Nonspecular x-ray-reflectivity study of partially correlated interface roughness of a Mo/Si multilayer

D. R. Lee
Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea

Y. J. Park
Pohang Accelerator Laboratory, Pohang University of Science and Technology, Pohang 790-784, Korea

D. Kim and Y. H. Jeong
Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea

K.-B. Lee*
Department of Physics and Pohang Accelerator Laboratory, Pohang University of Science and Technology, Pohang 790-784, Korea

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Nonspecular x-ray-reflectivity intensities were measured to characterize the interface morphology of a Mo/Si multilayer. Longitudinal off-specular scans and transverse scans at several multilayer peaks and valleys were carried out. For the analysis of the experimental data, a height cross-correlation function between different interfaces was derived for a model multilayer whose interfaces are partially correlated. The parameters related to the interface morphology were obtained by fitting the measured intensities within the distorted-wave Born approximation. The intermixing widths of the graded interfaces, the correlated interface roughness amplitude, and a vertical correlation length were obtained by analyzing the off-specular intensities.
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Specular and nonspecular x-ray reflectivity in grazing incidence angle have been effectively utilized to characterize interface morphology. While specular reflectivity yields a density profile perpendicular to the sample surface, nonspecular scattering yields lateral interface structures. Sinha *et al.* formulated a distorted-wave Born approximation (DWBA) formalism for a single rough surface to analyze x-ray-reflectivity intensities near the critical angle.[1] The extension of the DWBA calculation to layered structures were done by Holý and Baumbach[2] and others[3,4] for epitaxially grown heterostructures with relatively sharp interfaces. Though the intermixing of elements between layers is another crucial factor determining interface characteristics, especially in magnetic metallic multilayers, a DWBA analysis for nonspecular reflectivity intensities of multilayers with graded interfaces has not been reported earlier, to our knowledge.

In this work we carried out nonspecular x-ray-reflectivity studies of a Mo/Si multilayer which has graded interfaces. The experimental data have been analyzed within the DWBA to extract the parameters related to its interface morphology, including the intermixing widths. A simple model is presented to describe partially correlated interfaces in multilayers. The cross-correlation function, which has been widely used without derivation in order to characterize partially correlated interfaces of epitaxially grown multilayers,[4–6] is derived from the model, and has been applied to analyze our nonspecular x-ray-reflectivity data.

During deposition process of multilayers, a height profile $h_m(\vec{r})$ of the mth interface with respect to the average interface plane $\overline{z}_m$ is conformally transferred to the $(m+1)$th interface, while uncorrelated random noise generates a deviation from perfect correlated interfaces. For partially correlated interfaces, the interface profile can be related to the adjacent one by the following recursive equation:[7]

$$\hat{h}_{m+1}(\vec{r}) = [B_m \hat{\zeta}_{m+1}(\vec{r}) + A_m \hat{h}_m(\vec{r})], \tag{1}$$

where $\vec{r}$ is the lateral coordinate in the average interface plane. $\hat{h}_m(\vec{r})$ and $\hat{\zeta}_m(\vec{r})$ refer to the normalized profile of the mth interface and normalized deviation from the perfect replication of the $(m-1)$th interface, respectively. The first term on the right-hand side of Eq. (1) represents the deviation from a perfect correlated interface due to random noise at the mth interface. A_m is a replication factor representing the degree of conformality, and satisfies the relation $A_m^2 + B_m^2 = 1$. Then $\hat{h}_m(\vec{r})$ can be expressed as

$$\hat{h}_m(\vec{r}) = \sum_{i=1}^{m} B_{i-1} \left[\prod_{j=i}^{m-1} A_j \right] \hat{\zeta}_i(\vec{r}). \tag{2}$$

Assuming that A_i's and B_i's are independent of i, the self-correlation function of the interface roughness is

$$\begin{aligned}\langle h_m(\vec{r}) h_m(0) \rangle &= \sigma_m^2 \sum_{i=1}^{m} \sum_{j=1}^{m} B^2 A^{m-i} A^{m-j} \langle \hat{\zeta}_i(\vec{r}) \hat{\zeta}_j(0) \rangle \\ &= \sigma_m^2 (1 - A^{2m}) \langle \hat{\zeta}(\vec{r}) \hat{\zeta}(0) \rangle \approx \sigma_m^2 \langle \hat{\zeta}(\vec{r}) \hat{\zeta}(0) \rangle,\end{aligned} \tag{3}$$

