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X-ray multilayer coatings for use at energies above 100 keV

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ABSTRACT

We discuss the development of X-ray multilayer coatings for use as broad-band reflectors operating at energies above 100 keV. Such coatings can be used to produce hard X-ray telescopes that will make possible a variety of entirely new astronomical observations. We summarize our recent investigation into the growth, structure and hard X-ray performance of depth-graded W/Si multilayers, present follow-up information on Cu/Si multilayers, and discuss preliminary results obtained with Ni/Si, $Ni_{.8}Cr_{.2}$/Si, and $Ni_{.93}V_{.07}$/Si multilayers.

Keywords: multilayer coatings, hard X-ray optics

1. INTRODUCTION

Depth-graded multilayers are the enabling technology for a new generation of hard X-ray astronomical telescopes, including those being developed for the HEFT [1] and In-Focus [2] balloon instruments, and the Astro-G and Constellation-X satellite instruments [3]. These new telescopes, operating at photon energies in the range 20 – 100 keV, will provide an improvement in sensitivity by a factor of ~1000 or more over previous (non-focusing) instruments, and will undoubtedly lead to a number of important astronomical discoveries.

We describe here the development of depth-graded multilayer structures for use as hard X-ray reflectors operating at energies greater than 100 keV. With a high-resolution, high-energy focusing mission based on such technology a variety of new observations will become possible, including: measuring the time-history of the emission from ^{56}Ni (158 keV) in Type 1a SNe; investigating the sites of particle acceleration in young SNR; measuring the nuclear continuum and cutoff in Seyfert I galaxies; and detection of Compton backscatter radiation (170 keV) in Galactic black hole candidates.

Unlike periodic multilayers, which provide high reflectance only over a limited range of angles and X-ray energies, depth-graded multilayers are used for broadband reflectance: in a depth-graded multilayer structure the layer thicknesses vary with depth into the film stack, such that each bilayer is effectively tuned to a different X-ray wavelength. Depth-graded multilayer structures are particularly effective in the hard X-ray region ($E > 20$ keV) where hundreds or even thousands of bilayers can be used, in principle, as a result of the low absorption and consequently high penetration depth in the film at these energies. However, the performance of such structures depends critically on the achievable level of interface perfection: deviations from atomically smooth and chemically abrupt interfaces – i.e., interfacial roughness and interfacial diffuseness, respectively – will reduce the reflection coefficients at the interfaces, thereby reducing the overall reflectance of the multilayer stack. Interface imperfections can result from a variety of material- and/or growth-dependent mechanisms, including the formation of mixed-composition amorphous interlayers by diffusion or by mixing due to energetic bombardment during growth, and roughness resulting from surface stress or from low adatom surface mobility.

Promising material combinations found to have relatively small interface imperfections and suitable for use in depth-graded X-ray multilayers include Pt/C [4], Cu/Si [5], and W/Si [6]; we summarize below our recent work on depth-graded W/Si multilayers, present follow-up information on Cu/Si multilayers, and also present preliminary results obtained with Ni/Si, $Ni_{.8}Cr_{.2}$/Si, and $Ni_{.93}V_{.07}$/Si multilayers.

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2. W/Si MULTILAYERS

Depth-graded W/Si multilayers have already been shown to have good performance as broad-band X-ray reflectors up E~40 keV [6]. We have recently investigated in detail using X-ray scattering and high-resolution transmission electron microscopy (TEM) the growth and structure of periodic and depth-graded W/Si multilayers prepared by planar magnetron sputtering, and have measured the performance of optimized depth-graded structures up to E=212 keV. These new results are described in detail elsewhere [7]; we summarize here our most important findings with regard to the development of hard X-ray astronomical telescopes using these coatings.

