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Article

Journal of
Nanoscience and Nanotechnology
Vol. 19, 609–612, 2019
www.aspbs.com/jnn

Cr/C Reflective Multilayer for Wavelength of 44.8 Å

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The working wavelength of Ni-like, Ta soft X-ray laser is 44.8 Å, just near the "water window." High reflection multilayers are required for this kind of laser in China. In this work, we design and fabricate carbon-based multilayer reflective samples. The Cr/C multilayer was selected from proposed candidates such as Co/C, Ni/C, and CoCr/C material combinations. The period thickness is only 22.6 Å. Cr/C multilayers were deposited by the magnetron sputtering method. Multilayers with bi-layer numbers of 150, 200, 250 and 300 were deposited onto super polished silicon wafers. All multilayers have been characterized by grazing incidence X-ray reflectance (GIXRR). Then, near-normal incidence reflectance measurements were performed at beamline 3W1B, Beijing synchrotron radiation (BSRF). The highest reflectance of 13.2% is achieved with the bi-layer number of 300. Transmission electron microscopy measurements also clearly show the sharp Cr–C interfaces in the multilayer.

Keywords: Soft X-ray Laser, Multilayer, Magnetron Sputtering, Reflectance, Synchrotron Radiation.

1. INTRODUCTION

The spectral region in the longer side of the carbon K-edge (wavelength $\lambda \sim 43.6$ Å) holds several unique applications. It is the best wavelength choice for soft X-ray holography of carbon containing samples, and provides an opportunity for high contrast soft X-ray microscopy of biological specimens, ascribable to the highest transparency of carbon over all of the soft X-ray range.[1–5] Nickel (Ni)-like tantalum (Ta) soft X-ray lasers, which have been successfully demonstrated operating at 44.8 Å,[7–8] provide a possible source for these applications. High performance optical elements are demanded for these lasing experiments and applications. The common requirements for these applications are high spectral selectivity, good thermal stability and, most importantly, high reflectance. Multilayer (ML)-based reflective optics in the extreme ultraviolet (EUV) and soft X-ray regions have significantly progressed in the past three decades,[9–12] which advances applications such as EUV lithography, X-ray astronomy, soft X-ray microscopy, X-ray diagnostics of high-temperature plasma and synchrotron instrumentation. At wavelength $\lambda \sim 45$ Å, state-of-the-art multilayers based on Co/C, Cr/C and Fe/C coatings have achieved normal-incidence reflectance of 14.3%, 12.2% and 13.0%,[3, 13, 14] respectively. Nevertheless, this reflectance is still three times lower than the theoretical prediction. The main factors limiting the reflectance of these multilayers are connected with the short period thickness D and the large bi-layer number N needed for reflectance saturation at this short wavelength. Effects of interface defects and period fluctuation become more critical when period thickness D is reduced, while multilayers with large bi-layer number suffer from roughness growth by roughness replicating and layer thickness drifts relevant to long deposition times. Thus, increasing the bi-layer number may not provide enhanced reflectance for short period multilayers.

In this paper, Cr/C multilayers with various bi-layer numbers (N) of 150, 200, 250 and 300 were deposited by the magnetron sputtering technique. Grazing incidence X-ray reflectance (GIXRR), near-normal incidence soft X-ray reflectance (SXR) and transmission electron microscopy (TEM) were used to investigate interface roughness growth and layer thickness drift due to the increasing bi-layer number.

2. DESIGN AND FABRICATION OF THE MULTILAYERS

Carbon can be a natural spacing material due to its low absorption at wavelengths little longer than the K absorption edge, while theoretically $3d$-group transition metals

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can be selected as the scattering layer material to achieve high reflectivity. In 1992, Niibe et al.[15] fabricated and characterized Cr/C multilayer achieving the reflectivity of 7% at the incidence angle of 12°; Andreev et al.[13] demonstrated Cr/C, Co/C and Fe/C multilayers in 2000 and achieved reflectivities of 12.2%, 11.2% and 13.0% respectively; Artyukov et al.[3,14] achieved reflectivity of 14.3% by Co/C multilayer at normal incidence. In China, research on multilayers at wavelengths near the "water window" are also in process. Since 2006, Tongji University has been working on different multilayer material combinations and achieved reflectivity of 7.5% using a Cr/C multilayer at near-normal incidence.[16]

In this paper, we designed and fabricated Cr/C high-reflectivity multilayers working at 44.8 Å. All multilayers were designed with the same stack structure except for the bi-layer number. They were tuned to $\lambda = 44.8$ Å at 5° incident angle, leading to a period of $D = 22.6$ Å, thickness ratio of $\Gamma_C = 0.6$ (carbon layer thickness to period thickness, $\Gamma_C = d_C/D$) and saturated bi-layer number of $N_{\text{sat}} = 300$. In order to study the influence of bi-layer number N on the reflectivity, we deposited four Cr/C multilayers with bi-layer numbers $N = 150$, 200, 250 and 300, respectively. The optical constants are from the CXRO website.[17]

