Specular and diffuse x-ray scattering from tungsten/carbon multilayers having a high reflectivity at 10 keV

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ABSTRACT

X-ray scattering measurements at 10 keV from multilayers having a period of 24.8 Å and consisting of 100 W/C bilayers are reported. Specular scans revealed first order reflectivities in the range 73.5% to 78.0% with bandpasses in the range of 1.5% to 1.7%. Total roughness (or interface grading) values deduced from fitting were in the range 2.5 to 3.0 Å for the last-to-grow surface of the W layers. Diffuse scattering measurements were made in a novel geometry that permitted investigation of in-plane momentum transfers up to 0.2 Å⁻¹. This is roughly an order of magnitude larger than is possible in conventional rocking scans. A power law dependence of the diffuse scattering after integration over a 'Brillouin zone' is found. The exponent of this power law, 1.75, when interpreted using a logarithmic correlation function leads to a value of 1.0 Å for the correlated roughness.

1. Introduction

Multilayer optics are useful as monochromators and analyzers for synchrotron radiation beamlines. The angular acceptance and energy bandpass fall into an entirely different class when compared to those available with crystal optics. Typical values for multilayers are one thousand times larger compared to crystals.¹ The angular acceptance and energy bandpass can also be tailored by growing either fewer layers for wider bandpasses or more for narrow ones. Largely because of their roughness continuous sputtered films are limited to a thickness slightly less than 10 Å and for this reason, Bragg angles of the first order reflection for hard x-rays are a few degrees at most. As part of a project to produce a cooled monochromator optic for the Cornell High Energy Synchrotron Source (CHESS)², a detailed study of the scattering properties of a set of W/C multilayers was made at the CHESS F3 beamline. Multilayer coatings were DC magnetron sputtered in the deposition facility of the Advanced Photon Source (APS).^{3,4}

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2. Samples and Specular Scattering

Multilayers were grown on two Si substrates. One was 2.5 mm thick with a surface roughness in the range 4 to 5 Å, and the other was 0.5 mm thick with a surface roughness of in the range 2 to 3 Å. Roughnesses were measured with a WYKO TOPO 3D/2D instrument (5x objective) at the APS. The basic diffraction properties were quite similar, as shown in the Table.

Table. Performance of W/C multilayers (10 keV)

substrate	first-order reflectivity	bandpass
2.5 mm thick Si	74%	1.7%
0.5 mm thick Si	78%	1.5%

Other results for the two substrates were not substantially different and we will not distinguish them subsequently, although all data presented below are for the thin substrate. A specular scan is shown in Fig. 1 together with two fits to the data made using the IMD software package⁵. To obtain the fits, the data were normalized to the incident flux as measured with an ion chamber without a sample in place and were convolved with a gaussian resolution function corresponding to the angular divergence (vertical) from a bending magnet at CHESS, namely, $\sigma = 0.0024$ deg. This value of the resolution function was corroborated in the fitting since larger values broadened the first and second-order peaks (especially as seen on plots where the intensity is plotted on a linear scale). A value of 9.2 Å was deduced for the W layer thickness and 15.7 Å for the C layer thickness. The interface profile function used for all interfaces was an error function (see ref. 5). The fits were quite sensitive to the roughness of the W layers (i.e., at the top of the W layers or, in other words, at the last-to-grow surface) and were insensitive to roughness of the amorphous carbon layers. For the fit shown in Fig. 1a, a value of 3 Å was assumed for all interfaces. Dramatically poorer fits resulted for W layer roughnesses less than 2.5 Å or larger than 3.0 Å. As shown in Fig. 1b, almost the same fit is obtained by assuming that the C layers are rougher (increased from 3 Å to 4.5 Å) and compensating the effect of this increased roughness with a slightly decreased roughness for the W layers (deceased from 3 Å to 2.5 Å). Consequently, our results are not as definitive regarding the roughness of the last-to-grow surface of the C layers.

3. Diffuse scattering

Conventionally, diffuse scattering experiments are performed by rocking the sample in the diffraction plane^{6,7} (i.e., about an axis that is normal to the diffraction plane, as shown in Fig. 2a), and this method has been applied to the study of W/C multilayers.^{8,9} However, because either the incident beam or the scattered beam becomes grazing, this method is limited to lateral momentum transfers of ~ 0.02 Å⁻¹ (a value that applies to our samples for the second order). Analyses near the total external

