

Advancements in Hard X-ray Multilayers for X-ray Astronomy

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ABSTRACT

This paper is focused on recent progress in the development of broad-band multilayer coatings designed for hard X-ray energies, for use in future astronomical telescopes. We describe a new laboratory-based hard X-ray reflectometer for at-wavelength characterization of multilayer films, we present the results of an experimental comparison of the hard X-ray performance of several W-based periodic multilayer coatings, and we describe the optimization and experimental performance of new non-periodic Co-based multilayer coatings (both depth-graded and aperiodic), designed for continuous response through the W and Pt K-edges near 70 and 80 keV, respectively. We discuss future research directions in light of these new results.

Keywords: Multilayers, Hard X-ray Astronomy

1. INTRODUCTION

The success of the NuSTAR astronomy mission has paved the way for the use of broad-band multilayer coatings operating at hard X-ray wavelengths. Reflective multilayer coatings such as those used on NuSTAR, that can operate efficiently at grazing incidence angles that are larger than those required for use with single-layer coatings such as Ir, enable the construction of focusing telescopes operating at X-ray energies higher than ~ 10 keV, and having focal lengths that are sufficiently short for practical use in balloon and satellite instruments. Future astronomy missions targeting the hard X-ray band will likely have improved angular resolution and better sensitivity, and may operate up to X-ray energies that exceed those attainable with the NuSTAR instrument.

This paper is focused on the development of improved broad-band multilayer coatings operating in the hard X-ray band, specifically for use in future X-ray astronomy missions having better sensitivity and operating at higher energies. We describe recent progress in the development of the needed infrastructure for hard X-ray multilayer design, fabrication and at-wavelength characterization, and in the development of W- and Co-based broad-band multilayers.

In section 2 we outline how hard X-ray multilayers operate and how they are made, and we describe a new laboratory-based hard X-ray reflectometer for at-wavelength characterization. In section 3 we present recent results comparing the hard X-ray performance of a variety of periodic W-based multilayers, followed by the results of an investigation into the optimization of non-periodic CoCr/C multilayers, both depth-graded and aperiodic, designed for operation up to 100 keV or greater. We conclude with a summary of our results, and an outline for future research directions.

2. HARD X-RAY MULTILAYERS: OVERVIEW

Principle of Operation

Multilayer coatings use optical interference to efficiently reflect X-rays at graze angles that are significantly larger than those used for single-layer coatings such as Ir that rely on total external reflection for high efficiency. Multilayer coatings comprise a stack of layers of optically dissimilar materials designed so that the small reflections that occur at each interface in the stack add coherently, in phase, over some range of graze angles and photon energies. A “periodic” multilayer, as illustrated in Fig. 1a, is a film stack containing a number of repeating, identical bilayers (typically) of thickness d . Just as Bragg's law describes the condition for constructive interference of X-rays in a crystal, the same law describes the condition for constructive interference in a periodic multilayer film (albeit with small, yet important corrections for refraction within the layers, which we ignore for this simplified discussion): $n\lambda=2d\sin\theta$, where n is the Bragg order, θ is the grazing incidence angle and λ is the photon wavelength. At a given incidence angle, a periodic multilayer film will reflect X-rays over a relatively narrow range of energies, as illustrated, for example, in Fig. 1b, where we present the measured and calculated reflectance of a periodic W/Si multilayer film containing $N=100$ repetitions of a bilayer of thickness $d=4.2$ nm, as measured at graze angles ranging from $\theta=0.1^\circ$ to $\theta=0.5^\circ$, as labeled.

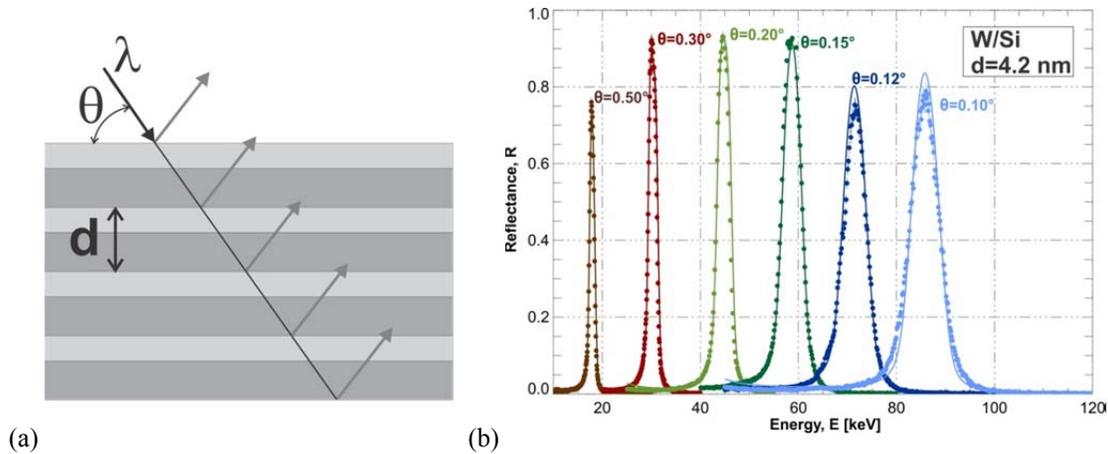


Figure 1. (a) Schematic diagram of a periodic multilayer. (b) Measured (symbols) and calculated (solid) reflectance of a periodic W/Si multilayer having a period $d=4.2$ nm, measured at graze angles in the range $\theta=0.1^\circ$ to $\theta=0.5^\circ$.

For X-ray astronomy, multilayer coatings having a broad spectral response are typically required, which necessitates the use of non-periodic coatings. That is, as illustrated in Fig. 2a, a broad-band multilayer comprises a stack of bilayers having a range of thicknesses. Several types of non-periodic coatings have been investigated over the years, including designs containing several periodic “blocks” of bilayers having a range of periods,¹ so-called “depth-graded” designs in which the distribution of bilayer thicknesses are described analytically,² and fully aperiodic structures that are designed numerically;³ aperiodic coatings can now be designed with the IMD software package⁴, for example. The coatings used for the NuSTAR instrument are depth-graded designs, comprising either W/Si bilayers or Pt/C bilayers.⁵ Shown in Fig. 2b is the measured and calculated reflectance of a depth-graded W/Si multilayer designed to operate efficiently up to the W K-edge near 70 keV; the Pt/C multilayers used on NuSTAR operate with similarly high efficiency up to the Pt K-edge near 80 keV.

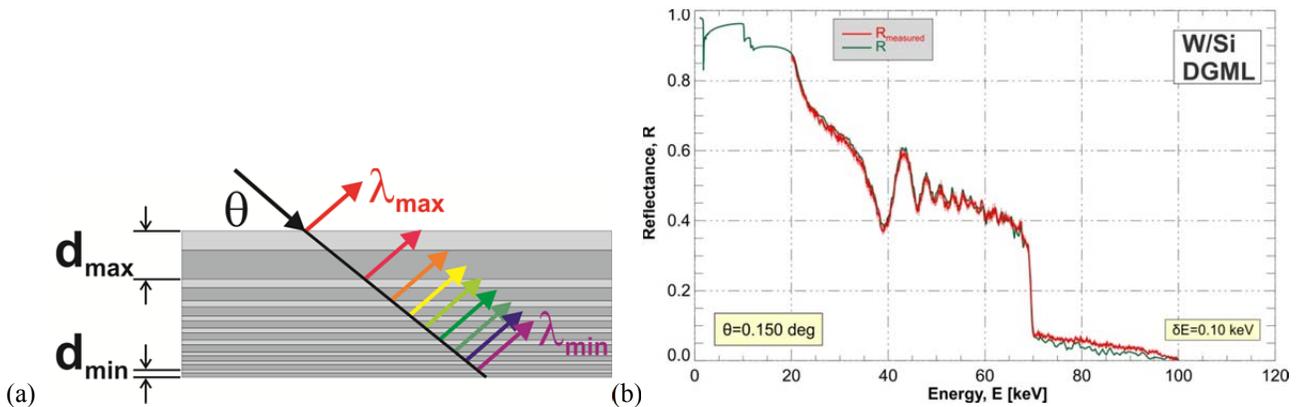


Figure 2. (a) Schematic diagram of a non-periodic multilayer coating containing a range of bilayer thicknesses. (b) Measured reflectance (red) of a non-periodic (depth-graded) W/Si multilayer having $N=291$ repetitions, and a fit (green) to the measured data.