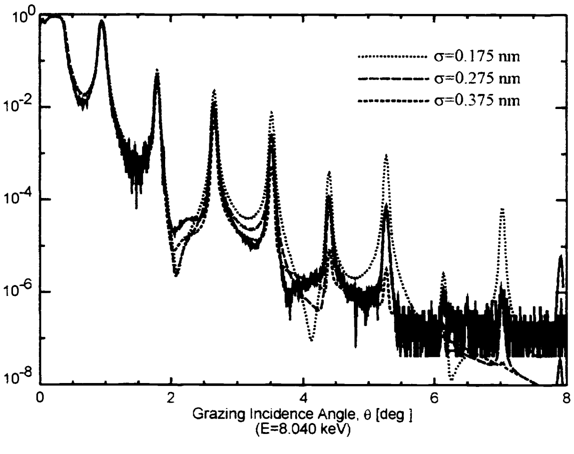

Figure 1. Specular X-ray (E=8.04 keV) reflectance of a periodic W/Si multilayer containing 20 bilayers. Calculations made using various interface widths as discussed in the text are indicated.

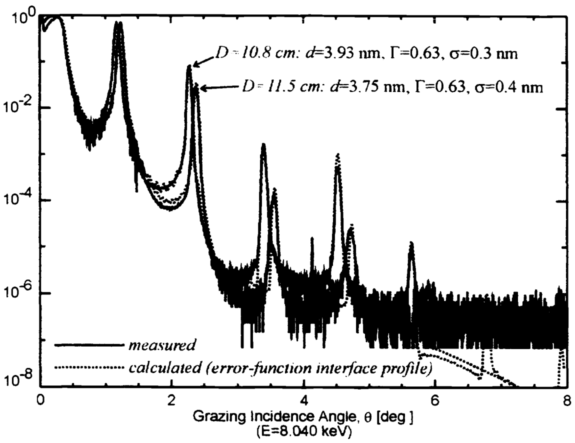

Figure 2. Specular X-ray (E=8.04 keV) reflectance for periodic W/Si multilayers containing 100 bilayers, deposited at $P_{Ar}=2$ mTorr, and with target-to-substrate distances D=10.8 cm and D=11.5 cm.

From X-ray analysis of periodic W/Si multilayers we find interface widths in the range σ=0.275 – 0.35 nm for films deposited at low argon pressure (with a slight increase in interface width for multilayers having periods greater than ~20 nm, possibly due to the transition from amorphous to polycrystalline metal layers identified by TEM and SAED), and somewhat larger interface widths (i.e., σ=0.35 – 0.4 nm) for structures grown at higher Ar pressures, higher background pressures, or with larger target-to-substrate distances. We illustrate some of these findings in Figures 1 and 2. Shown in Fig. 1 is the grazing incidence specular X-ray (8.04 keV) reflectance of a periodic W/Si multilayer containing 20 bilayers, deposited at an argon pressure $P_{Ar}=1.5$ mTorr; also shown for comparison in Fig. 1 are calculated reflectance curves computed with identical W-on-Si and Si-on-W interface widths σ=0.175 nm, σ=0.275 nm, and σ=0.375 nm, assuming an error-function interface profile. The experimental Bragg peak intensities are best matched for the calculation using σ=0.275 nm, particularly at the largest graze angles where the sensitivity is greatest for reflectance changes resulting from small changes in σ. By matching the positions and relative intensities of the Bragg peaks, the apparent layer thicknesses were determined to be $d_W=2.15$ nm and $d_{Si}=2.90$ nm. Shown in Fig. 2 are the specular reflectance measurements for otherwise identical periodic W/Si films containing 100 bilayers, deposited at an argon pressure $P_{Ar}=2$ mTorr, but with target-to-substrate distances D=10.8 cm and D=11.5 cm. From these data we estimate comparable periods for the two films ($d=3.925$ nm vs. $d=3.750$ nm), but the interface properties are evidently different: the Bragg peak intensities for graze angles greater than θ ~4° are markedly smaller for the film deposited with D=11.5 cm, and we thus infer σ=0.40 nm for this film vs. σ=0.30 nm for the film having D=10.8 cm.

Further analysis of periodic W/Si multilayers using non-specular X-ray reflectance analysis and TEM suggest that the dominant interface imperfection in these films is interfacial diffuseness; interfacial roughness is minimal (σ_r ~ 0.175 nm) in structures prepared under optimal conditions, but can increase under conditions in which the beneficial effects of energetic bombardment during growth are compromised (e.g., high Ar pressure, high background pressure, large target-to-substrate distance, etc.)