where A^{2m} is negligible and $\langle \hat{\zeta}_i(\vec{r}) \hat{\zeta}_j(0) \rangle$ vanishes for $i \neq j$ because they are uncorrelated to each other. Here σ_m is the rms amplitude of the correlated roughness of the mth interface. Then the morphologies of the interfaces are similar to each other, and can be expressed with the same parameters. If they can be described as self-affine, the self-correlation function is approximately proportional to $\sigma_m^2 e^{-(r/\xi_x)^{2H}}$.[1] Here r and H are the distance along the interface plane and the roughness exponent describing the jaggedness of the interface, respectively. ξ_x is the lateral cutoff length. The cross-correlation function between the mth and nth interfaces ($m>n$) can be expressed as

$$C_{mn}(\vec{r}) = \langle h_m(\vec{r}) h_n(0)\rangle = A^{m-n}\langle h_n(\vec{r}) h_n(0)\rangle \approx \sigma_m \sigma_n e^{-(r/\xi_x)^{2H}} e^{-|\bar{z}_m - \bar{z}_n|/\xi_z} \quad (4)$$

for $A^{m-n} = e^{-|\bar{z}_m - \bar{z}_n|/\xi_z}$. ξ_z reperesents the vertical correlation length along the layer growth direction. Here we note that roughness components of different wavelengths have the same replication factor A resulting in a constant correlation length ξ_z. If A depends on the wavelength of the interface roughness,[8] the correlation functions in Eqs. (3) and (4) should be expressed as a convoluted function of the replication factor and the random noise. Then ξ_z is a function of lateral component ($\vec{q}_{xy}$) of the momentum transfer of scattered x rays.

With the cross-correlation function given above, scattered x-ray intensities from a multilayer with N interfaces can be expressed in the DWBA as[2,3]

$$I(\vec{q})/I_0 = |\tilde{R}(q_z)|^2 \delta(\vec{q}_{xy}) + \frac{1}{l_x \times l_y}\int \left(\frac{d\sigma}{d\Omega}\right)_{\rm diff} d\Omega,$$

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm diff} = \frac{k_o^4}{16\pi^2}\sum_{i,j=1}^{N} (n_i^2 - n_{i+1}^2)(n_j^2 - n_{j+1}^2)^* \times \sum_{m,n=0}^{3} S\frac{D_m^{i+1} D_n^{j+1*}}{q_{mz}^{i+1} q_{nz}^{j+1*}} e^{-1/2[\sigma_{i,t}^2 (q_{mz}^{i+1})^2 + \sigma_{j,t}^2 (q_{nz}^{j+1*})^2]} \times \int d^2\vec{r}[e^{q_{mz}^{i+1} q_{nz}^{j+1*} C_{ij}(\vec{r})} - 1] e^{i\vec{q}_{xy}\cdot\vec{r}}, \quad (5)$$

where $I(\vec{q})$, I_0, $|\tilde{R}(q_z)|$, and $(d\sigma/d\Omega)_{\rm diff}$ are the scattered intensity with momentum transfer of $\vec{q}$, the incident intensity, the specular reflectivity, and the differential cross section of the diffuse scattering, respectively. $l_x \times l_y$ is the beam cross section, while S represents the illuminated area by incident x rays. k_o is the wave-vector length in vacuum, and the n_i's are refractive indices in the ith layer. D_m's and q_{mz}'s for each layer are denoted by[3]

$$D_0 = T_1 T_2, \quad D_1 = T_1 R_2, \quad D_2 = R_1 T_2, \quad D_3 = R_1 R_2,$$

$$q_{0z} = k_{1z} + k_{2z}, \quad q_{1z} = k_{1z} - k_{2z}, \quad q_{2z} = -q_{1z}, \quad q_{3z} = -q_{0z}, \quad (6)$$

where the amplitudes $T_{1,2}$ and $R_{1,2}$ of transmitted and reflected waves in a multilayer with ideally smooth interfaces are defined for an incident wave vector k_1 and for a scattered wave vector k_2, respectively. $\sigma_{i,t}$ represents a total interfacial width for the ith graded interface, containing both its interface roughness amplitude and intermixing width.[9]