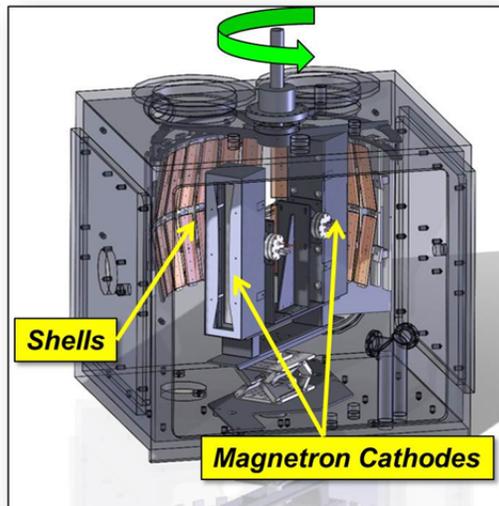
The X-ray performance of a multilayer coating is determined by the optical properties of the materials and the quality of the interfaces. Optimal performance is achieved with materials that (a) have large differences in their complex indices of refraction, so that the reflection coefficient at each interface is maximal, (b) have low absorption, so that the incident radiation penetrates deeply into the layer stack and reflects from as many interfaces as possible, and (c) form smooth, sharp (i.e., chemically abrupt) interfaces, so as to minimize both non-specular scattering from interfacial roughness, and increased transmission at an interface that is diffuse because of chemical reaction or atomic diffusion between adjacent layers.⁶ Furthermore, the high-energy cutoff of a non-periodic coating is constrained by the minimum bilayer thickness that can be fabricated in practice, which is in turn limited by interfacial diffusion and roughness, and is highly dependent on the constituent layer materials and the deposition conditions. While the identification of material pairs that meet the optical requirements is straightforward (assuming that accurate optical constants are available), the ability to grow nm-

scale bilayers having sufficiently small periods, and sufficiently small interface imperfections must be investigated experimentally, in general.

Fabrication

Magnetron sputtering is the most widely used technique for the deposition of hard X-ray multilayers for astronomical optics. The magnetron cathode consists of a solid target of the material to be deposited, fixed in place in front of an array of strong, permanent magnets. The cathode is mounted in a vacuum chamber that is evacuated to a low base pressure and then back-filled with the chosen working gas (e.g., Ar). A voltage is applied to the target relative to the grounded anode so as to create a plasma that is concentrated by the magnetic field in a region close to the target surface. Ionized gas atoms in the plasma are accelerated to the target surface where they can collide with and eject target atoms; the ejected target atoms then migrate to the substrate surface where they condense to form the growing film. The power applied to the magnetron cathode, and the gas flow rate, are typically controlled with high precision, in order to achieve high temporal stability during the deposition run, and good run-to-run repeatability. Magnetron sputtering is easily scaled to large areas, making it an attractive choice for coating large numbers of the cylindrical telescope mirror substrates that are used in the construction of nested, hard X-ray telescopes like NuSTAR. Magnetron sputtering also provides significant control over the distribution of energetic particles that bombard the growing film, a beneficial mechanism that can reduce surface and interface roughness in nm-scale multilayer films by increasing the effective surface mobility of adatoms.⁷ Control of particle energetics can be achieved, for example, by adjusting the working gas pressure and composition, by adjusting the power applied to the magnetron, by the application of a bias voltage applied to the substrate relative to the cathode, and through other methods.⁸

Some of the W/Si multilayers used in the construction of the NuSTAR instrument were grown by magnetron sputtering in argon using the large coating system shown in Fig. 3 that utilizes planar, rectangular magnetrons that are 50 cm in length.⁹ Two magnetron cathodes (one for W, one for Si) are mounted vertically, facing outward, while the inward-facing substrates are mounted in a rotating cylindrical fixture. Shaped baffles placed in front of each cathode are used to control axial coating uniformity. The multilayer stack is grown layer-by-layer, by rotating the substrates past each cathode in turn, building up the coating one layer at a time. The rotational velocity of the substrates is computer-controlled so that the layer thicknesses can be precisely adjusted according to the multilayer design, with sub-Å control.



(a)



(b)

Figure 3. (a) RXO's large planar magnetron system, as configured for coating cylindrical mirror shells. (b) NuSTAR shell segments after multilayer coating.

At-Wavelength Reflectometry

We have recently completed the development of a dedicated hard X-ray reflectometer (HXRR) facility¹⁰ for 'at-wavelength' characterization of X-ray mirrors in the energy range $\sim 15\text{--}150$ keV. Up until now we have been able to make only infrequent hard X-ray measurements of the coatings that we develop, using synchrotron radiation, typically at

the ESRF in France.¹¹ The new HXRR facility, which provides near-immediate feedback during (rather than long after) coating development, allows us to better investigate and refine our deposition techniques, and thus more effectively improve coating performance.

A schematic diagram and CAD model of the new HXRR facility are shown in Fig. 4. A sealed X-ray tube with a W anode (Comet MXR-160HP/11) operating at up to 160 kVp generates a continuum of X-rays. Two slit assemblies (JJ X-ray AT-F7-Air) spaced 4.1 m apart, each comprising horizontal and vertical slit blades (10-mm-thick WC) with motorized translations and tilts, are used to produce a low-divergence, small-diameter pencil beam. With 40 μm horizontal slit widths the beam divergence is ~ 8 arc-sec, and the incident count rate is $\sim 5,000$ cps with the tube at full power. The mirror under test is held flat with a vacuum chuck that is mounted on a 4-circle (Huber) goniometer that provides incidence angle adjustment with 0.0001° resolution. Motorized translation stages are used to position the mirror both horizontally (10 cm travel) and vertically (15 cm travel) as well, with $0.1 \mu\text{m}$ resolution. The CdTe detector (Amptek XR100T), located behind a third slit, rotates concentrically with the mirror in order to measure the spectrum of both the incident and reflected X-ray beams. The goniometer is housed in a HEPA-filtered radiation enclosure to accommodate flight hardware. Incident and reflected spectra are typically acquired with 15-min integration times. The reflectance is computed as the reflected spectrum divided by the incident spectrum.

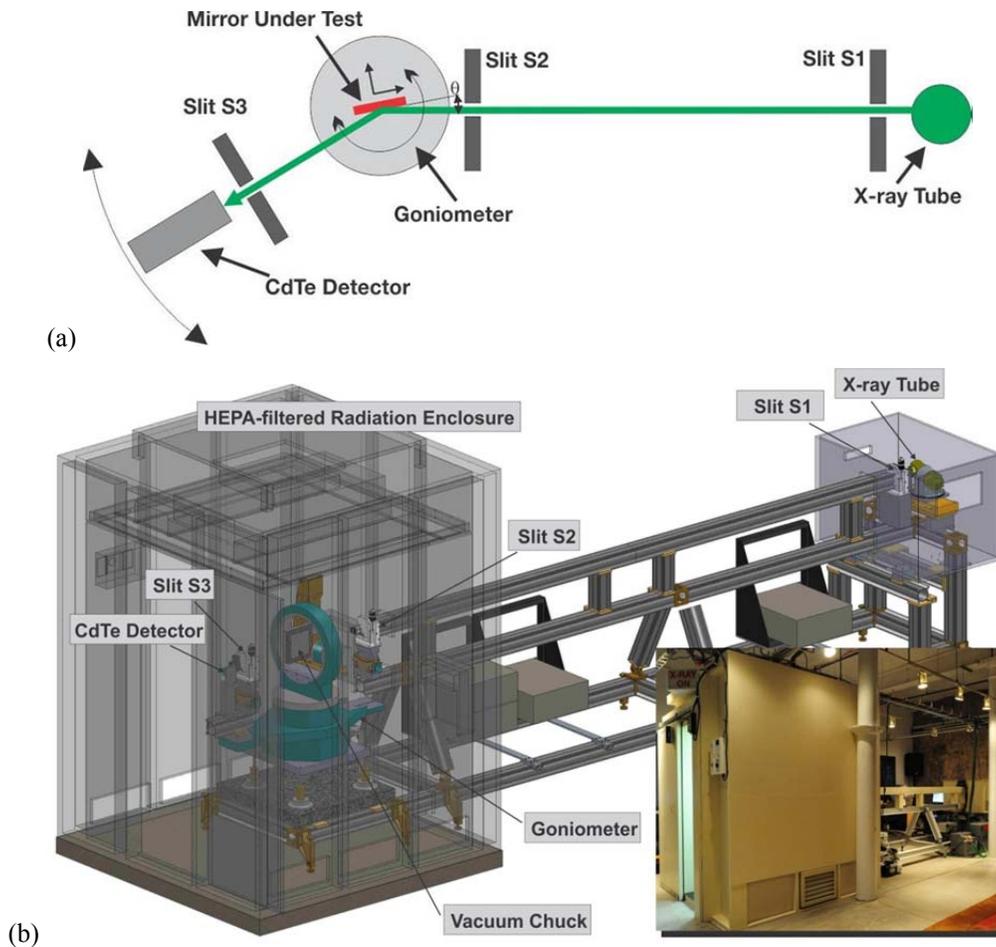


Figure 4. (a) Schematic diagram, and (b) CAD model and photo (inset) of the RXO Hard X-ray Reflectometer.

