High-efficiency diffractive x-ray optics from sectioned multilayers

H. C. Kang and G. B. Stephenson

Center for Nanoscale Materials and Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

C. Liu, R. Conley, A. T. Macrander, and J. Maser

Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439

S. Bajt and H. N. Chapman

Lawrence Livermore National Laboratory, Livermore, California 94550

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We investigate the diffraction properties of sectioned multilayers in Laue (transmission) geometry, at hard x-ray energies (9.5 and 19.5 keV). Two samples are studied, a W/Si multilayer of 200 \times (29 nm) periods, and a Mo/Si multilayer of 2020 \times (7 nm) periods, with cross-section depths ranging from 2 to 17 \mu m. Reflectivities as high as 70% are observed. This exceeds the theoretical limit for standard zone plates operating in the multibeam regime, demonstrating that all of the intensity can be directed into a single diffraction order in small-period structures. © 2005 American Institute of Physics. [DOI: 10.1063/1.1897061]

Breakthroughs in efficient, high-resolution diffractive optics for hard x rays (e.g., to focus to nanometer-scale spots, or to compress to femtosecond-scale pulses) are technically challenging but theoretically very promising. High spatial resolution requires correspondingly thin structures transverse to the beam direction, while high efficiency requires a relatively large depth along the beam direction. Fabrication of such high-aspect-ratio nanostructures challenges the limits of lithographic techniques. An alternative approach is to make a cross section of a multilayer film, as in “sputter/slice” zone plates. Although very high aspect ratios can be produced, the challenge in this case is to deposit a multilayer with hundreds or thousands of accurately placed layers, and to maintain the structure during sectioning. The optical properties of sectioned multilayers in the Laue (transmission) geometry, also known as volume gratings, have been calculated and measured in the soft x-ray region, but are relatively unexplored in the hard x-ray region. In theory, the properties are very promising: as the layer thickness becomes smaller, the efficiency should approach unity because all of the intensity can be directed into a single diffraction order.

We have been exploring the fabrication and hard x-ray diffraction properties of sectioned multilayers in Laue geometry. Results have been obtained for two multilayers of differing period and composition that bridge the gap between large and small layer thickness behavior. In a preliminary publication, we reported x-ray rocking curves showing fringes that allow us to determine the depth of the sections. Here we report measurements of the peak reflectivity as a function section depth, which demonstrate that high efficiency can be obtained at small periods. We find reasonable agreement between measurements and dynamical diffraction theory, providing a solid foundation for both the design and fabrication of novel hard x-ray optics.

Two constant-period multilayers were grown on Si (001) substrates by dc magnetron sputter deposition techniques, as described elsewhere. Multilayer periods and layer volume fractions were determined by modeling Bragg-geometry x-ray reflectivity data. Multilayer A has 200 \times (29 nm) W/Si periods, a total thickness of 6 \mu m, and W volume fraction of 60%. Multilayer B has 2020 \times (7 nm) Mo/Si periods, a total thickness of 14 \mu m, and Mo volume fraction of 50%. These may be the thickest x-ray-optical-quality multilayer films produced to date. As the outermost zones of a focusing structure, the 7 nm period of Multilayer B would correspond to a 4.2 nm diffraction-limited focal size.

To study the Laue-geometry diffraction behavior as a function of section depth using a single sample, we produced cross sections with depths intentionally varying by \sim 10 \mu m across a 2 mm length. The Laue diffraction geometry used is shown in Fig. 1 (inset). Here we define z as the layer stacking direction (“thickness”), x perpendicular to a cross-sectioned surface (“depth”), and y along the wedge (“length”). The x-ray scattering was mapped by scanning the scattering vector in the Q_x and Q_y directions. The dependence on section depth was determined by translating the 50 \mu m illuminated region across the sample in the y direction. Absolute reflectivity at the Bragg peak was measured using an incident beam with a z dimension equal to the multilayer thickness. Measurements were performed at beamline 12BM of the Advanced Photon Source. Using a

\begin{figure}[h]
\centering
\includegraphics{fig1}
\caption{Radial scan for multilayer B, showing positive and negative orders of Bragg peaks (E=19.5 keV, w=10 \mu m). Inset: Schematic of Laue geometry used, with sectioned multilayer illuminated through side.}
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\textsuperscript{a}Author to whom correspondence should be addressed. Electronic mail: stephenson@anl.gov
Si(111) monochromator, an x-ray energy $E$ of either 9.5 or 19.5 keV ($\lambda = 1.31$ or 0.64 Å) was selected.

