14.5% near-normal incidence reflectance of Cr/Sc x-ray multilayer mirrors for the water window

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Cr/Sc multilayer mirrors, synthesized by ion-assisted magnetron sputter deposition, are proved to have a high near-normal reflectivity of $R = 14.5\%$ at a grazing angle of 87.5° measured at the wavelength $\lambda = 3.11$ nm, which is an improvement of more than 31% compared with previously published results. Elastic recoil detection analyses show that the mirrors contained as much as 15 at. % of N and traces of C and O. Soft x-ray reflectivity simulations reveal interface widths of $\sigma = 0.34$ nm and an exceptionally small layer thickness drift of $\sim 1.6 \times 10^{-5}$ nm/multilayer period throughout the stack. Simulations show that a reflectivity of $R = 25.6\%$ is attainable if impurities and layer thickness drift can be eliminated. The abrupt interfaces are achieved with ion assistance with a low ion energy of 24 eV and high ion-to-metal flux ratios of 7.1 and 23.1 during Cr and Sc sputter deposition, respectively. In addition, a near-normal incidence reflectivity of 5.5% for the C VI emission line ($\lambda = 3.374$ nm) from a laser plasma source was verified.

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Much effort is being put into realizing highly reflective mirror optics for the water window wavelength range (2.4–4.4 nm).[1–5] The driving force is prospects of high-resolution microscopy,[6,7] deep-space telescopy,[8] new optics for free-electron x-ray lasers,[9] time-resolved x-ray spectroscopy,[10] wavelength-dispersive detection of light-element x-ray emission,[11] and soft x-ray polarimetry.[12]

The maximal theoretical normal-incidence reflectivity,[13] R, in the water window is 64%, which should occur as a sharp peak at the Sc $2p$ absorption edge (photon energy of $h\nu = 398.8$ eV and wavelength of $\lambda = 3.11$ nm) for a Cr/Sc multilayer mirror consisting of more than 600 bilayers with individual layer thicknesses of $d_{\mathrm{Cr}} = 0.59$ nm and $d_{\mathrm{Sc}} = 0.97$ nm. However, for practical reasons the results presented in this Letter are obtained a few degrees below normal (near-normal) incidence. The highest reported near-normal incidence reflectivity is, until now, only 11%.[5]

The progress in making normal-incidence multilayer mirrors for the water window is thus not as rapid as it has been for the extreme ultraviolet range, where reflectivities approaching the theoretical limit now are reported.[14] The reason is that several physical limitations come into play as the x-ray regime is approached; the most important limitation is the extremely high sensitivity to interface imperfections that is associated with the short repetition periods required. For example, an interface width, σ, of only 0.3 nm decreases the theoretical reflectivity from 64% to 31.7%, and a σ of 0.5 nm yields 2.6% reflectivity.[15] Another limitation is the long-term stability of the deposition process that is required to deposit several hundreds of consecutive periods without layer thickness variations on the 0.01-nm scale. In addition, since the maximal reflectivity in the water window is achieved at the absorption edge of one of the constituting ($3d$ transition) elements, the maximal reflectivity is achieved within only a narrow bandwidth, and the multilayer must have a period, Λ, that exactly matches the absorption edge with an absolute accuracy of ~ 0.01 nm. A practical drawback of this feature is that only tunable synchrotron-based instrumentation can utilize the maximal performance of such mirrors. Instrumentation based on line sources, e.g., a laser-produced plasma (LPP) x-ray source,[6,7] therefore provides additional challenges in

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producing suitable mirror optics. For example, the line nature of the x-ray emission makes it impossible to match the radiation to the maximal obtainable mirror reflectivity at an absorption edge, and it also places high demands on the absolute accuracy of the period Λ. The highest reported near-normal peak reflectivity for any atomic emission line in the water window is only 5.5%.[2]

We previously reported on a sputter deposition process for Cr/Sc multilayer mirrors that reduced both high and low lateral spatial frequency interface roughness as well as a transverse accumulated roughness through high-flux low-energy ion assistance during growth.[2] The process utilizes the fact that a high flux of ions with kinetic energies lower than the bulk displacement energies of the multilayer constituents stimulate ad-atom mobility without recoil mixing.[16]