X-ray reflectance analysis was also used to measure the variation in the W and Si deposition rates with bilayer thickness: we find that the W and Si layer thicknesses are non-linear with the deposition times, as shown in Figure 3. This non-linearity can be explained by a variety of mechanisms, including diffusion, W-Si interlayer formation, and re-sputtering during growth. In any case the thicknesses can be described as a bi-linear function of time, and such fits to the data (straight

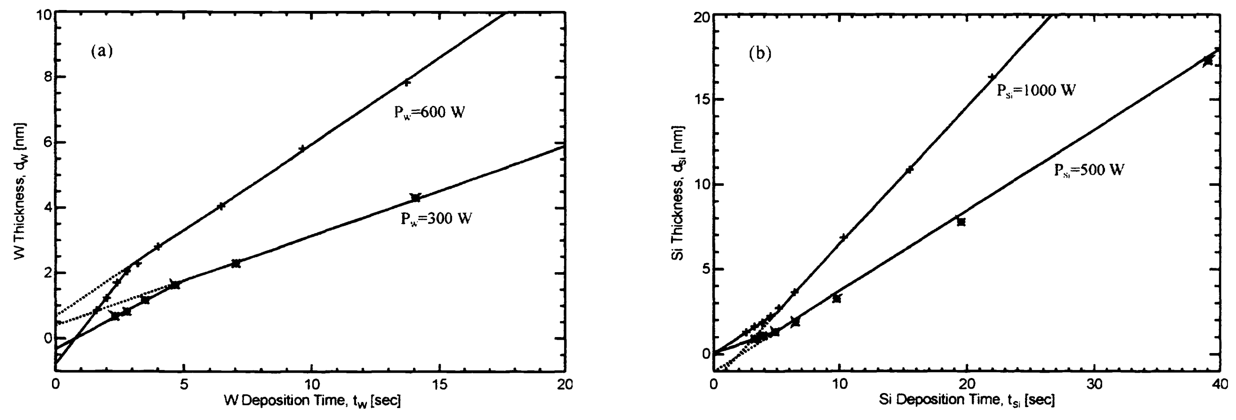

Figure 3. Layer thickness vs. deposition time for W (a) and Si (b) determined from X-ray reflectance analysis of multilayer films deposited at the magnetron powers indicated. Also shown are bi-linear fits to these data.

lines in Fig. 3) can be used to predict the measured multilayer periods with an accuracy of better than 0.05 nm, which is sufficient for the deposition of depth-graded multilayers for use as hard X-ray mirrors.

Hard X-ray reflectance measurements made with synchrotron radiation at ESRF were used to quantify the performance of optimized depth-graded W/Si structures over the photon energy range from 18 – 212 keV. Shown in Figure 4 is a cross-sectional TEM image of the top portion of a depth-graded W/Si multilayer structure containing 150 bilayers, and designed for high reflectance over the range 20 keV < E < 70 keV. Shown in Figs. 5(a) and 5(b) are plots of the reflectance vs. energy of an identical depth-graded film, measured over the range 18 keV < E < 170 keV, at grazing incidence angles θ=0.095° and θ=0.075°, respectively; also shown are the calculated reflectance curves, made using a depth-graded distribution of interface widths in the range σ=0.275 – 0.35 nm (as suggested by 8 keV X-ray and TEM analysis), which agree favorably with the measurements. The reflectance of this film drops sharply above the W K-edge (E~69.4 keV), where absorption in the film is high; the reflectance at the design incidence angle [i.e., θ=0.095°, Fig. 5(a)] is only a few per cent for E>100 keV, however at θ=0.075° [Fig. 5(b)] the reflectance is in excess of 10% up to E=170 keV. Depth-graded coatings identical to those shown in Fig. 5 – as well as other coatings designed to operate at smaller graze angles – have been deposited onto the curved glass substrates that will be used for the HEFT hard X-ray telescope [8], and high energy synchrotron reflectance measurements reveal equivalent performance at the operational graze angles [9].

