Small d-spacing WSi_2/Si narrow bandpass multilayers

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ABSTRACT

To develop narrow-bandpass multilayer monochromators, we have studied small d-spacing WSi_2/Si multilayers. We found that WSi_2/Si is an excellent multilayer system for achieving both the desired spectral resolution and peak reflectivity. Compared to other traditional multilayer systems such as W/Si, WSi_2/Si not only has a lower density and lower absorption, but also is a chemically more stable system, since WSi_2 is already a silicide. One thus expects better thermal stability and sharper interfaces for WSi_2/Si multilayers. There are two approaches to achieve high-resolution multilayers: either decrease the d spacing or use low absorption materials. By using WSi_2/Si, we can utilize both approaches in the same system to achieve good energy resolution and peak reflectivity. Another advantage of this system is that the sputtering rate for Si is much higher than other traditional low-Z materials. Several WSi_2/Si multilayers have been fabricated at the Advanced Photon Source (APS) deposition lab using dc magnetron sputtering with constant currents of 0.5 A in Ar at a pressure of 2.3 mTorr. A test sample of [9.65Å-WSi_2/10.05Å-Si] × 300 was studied at four institutions: using laboratory x-ray diffractometers with Cu Kα (8.048 keV) wavelength at the APS x-ray lab and at European Synchrotron Radiation Facility (ESRF), and using synchrotron undulator x-rays at 10 keV at MHATT-CAT and at 25 keV at ChemMatCARS-CAT of the APS. The measured first-order reflectivity was 54% with a bandpass of 0.46% at 10 keV and 66% reflectivity with a bandpass of 0.67% at 25 keV of undulator x-rays. Similar results were obtained from Cu Kα x-rays. This result is very attractive for the design of a multilayer monochromator for the ChemMatCARS-CAT to be used in the 20 to 25 keV range. Other small d-spacing multilayers are being studied. Comparison between WSi_2/Si and W/Si multilayers will be discussed.

Keywords: x-ray optics, multilayer mirrors, sputter deposition

1. INTRODUCTION

Narrow-bandpass multilayers have attracted much attention in the past few years.[1-2] Recently, scientists at the ChemMatCARS-CAT at the APS found out that when the bandwidth of the synchrotron x-ray beam gets too large, their crystal-refinement programs do not work well for their crystallography experiments. A multilayer monochromator with a bandpass around 0.5% is more ideal for crystallography studies.

From Bragg optics, the width of the bandpass is determined by the number of diffracting planes N, i.e., $dE/E \propto 1/N$. For multilayers, N is limited by the penetration depth of the x-rays, as well as the capability of manufacturing processes. The bandpass of Bragg reflection for multilayers is thus much greater than crystals. The x-ray flux, however, is much greater for multilayer optics than crystal optics. To decrease the width of the bandpass, one may use low absorption material or use small d-spacing to increase N. A typical limitation for using low absorption materials is that the growth rates of most of these materials, such as Al_2O_3, B_4C, and C etc., are very low. The low growth rate, combined with a large N to grow, not only prolongs the manufacturing process but also presents a major challenge of stability for the deposition system. A multilayer system with a medium absorption and a high growth rate combined with a small d-spacing would be ideal for applications like the monochromator for the ChemMatCARS-CAT as mentioned earlier. In other applications, such as x-ray fluorescence detection and large-incident-angle x-ray monochromators, it is also desirable to have multilayers with a small d-spacing to decrease the absorption of x-rays and to increase the Bragg angle.

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However, fabricating small d-spacing multilayers is also a challenge. As d decreases, the interfacial roughness becomes more dominant and the reflectivity decreases. The interfacial roughness is related to the substrate roughness, as well as to interlayer diffusion/chemical reaction at the interface. In searching for a suitable narrow bandpass multilayer system for the monochromator for the ChemMatCARS-CAT, we found that WSi_2/Si not only has a lower density and lower absorption, but also is a chemically more stable system. Since WSi_2 is already a silicide, one would expect a sharper interface between WSi_2 and Si than that between W and Si. Under the same growth conditions, Si grows ~8 times faster than C or B_4C. WSi_2 also has a higher deposition rate than W. WSi_2/Si is thus an interesting system to study. Previously, WSi_2/Si multilayers have been studied for applications of curved graded multilayer mirrors for laboratory x-ray diffractometers.[3] In this paper we study the application of WSi_2/Si multilayers for narrow bandpass mirrors for synchrotron radiation and present the results in comparison with the W/Si system.