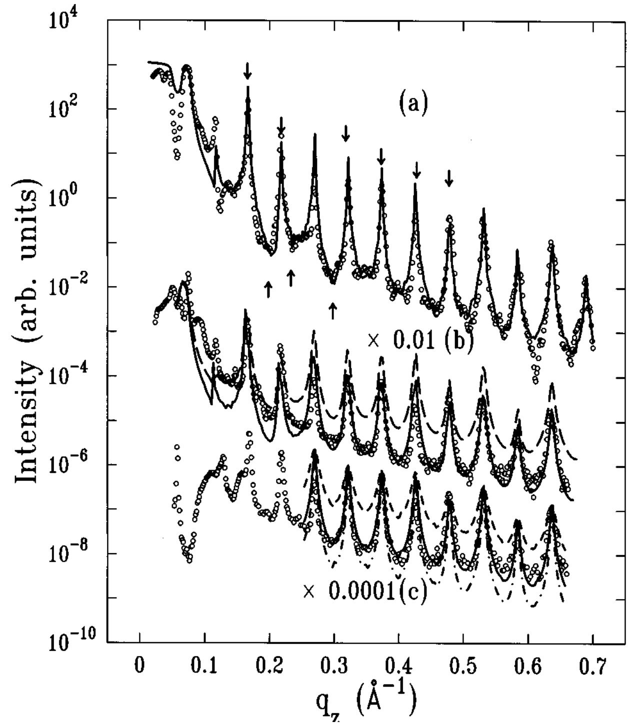

FIG. 1. Specular scan and off-specular scans at two diffferent offset angles: (a) the measured specular intensities (circles) and the result of fitting (solid line). Arrows indicate the values of q_z, where rocking scans have been performed. (b) The measured off-specular intensities at an offset angle of 0.055° (circles) and the results of the calculation for sharp interfaces without any intermixing (dashed line). The solid line represents the best fit for the graded interfaces which have both correlated roughness and intermixing. (c) The measured off-specular intensities at an offset angle of 0.6° (circles), and the results of calculations with different values of ξ_z: 300 Å (dashed line), 600 Å (solid line), and 1200 Å (dash-dotted line).

The sample of this study is a $(Mo/Si)_{30}$ multilayer prepared by the rf magnetron sputtering method as described elsewhere.[10] X-ray-reflectivity measurements were carried out at a bending magnet beamline 3C2 at the Pohang Light Source (PLS).[11] X rays of $\lambda = 1.608$ Å monochromatized by a Si(111) double-crystal monochromator were focused at the sample position by a torroidal premirror. Three different types of scans were conducted with a Huber four-circle diffractometer: $\theta - 2\theta$ scans for specular reflectivity, offset $\theta - 2\theta$ scans for off-specular reflectivity, and transverse rocking scans.

Figure 1 shows the results of the x-ray-reflectivity measurements from the sample. Specular reflectivity intensities show peaks corresponding to a bilayer thickness of 118.8 Å in Fig. 1(a). Two off-specular scan intensities with offset angles of 0.055° and 0.6°, as shown in Figs. 1(b) and 1(c), also show sharp resonant diffuse scattering (RDS) peaks indicating strong vertical correlation in the interface roughness.[2] The measured intensities for $q_x = 0$ in Fig. 1(a) have been fitted, after subtracting diffuse intensities, using Parratt's recursive relation[12] modified for rough interfaces with error function interface profiles. Later the parameters were refined by fitting the measured intensities with the full expression of Eq. (5), as described below. The broadening of the peak widths at high q_z's can be explained by incoherent random errors in each layer thickness,[13] and the fluctuation in the thickness was estimated to be 1.2 Å. The solid lines represent the fit, while the circles represent the measured intensities. Fitting gives the total interfacial widths σ_t's of 2.4 and 3.5 Å for Si-on-Mo interfaces and Mo-on-Si interfaces, respectively. However, there is an ambiguity in the estimates of σ_t's because the calculations give almost the same results when the values of two σ_t's are exchanged. This appears to be a common difficulty in the analysis of multilayers.[14] It has been revealed by TEM studies[15] that amorphous intermixed layers are formed on Mo-on-Si interfaces and these interfaces have thicker interfacial widths than Si-on-Mo interfaces, in both sputter deposited and evaporated multilayers. Therefore, we assign the larger value of σ_t to the interfacial width of Mo-on-Si interfaces of our sample. Other parameters can be determined from an analysis of nonspecular reflectivity intensities, as described below.

If elements are intermixed in a certain range across the interfaces, the interfaces are graded ones which have both interface roughness and interfacial widths due to intermixing. For these graded interfaces, it is difficult to estimate interface roughness amplitudes σ_c's and intermixing widths σ_d's by analyzing the specular reflectivity data which provides only σ_t's. We note that $C_{ij}(\vec{r})$ in Eq. (5) is the only term containing σ_c's explicitly, and, therefore, an analysis of diffuse scattering intensities is necessary to estimate the parameters related to the interface roughness.