3. RECENT RESULTS

Periodic W-based Multilayers

A variety of high-quality hard X-ray multilayers containing W layers have been demonstrated over the years, including W/Si,¹² W/B₄C,¹³ W/SiC,¹¹ and others. Multilayers grown by reactive sputtering, using an Ar-N₂ mixture in place of pure

Ar as the working gas, have been shown to produce films having smoother interfaces and lower stress in many cases. Reactively-sputtered W/B₄C films¹⁴ are particularly attractive: W/B₄C multilayers with periods as small as d=0.8 nm have been demonstrated for use at normal incidence in the soft X-ray band,¹³ however the W/B₄C system also has very high stress. High film stress can lead to both catastrophic coating adhesion failure and substrate distortion that degrades angular resolution, particularly in the case of thin-shell cylindrical mirror substrates. Low-stress, reactively-sputtered W/B₄C:N₂ multilayer films may thus prove to be a good choice for use in future hard X-ray instruments targeting higher X-ray energies than NuSTAR, where very small bilayer thicknesses may be required.

We have recently compared the hard X-ray reflectance up to E~100 keV of a variety of periodic W-based multilayers. Shown in Fig. 5 are the measured reflectance-vs-energy curves around the 1st order Bragg peaks for periodic multilayers having d~4 nm, comprising N=100 bilayers of either W/Si, W/SiC, or W/B₄C, or low-stress, reactively-sputtered W/SiC:N₂ and W/B₄C:N₂. Each film was measured at six graze angles from $\theta=0.1^\circ$ to $\theta=0.5^\circ$, as labeled. We find that all these films have nearly identical X-ray reflectance (keeping in mind that there is a small variation in period from sample to sample), with only slight variations in spectral band-pass and peak reflectance; the small variations in peak reflectance at a given graze angle that we do observe could be due to variations in substrate flatness, but in any case these variations are within the experimental uncertainty of the measurements. In short, it should be possible to use any of these W-based multilayer systems for the fabrication of efficient broad-band, non-periodic coatings, at least for operation up to E~100 keV; the smallest bilayer thicknesses can be achieved with the W/SiC and W/B₄C systems, however, and so these material combinations may prove to be superior for the development of coatings operating up at higher X-ray energies.

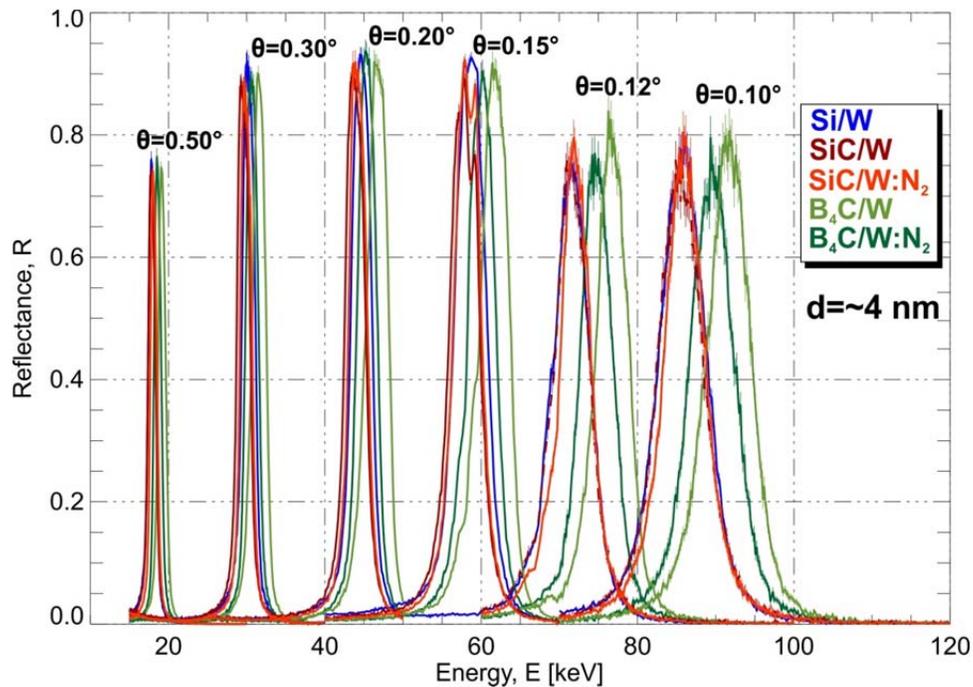


Figure 5. Experimental reflectance-vs-energy of five different periodic W-based multilayers, measured at incidence angles in the range $\theta=0.1^\circ$ to $\theta=0.5^\circ$. The coatings all contain N=100 periods, with a bilayer thickness of d~4 nm.

Depth-Graded CoCr/C Multilayers

The W/Si and Pt/C coatings used on NuSTAR were designed to operate most efficiently up to the W and Pt K-edges near 70 and 80 keV, respectively, as shown in Fig. 6 where we plot theoretical reflectance-vs-energy curves for both systems. Multilayer coatings constructed from light elements such as Ni and Co, also shown in Fig. 6, have already been identified as offering the possibility of continuous energy coverage without discontinuities up to X-ray energies that are higher than the W and Pt K-edges.^{15,16,17} After many years of research, however, the challenge remains of producing Ni- or Co-based coatings by magnetron sputtering that have sufficiently small bilayers, and sufficiently smooth, sharp interfaces. The difficulty in producing high-quality interfaces in short-period Ni or Co multilayers is due in large part to the relative masses of these atoms as compared to the mass of the Ar atoms typically used as the working gas in magnetron sputtering. That is, as mentioned above, bombardment during film growth by energetic particles in magnetron

sputtering can reduce interfacial roughness, and is largely responsible for the high-quality interfaces that can be achieved in many material systems through this deposition technique. In the case of heavy atoms such as W or Pt, most of the beneficial particle bombardment comes from neutral Ar atoms that are reflected by the W or Pt target;¹⁸ these neutral Ar atoms can arrive at the surface of the growing film with a large amount of energy and momentum, as a result of the large mass difference relative to the target atoms with which they collide, and are thereby able to transfer significant energy to migrating adatoms. In the case of Ni and Co, however, this mass difference (i.e., between the working gas atoms and the target atoms) is greatly reduced, and consequently, so is the beneficial bombardment of the growing film by energetic neutral Ar atoms, as the reflected Ar atoms carry much less energy and momentum. Without this bombardment, these particular light elements apparently do not have sufficient surface mobility to overcome the detrimental effects of self-shadowing.¹⁹