2. EXPERIMENT AND RESULTS

The multilayer films were prepared by dc magnetron sputtering in our large deposition system that has been described previously.[4] The system consists of four large vacuum chambers in series, with three CTI model CT-8 cryo pumps and an Alcatel ADP 81 dry pump providing a base pressure of $< 2 \times 10^{-8}$ Torr for the system. The multilayer films were grown in argon at 2.3 mTorr and ambient temperatures onto superpolished Si substrates with dimensions of $25 \times 50 \times 2.5$ mm^3. The substrates were purchased from Wave Precision and have typical rms surface roughness of < 1 Å.[5] These superpolished flat substrates were successfully used for fabricating elliptical Kirkpatrick-Baez mirrors using a profile coating technique.[6] For multilayer deposition, substrates were loaded on a carrier with the polished surface facing down, and were alternately moving linearly back and forth over two 3-inch-diameter planar sputter guns during deposition. Laterally uniform depositions were achieved through the design of shaped apertures above sputter guns.[7] The sputter guns were operated at a constant current of 0.5 A and the film thicknesses were determined by the moving speeds according to growth rate calibrations. The guns were programmed to be turned on 7 sec before the substrate was moved in and turned off when a desired layer was deposited. This procedure reduces the usage of the target material, lowers the target temperature, and ensures comparable growth conditions for each sequential layer growth.

To calibrate the growth rates, two 0.5” × 1” × 0.5mm Si test substrates (cut from an ordinary wafer) were loaded on the substrate holder ~15” apart. Two different [WSi_2/Si] × 15 multilayers were grown on these substrates with certain fixed moving speeds when they were passing the sputter guns. For substrate A, three loops over the WSi_2 gun and two loops over the Si gun were used to complete a bilayer. For substrate B, two loops over the WSi_2 gun and three loops over the Si gun were used to complete a bilayer. These two multilayers were characterized at the APS x-ray lab. Reflectivity measurements were made using Cu-Kα_1 x-rays with a collimating multilayer optic followed by a Ge crystal monochromator. The experimental data are presented in Figure 1, together with the fitted data using the IMD computer software.[8] The calibration results indicate that there is a very good agreement for the expected gamma (the ratio of WSi_2 thickness to the period d) for the WSi_2/Si system. The fitted thickness for Si is 50.28 Å for sample A and 75.42 Å for sample B, and for WSi_2 is 36.12 Å for A and 24.08 Å for B. A SiO_2 layer of 32 Å was added to the substrate surface and a layer of 8 Å to the topmost Si layer in the fitting model. Subsequently the film thickness was controlled by scaling the substrate moving speed and the number of loops (one loop for each layer for the present study).

A [9.65Å-WSi_2/10.05Å-Si] × 300 multilayer was grown on a 1” × 2” × 2.5 mm^3 superpolished substrate and measured using both the lab x-ray and synchrotron radiation x-ray beams. Figure 2 summarizes reflectivity measurements at four institutions. Figure 2(a) is from the MHATT-CAT of the APS and measured at 10 keV on 7-ID beamline. Figure 2(b) is from the ChemMatCARS-CAT of the APS at 25 keV on 15-ID beamline. Figure 2(c) is from the ESRF measured with a laboratory x-ray diffractometer at 8.048 keV. Figure 2(d) was measured at the APS x-ray lab. Figure 2(a) shows a first order reflectivity of 54% with a bandpass of 0.46%. Figure 2(b) shows a 66% reflectivity with a bandpass of 0.67% at a higher energy of 25 keV. For lab x-rays, Fig. 2(c) shows a 50.8% reflectivity with a bandpass of 0.575%, and Fig. 2(d) shows a 45% reflectivity with a bandpass of 0.575%. The slightly lower reflectivity measured at the APS x-ray lab may be due to incident beam intensity calibration problems associated with the sagittal divergence of the x-ray beam. Figures 2(c) and (d) demonstrate that lab x-ray diffractometers, when well adjusted and aligned, can also give good results for narrow-bandpass studies.