In order to calculate nonspecular scattering intensities, we have used the whole expression of Eq. (5) without a small-q_z approximation, because it has been shown by others[6] that there is a significant discrepancy in the region of $q_z \sim 1$ between the results of calculations with the full expression and with the approximation. However, some parameters are more sensitive to specific scans. For example, a longitudinal off-specular scan is sensitive to correlated roughness amplitudes σ_c's, because it scans the intensities for different q_z's while $\vec{q}_{xy} \neq 0$. Therefore, the parameters were roughly estimated first with data of the corresponding scans to avoid the dangers of becoming trapped in local minima in the fitting process. The lateral coherence length was roughly estimated from the full width at half maximum of the diffuse part of rocking curves at multilayer peaks. Then the coherence length and roughness exponent were evaluated together by fitting the shapes of diffuse parts of the rocking curves at peaks with large-q_z values. In this study we have used an averaged value of correlated roughness amplitudes for all interfaces, with an assumption that the major features of correlations between different interfaces in our sample can be described by averaged values for correlated roughness amplitudes and vertical correlation lengths. Since the shapes of rocking curves at fixed q_z values do not depend strongly on a correlated roughness amplitude, the amplitude was estimated from the slope of resonant diffuse peaks in longitudinal off-specular intensities. The vertical correlation length was evaluated from the ratios bewteen the intensities of the peaks and valleys of off-specular scans. To refine the parameters, the whole set of the x-ray-reflectivity data including specular intensities were fitted simultaneously to minimize possible errors due to the interplay between various parameters used in the analysis.[4]

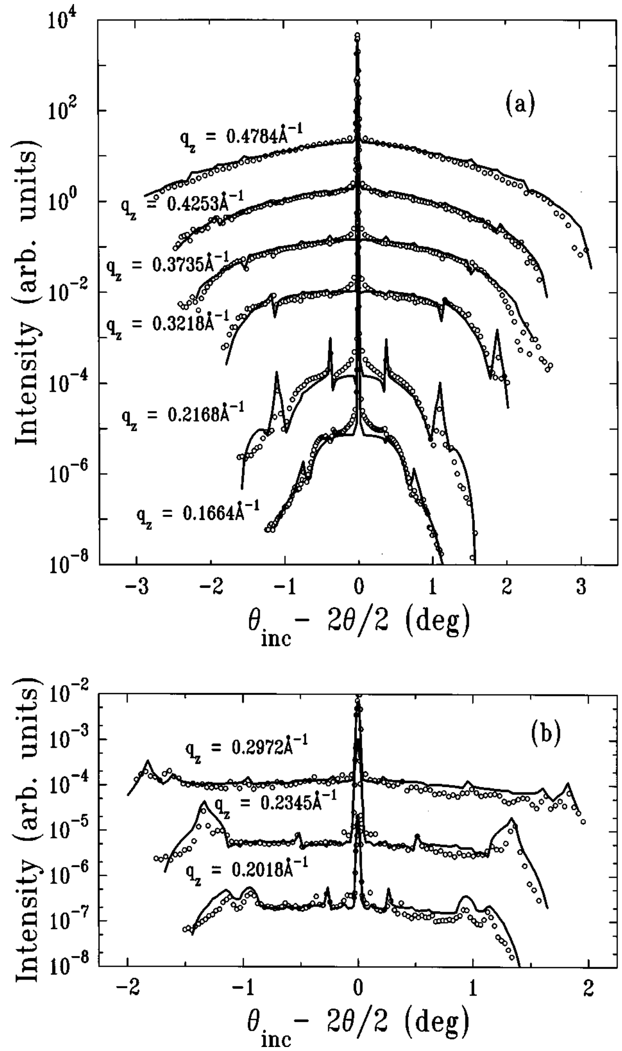

FIG. 2. Rocking curves at multilayer Bragg peaks (a) and at their valleys (b). Solid lines represent fits using the correlation function and the parameters explained in the text. The intensities are shown as a function of $[\theta_{\rm inc}-(2\theta/2)]$ in order to illustrate the low q_z scans better while $q_x = q_z \times \sin[\theta_{\rm inc}-(2\theta/2)]$.