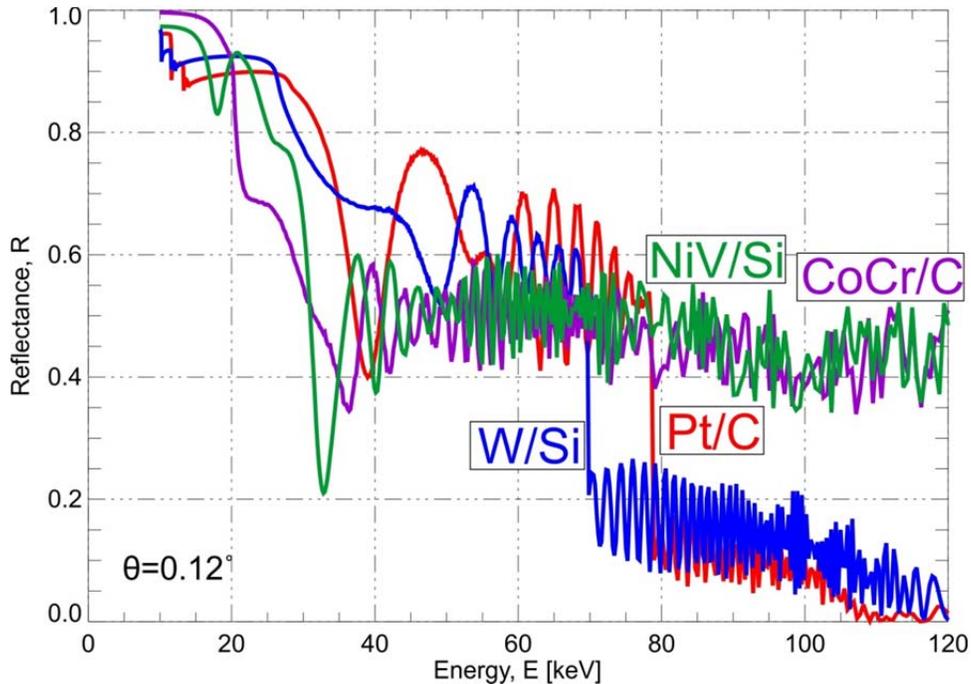


Figure 6. Theoretical reflectance of depth-graded W/Si, Pt/C, NiV/Si, and CoCr/C multilayers. Ni- and Co-based multilayers offer the promise of continuous energy response beyond the W and Pt K-edges near 70 and 80 keV, respectively.

In an effort to improve interface quality in spite of the lack of beneficial bombardment by energetic Ar atoms when sputtering Ni or Co, we have investigated the use of reactive sputtering in the growth of both Ni- and Co-based hard X-ray multilayer structures.¹⁶ While we have explored reactive sputtering (along with a variety of other deposition techniques) with both materials, here we focus exclusively on the recent progress made with Co-based multilayers. Specifically, we have used a sputtering target made of a non-magnetic Co alloy containing 20% Cr (ferromagnetic targets are generally problematic for planar magnetron cathodes), and have fabricated multilayer films comprising CoCr/C bilayers. Through a systematic investigation, we found that reactive sputtering with an Ar-N₂ mixture (~25% N₂ by volume) greatly reduces interfacial roughness (and film stress) in these films. To illustrate, shown in Fig. 7 are high-resolution transmission electron micrographs (HRTEM) of periodic CoCr/C films deposited using both reactive sputtering and non-reactive sputtering using pure Ar as the working gas, showing an obvious improvement in interfacial roughness in the case of the film deposited by reactive sputtering. The reason for this improvement, and in the associated reduction in film stress, is an open question.

In spite of the improvement in interface quality observed in periodic CoCr/C films deposited by reactive sputtering, depth-graded CoCr/C films nevertheless show hard X-ray performance that is considerably worse than expected theoretically. Shown in Fig. 8 are the measured and calculated reflectance-vs-energy curves for a depth-graded CoCr/C:N₂ film at three different graze angles: $\theta=0.10^\circ$, $\theta=0.15^\circ$, and $\theta=0.20^\circ$. This particular film contains N=1100 bilayers, with bilayer thicknesses in the range $2.5 \text{ nm} < d < 26.5 \text{ nm}$. The theoretical curves were computed using IMD, assuming interface widths of $\sigma=0.6 \text{ nm}$. The large disparity between the measured and calculated reflectance curves

suggest interface imperfections that are significantly larger than 0.6 nm; it may be the case that the interfacial roughness increases during film growth from the bottom to the top of the film, which is certainly plausible given that the total film thickness is approximately 3.5 μm . (In comparison, the depth-graded W/Si film shown in Fig. 2b, for example, has a total film thickness of less than 1 μm .)

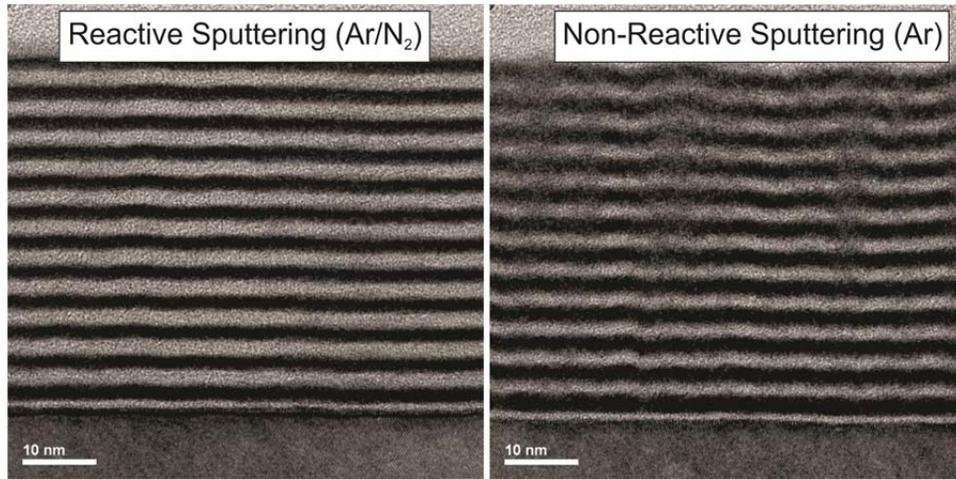


Figure 7. HRTEM images of periodic CoCr/C films deposited using reactive sputtering with an Ar-N₂ gas mixture, and non-reactive sputtering using pure Ar. Obvious signs of columnar growth are evident in the film deposited using non-reactive sputtering; the reactively-sputtered film has significantly better interface quality.

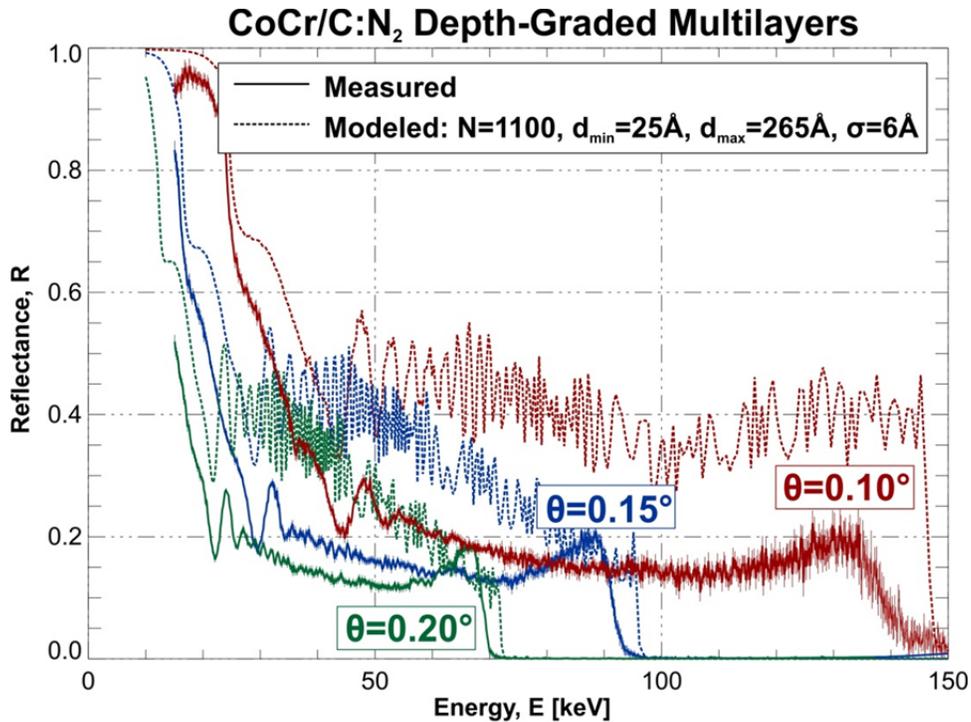


Figure 8. Measured (solid) and calculated (dotted) reflectance of a depth-graded CoCr/C:N₂ film containing N=1100 bilayers, with bilayer thicknesses in the range $d=2.5\text{--}26.5\text{ nm}$, at incidence angles $\theta=0.10^\circ$, 0.15° , and 0.20° as indicated. The calculated reflectance assumes 0.6 nm interface widths, however the experimental performance suggests much larger interface imperfections.