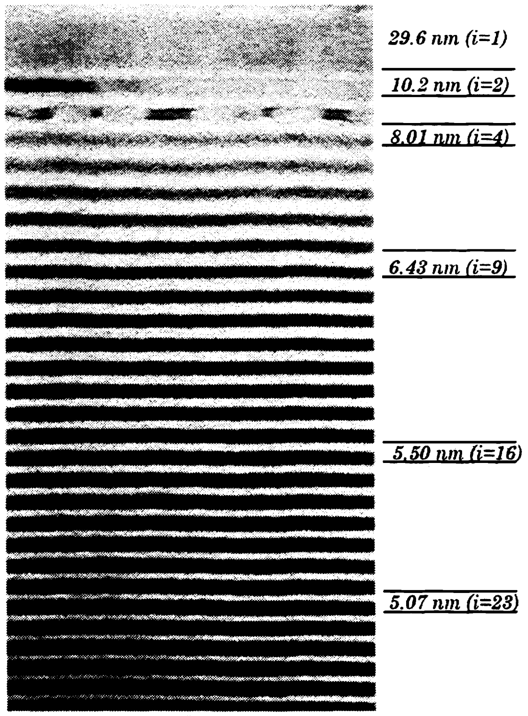

Figure 4. Cross-sectional transmission electron micrograph of the top portion (i.e., the top ~28 bilayers) of a depth-graded W/Si multilayer structure. Selected bilayer indices and thicknesses are indicated. The W (dark bands) and Si (light bands) layers are separated by thin amorphous W-Si interlayers (grey bands). Note that the topmost Si layer is not completely visible in this image.

In spite of the large absorption above the W K-edge, W/Si multilayers can still be used effectively for hard X-ray mirrors that operate at energies substantially greater than 100 keV. For example, shown in Figure 6 is the measured reflectance vs. energy (at an incidence angle θ=0.105°) for a W/Si multilayer having a slight gradation in bilayer thickness with depth: this film contains 800 bilayers, with bilayer thicknesses in the range 2.09 < *d* < 2.25 nm. The peak reflectance near the center of the Bragg peak (which is only slightly broadened due to the small depth gradation) at E=160 keV is R=78.7±4%, which agrees with the expected reflectance

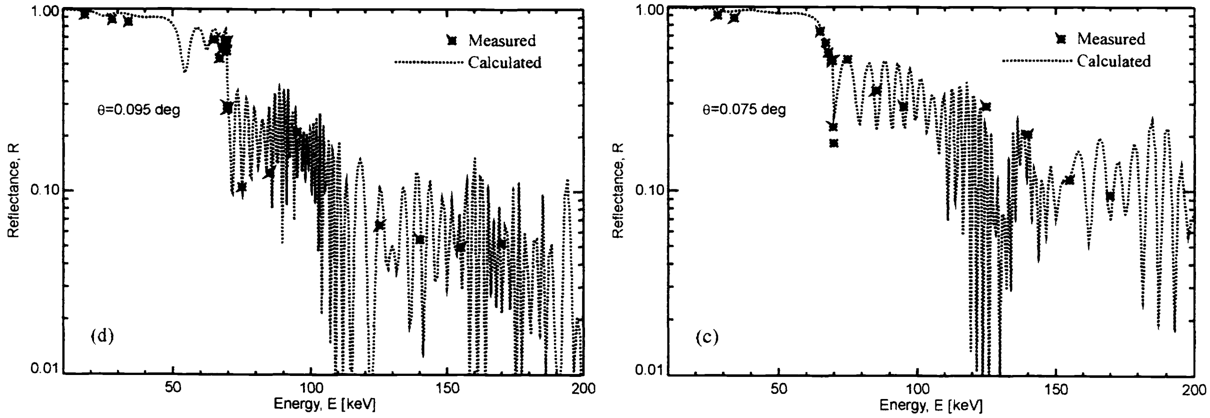

Figure 5. Reflectance vs. energy data for a depth-graded W/Si multilayer, identical to the one shown in Fig. 4, measured at θ=0.095° (a) and θ=0.075° (b). Shown for comparison are calculations made using a depth-graded distribution of interface widths in the range σ=0.275 – 0.35 nm.

