Characterization of roughness correlations in W/Si multilayers by diffuse x-ray scattering

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Abstract: We present diffuse, nonspecular x-ray measurements of the interface height-height self-correlations in an amorphous W/Si multilayer. The measurements have been performed in a scattering geometry that is placed out of the plane of reflection, giving access to a much larger range in parallel momentum transfer than in conventional nonspecular geometries. For the sample studied a logarithmic form of the correlation function is found.

1. INTRODUCTION

During recent years, a lot of nonspecular x-ray studies of thin film surface and interface structures have been reported [1-4], aiming for a better statistical description of interface roughness than available by other more local methods, e.g. electron or STM/AFM microscopy. For most experiments, a partially self-affine, isotropic height-height correlation of the interface

$$C(R) := \langle z(\mathbf{R}') z(\mathbf{R}'+\mathbf{R}) \rangle = \sigma^2 \exp[-(R/\xi)^{2h}], \quad \mathbf{R} = (X, Y) \quad (1)$$

was assumed, where σ denotes the rms-roughness that is also accessible from specular reflectivity measurements, h is the roughness exponent corresponding to a certain universality class of non-equilibrium growth, and ξ is an upper cutoff to the self-affine behaviour. σ , h , and ξ are usually extracted by comparing the nonspecular intensity $I \sim |T_i(\mathbf{Q})|^2 |T_f(\mathbf{Q})|^2 S(\mathbf{Q})$ to the structure factor $S(\mathbf{Q})$ of a rough surface which is given by

$$S(\mathbf{Q}) = \exp[-1/2 |\mathbf{Q}_z|^2 \sigma^2] / |\mathbf{Q}_z|^2 \iint dX dY (\exp[|\mathbf{Q}_z|^2 C(R)] - 1) \exp[i(\mathbf{Q}_x X + \mathbf{Q}_y Y)] \quad (2)$$

obtained within the distorted-wave Born approximation [5]. $\mathbf{Q}$ is the scattering vector in the medium, and $T_i(\mathbf{Q})$, $T_f(\mathbf{Q})$ are the Fresnel transmission functions of the incident and exit beam, respectively, that lead to intensity (Yoneda) peaks, whenever the angle of incidence α_i or the exit angle α_f equals the critical angle α_c .

Two major difficulties arise in the evaluation of the experimental data. Firstly, the nonspecular intensity of equation (2) is difficult to separate experimentally from the specularly reflected beam. The incident beam of any real x-ray source has a finite divergence that is reflected from the rough surface to zero order like from a planar mirror, thereby contaminating the pure nonspecular scattering due to the roughness. These intensity tails of the primary beam have to be deconvoluted like in a small angle scattering experiment in order to obtain correct results. Extracting correlations up to 1000 Å or higher, e.g. a typical value for ξ , which is obtained from the intensity very close to the specular condition, may require special conditions to suppress scattering from air and imperfect slits, if the nonspecular intensity is not particularly high.

The second difficulty is due to the fact, that in all conventional nonspecular x-ray experiments cited above, the necessary parallel component of the momentum transfer $Q_{\parallel}$ lies in the xz-plane of reflection

Fig.1. The Ewald semi-circle: Only the lightly hatched area is accessible if the detector is kept in the plane of reflection. The Bragg sheets are indicated by the lines parallel to $Q_{||}$. The darkly hatched area was probed in the present experiment.

Fig.2. The scattering geometry in real space.

and is thus only accessible in a limited range. If the sample is rocked around the specular position $\alpha_i = \alpha_f$, while keeping the detector fixed (*rocking scan*), the incidence and exit angle α_i and α_f can only be varied within the limits given by the sample surface. If the sample is held fixed and the detector is moved around the specular condition (*detector scan*), α_f can be increased without any constraint of the sample surface, but a larger exit angle will automatically reduce the nonspecular intensity due to the exponential decay with Q_z , see eqn.(2). Apart from the limited length scale accessible, it is of further disadvantage that the Fresnel transmission functions influence the distribution of the nonspecular intensity via the changing α_i and α_f .