All multilayers were deposited by an ultrahigh vacuum, direct current magnetron sputtering system. The vacuum chamber was evacuated by a turbo-molecular pump, and the base pressure was 5.0E–5 Pa. The sputter gas was argon (99.999% purity) at a constant pressure of 1.0 mTorr (0.133 Pa). Circular targets with 100 mm diameter were used. The cathodes were operated in regulated power mode, with 20 W and 120 W applied to Cr (99.95% purity) and C (99.999% purity) targets respectively. All multilayers were deposited onto sliced polished silicon (100) wafer (sliced to size of 30×40 mm^2), the RMS (root mean square) roughness of which is about 3 Å. The substrate was mounted 80 mm above the target. The deposition rate was preliminarily calibrated to be 1.2 Å/s and 0.4 Å/s for Cr and C, respectively. Individual layer thicknesses were adjusted by varying the exposure time of the substrate as it stays over each magnetron cathode.

3. CHARACTERIZATION OF MULTILAYERS

The structural analysis of multilayers was investigated by grazing incidence X-ray reflectivity (GIXRR) using a 4-circle X-ray diffractometer having a sealed Cu X-ray tube and a Si (220) crystal monochromator tuned to the Cu K-α line ($\lambda = 0.154$ nm, $E = 8.05$ keV). The angular divergence of this system was estimated to be ~0.007°. Figure 1 shows the measured results of four multilayers with different bi-layer number N. For the convenience of comparison, the reflection curves were, in turn, translated vertically by three orders of magnitude. All curves show sharp and intense Bragg peaks up to 4° of grazing incidence angle, which indicated good interfaces with small

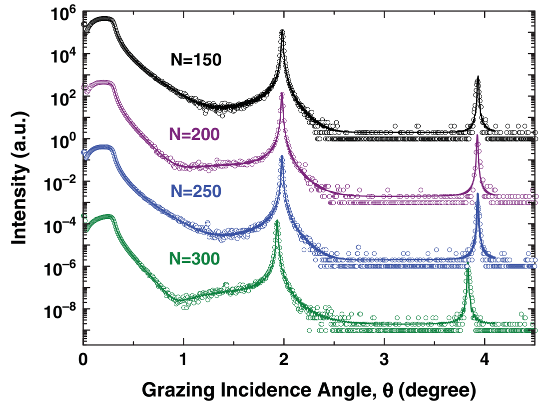

Figure 1. Grazing incidence X-ray reflectance of multilayer samples with different bi-layer number N: Dots for measured data, solid line for fitted curves.

roughness or inter-diffusion and confirmed great stability during fabrication. The measured GIXRR curves have been fitted in Bede Refs software using a "differential evolution" algorithm.[18] The individual layer thickness d, layer density ρ (in % of bulk density) and interface width σ are the fitting parameters. In the fitting procedure, the first layer of carbon near air ambience has been separated from the layer stack as independent variable, in consideration of surface oxidation or contamination. The fitted curves were also shown in Figure 1 as solid lines. The fitted period thicknesses were listed in Table I. The interface width of four samples is about 3 Å, and no perceptible roughness growth exists with increasing bi-layer number. The Cr layer and C layer densities are about 85–90% of bulk density, which is typical in sputtered films. Comparing to the fitted curves, all measured ones have a little broader Bragg peaks, which is more evident in the second order peak. This suggests a small deviation from a uniformly periodic stack, but it cannot be determined if it is due to random fluctuation or monotonic drift from the GIXRR data.

The near-normal incidence reflectance measurements were performed at beamline 3W1B, Beijing synchrotron radiation facility (BSRF).[19] The measured reflectivity curves of multilayers with different bi-layer numbers were shown in Figure 2. Detailed data is given in Table II. The peak reflectivities of $N = 150$, 200, 250 and 300 samples are 9.1%, 10.3%, 12.4% and 13.2%, respectively. The reflectivity increasing with increased bi-layer number, indicates no significant roughness growth. All peaks

Table I. Fitted multilayer period thicknesses from GIXRR measurements shown in Figure 1 and synchrotron radiation (SR) measurements shown in Figure 2.