(shown as the dotted line in Fig. 6) within the experimental uncertainty. This same film was also measured at E=212 keV, where the peak reflectance was found to be R=76.5±4% at θ=0.08°, again in agreement with the expected value.

3. Cu/Si, Ni/Si, Ni$_{.8}$Cr$_{.2}$/Si AND Ni$_{.93}$V$_{.07}$/Si MULTILAYERS

Ni- and Cu-based multilayer structures offer the advantage over, e.g., W- or Pt-based films of having much lower absorption at energies above 100 keV; as a result of the lower absorption, the X-ray penetration depth into the film is greater which in turn results in enhanced broad-band performance. To illustrate, shown in Figure 7 are theoretical reflectance curves for Ni/Si, Cu/Si, W/Si, and Pt/C multilayers optimized for use in the 145 – 175 keV energy band.

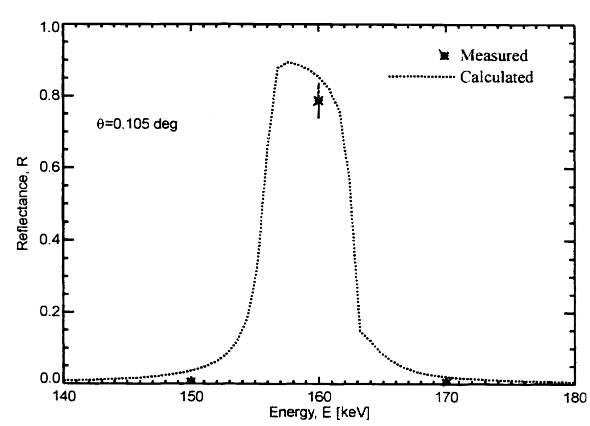

Figure 6. Measured reflectance vs. energy at θ=0.105°for a W/Si multilayer containing 800 bilayers, with bilayer thicknesses in the range 2.09 < *d* < 2.25 nm. The calculated reflectance using σ=0.3 nm interface widths is shown for comparison.

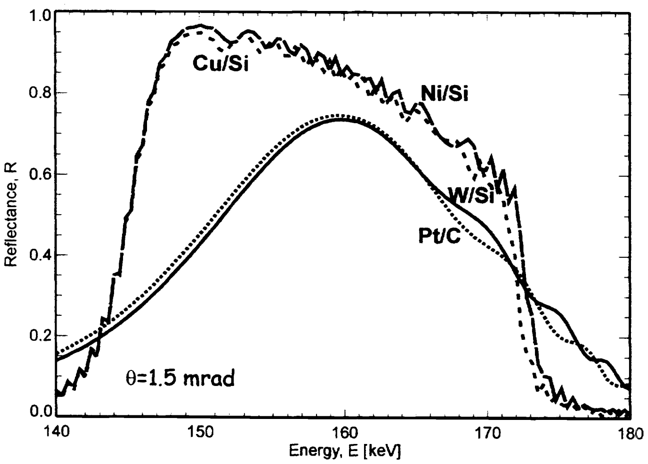

Figure 7. Theoretical reflectance curves for Ni/Si (long dashed), Cu/Si (dashed), W/Si (solid) and Pt/C (dotted) depth-graded multilayers containing 600 bilayers, optimized for use over the energy range from 145 < E < 175 keV.

We have investigated previously the growth and structure of Cu/Si multilayers [5], and found small interface widths (i.e., σ =0.23 – 0.3 nm) and good room-temperature stability over a period of several months. However, subsequent observations revealed that these structures are in fact unstable at room temperature: the films showed visible degradation (i.e., crazing and discoloration) after periods of 10 – 20 months (depending on the multilayer period). This degradation is likely due to Si diffusion resulting in copper-silicide formation. But whatever the cause, as a result of the long-term instability of Cu/Si multilayers, these structures are apparently unsuitable for use in space-borne hard X-ray telescopes.