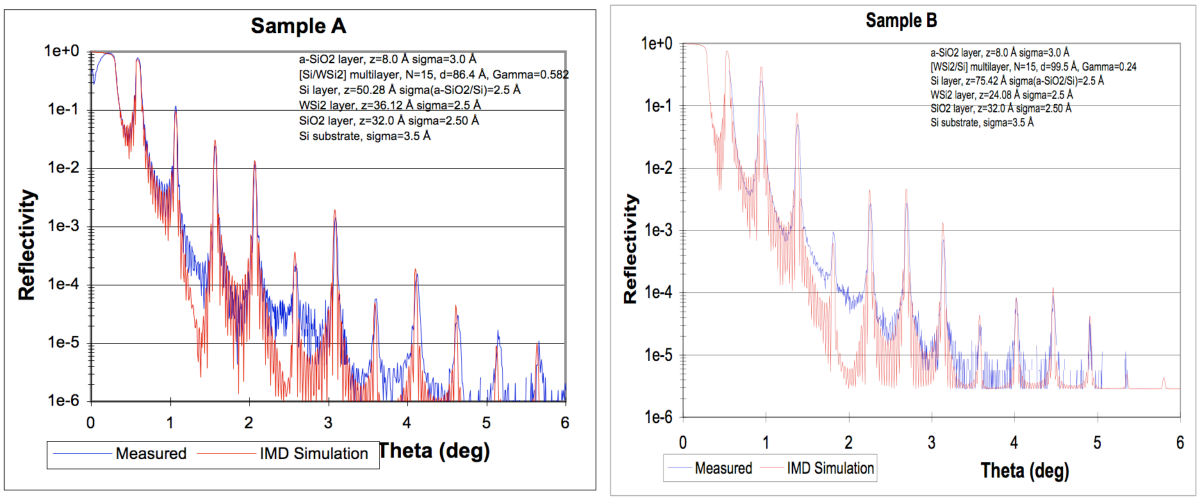

Fig. 1. Reflectivity of samples A and B at 8.048 keV together with the fitted data using IMD software. The model used in the fitting is shown in the insert.

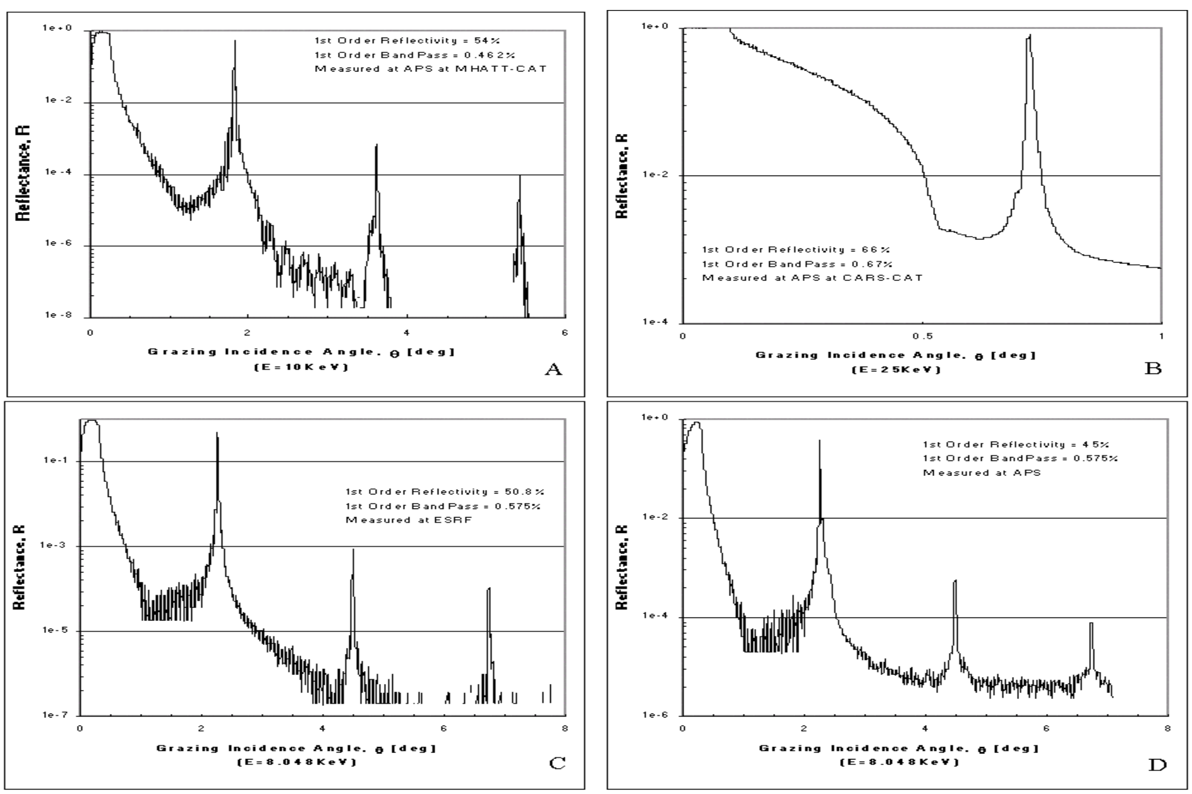

Fig. 2. Reflectivity scans for a [9.65Å-WSi_2/10.05Å-Si] × 300 multilayer carried out at: (a) the MHATT-CAT of the APS at 10 keV, b) the ChemMatCARS-CAT at 25 keV, (c) the ESRF x-ray lab, and (d) the APS x-ray lab.