The areas of reciprocal space probed in the conventional nonspecular scattering geometries are sketched in Fig.1. The semi-circles indicate the limits due to the sample surface, that are typical for scans in the plane of reflection. Experimentally, the high Q_z region is difficult to measure due to the exponential decay in intensity. It is therefore of advantage to measure at low Q_z , but this implies limits in $Q_{||}$, if the a scattering geometry in the plane of reflection is chosen. The $Q_{||}$ range accessible in our scattering geometry is indicated as the darkly hatched area in figure 1 (see next section).

2. DIFFUSE SCATTERING FROM ROUGH MULTILAYER INTERFACES

We have recently proposed a scattering geometry for nonspecular diffuse scattering from rough interfaces, where the detector is moved out of the plane of reflection at constant angles of incidence and exit, like in grazing incidence diffraction [6]. Here, $Q_{||}$ is increased at constant Q_z . The advantages of this method are obvious: The experimental result can be extracted without constraint in the range of parallel momentum transfer and without any dependence on transmission effects or exponential decay other than an overall scaling factor.

In this work we will show how to obtain reliable values of the roughness exponent h by measuring over a wide dynamical range in nonspecular intensity and momentum transfer, see the hatched region in Fig.1. $Q_{||} = 2\pi/\lambda$ corresponds to $2\theta = 90^\circ$ (see figure 2). To enhance the nonspecular scattering versus the specularly reflected beam, we used a multilayer sample and average the self-correlations of the top $N=60$ interfaces. The parameter ξ could not be determined, because of the finite divergence of the primary beam. Here, a reliable experiment would require either an absolute intensity measurement like performed in [1] or an experimental setup of small-angle scattering, that allows for measurements in the low Q range.

The propagation of the primary and scattered x-ray amplitudes within the multilayer is rather complex. Under certain conditions the scattered amplitudes interfere coherently giving rise to intensity modulations along Q_z , the so called "Bragg sheets" at $Q_z = n \cdot 2\pi/d$, if the roughness is conformal, e.g. if the

fluctuations of different interfaces are correlated [3]. For nearly perfect interfaces additional modulations may result from multiple scattering effects [7]. However, if the scattering intensity is integrated over a sufficiently large range of Q_z around $2\pi/d$ (first Bragg sheet), these intensity modulations corresponding to correlated and non-correlated components of the roughness are averaged over, and the integrated intensity should give simply N times the result of a single interface.

We have measured the nonspecular intensity with a one-dimensional position sensitive detector (PSD) directed along the surface normal (z -axis), covering a range of exit angles, while the angle of incidence is held fixed. Starting in the plane of reflection, the PSD is now moved out of this plane by an angle 2θ , thereby increasing $Q_{||}$ without any constraint. The experimental setup is sketched in Fig.2.

The experiment was performed at the wiggler beamline W1 at the DORIS storage ring at HASYLAB / DESY in Hamburg using a wavelength of $\lambda = 1.44 \text{ \AA}$. The important beam collimations both in and out of the plane of reflection were set to $\Delta\alpha_i = 1 \text{ mrad}$, $\Delta\alpha_f = 0.1 \text{ mrad}$, and $\Delta 2\theta = 2 \text{ mrad}$. The PSD has a sensitive length of 45 mm and covered a range of $\Delta\alpha_f = 2.7^\circ$ at a distance of 959 mm behind the sample. As a sample we chose an amorphous tungsten/silicon (W/Si) multilayer as grown by a computer controlled magnetron sputtering machine, using a well polished silicon wafer as a substrate. The multilayer consisted of 150 bilayers of periodicity $d=22.8 \text{ \AA}$. The individual W and Si layers were $d_W = 10.8 \text{ \AA}$ and $d_{Si} = 12.0 \text{ \AA}$, respectively. An average interface rms-roughness of $3 \text{ \AA}$ had been determined in a previous reflectivity measurement. The first Bragg sheet of the multilayer is expected to occur at $Q_z = 0.276 \text{ \AA}^{-1}$. In order to place the center of the PSD on the first diffuse Bragg sheet, we chose $\alpha_i = 0.98^\circ = 3.15 \alpha_c$. Since this incidence angle is smaller than the first Bragg angle, α_f has to be set to 2.64° to meet the condition $Q_z = 2\pi/d$. The corresponding scattering depth, which is a function of α_i and α_f [6], is then basically absorption controlled and corresponds roughly to the top 60 interfaces.