The highest structural order of the multilayers were obtained with a kinetic energy of the Ar ions of 24 eV and ion-to-metal flux ratios of 7.1 and 23.1 for Cr and Sc, respectively. Characterization of the mirrors using a reflectometer based on a LPP C VI line ($\lambda = 3.374$ nm) source[17] showed a reflectivity of 5.5%. Simulations[13] of the reflectivity data implied interface widths of only 0.425 nm,[2] which in turn predicted that a normal-incidence reflectivity of 14.6% should be achievable for mirrors optimized for the Sc $2p$ absorption edge.[15] This would correspond to an increase in reflectivity of >30% as compared with previously published data.[5]

However, the theoretical predictions are based on a few assumptions that must be investigated. First, because experimental values of the optical constants for Sc in the water window are not available, the simulations have been based on an estimate using the density and atomic number,[18] which may give an error in absolute reflectivity. Second, impurities such as C, N, and O incorporated during the deposition process lead to a dilution of the layer materials, with a smaller optical contrast and increased absorption as a consequence. Third, the simulations were based on the assumption that the multilayers had a constant period throughout the multilayer stack. However, more advanced simulations indicate that a drift of the deposition rates, causing a change in the period as small as $<1 \times 10^{-5}$ nm/period, will significantly reduce and alter the shape of the reflectivity peak. This, in turn, may have caused an overestimation of the interface widths. Even magnetron sputter sources, which are known to be extremely stable, have such small drifts due to the continuously changing magnetic field in the erosion tracks. The tunability of synchrotron radiation also makes it possible to check the accuracy of the LPP-based reflectometer that gave input data for our predictions.[15] To prove the capabilities of our deposition technique and to investigate the possible influence of the issues mentioned above, it is thus essential to experimentally verify the predicted performance for the Sc $2p$ absorption edge ($\lambda = 3.11$ nm) using synchrotron radiation.

In this Letter we report experimental evidence of the performance, measured by synchrotron radiation, of two specifically designed Cr/Sc multilayer mirrors, one with $N = 600$ bilayers and a period ($\Lambda = d_{\mathrm{Cr}} + d_{\mathrm{Sc}}$) of $\Lambda = 1.56$ nm that is designed for near-normal reflection of the wavelength $\lambda = 3.11$ nm (Sc $2p$ absorption edge) and one with $N = 400$ bilayers and a period of $\Lambda = 1.75$ nm that is optimized for $\lambda = 3.374$ nm (C VI emission line). Both multilayers have a nominal layer thickness ratio of $\Gamma = d_{\mathrm{Cr}}/\Lambda = 0.47$. The latter multilayer was made to verify the absolute calibration of our LPP-based reflectometer. Using elastic recoil detection analyses it was found that the multilayers contained a significant amount of impurities: 15 at.% of N, 3 at.% of O, and 0.5 at.% of C.

Soft x-ray reflectivity measurements were performed with synchrotron radiation at the Calibration and Standards bending magnet beamline 6.3.2 at the Advanced Light Source (ALS).[19] High spectral resolution was obtained using a varied-line-spacing plane-grating monochromator, and the reflectivity was monitored by a CCD detector. The reflectivity of the multilayer containing 600 periods was measured across the Sc absorption edge, as shown in Fig. 1. The peak reflectivity, which is 14.5% at an incidence angle of 87.5°, is more than 31% higher than any previously reported data[1,5,20–22] and verifies the predicted performance of 14.6%, which was based on LPP-based reflectometry data of other samples using the C VI emission line.[2,15] The spectral resolution, $\Delta\lambda/\lambda$, is only 3.6×10^{-3}, showing that the mirror has minimal layer thickness fluctuations.

However, the small peak on the short-wavelength flank (marked by *) indicates that the multilayer is not quite perfect. It is possible to assign such an extra peak to a constant drift in the material fluxes during the deposition. The dashed curve in Fig. 1 shows a simulation of the synchrotron data using the IMD software,[13] where a drift of $\delta = 1.6 \times 10^{-5}$ nm per bilayer thickness and the composition determined by elastic recall detection analysis was taken into account along with an interface width of 0.34 nm. The fit to the experimental curve is very good, and the extra peak is qualitatively reproduced. Although such a small drift might seem negligible, it clearly influences the reflectance of the multilayers. The dotted curve in Fig. 1 shows a simulation assuming no impurities and no drift using the same interface width. As can be seen, the reflectivity would increase to $R = 25.6\%$, i.e., a considerable increase of 76.6%.

