Soft x-ray (97-eV) phase retardation using transmission multilayers

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(Received 18 November 1991; accepted for publication 30 March 1992)

Phase retardation as a function of incidence angle of 97-eV soft x rays from a laser plasma source on transmission through a free-standing molybdenum/silicon multilayer was measured using a multilayer polarizer and a polarization analyzer. The maximum retardation of 49° between $\sigma$ and $\pi$ components is over 2/3 that calculated for an ideal structure. At maximum retardation the transmittance ratio of $\sigma$- to $\pi$-amplitudes was 0.66 and the intensity transmittance, averaged for both components, was 20%. These multilayer structures will be useful in soft x-ray polarization applications.

Experimental control of the polarization state of soft x-ray beams can be obtained with optical elements such as linear polarizers and phase retarders. Together such elements can generate beams with specific polarization states (making sources of circularly polarized photons unnecessary), and analyze complex polarization states produced by, for example, sample interaction. The nature of the optical constants of materials in the soft x-ray range ($n \approx 1$ for all materials) has hampered the realization of optics for polarization control. Practical linear polarizers based on the polarization dependence of Thompson (charge) scattering have been demonstrated in the form of multilayer reflectors having reflectance maxima at a 90° scattering angle. Phased retarders using the relative phase change accumulated during multiple total reflections have been demonstrated at energies up to 30 eV in the extreme ultraviolet, though significant retardation becomes more difficult to achieve as photon energy increases.

Calculations have shown that soft x-ray multilayer structures can produce useful values of phase retardation in both specular reflection and transmission geometries. This birefringence and resulting phase retardation originates from the unequal response of $\sigma$ and $\pi$ field components to charge scattering when the scattering vector is near the multilayer Bragg interference condition, and is especially large when this condition is satisfied near 45° incidence angle (total scattering angle near 90°), where the two components have the largest difference in scattering cross section. At these conditions the $\sigma$-field component exists as strong standing wave whose position with respect to the multilayer unit cell varies rapidly with angle across the Bragg peak, resulting in a strong resonance in the effective index of refraction experienced by the $\sigma$ field. The $\pi$-field component experiences only weak scattering near 90° scattering angle and hence weak modulation in effective refractive index. The difference in phase between the two components (retardation) can be significant in both the reflection and transmission cases. The predicted phase retardation in the reflection geometry has recently been demonstrated at 97 eV. In the present letter, measurements of phase retardation in the transmission geometry are reported.

Free-standing transmission structures were obtained by sputter depositing multilayers onto photosensitive coated Si wafers. A rigid frame was then bonded to the free surface of the multilayer and the photoresist was dissolved, leaving a free-standing multilayer supported by the frame. Some surface contamination was left on the multilayer. The free-standing multilayers thus obtained are not perfectly flat, though portions within the roughly 2-mm² area had slope errors small compared to the angular widths of the interference features of interest, enabling high-resolution measurements of these features to be made. The multilayer studied here consisted of Mo/Si bilayers repeated for 20 periods with each bilayer 8.75 nm thick and with the Mo layer comprising roughly 1/3 of each bilayer.

Molybdenum/silicon multilayers and 97 eV photon energy were chosen for initial measurements because reasonably ideal Mo/Si multilayers having roughly a 9-nm period are easily fabricated. These structures show especially strong interference effects at photon energies just below the Si L$_{III}$ edge ($\approx 99.5$ eV) where the contrast between the optical constants of Si and Mo is large. Linear multilayer polarizers, which are necessary for these measurements, had already been characterized at 97 eV.

The phase retardation measurement used radiation from an unpolarized laser-plasma source that was monochromated and focused by a grating and a mirror, as in Fig. 1. The monochromator was tuned to 97 eV ($\lambda = 12.8$ nm). Identical Ru/Si multilayers with interference peaks...
designed to reflect at 90° total scattering angle were used as a linear polarizer and a polarization analyzer. The detector was a microchannel plate. The transmission multilayer retarder was positioned between the polarizer and analyzer with its azimuthal orientation selected to give equal amounts of \( \sigma \) and \( \pi \) components incident on the multilayer. This corresponds to orienting the multilayer scattering plane (containing the incident ray, multilayer normal, and specularly reflected ray) 45° out of the plane of Fig. 1. The incidence angle \( \theta \) on the retarder (measured from the surface) was varied with a vacuum stepper motor.

Quantities of interest are the complex amplitude transmittance of the two components, \( t_\sigma \) and \( t_\pi \). The retardation, \( \Delta \), can be expressed as \( t_\sigma / t_\pi = |t_\sigma / t_\pi|e^{i\Delta} \). The experimental procedure was to rotate the analyzer azimuthally about the beam transmitted through the retarder, thus measuring intensity curves showing sinusoidal variation. The minimum and maximum intensities and azimuthal positions of the extrema were obtained from fits to the data, and are the observables from which the ellipticity angle and the orientation of the polarization ellipse are first determined using standard rotating-analyzer ellipsometric techniques. From these quantities \( |t_\sigma / t_\pi| \) and \( |\Delta| \) are then derived.

The degree of linear polarization of the beam after the polarizer, or alternatively the polarization of the analyzer, was determined with the transmission phase retarder normal to the beam, where \( \sigma \) and \( \pi \) components are equivalent. The analyzer scan thus obtained is the curve labeled 90° in Fig. 2, from which the polarization of the reflecting multilayers was determined to be 0.97. The beam incident on the phase retarder is thus highly linearly polarized.