reflection angle are complicated because it becomes necessary to use the distorted wave Born approximation (DWBA) instead of the simpler Born approximation (BA).⁶ Also, the occurrence of multiple scattering in such rocking curves makes the use of the DWBA necessary.¹⁰ These complications can be avoided and momentum transfers roughly an order of magnitude larger can be investigated if one uses a scattering geometry whereby the sample is rocked out of the diffraction plane by an angle χ about an axis that lies in the diffraction plane and in the surface plane of the sample, as shown in Fig. 2b. We note that for the case of diffuse scattering studies done by taking rocking curves (see Fig. 2a) in a vertical scattering plane, slits for the horizontal direction are typically kept wide open so that one must integrate over q_y before assessing the power law dependence. This integral results in a change of unity in the power law deduced from the data. However, for the presently used geometry the reciprocal space directions are reversed. Since the resolution in q_x in our case is very good (0.00006 deg), we have a point-like resolution and the power law dependence can be assessed directly from the data.

Scanning in reciprocal space was done with SPEC software¹¹ using the relations listed in the Appendix. We note that the angle of incidence, α_{inc} , and the angle of scattering, α_{scatt} , (both defined with respect to the plane of the surface) were kept constant during H (i.e., q_y) scans. This geometry is different than that used by Salditt et al.^{12,13} for W/Si multilayers to measure scattering at large in-plane momentum transfers. These workers used a specialized goniometer to rotate a vertical position-sensitive-detector azimuthally with the multilayer kept stationary. L (i.e., q_z) scanning in that case was accomplished by reading out the position dependent detector readings. L scanning in our case was accomplished by a conventional θ - 2θ scanning procedure where θ and 2θ are defined within the diffraction plane. These scans were always done for a symmetrical arrangement, i.e., the scattering angle, (2θ), was always equal to twice the θ angle. Our geometry was implemented using a conventional Eulerian cradle. The resolution volume was calculated and also measured. At low χ 's (i.e., at low H 's), L resolution was determined by the vertical divergence from the bending magnet, which was characterized by a gaussian with a value of 0.0024 deg for σ as discussed above. At low χ 's, H resolution was determined by the horizontal divergence and by the slit settings, and we calculate a value of $0.009 \text{ \AA}^{-1}$. This value is consistent with a measured value of $0.01 \text{ \AA}^{-1}$ obtained from H scans. A clear change from a specular scattering regime to a diffuse scattering regime was visible in H scans at $H = 0.01 \text{ \AA}^{-1}$. At large χ 's, L and H resolutions become interchanged from those at low χ 's.

L scans at various H 's were performed for both first and second orders. The first order data contained a feature due to multiple scattering, and we concentrated instead on the second order. Data for the second order are shown in Fig. 3. Diffuse scattering from correlated interface roughness was observed between H values from $0.02 \text{ \AA}^{-1}$ and $\sim 0.18 \text{ \AA}^{-1}$. Diffuse scattering at larger H values was also observed. The origin of this scattering has not been determined, but may be due to uncorrelated interface roughness. Another possibility is scattering from correlated charge density fluctuations with repeat spacings less than the multilayer period as suggested by Salditt et al.¹⁴. The data were fit to a Lorentzian function for the correlated part plus a linear term to represent the remaining diffuse scattering. An example fit is shown in Fig. 4. As proposed by Salditt et al.¹², the integral of these Lorentzians over one Brillouin zone (i.e., from the middle

point between the first and second orders to the middle point between the second and third orders, which in our case corresponds to $0.376 \text{ \AA}^{-1} < L < 0.626 \text{ \AA}^{-1}$) is the appropriate way to test for a power law dependence, which can then be used to infer a value for the correlated roughness. Our results are shown in Fig. 5, and do indeed reveal a power law dependence. The exponent of the power law is 1.75, and following Ref. 12, we equate this exponent to $2 - (q_z \sigma)^2$. With the value $q_z = 0.50 \text{ \AA}^{-1}$, which corresponds to the second order for our multilayer, we obtain a value of $1.0 \text{ \AA}^{-1}$ for the correlated roughness. We note that this value is less than the total roughnesses used for fitting to the specular scans. The difference is either due to the uncorrelated physical roughness or to a grading in the x-ray optical density which reduces the reflectivity just as actual physical roughness would¹⁵. We note that this inability to distinguish grading (e.g., from an interdiffused region) from physical roughness in its effect on specular reflectivity data allows for an explanation of an alternating roughness over a multilayer period based on such grading.