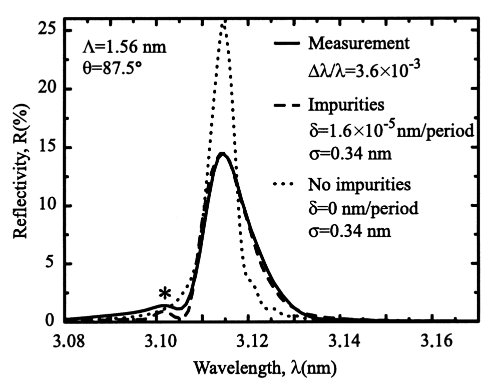

Fig. 1. Measured soft x-ray reflectivity of a Cr/Sc multilayer at the Sc absorption edge.

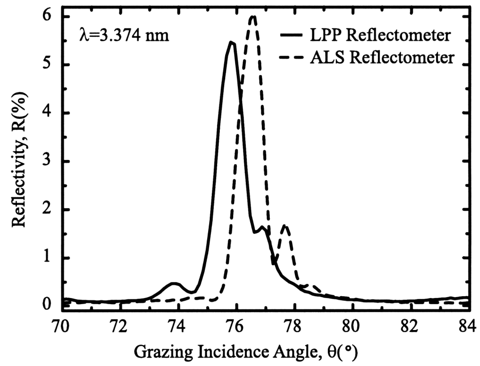

Fig. 2. Soft x-ray reflectivity measurements of a multilayer for a fixed wavelength, $\lambda = 3.374$ nm, for the LPP-based reflectometer utilizing the C VI emission line (solid curve) and for the synchrotron-based reflectometer at the ALS (dashed curve).

The at-wavelength reflectometer used for our predicted performance[15] is based on a high-brightness line-emitting LPP source utilizing an ethanol liquid-jet target emitting mainly the C VI emission line at $\lambda = 3.374$ nm.[17,23] Absolute reflectivity measurements were performed by using a Cr/Sc multilayer with known reflectivity as a calibration standard. A multilayer mirror, designed as a condenser mirror for an LPP-based microscope, containing 400 bilayers with a period of $\Lambda = 1.75$ nm, was investigated using both at-wavelength (measured with the LPP-based instrument) and synchrotron radiation reflectivity measurements in order to verify the LPP reflectometer accuracy. The results of these measurements are shown in Fig. 2.

As seen in the figure, the at-wavelength reflectivity is 5.5% at a grazing incidence angle $\theta = 75.8°$, whereas the synchrotron radiation reflectivity is 6.1% at $\theta = 76.6°$. The difference in reflecting angle ($\Delta\theta = 0.8°$) between the two measurements is due to a slight difference in the multilayer period ($\Delta\Lambda = 0.0061$ nm) between the measured spots on the sample. The fringes appearing on each side of the main Bragg peak in both measurements are caused by the continuous drift of the deposition rate mentioned earlier. The measured difference in the absolute reflectivity is 0.6%. However, it should be noted that the absolute value of the reflectivity may depend on the optical setup, the energy as well as the angular spread of the incoming beam, etc. We therefore conclude that the synchrotron measurement, to a relative accuracy $\Delta R/R$ of 0.1, confirms our previous LPP measurements.

In conclusion, we have used high-flux, low-energy ion-assisted sputter deposition to make Cr/Sc multilayer x-ray mirrors with minimal roughness and layer thickness fluctuations in combination with very abrupt interfaces. The mirrors exhibit high spectral resolution, $\Delta\lambda/\lambda = 3.6 \times 10^{-3}$, and, although far from the theoretical limit, the soft x-ray near-normal reflectance measured at the Sc $2p$ absorption edge exhibits an excellent value of $R = 14.5\%$, which is in very good agreement with predicted performance.[15]

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