N (Å)	150	200	250	300
D_{GIXRR}	22.55	22.59	22.54	23.15
D_{SR}	22.65	22.68	22.60	23.17

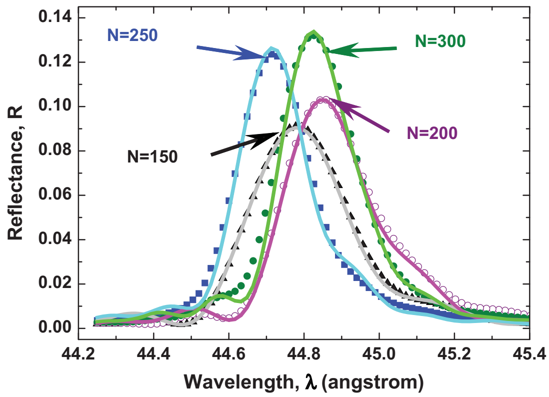

Figure 2. Near-normal incidence reflectance of Cr/C multilayer samples with different bi-layer numbers N: Dots for measured data, solid line for fitted curves.

of the four curves exhibit common characteristics of a rising hump at the long wavelength side and oscillation at the short wavelength side. This is an evident clue for an increasing period thickness from substrate to air ambience. Taking into consideration this period thickness drift as fitting parameter, we performed fitting on the SXR data employing Levenberg-Marquardt algorithm[20] and the best-fit parameter from GIXRR as initial value. The fitted curves are solid lines shown in Figure 2, and the fitting period thicknesses D_{SR} were also listed in Table I, consisting with the GIXRR fitting D_{GIXRR}. The period thickness drift ΔD is also obtained in the fitting. The drift values for $N = 150$, 200, 250 and 300 were 0.07, 0.08, 0.08 and 0.11 Å, respectively. It was shown that period thickness fluctuation should not exceed $\Delta D/D \sim 1/N$, which gives $\Delta D < 0.08$ Å for $N = 300$. Thus, in the $N = 300$ case, period thickness drift becomes an important drawback in the pursuit of high reflectivity.

Figure 3 shows the cross-sectional TEM image and selected-area electron diffraction (SAED) pattern (inset) of the $N = 300$ sample. The multilayer modulation was clearly observed at the upper right part of the micrograph. The bright line represents C layers while the dark one represents the Cr layers. The quality of the multilayer appears to be fairly good. The interfaces of Cr and C are flat and abrupt. There is no indication of increasing interface roughness or columnar structure. The bright line between the multilayer (ML) and Si substrate (lower left corner) shows the native oxide of the Si wafer, the thickness of which was deduced to be about 4.8 nm from the TEM

Table II. Detailed data from SR measurement shown in Figure 2.

N	Reflectivity (%)	Peak position (Å)	FWHM (Å)
150	9.1	44.78	0.292
200	10.3	44.84	0.260
250	12.6	44.71	0.210
300	13.2	44.83	0.243

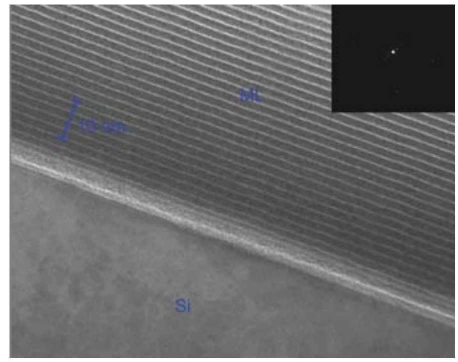

Figure 3. Cross-sectional TEM image and SAED pattern (inset) of the Cr/C multilayer with $N = 300$.

image. The SAED pattern in the inset exhibits only the Si reflection and multilayer reflection, no crystalline phase of Cr or C was indicated. The amorphous nature of Mo/Si multilayers has also been reported in previous research,[21] confirming the slim possibility of crystallization in such short period metal film.

4. CONCLUSIONS

We have successfully fabricated Cr/C high-reflectivity multilayers with bi-layer number of 150, 200, 250, and 300 at wavelength of 44.8 Å. All the multilayers were deposited by the DC magnetron sputtering method. These multilayers were characterized by grazing incidence X-ray reflectance, soft X-ray reflectance and transmission electron microscopy. The results show that the interface roughness of Cr/C multilayers does not grow with increasing bi-layer number, while the period thickness drift scales with bi-layer number. TEM clearly shows the sharp Cr–C interfaces in the multilayer. All multilayers exhibit good quality and marvelous optical performance. The highest reflectance of 13.2% was obtained by the 300 bi-layer sample.

Acknowledgments: The authors gratefully acknowledge Dr. Franz Schafers for his kindly assistance for the measurement at BESSY synchrotron facility for our calibration measurement. The authors are indebted to Professor Zhanshan Wang in Tongji University. This work was supported by the National Natural Science Foundation of China (contract numbers 11375131 and U1432244), by the Royal Society—NSFC International Exchanges funding (reference IE141043 and 11511130051) and the Fundamental Research Funds for Central Universities.

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Received: 14 May 2017. Accepted: 9 February 2018.