As it happens, CoCr/C films also work efficiently when operating near normal incidence in the soft X-ray band at energies below the C K-edge near 0.25 keV. Indeed, we have recently developed periodic CoCr/C multilayers for use as

efficient reflective polarizers operating at 45° incidence, in support of the development of a new type of astronomical soft X-ray polarimeter instrument.²⁰ In order to optimize the soft X-ray reflectance of these structures, we deposited a series of periodic CoCr/C:N₂ films by reactive sputtering, and we systematically varied the bias voltage applied to the substrate, from -10V to -300V. We also deposited films with a floating (unbiased) substrate potential. The soft X-ray reflectance of these films was measured at 45° incidence using synchrotron radiation at the ALS (courtesy of E. Gullikson). The results are shown in Fig. 9: we plot the reflectance-vs-wavelength as a function of substrate bias voltage in Fig. 9a, and the peak reflectance vs. bias voltage in Fig. 9b. We find that a substrate bias voltage of -100V produces the highest peak reflectance in this energy band. (Laterally-graded CoCr/C:N₂ multilayers were fabricated using this bias voltage, and the results are included in reference [20].) Presumably the applied substrate bias voltage affects the energetics of charged particles bombarding the growing film in a way that reduces roughness, although the detailed mechanisms responsible for this performance improvement are unclear at present.

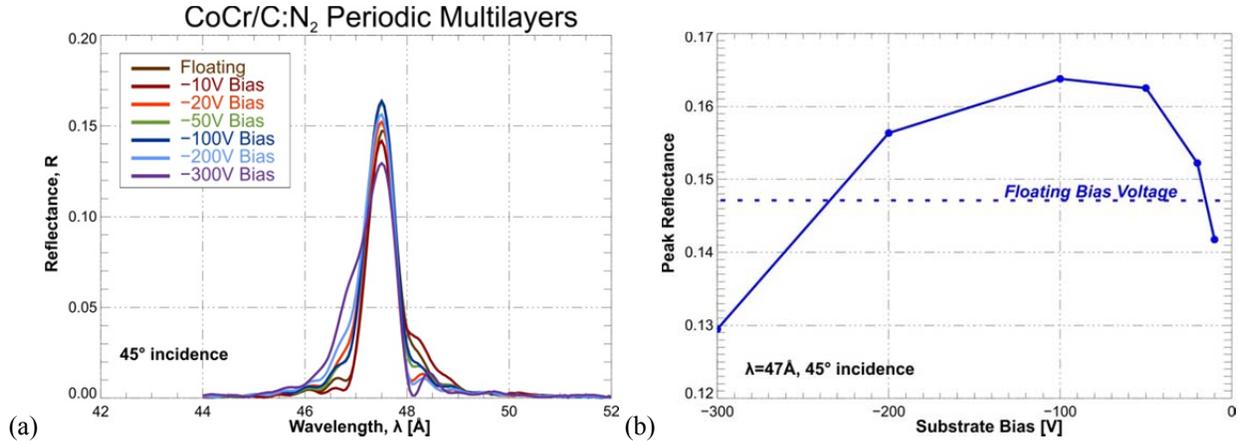


Figure 9. (a) Soft X-ray reflectance of periodic CoCr/C:N₂ multilayers, measured at 45° incidence using synchrotron radiation at the ALS, as a function of applied substrate bias voltage. (b) Peak reflectance vs. substrate bias voltage. The highest soft X-ray reflectance was obtained with a substrate bias of -100V.

Based on the good soft X-ray performance obtained with periodic CoCr/C:N₂ multilayers deposited with -100V bias, we used the same deposition conditions to fabricate a new depth-graded CoCr/C:N₂ multilayer that follows the same design as the film shown in Fig. 8. The hard X-ray performance of this film is shown in Fig. 10: as compared to the film deposited using a floating substrate voltage (also shown in Fig. 10), the new film provides significantly higher reflectance. For example, at a graze angle of $\theta=0.10^\circ$ we obtain reflectance of $\sim 20\% - 40\%$ in the energy range $50 < E < 130$ keV.

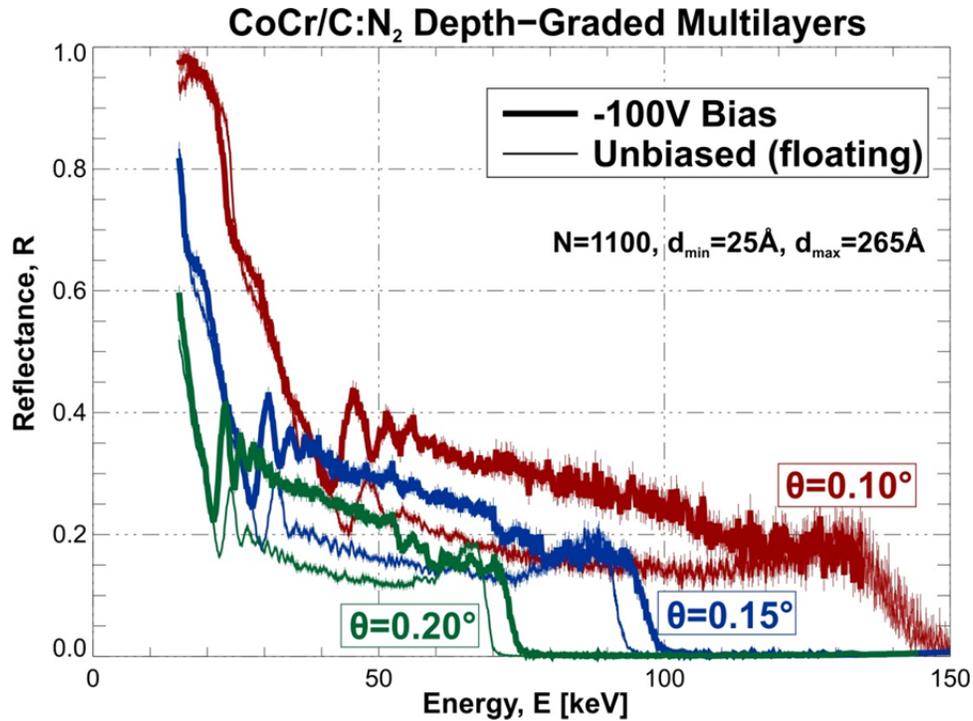


Figure 10. Measured reflectance of depth-graded CoCr/C:N₂ films, deposited using a floating substrate voltage (thin lines) or a bias voltage of -100V (thick lines), at incidence angles $\theta=0.10^\circ$, 0.15° , and 0.20° as indicated.

Aperiodic CoCr/C Multilayers

Aperiodic coating designs, in which the distribution of bilayer thicknesses is determined numerically rather than analytically, offer the promise of films having smooth, flat response over a wide energy range.^{3,21,22} We have begun to investigate the design and performance of aperiodic CoCr/C:N₂ multilayers. Shown in Fig. 11 are theoretical reflectance curves for one aperiodic design containing $N=800$ bilayers, and having a total film thickness of $2.7\ \mu\text{m}$, along with the theoretical response of the depth-graded design shown above in Figs. 8 and 10, which is somewhat thicker ($3.5\ \mu\text{m}$.) We (optimistically) assume $0.3\ \text{nm}$ interface widths in both cases. This particular aperiodic coating was designed (using a genetic algorithm in IMD) to have flat response from 40 to 110 keV, and smooth response from 20 to 40 keV, at an incidence angle of $\theta=0.135^\circ$. The distribution of layer thicknesses vs film depth for the two designs are shown in Fig. 12.