Figure 1 shows a typical radial scan along $Q_z$ in the specular direction ($Q_x = 0$), Bragg reflections from the fundamental multilayer period and higher harmonics are observed. The pattern is symmetric about zero, which illustrates a feature of the Laue geometry: diffraction from the “top” or “bottom” of the multilayer is equivalent. Figure 2 shows typical transverse (rocking) scans along $Q_x$ at the first-order Bragg reflection in $Q_z$, for $E = 9.5$ keV and various values of section depth. The scattering patterns exhibit interference fringes around the central peak. The fringe spacing $Q_0$ gives the depth of the second-order peak for multilayer A is significant. It is interesting to consider the Borrmann effect for samples with large section depth. 

An analytical solution for the reflectivity of small-period multilayers can be developed by extending the 2-beam dynamical theory for crystals. For a centrosymmetric crystal in the symmetric Laue geometry, in the limit of small diffraction angle, the reflectivity is given by

$$R = \exp \left[ \frac{2\pi w}{\lambda} \text{Im}(\psi_0) \right] \sin \left( \frac{\pi w \psi_H}{\lambda} \sqrt{1 + \eta^2} \right)^2 \left[ 1 + \eta^2 \right]^{-1},$$

where $\eta$ is related to the transverse wave number $Q_z$ by $\eta = Q_z \lambda / 2 \pi \psi_H$. For a binary multilayer, the normalized structure factors in the forward and diffracted-beam directions, $\psi_0$ and $\psi_H$, are given by $\psi_H = 2 n \sin(\pi Lx) / \pi L$, $\psi_0 = 2(\langle n \rangle - 1)$. Here $n$ is the refractive index, $\langle \rangle$ represents the mean of the two layers in the multilayer, e.g., $\langle n \rangle = x n_1 + (1-x) n_2$, $\Delta$ represents the difference between the two layers, e.g., $\Delta n = n_1 - n_2$, $x$ is the volume fraction of layer 1, and $L$ is the order of the 00L Bragg peak. The reflective indices of each of the layers are complex numbers given by $n = 1 - \delta - i \beta$, where $\delta$ and $\beta$ depend on material and photon energy. At the center of the rocking curve, where $\eta = 0$, the specular reflectivity as a function of section depth $w$ can be expressed as

$$R = \exp \left[ \frac{-4\pi w \langle \beta \rangle}{\lambda} \right] \sin^2 \left[ \frac{2w \sin(\pi Lx) \Delta \delta}{\lambda L} \right] + \sinh^2 \left[ \frac{2w \sin(\pi Lx) \Delta \beta}{\lambda L} \right].$$

The $\sin^2$ term gives the oscillatory pendellösung component, with a period $w_0 = \pi \lambda L |2 \Delta \delta \sin(\pi Lx)|^{-1}$, while the sinh² term gives the anomalous transmission (Borrmann effect) for samples with large section depth.

Dashed curves in Fig. 3 show reflectivities calculated using the 2-beam expression, Eq. (2), with standard elemental densities for the W, Mo, and Si layers. For the first-order peaks, there is qualitative agreement with the observed depth dependence, while the predicted oscillation period with depth of the second-order peak for multilayer A is significantly larger than observed. It is interesting to consider the predicted dependence of the pendellösung period on sample composition and x-ray wavelength obtained from the 2-beam expression. For a multilayer with equal thickness layers ($x$...
\(= 0.5\), the period for \(L = 1\) is given by \(w_0 = \pi \lambda / 2 \Delta \delta\). This is a factor of \(\pi / 2\) larger than for a phase zone plate.) Ignoring anomalous scattering effects near absorption edges, the real part of the refractive index can be expressed using 
\[
\delta = r_e \rho_e \lambda^2 / 2\pi, \quad \text{where} \quad r_e = 2.82 \times 10^{-13} \text{cm}\]
the Thomson radius of the electron and \(\rho_e\) is the electron density.\(^{15}\) This gives \(w_0 = \pi^2 / r_e \Delta \rho \lambda\), which is inversely proportional to the density difference between the layers and to photon wavelength. We confirmed this wavelength dependence, finding that the density giving maximum reflectivity increased by approximately a factor of 2 when the wavelength was decreased from 1.31 to 0.64 Å. The observed period for multilayer B in Fig. 3(c) is \(\sim 20%\) larger than predicted, which may indicate that the density difference in the sample is lower than that between pure Mo and Si, owing to interdiffusion and/or reaction to form MoSi\(_2\).\(^{16}\) The relatively shallow minimum in the observed reflectivity from multilayer B at larger depths is likely due to the finite transverse resolution of the measurement, which can be seen in Fig. 2(b).