Off-specular intensities were calculated for both models with and without graded interfaces, which is an extension of the work of Wormington *et al.* for a single graded surface[9] to a multilayer with graded interfaces. Figure 1(b) presents the measured off-specular scan intensities as well as the results of calculations. For a multilayer with sharp interfaces, the values of the total interfacial widths estimated above from the analysis of the specular scan intensities were used both for $\sigma_{i,t}$'s and σ_c's in $C_{ij}(\vec{r})$ of Eq. (5). The results of the calculation are presented as a dashed line. At high q_z's the calculation gives higher intensities than the measurements, and this is explained by the overestimated σ_c's, resulting in high diffuse scattering intensities. For a multilayer with graded interfaces, the total interfacial width σ_t can be expressed as $\sigma_t^2 = \sigma_c^2 + \sigma_d^2$, where σ_c and σ_d represent the correlated roughness amplitude and the intermixing width, respectively. We have also calculated the diffuse scattering intensities for these graded interfaces, and the best fit yields σ_c of 1.8 Å. Then σ_d's for Si-on-Mo and Mo-on-Si interfaces were estimated as 1.6 and 3.0 Å, respectively. The result of the calculation is presented as a solid line, and it shows good agreement with the measured intensities. We have also simulated, for a model of accumulative interface roughness, $\sigma_{c,j}^2=\sigma_0^2+j\times\delta^2$, and found that it shows the same tendency.

In off-specular scans of partially correlated systems, relative intensities at the valleys with respect to those of the RDS peaks are sensitive to the value of ξ_z, because the extent of the interference depends on the vertical correlation. For $CoSi_2$/Si/$CoSi_2$ layers on a Si(111) substrate, ξ_z has been estimated from the amplitude of the oscillations at high q_z by others.[4] Figure 1(c) shows the measured intensities with an offset angle of 0.6° with the results of the calculation with three different values of ξ_z: 300, 600, and 1200 Å. The vertical correlation length was estimated to be 600 Å, which is about one-sixth of the total thickness of the multilayer.

Figure 2 shows the measured rocking curves at multilayer peaks [Fig. 2(a)] and at the valleys between them [Fig. 2(b)]. The rocking curves around the valleys in Fig. 2(b) show different shapes of diffuse scattering from those of the multilayer peaks in Fig. 2(a). The diffuse scattering intensities at the valleys are relatively flat with respect to q_x, and the ''Yoneda wings'' are more apparent compared to those in the rocking curves in Fig. 2(a). This is because the transverse rocking scans at the peaks are the scans along the RDS streak which have broad humplike peaks around specular peaks.[2] The measured intensities have been compared with the calculation for $I(q_x)$, which is the integrated intensity of $I(\vec{q})$ along the y direction due to the wide slitwidth in the perpendicular direction to the diffraction plane. The best fit gives 70 Å and 0.4 for ξ_x and the H exponent, respectively. Solid lines in Figs. 2(a) and 2(b) represent the results of the fitting. Though the vertical correlation length ξ_z is estimated as 600 Å, corresponding to one-sixth of the total film thickness, calculated intensities with the same set of parameters show main features including Bragg-like peaks at corresponding angles in the rocking curves of many different order multilayer peaks and their valleys. This implies that our estimates properly represent the average values of the parameters for the whole film.

In this work we have assumed the same replication factor for roughness components of different wavelengths, which results in constant ξ_z. Generally, it is expected that the combined effects of random noises from deposition and thermodynamic kinetics on growing surfaces result in a wavelength-dependent replication factor $A(q_x)$, and generate surfaces whose correlation lengths and roughness amplitude change as functions of time or thickness before they saturate.[16] In multilayers the interfaces manifest such evolutions in the form of broadening in off-specular peaks, and observations of such evolutions have been reported recently.[17–19] In this study, off-specular x-ray-reflectivity measurements have been performed, as shown in Figs. 1(b) and 1(c). Off-specular peaks become broader at higher q_z's. However, the broadening in off-specular peaks is also attributed mainly to the incoherent fluctuations in layer thickness, because the peak widths at the same q_z do not show any significant q_x dependence up to a value of q_x of 0.03 $Å^{-1}$, and RDS of broader specular peaks at higher q_z's also gives broader off-specular peaks of the same peak widths. These results imply that the replication factor for the roughness is constant for a length scale longer than 200 Å, which is consistent with the estimate for lateral cutoff length 70 Å. Further studies in different diffraction geometries are required for the extended range of the lateral momentum transfer[18,19] to observe the transition from the conformal behavior.

In summary, we presented a comprehensive x-ray-reflectivity analysis for a Mo/Si multilayer sample with graded interfaces. A simple model which describes partially correlated interfaces was presented, and the correlation function derived from the model was applied to analyze the measured intensities with the DWBA calculations. The parameters related to the morphology of the graded interfaces were extracted from the analysis. It was also demonstrated that the analysis of the off-specular reflectivity intensities is effective to separate the correlated roughness amplitude and the intermixing widths of the graded interfaces.

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*Electronic address: kibong@postech.ac.kr

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