduce absorption from the SiN layers by adjusting the chemical composition of the SiN alloy are underway. Reflectance measurement over a period of six months confirmed that Si/Gd multilayers were stable, with constant peak wavelengths and less than 1% absolute decreases in the peak reflectances. More experiments are planned to validate long-term stability of standard and interface-engineered Si/Gd multilayers, including those that have undergone high-temperature annealing, to determine their suitability for solar imaging applications.

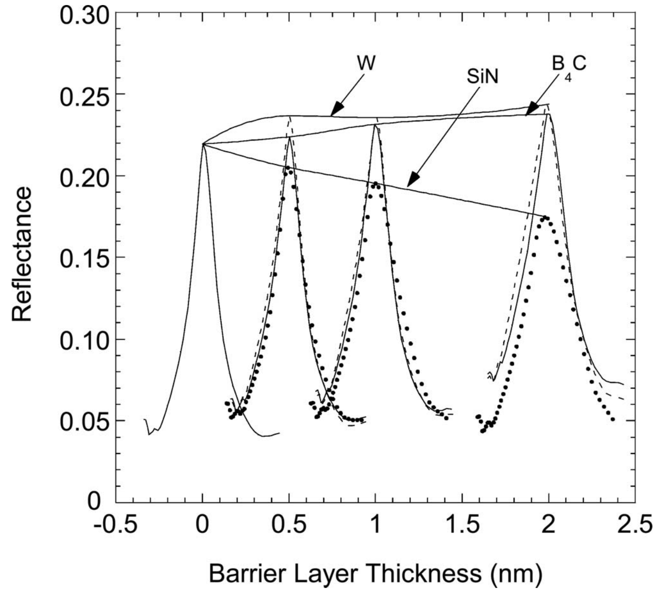

Fig. 3. Measured EUV reflectance of interface-engineered Si/Gd multilayers as a function of B_4C, W, and SiN barrier layer thicknesses.

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