3. RESULTS AND DISCUSSION

In Fig.3 the nonspecular intensity in the vicinity of the first Bragg peak integrated over all PSD channels is shown as a function of parallel momentum transfer $Q_{||} = 4\pi \sin\theta / \lambda$ on a double-logarithmic scale. The normalized decay of the specularly reflected beam as a function of $Q_{||}$ is plotted for comparison. Here we have kept α_i , α_f constant, and moved the detector from $2\theta = 0^\circ$ to 90° . In this range the intensity decreases by about four orders of magnitude, reflecting the decay of the height-height self-correlation of the interfaces. At $Q_{||} = 1 \text{ \AA}^{-1}$ corresponding to $2\theta = 13^\circ$ the intensity decay leads over to a value, that is determined by amorphous scattering. In the high angle region $2\theta > 13^\circ$ we observe an incoherent superposition of the amorphous structure factor of the W and Si layers, that give rise to characteristic peaks. Because of the higher density and larger atomic form factor of the W layers, the intensity is dominated by the structure factor of the W layers.

Thus, the scattering geometry used in this experiment allows for a quantitative distinction between regions in Q -space dominated by the divergence of the primary beam, the interface roughness and the amorphous scattering, respectively [8]. In order to extract the roughness exponent h , we need to specify a "window" in Q -space, where the measured intensity is dominated by scattering from interface roughness. Towards low $Q_{||}$ this window is limited by the parasitic scattering of the primary (and hence the specular) beam, towards high $Q_{||}$ by amorphous scattering. However, from Fig.2 it can be seen, that in between these limits a reasonable comparison to theoretical curves can be made. The slope of the intensity decay gives access to the roughness parameter h . Curves calculated from eqn.(2) are drawn for values of $h=0.1$ and 0.3 . They obviously do not explain our experimental results. However, if a logarithmic roughness characterized by a correlation function $C(R) := A - B \ln(R)$ is assumed instead of the power law behaviour of eqn.(1), where A and B are constants, we find reasonable agreement between theory and experiment. This curve is indicated by " $h=0$ " in Fig.3.

Logarithmic roughness is expected for systems growing according to the linear version of the growth equation of Kardar, Parisi, and Zhang (KPZ) in 2+1 dimensions [9]. The results of the Q_z -resolved data also indicate that the growth of the multilayer was governed by the KPZ equation. It has been analysed in a previous paper [10]. The Q_z -resolved data contains information on the degree of conformality, i.e. on how correlated the fluctuations of different interfaces are as a function of the spatial Fourier component. Roughly speaking, it is found that the interfaces are correlated at long wavelength, but uncorrelated at short wavelength of the fluctuations.

Fig. 3: The Q_z -integrated intensity as a function of $Q_{||}$. At low $Q_{||}$ the finite width of the specularly reflected beam and the instrument dominate the measurement. At high $Q_{||}$ scattering from the amorphous state is the leading contribution. In between these limits the curve reflects the statistical properties of the interface roughness. With a logarithmic correlation function ($h=0$) a good data fit can be obtained, whereas a power law behaviour can clearly be excluded. For comparison, curves for $h=0.1$ and $h=0.5$ are drawn.

4. SUMMARY

In summary, we have shown that nonspecular scattering can be measured in a scattering geometry that is not constrained within the plane of reflection. Thus the full "roughness dominated window" in Q -space can be exploited and reliable results for the roughness exponent h can be obtained. In the case of the W/Si multilayer studied here, a logarithmic height-height correlation function is found. This behaviour is predicted by the linear version of the KPZ theory, if the statistical fluctuations of the deposition process during growth are smoothened by the local curvature of the interface.

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