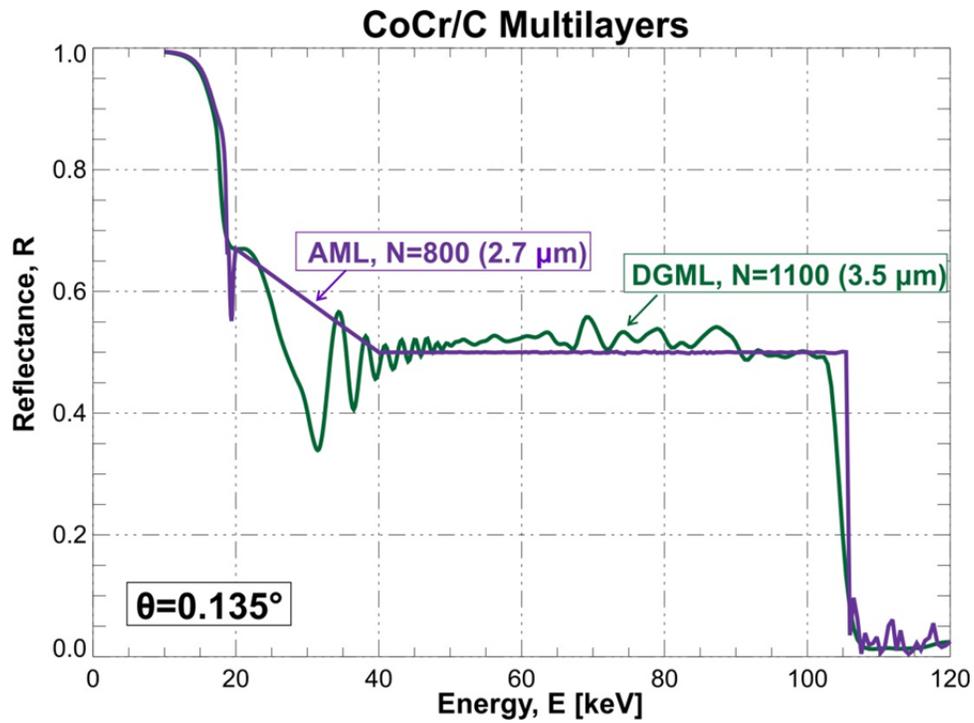


Figure 11. Theoretical reflectance curves for depth-graded (DGML) and aperiodic (AML) CoCr/C multilayers.

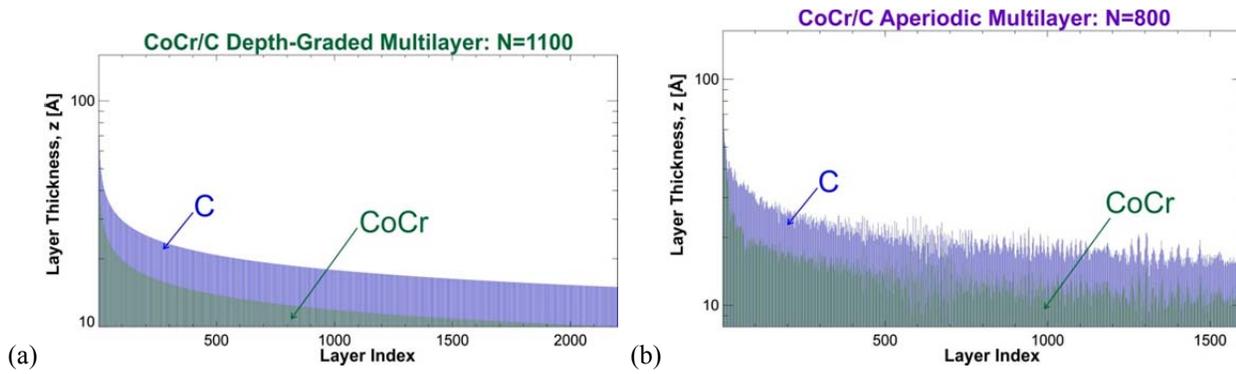


Figure 12. Layer thickness distributions for the DGML and AML coatings shown in Fig. 11.

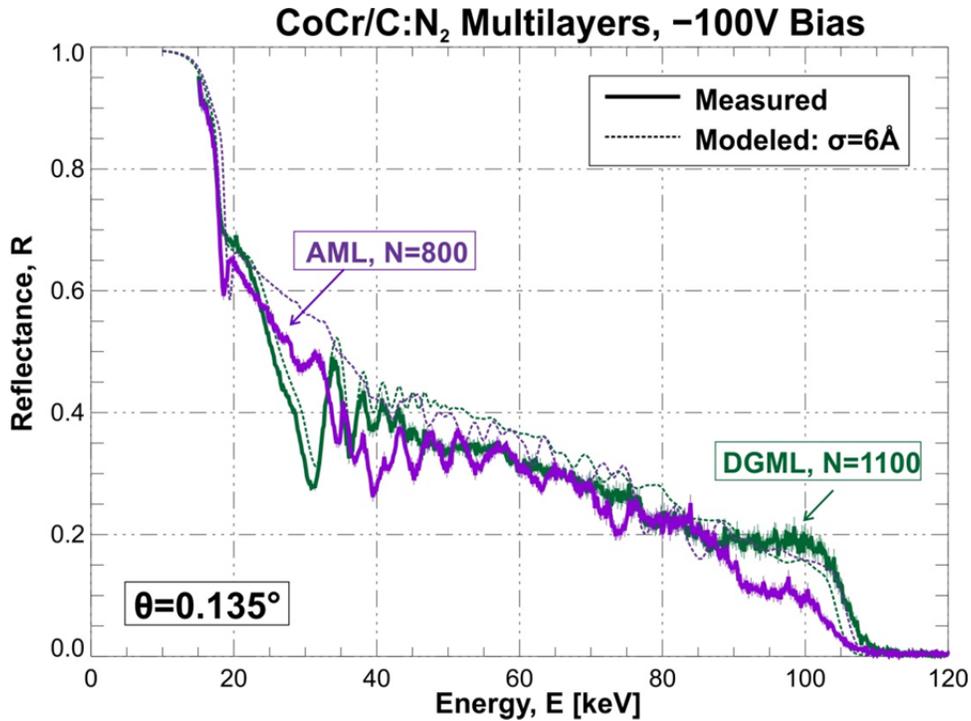


Figure 13. Measured and calculated reflectance of the DGML and AML CoCr/C:N₂ coatings shown in Fig. 11. The calculations use 0.6-nm-thick interface widths in both cases.

The aperiodic CoCr/C:N₂ film was fabricated using reactive sputtering and -100V substrate bias. The measured hard X-ray reflectance is shown in Fig. 13, along with reflectance of the depth-graded film already presented above that was deposited under identical deposition conditions. The calculated response curves of the two films are also shown in Fig. 13, assuming interface widths of $\sigma=0.6$ nm. The calculations using this interface width agree reasonably well with the measurements in this case (and obviously the 0.3 nm interface widths assumed in the coating designs shown in Fig. 11 are incorrect), although the disparity between measurement and calculation is greater in the case of the aperiodic film. This disparity is likely due to layer thickness errors stemming from inaccurate deposition rate calibrations, and/or the growth of CoCr-C interlayers at each interface that were not taken into account in the coating design. While both structures likely suffer from layer thickness errors to some extent, the reflectance of the aperiodic structure is far more sensitive to such errors than is the depth-graded design. In any case, aperiodic film designs show promise, but more work is needed to reduce layer thickness errors and interface imperfections in order to realize experimental performance that is even closer to the design.

4. SUMMARY AND CONCLUSIONS

We have described the development of a new laboratory-based Hard X-ray Reflectometer system used for at-wavelength characterization of hard X-ray multilayers for astronomy. We have used this system to compare the performance of a variety of periodic W-based multilayers at hard X-ray energies. We find that periodic W/Si, W/SiC, and W/B₄C multilayers, as well as low-stress W/SiC:N₂ and W/B₄C:N₂ multilayers prepared by reactive sputtering in an Ar-N₂ gas mixture, all have about the same hard X-ray performance. Based on these results, we conclude that there will likely be little difference in the performance of depth-graded multilayers fabricated from any of these material combinations, at least in the case of films designed for energies below ~100 keV. However, smaller bilayer thicknesses are possible with the W/SiC and W/B₄C systems (and presumably with low-stress W/SiC:N₂ and W/B₄C:N₂ multilayers fabricated by reactive sputtering as well), and so these material combinations may be better suited to W-based multilayers designed for higher X-ray energies, well above the W K-edge, where very small bilayer thicknesses may be needed.