We have recently begun to investigate Ni-based multilayers, which offer theoretical performance equivalent to Cu/Si, and some of these structures show small interface widths and good stability not just at room temperature but at elevated temperatures as well, as described below. However, because Ni is ferromagnetic, pure Ni films are more difficult to deposit by planar magnetron sputtering: in the planar sputtering geometry (which is ideally suited for use in large-area deposition) the magnetic field strength above the surface of a ferromagnetic target is greatly reduced, and so it is difficult to sustain an argon plasma at argon pressures low enough to produce smooth, high-density films. We have therefore investigated the growth and structure of multilayers containing silicon and either of the non-magnetic Ni alloys $Ni_{.8}Cr_{.2}$ and $Ni_{.93}V_{.07}$, which both can be grown easily by planar magnetron sputtering. (The difference in the theoretical hard X-ray reflectance between Ni/Si and $Ni_{.8}Cr_{.2}$/Si or $Ni_{.93}V_{.07}$/Si multilayers is negligible.) For these initial investigations, we have used an S-Gun deposition system to deposit periodic multilayers. Although the S-Gun geometry [10] does not easily scale to afford deposition over the large substrate areas that would be required for practical hard X-ray telescopes, there is little difficulty depositing ferromagnetic films in the S-Gun geometry, so it was thus possible to compare directly (non-magnetic) Ni-alloy/Si multilayer films with (magnetic) Ni/Si multilayers.