Analyzer scans were made at a series of different incidence angles \( \theta \) on the multilayer, three of which are shown in Fig. 2. Several features are evident in the analyzer scans as \( \theta \) decreases from 90°. The reduction in intensity is explained partly by the decreased transmissivity as the effective thickness of the sample increases. The contrast of the analyzer scans, \( (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}}) \), decreases at certain \( \theta \) values. If the retarder produced \( \Delta \approx 90° \) between the components of the incident linearly polarized beam and \( |t_\sigma / t_\pi| = 1 \), the analyzer scan would be flat (zero contrast) indicating perfect circular polarization. \( \Delta \neq 90° \) yields elliptical polarization. Also observed in Fig. 2 are shifts with \( \theta \) in positions of the extrema of the analyzer scans. Such shifts with no change in contrast would indicate a rotation of the plane of linear polarization, and accompanied by a change in contrast, as observed here, indicate a rotation of the major axis of the polarization ellipse away from the plane of incident linear polarization by an amount equal to this angular shift.

Analysis of the scans in Fig. 2 and at other \( \theta \) values yields the \( \Delta \) and \( |t_\sigma / t_\pi| \)-vs-\( \theta \) results in Fig. 3. The estimated error in \( \Delta \) is largest near \( \Delta = 0 \) where errors in fitting the analyzer scans are amplified in the analysis, and is small as \( \Delta \) increases. Also plotted in Fig. 3 are the \( \Delta \) and \( |t_\sigma / t_\pi| \) curves calculated assuming an ideal multilayer (no intermixing across interfaces or lateral interfacial roughness) using the following optical constants from Ref. 10; \( \delta_{\text{Si}} = -0.0037 \), \( \beta_{\text{Si}} = 0.0020 \), \( \delta_{\text{Mo}} = 0.068 \), \( \beta_{\text{Mo}} = 0.0059 \). In the calculation the Mo layers were assumed to have \( 1/3 \) of the bilayer thickness, based on deposition rates, and the bilayer thickness was varied to position the resonance in \( \Delta \) at the observed position, with the resulting 8.75-nm thickness in good agreement with that measured from the witness samples. The trends in the measured \( \Delta \) and \( |t_\sigma / t_\pi| \) with \( \theta \) are in reasonable agreement with those calculated for this ideal structure. The calculated quantities are sensitive to several factors which are not precisely known, including the relative thickness of the Mo and Si layers and some amount of interfacial roughness or interdiffusion which are known to exist in Mo/Si multilayers. The calculations are also sensitive to the optical constants which may contain uncertainties, especially for Si very close to its LIII edges. The effect of interfacial roughness was simulated by multiplying the reflected amplitude at each interface by exp( \( - \sigma q^2 \)), where \( \sigma \) represents a rms roughness and \( q = 4\pi \sin \theta /\lambda \). An assumed interfacial roughness of \( \sigma = 6.5 \) Å brings the calculated retardation into good agreement with that measured, although we do not rule out other factors as possibly contributing to the discrepancy between ideal calculation and measurement. At maximum retardation \( |t_\sigma / t_\pi| = 0.66 \). The ellipticity angle at maximum retardation is 22.2°, the tangent of which (0.41) gives the ratio of minor-to-major axes of the ellipse traced out by the electric vector in the plane normal to
propagation. The major axis of this ellipse is rotated by $-26.5^\circ$ from the plane of incident linear polarization. The transmitted intensity was measured in a different geometry using unpolarized incident light to be roughly 2/3 of that calculated for an ideal structure. This reduction may result from residual contamination left on the free-standing sample after preparation, or from uncertainties in the optical constants. At the maximum retardation setting, 20% of the unpolarized incident beam was transmitted.

In summary, these measurements confirm the predicted phase retardation behavior of free-standing transmission multilayers. The measured retardation and transmission values are in reasonable agreement with calculations for ideal structures, and are large enough to be of interest for various applications. These and other measurements\textsuperscript{4,7} demonstrate that standard optical techniques combining linear polarizers and retarders to generate and analyze specific polarization states can be extended into at least part of the soft x-ray region. Such optics will enable a variety of experiments in which the polarization dependence of x-ray interactions with matter will provide new information.

The Mo/Si multilayer studied here was not optimized for maximum retardation at 97 eV, and improvements in performance of Mo/Si retarders are expected for $h\nu < 100$ eV. At higher photon energies, materials other than Mo and Si yield the strongest multilayer interference effects. Two issues will limit the maximum photon energy at which multilayer retarders are useful. First, as $h\nu$ increases in the soft x-ray region, the complex index of refraction of all materials tends to unity, decreasing the retardation obtainable from ideal multilayer structures. Second, the multilayer bilayer thickness scales with $\lambda$, and ideal multilayers become harder to realize as the individual layers approach 1 nm in thickness. This results in part because the effects of structural imperfections increase as the bilayer thickness decreases. At the current state of soft x-ray multilayer development, it is not clear that significant retardation values will be obtained by this technique at photon energies above the carbon $K$ edge (284 eV). The continuing development of soft x-ray multilayers will ultimately determine the high-energy limit at which useful values of retardation can be obtained.

The authors thank T. Namioka for making these experiments possible. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and by a Grant-in-Aid for Scientific Research on Priority Areas, "X-ray Imaging Optics," No. 0223102 from the Ministry of Education, Science, and Culture, Japan.