We have made significant progress in the development of non-periodic hard X-ray CoCr/C films designed to provide continuous response above the W and Pt K-edges. We have determined that CoCr/C films deposited by reactive sputtering have smaller interface imperfections than films deposited using conventional sputtering using pure Ar; even

better performance can be obtained with reactively-sputtered CoCr/C films deposited with an optimized substrate bias voltage of -100V. We have fabricated and tested both depth-graded and aperiodic CoCr/C films using these optimized deposition conditions, and have demonstrated that these films can provide hard X-ray response in the range of 20-40% at $\theta=0.10^\circ$ incidence for X-ray energies from $\sim 50 - 130$ keV.

In spite of the recent progress made in the development of non-periodic CoCr/C multilayers as just summarized, without significant improvements in interface quality it is not clear that these coatings can provide a significant advantage over W-based multilayers for use in astronomical X-ray telescopes. Fig. 14 compares the measured reflectance-vs-energy curves for a depth-graded W/Si coating and a depth-graded CoCr/C coating, as measured at $\theta=0.10^\circ$ and $\theta=0.15^\circ$. While the CoCr/C film does provide higher reflectance than the W/Si film above the W K-edge, the reflectance of the CoCr/C film below the W K-edge is significantly lower than that of the W/Si film. (Furthermore, CoCr/C provides only a slight increase in reflectance above the K-edge at $\theta=0.10^\circ$.) Improved aperiodic CoCr/C designs, and perhaps a reduction in interface imperfections, may help with the performance at lower X-ray energies to some extent. Nevertheless, efficient CoCr/C designs are typically at least three times thicker than comparable W/Si coatings; the time (and hence the cost) required to deposit such films scales with thickness, while substrate distortions due to film stress will increase with coating thickness as well as with stress. Consequently, it may be more practical and cost-effective to use W-based coatings that are specifically designed to work above the W K-edge.

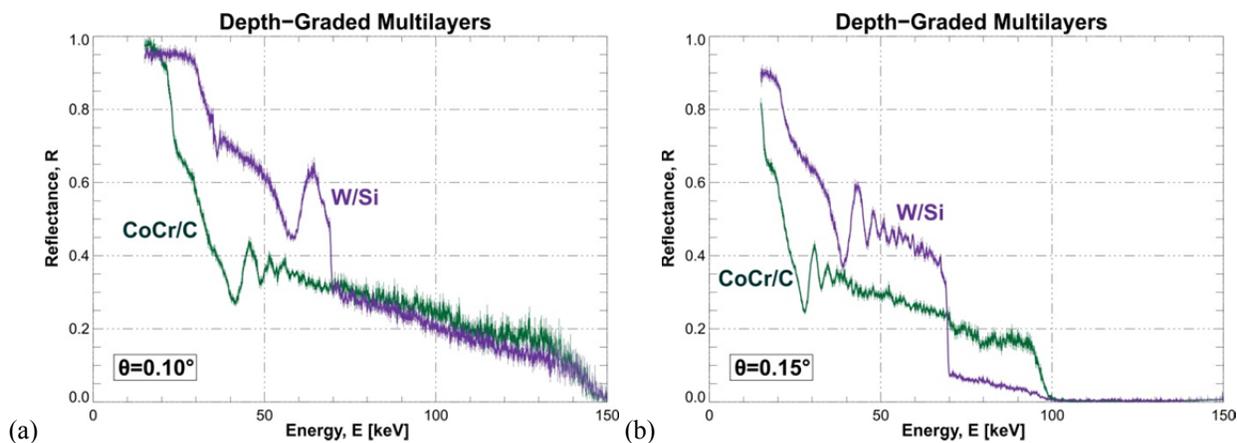


Figure 14. Experimental reflectance of depth-graded W/Si and CoCr/C:N₂ multilayers, measured at (a) $\theta=0.10^\circ$ and (b) $\theta=0.15^\circ$ incidence.

As shown in Fig. 15, depth-graded W/Si and W/SiC coatings have already been demonstrated to work efficiently in the 150–170 keV range,¹¹ while recent work on a new WC/SiC multilayer system has demonstrated that such multilayers can work efficiently up to energies at least as high as ~ 400 keV.²³ Therefore, future X-ray astronomy instruments targeting energies higher than the W and Pt K-edges might utilize only W-based coatings. For example, shown in Fig. 16 are two W/Si coatings designed specifically for relatively smooth response from $\sim 60-120$ keV: one is a depth-graded multilayer design containing $N=200$ bilayers, the other is an aperiodic multilayer containing $N=100$ bilayers. These coatings have total thicknesses of only 0.5 and 0.3 μm , respectively, and so they can be deposited much more quickly, and thus at lower cost, than Co- or Ni-based coatings that provide similar response in the 60–120 keV band, and that are several times thicker. Consequently, the most cost-effective construction of future, hard X-ray, nested telescopes targeting high X-ray energies might require only W-based coatings; the inner shells, having the smallest graze angles, could use high energy coating designs similar to those shown in Fig. 15, while the outer shells could use W-based coating designs similar to those used on NuSTAR, as shown, for example, in Fig. 2b, that operate with high efficiency below the W K-edge. Future research should be directed towards the further development of W-based coatings that operate above the W K-edge.

5. ACKNOWLEDGEMENTS

We kindly thank Eric Gullikson for the soft X-ray reflectance measurements presented here, and Herman Marshall for his support and encouragement in the work focused on the soft X-ray performance optimization of CoCr/C multilayers. This work was funded by NASA grant numbers NNXG06WC15G, NNX09AC45G, and NNX13AC54G.

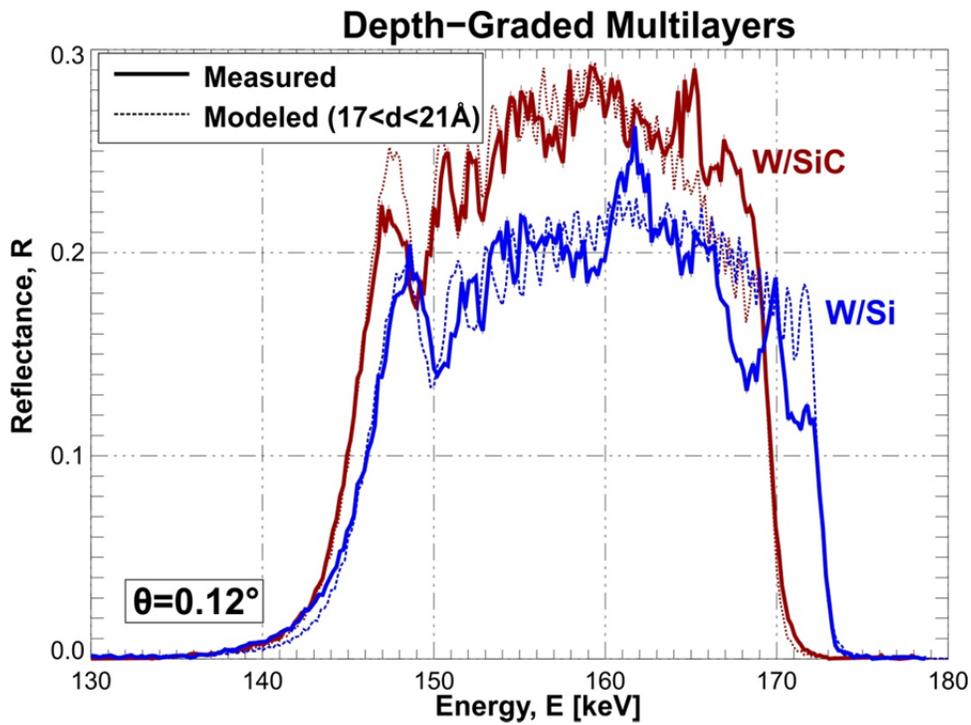


Figure 15. Measured (solid) and calculated (dotted) reflectance vs. energy of depth-graded W/Si and W/SiC multilayer films, measured using synchrotron radiation at the ESRF, from reference [11].