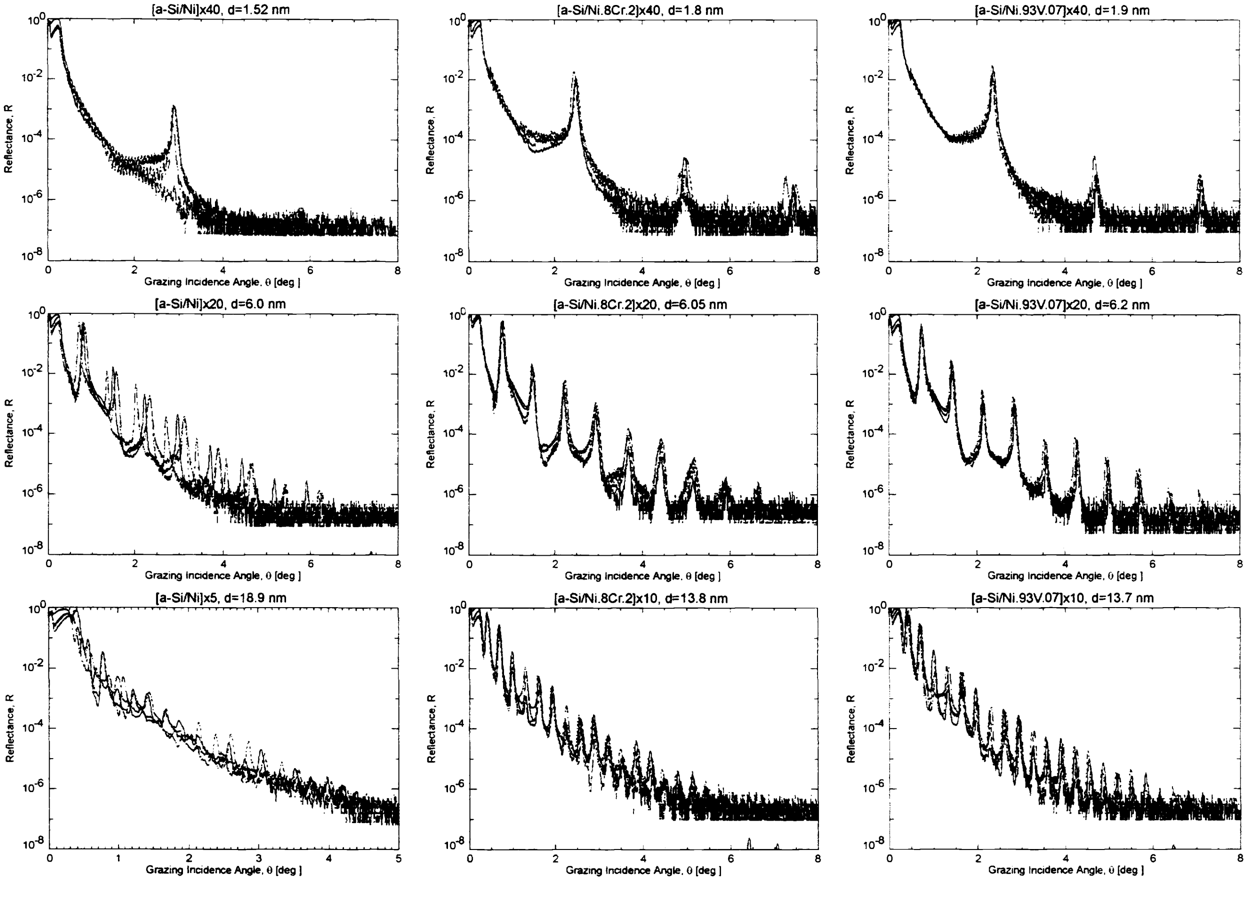

Figure 8. X-ray reflectance measured for periodic Ni/Si, $Ni_{.8}Cr_{.2}$/Si and $Ni_{.93}V_{.07}$/Si multilayers. Curves are shown for as-deposited (solid) samples, as well as samples measured after 1-hour thermal anneal in air at 100°C (dotted), 200°C (dashed) and 300°C (dot-dashed). The number of bilayers are indicated, as are the multilayer periods (d) determined for the as-deposited samples.

Shown in Figure 8 are the measured X-ray (8.04 keV) reflectance curves for a variety of periodic Ni/Si, $Ni_{.8}Cr_{.2}$/Si, and $Ni_{.93}V_{.07}$/Si multilayers, with periods ranging from ~1.5 nm – 19 nm (as will be required for use in depth-graded structures designed for operation above 100 keV.) From fits to these data, small interface widths, i.e., of order σ~0.3 nm, were inferred for all as-deposited films. Also shown in Fig. 8 for each of the nine samples are the X-ray reflectance curves measured after 1-hour thermal anneals made in air at temperatures 100°C, 200°C and 300°C. In the case of Ni/Si, large reductions in the Bragg peak intensities and shifts in their positions are observed at T=200°C and T=300°C for all three samples; these results indicate that significant structural changes (i.e., period shifts and increased interface widths) have taken place as a result of thermal anneal, probably due to diffusion and/or silicide formation, and suggest that Ni/Si might not be stable at room temperature over long periods of time. Much smaller changes in the Bragg peak intensities and positions are seen in the $Ni_{.8}Cr_{.2}/Si$ multilayers, while the $Ni_{.93}V_{.07}/Si$ films show even smaller changes in X-ray reflectance after thermal anneal; the small shifts in the Bragg peak positions in these $Ni_{.93}V_{.07}/Si$ multilayers indicate insignificant changes in the multilayer period in all cases, while increased interface widths can be inferred only for the d=13.7 nm films annealed at 200°C and 300°C. Multilayers containing Si and these non-magnetic Ni-alloys are thus especially promising for use as broad-band hard X-ray reflectors operating above 100 keV.

4. CONCLUSION

We have summarized our recent results on the development of depth-graded W/Si multilayer structures. Our results showing high reflectance at hard X-ray energies up to ~200 keV indicate that depth-graded W/Si multilayer coatings can be used for practical hard X-ray reflectors, and in particular for the new generation of hard X-ray astronomical telescopes currently being developed [1 – 4] that are designed to operate at energies below 100 keV, and possibly for future telescopes designed to operate at energies in excess of 100 keV. High quality films can be deposited by planar magnetron sputtering, a deposition technique that is easily scaled for large area coatings.

We have also described how Cu/Si multilayer films, although initially promising, appear to be unstable and therefore unsuitable for use in hard X-ray telescopes.

Finally, we have presented preliminary results obtained with periodic Ni/Si, $Ni_{.8}Cr_{.2}/Si$, and $Ni_{.93}V_{.07}/Si$ multilayers prepared by magnetron sputtering using an S-Gun deposition system. As a result of the especially encouraging results obtained with multilayers containing Si and non-magnetic Ni-alloys, showing small interface widths and good high-temperature stability, our future work will likely focus on the development of depth-graded $Ni_{.8}Cr_{.2}/Si$ and $Ni_{.93}V_{.07}/Si$ multilayers (prepared by planar magnetron sputtering) optimized for use above 100 keV.

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