An energy scan of this WSi_2/Si multilayer sample was also carried out at the ChemMatCARS-CAT beamline, as shown in Figure 3. The energy scan also shows a bandpass of 0.67%.

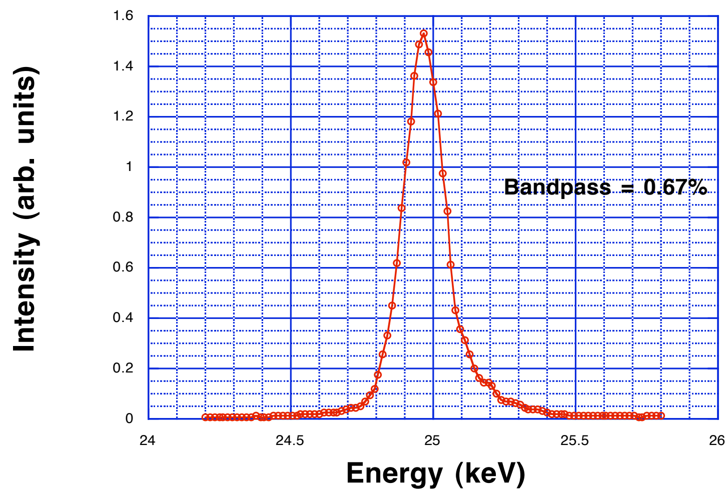

Fig. 3. An x-ray energy scan for a [9.65Å-WSi_2/10.05Å-Si] × 300 multilayer carried out at the ChemMatCARS-CAT.

These measurements show a trend of increased reflectivity with increased x-ray energy, in agreement with IMD simulations. For some reason, the bandpass measured at 10 keV is the lowest, which was also confirmed in IMD simulations. Figure 4 shows the reflectivity and bandpass for the WSi_2/Si multilayer at different energies using the IMD software. The reflectivity goes up with energy except a dip due to the W absorption edge near 10.24 keV. The reflectivity vs. energy curve of the WSi_2/Si multilayer has the same shape of the attenuation length vs. energy curve of W. The bandpass vs. energy curve shows oscillatory behavior for which we do not have an explanation at this time.

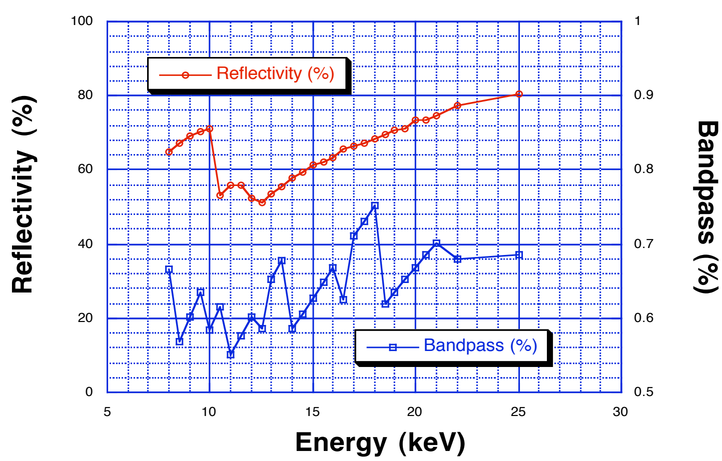

Fig. 4. IMD simulation of reflectivity and bandpass as a function of x-ray energy for a [9.65Å-WSi_2/10.05Å-Si] × 300 multilayer.

The relatively high reflectivity and narrow bandpass of the WSi_2/Si multilayer are very attractive for the design of a multilayer monochromator for the ChemMatCARS-CAT to be used in the 20 to 25 keV range. At these high energies, a larger N of more than 300 can also be applied to further increase the peak reflectivity and decrease the bandpass. WSi_2/Si multilayers have a better thermal stability than other multilayer systems such as W/Si. [9] A large N can be deposited with a stable structure.

3. DISCUSSIONS AND COMPARISON WITH W/SI MULTILAYERS

To compare WSi_2/Si with W/Si, a W/Si multilayer with similar layer thicknesses was grown on another superpolished Si substrate and measured in the APS x-ray lab. Figure 5 shows the reflectivity measurement data together with the IMD fitted data. The best fit gives a W thickness of 10.00 Å and Si thickness of 9.23 Å. A lower reflectivity of 36.2% and larger bandpass of 0.67% were obtained for this sample compared to the WSi_2/Si sample. One noticeable difference in the fitting parameters is that the interface roughness for W/Si is much higher than for WSi_2/Si (4.7 Å vs. 2.5 Å). This increased interface roughness and interface width might be the reason for the lower reflectivity and larger bandpass for the W/Si multilayer.