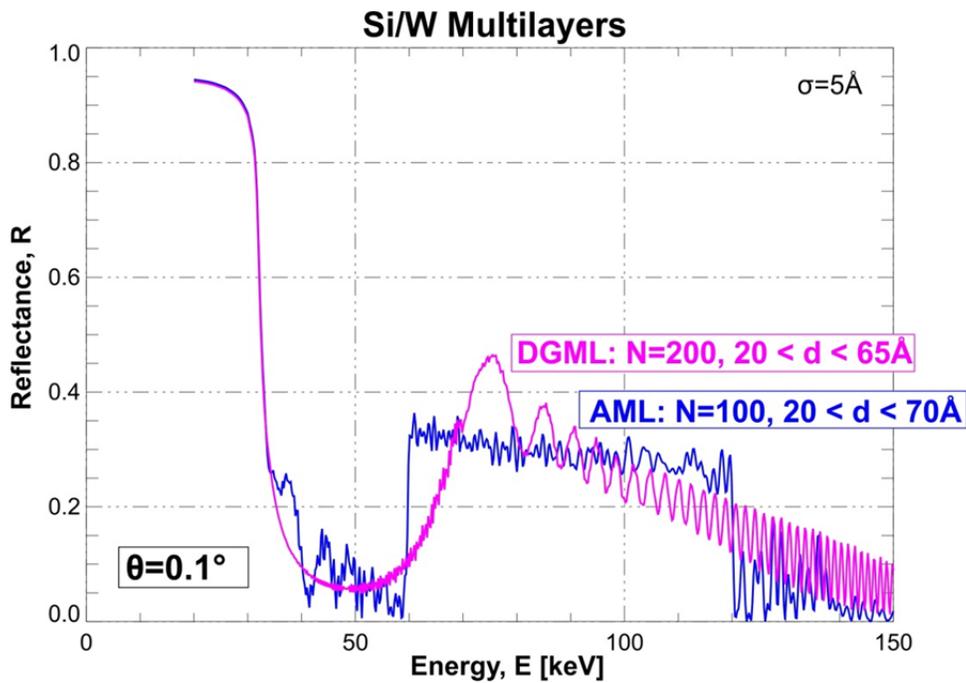


Figure 16. Theoretical reflectance of two W/Si multilayers, one depth-graded, one aperiodic, both designed for continuous response from ~60–120 keV.

REFERENCES

- [1] K. Yamashita, P. J. Serlemitsos, J. Tueller, S. D. Barthelmy, L. M. Bartlett, K.-W. Chan, A. Furuzawa, N. Gehrels, K. Haga, H. Kunieda, P. Kruczynski, G. Lodha, N. Nakajo, N. Nakamura, Y. Namba, Y. Ogasaka, T. Okajima, D. Palmer, A. Parsons, Y. Soong, C. M. Stahl, H. Takata, K. Tamura, Y. Tawara, and B. Teegarden, 'Supermirror hard-x-ray telescope', *App. Op.*, 37, 8067–8073 (1998); doi:10.1364/AO.37.008067
- [2] K. D. Joensen, P. Voutov, A. Szentgyorgyi, J. Roll, P. Gorenstein, P. Høghøj, and F. E. Christensen, 'Design of grazing-incidence multilayer supermirrors for hard-x-ray reflectors', *App. Opt.*, 34, 7935–7944 (1995)
- [3] I. V. Kozhevnikov, I. N. Bukreeva, E. Ziegler, 'Design of X-ray supermirrors', *Nucl. Inst. Meth., A*, 460, 424 – 443 (2001)
- [4] D. L. Windt, 'IMD - Software for modeling the optical properties of multilayer films', *Computers in Physics*, 12, 360-370 (1998); The current version of IMD can be downloaded at <http://www.rxollc.com/imd>
- [5] F. E. Christensen, A. C. Jakobsen, N. F. Brejnholt, K. K. Madsen, A. Hornstrup, N. J. Westergaard, J. Momberg, J. Koglin, A. M. Fabricant, M. Stern, W. W. Craig, M. J. Pivovarov, D. Windt, 'Coatings for the NuSTAR mission', *Proc. SPIE*, 8147, 81470U (2011)
- [6] D. G. Stearns, 'X-ray scattering from interfacial roughness in multilayer structures', *J. Appl. Phys.*, 71, 4286-4298 (1992)
- [7] F. M. D'Heurle, 'Aluminum films deposited by RF sputtering', *Metall. Transactions*, 1, 725-732 (1970)
- [8] J. A. Thornton, 'Influence of apparatus geometry and deposition conditions on the structure and topography of thick sputtered coatings', *J. Vac. Sci. Technol.*, 11, 666-670 (1974)
- [9] D. L. Windt and W. K. Waskiewicz, 'Multilayer facilities for EUV lithography', *J. Vac. Sci. Technol. B*, 12, 3826-3832 (1994)
- [10] D. L. Windt, 'Laboratory-based x-ray reflectometer for multilayer characterization in the 15-150 keV energy band', *Rev. Sci. Inst.*, 86, 043107 (2015); doi: 10.1063/1.4916737
- [11] D. L. Windt, S. Donguy, C. J. Hailey, J. Koglin, V. Honkimäki, E. Ziegler, F. E. Christensen, C. M. H. Chen, F. A. Harrison, W. W. Craig, 'W/SiC X-ray multilayers optimized for use above 100 keV', *App. Opt.*, 42, 2415 – 2421 (2003)
- [12] D. L. Windt, F. E. Christensen, W. W. Craig, C. Hailey, F. A. Harrison, M. Jimenez-Garate, R. Kalyanaraman, and P. H. Mao, 'Growth, structure and performance of depth-graded W/Si multilayers for hard X-ray optics', *J. Appl. Phys.*, 88, 460 – 470 (2000)
- [13] D. L. Windt, E. M. Gullikson, C. C. Walton, 'Normal-incidence reflectance of optimized W/B₄C multilayers in the range 1.4 nm λ <math>< 2.4\text{ nm}</math>', *Opt. Lett.*, 27, 2212-2214 (2002)
- [14] D. L. Windt, 'Reduction of stress and roughness by reactive sputtering in W/B₄C multilayer films', *Proc. SPIE* 6688, 66880R (2007)
- [15] D. L. Windt, F. Christensen, W. Craig, C. Hailey, F. Harrison, M. Jimenez-Garate, R. Kalyanaraman, and P. Mao, 'X-ray multilayer coatings for use at energies above 100 keV', *Proc. SPIE*, 4012, 442-447 (2000)
- [16] J. A. Bellotti and D. L. Windt, 'Depth-graded Co/C multilayers prepared by reactive sputtering', *Proc. SPIE*, 7437-743715 (2009)
- [17] D. Girou, E. Sleire, S. Massahi, F. E. Christensen, and A. C. Jakobsen, 'Development of Ni-based multilayers for future focusing soft gamma ray telescopes', *Proc. SPIE* 9603-43 (2015) *these proceedings*
- [18] H. Windischmann, 'Intrinsic stress in sputtered thin films', *J. Vac. Sci. Technol. A*, 9, 2431–2436 (1991)
- [19] G. S. Bales and A. Zangwill, 'Macroscopic model for columnar growth of amorphous films by sputter depositions', *J. Vac. Sci. Tech. A*, 9, 145 (1991); doi: 10.1116/1.577116
- [20] H. L. Marshall, N. S. Schulz, D. L. Windt, E. M. Gullikson, E. Blake, D. Getty, Z. McInturff, 'The use of laterally-graded multilayer mirrors for soft X-ray polarimetry', *Proc. SPIE*, 9144 (2014)
- [21] C. Morawe, E. Ziegler, J.-C. Peffen, and I. V. Kozhevnikov, 'Design and fabrication of depth-graded X-ray multilayers', *Nucl. Inst. Meth. A*, 493, 189 – 198 (2002)
- [22] A. L. Aquila, F. Salmassi, F. Dollar, Y. Liu, and E. M. Gullikson, 'Developments in realistic design for aperiodic Mo/Si multilayer mirrors', *Opt. Exp.*, 14, 10073 (2006)
- [23] M. Fernández-Perea, M. J. Pivovarov, R. Soufli, J. Alameda, P. Mirkarimi, M.-A. Descalle, S. L. Baker, T. McCarville, K. Ziock, D. Hornback, S. Romaine, R. Bruni, Z. Zhong, V. Honkimäki, E. Ziegler, F. E. Christensen, and A. C. Jakobsen, 'Ultra-short-period WC/SiC multilayer coatings for x-ray applications', *Nucl. Inst. Meth. A*, 710, 114 – 119 (2013)