Effects of scattering into multiple directions are not included in the 2-beam solution. One such effect can be seen in the rocking curves in Fig. 2. For multilayer B, the features at \(Q_z = \pm 0.0017 \text{Å}^{-1}\) occur when either the incident or exit beam makes the Bragg angle with the fundamental \(L = \pm 1\) reflection. For multilayer A, this condition occurs at \(Q_z = \pm 0.0001 \text{Å}^{-1}\), so that the features are part of the complex shape of the central peak. One expects multi-beam effects to contribute to the specular reflectivity whenever the section depth is small enough to broaden the specular peak by an amount comparable to the spacing between adjacent Bragg peaks. Conversely, the 2-beam expression \(2\) is valid and multibeam effects are negligible for the \(L = 1\) peak when the sample depth \(w\) is large enough so that the rocking width of the \(L = 1\) peak, \(|\Delta \theta| = d / 2w\), is smaller than its distance from the \(L = 2\) peak, \(\Delta \theta_1 = \lambda / 2d\), where \(d\) is the multilayer period. This condition can be written as
\[
d < (\lambda \omega)^{1/2},
\]
which is satisfied for multilayer B, but not always for multilayer A, in this study. For periods above this limit, multiple diffraction orders are simultaneously excited, so that the efficiency into any one order is limited. For example, the maximum theoretical efficiency is \(\sim 40%\) for an ideal binary zone plate in the multibeam regime.\(^{17}\) For periods satisfying relation \(3\), all of the intensity can be directed into one diffraction order by choice of incidence angle, giving a peak reflectivity near unity for hard x rays with low absorption. Substituting the \(x = 0.5\) expression for optimum sample depth \(w = w_0 / 2\) into condition \(3\) cancels the wavelength dependence, giving \(d < (\pi / (2r_e \Delta \rho))^2\). Thus the condition depends only on the multilayer materials. For Mo/Si and W/ Si, the limit occurs at \(d = 30\) and 21 nm, respectively. Multibeam effects are expected to occur only away from the center of the rocking curve for 7 nm Mo/Si multilayer B, but at the center of the rocking curve for 29 nm W/Si multilayer A.

Model multibeam scattering, we have carried out calculations using a version of coupled-wave (CW) theory.\(^{18,19}\) The relationship between the complex dielectric constant \(\varepsilon = n^2\) used in CW theory and the optical constants used in the 2-beam theory is \(\Delta \varepsilon = 2\Delta \delta - 2i\Delta \beta\). Reflectivity results from CW theory are displayed as solid curves in Fig. 3. For multilayer B, the multibeam solution differs little from the 2-beam solution. For multilayer A, the difference is large. The lower peak reflectivity and more complex functional form of the multibeam solution for the first-order peak agree better with the experimental results, as do the positions of the maxima and minima for the second-order peak. The differences between the experimental data and the multibeam theory could be due to interfacial roughness and experimental resolution not included in the calculation.

In summary, we have successfully fabricated sectioned multilayers that give high reflectivities for hard x rays in the Laue geometry. These structures have the high ratio of section depth to layer thickness (e.g., 2000 at the reflectivity maximum for multilayer B) needed to produce efficient hard x-ray optics with sub-10-nm spatial resolution. We observe oscillatory reflectivity as a function of section depth, with high diffraction efficiency at small periods. These observations verify the conceptual basis of dynamical diffraction theory for multilayers in the hard x-ray region, and indicate that the sectioned multilayer samples are of sufficient structural quality to produce dynamical effects. This provides a promising demonstration of the techniques and principles for the design of novel hard x-ray optics using sectioned multilayers.

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