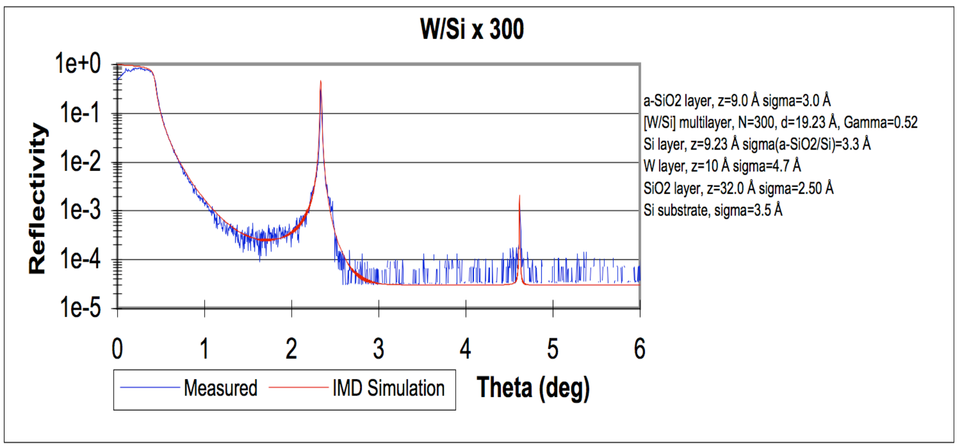

Fig. 5. Reflectivity of a W/Si multilayer at 8.048 keV together with the fitted data using IMD software. The model used in the fitting is shown on the left of the figure.

It is well known that multilayers consisting of chemically interacting materials (such as W/Si and Mo/Si) suffer more interdiffusion mixing between these materials and are less stable than multilayers consisting of noninteracting materials (such as WSi_2/Si and $MoSi_2/Si$). [10-11] The interdiffusion mixing in these multilayers is the dominant factor for interfacial imperfection. Our experiments demonstrate that WSi_2/Si multilayers have sharper interfaces than W/Si multilayers.

In the study of W/Si multilayer growth, we found that the film thicknesses are not scalable to the substrate moving speed (which determines the exposure time of the substrate to the sputtered flux), in agreement with the observation by Windt et al. [12] The nonlinearity of layer thickness with deposition time might also be attributed to the interdiffusion between W and Si. We found the nonlinearity is more pronounced in the small d-spacing multilayers ($d<3$ nm). While for WSi_2/Si multilayers, the growth rate can be scaled to the substrate moving speed more easily.

While WSi_2/Si multilayers have sharper interfaces than W/Si multilayers, we could not obtain a much smaller d-spacing than 19 Å without sacrificing reflectivity. When either WSi_2 or Si gets thinner than 8 Å , the reflectivity of a 300-bilayer WSi_2/Si multilayer decreases to below 30%.

Since Si oxidizes easily, it is important to have a very low oxygen content in the vacuum chamber. [13] To ensure a clean deposition, we pre-sputter the targets on a blank plate for 5 min at the beginning of a multilayer deposition with a base pressure better than 2×10^{-8} Torr in the vacuum chamber. The oxidization of Si can still be a serious problem for the WSi_2/Si multilayer system.

WSi_2/Si multilayers can be grown very fast and have a sharp interface. A very large number of bilayers can be grown with well-controlled layer thicknesses and structural stability. These properties make them promising candidates for other applications. Very recently, depth-graded WSi_2/Si multilayers with a zone plate structure have been made at the APS deposition lab for hard x-ray focusing applications. [14] A test multilayer has a total of 470 alternating layers with thickness gradually increasing from ~15 nm to ~60 nm. It took ~45 h to grow a total of ~11.27 μm multilayer on a Si substrate.[15] An one-dimensional hard x-ray focusing experiment with a linear zone-plate sectioned from this multilayer demonstrated good x-ray focusing capabilities.[16]

4. SUMMARY

WSi_2/Si is an attractive multilayer system for narrow-bandpass x-ray monochrometor applications. It can be grown with a high growth rate and has a sharp interface between layers. A bandpass of ~0.5% with reflectivity over 50% can be easily achievable with WSi_2/Si multilayers. WSi_2/Si is also a promising candidate for growing linear zone-plate multilayers. However, a low oxygen content in the vacuum chamber is required to grow these multilayers.

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