The situation regarding published reports of interfacial roughnesses for the W layers (i.e., C on W) and for the C layers (i.e., W on C) was summarized by Savage et al.⁸ in a report which dates to 1992. The conclusions of various groups differ. Petford-Long et al. (see Ref. 8) using high-resolution cross-section transmission microscopy on ion-beam deposited multilayers found evidence that the W on C interface was rougher than the C on W interface, in agreement with our findings. However, other workers using *in situ* ellipsometry and grazing incidence x-ray diffraction on rf-sputter deposited films report that the W on C interface is abrupt while the C on W one is diffuse over $4 \text{ \AA}$ ¹⁶. Savage et al.⁸ report good fits to their x-ray data taken at 8 keV for differences of only about 20%. Our samples were prepared in the same manner employed by Savage et al. employed, i.e., by dc magnetron sputtering. They also report that the presence of a $500 \text{ \AA}$ sputtered C buffer layer greatly increases both the correlated as well as the total roughness. This implies that thick C layers do indeed produce a roughened effect. Based on their studies, they suggest that interfaces can affect the evolution of surface roughness.⁸

4. Summary

Tungsten/ carbon multilayers with high reflectivities of 73.5% and 78% and with bandpasses of 1.5% and 1.7% have been grown in the deposition facility located at the Advanced Photon Source. Total roughnesses in the range $2.5\text{--}3.0 \text{ \AA}$ for the amorphous W layers (i.e., at the top or, in other words, the last-to-grow surface of the W layers) were obtained from fits to specular diffraction data. Because of the insensitivity to the roughness of the C layers, a similarly definitive value for the roughness for the C layers was not inferred. Diffuse scattering data were obtained in a novel geometry implemented using a conventional Eulerian cradle out to in-plane momentum transfers of $0.2 \text{ \AA}^{-1}$. The diffuse scattering data revealed a power law dependence for the scattering integrated over one Brillouin zone of the multilayer. From these data, a value of $\sigma = 1.0 \text{ \AA}$ was inferred for the correlated roughness by invoking a logarithmic correlation function. These results indicate that the correlated interface roughness present in these multilayers is significantly less than the total roughness.

5. Acknowledgement

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6. Appendix

Summary of formulas used by SPEC:

$$\sin\alpha_{\text{inc}} = \sin\theta \cos\chi$$

$$\sin\alpha_{\text{scatt}} = \sin((2\theta)-\theta) \cos\chi$$

$$L = q_z = \frac{2\pi}{\lambda} (\sin\theta + \sin((2\theta) - \theta)) \cos\chi$$

$$K = q_x = \frac{2\pi}{\lambda} (\cos\theta - \cos((2\theta) - \theta))$$

$$H = q_y = \frac{2\pi}{\lambda} (\sin\theta + \sin((2\theta) - \theta)) \sin\chi$$

7. Figure Captions

Fig. 1. Specular (q_z) scan at 10 keV for the 100-period W/C multilayer showing the total external reflection region as well as both first- and second-order peaks. The same data are shown as filled symbols in both (a) and (b). Fits are shown as lines. The fit in (a) was made for the following parameters: bilayer period=24.85Å, W layer thickness= 9.19Å, C layer thickness=15.66Å, Si substrate roughness= 3.0Å, roughness of the W layers (the last-to-grow surface) = 3.0Å, roughness of the C layers (last-to-grow surface) = 3.0Å. The fit in (b) was made with the same parameters as in (a) but with the roughness of the W layers = 2.5Å, and the roughness of the C layers = 4.5Å.

Fig.2. Diffractometer arrangements used to measure diffuse scattering. (a) Conventional rocking method. The view shown is a side view of the diffraction plane in which the sample is rocked. q_x is the component of the in-plane momentum transfer that is measured. The range that is accessible is limited because either the incident beam, k_{inc} , or the scattered beam, k_{scatt} , quickly reaches a grazing condition. (b) The geometry used currently. The diffraction plane is viewed end-on, and the multilayer is rotated by an angle χ using a Eulerian cradle.

Fig.3 L (i.e., q_z) scan data for H (i.e., q_y) values of 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 0.08, 0.10, 0.12, 0.16, 0.18, 0.19 Å⁻¹. For clarity the data points are not shown but the lines connecting them are. The peak intensity decreased monotonically with increasing H. The fact that specular scattering is included for H=0.01 Å⁻¹ but not for H=0.02 Å⁻¹ corroborates our separate L resolution measurement of 0.01 Å⁻¹. Scattering from correlated interface roughness is no longer observed at H=0.19 Å⁻¹.

Fig.4. L scan for H=0.08 Å⁻¹. A fitted function comprised of a Lorentzian on top of a line is also shown.

Fig. 5. The area of the Lorentzian fits over one Brillouin zone around the second order as function of H. A power law decay of the amplitude is evident up to 0.1 Å⁻¹. The value of the exponent is 1.75.

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Fig. 1

(a)

(b)

Fig. 2

APS W/C MULTILAYER

2nd order

Fig. 3

Fig. 4

Fig. 5