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Diffuse scattering of hard x rays from rough surfaces

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The diffuse scattering of hard x rays from rough solid surfaces has been measured and described quantitatively in terms of an improved distorted-wave Born approximation. The rough surface is characterized by the rms roughness σ, the height-height correlation length ξ, and the roughness exponent h. The value for σ is in excellent agreement with that deduced from reflectivity. The significance of the parameters σ, ξ, and h is tested by comparison with the results obtained from scanning force microscopy.

The scattering of electromagnetic waves by rough surfaces or interfaces has been the object of research over many years (Refs. 1–6 and references quoted therein). The range of roughness which can be studied depends on the wavelength of the radiation. So, hard x rays with a wavelength in the angstrom range are especially suited for the investigation of flat surfaces with roughness in the nanometer regime. In most investigations done so far the specularly reflected contribution of the scattering has been used in order to deduce the rms roughness σ. Roughness exponentially reduces the Fresnel reflectivity by a Debye-Waller–type damping factor. Reflectivity measurements give no information on the correlation length ξ with which the roughness decays laterally. This information can be obtained from the diffuse component of scattering which is observed in a wide angular range around the specularly reflected beam. Sinha *et al.*[5] have described the diffuse scattering in the first distorted-wave Born approximation. Surface roughness is treated as a small perturbation of the smooth surface for which the exact solution is known from Fresnel theory. We will show in this paper that the first Born approximation, as given by Sinha *et al.*, is not able to describe the data quantitatively. We will present an extension of the theory and experimental data which are explained quantitatively by this improved theory. The extension of the theory consists of replacing the transmitted amplitude, which in the first Born approximation is that of the smooth surface, by its corresponding expression for the rough surface. It turned out from the comparison between theory and experiment that the first Born approximation fails when σ exceeds about 30 Å. Then the roughness can no longer be treated as a small perturbation. The results obtained by diffuse scattering on a number of metallic films are compared with those obtained by scanning force microscopy. This is another technique able to give the rms roughness σ and the correlation length. The finite radius of curvature of the tip is shown to feign a roughness smaller than it is in reality. To our knowledge it is the first time that a quantitative description of diffuse scattering in the hard x-ray range has been obtained and that the results have been compared to data obtained on the same samples by a completely different technique, like scanning force microscopy.

Specular reflection and diffuse scattering were measured at the beam line RÖMO 1 of the Hamburger Synchrotronstrahlungslabor. Details of the experimental setup have been described in Ref. 7. The scattered photons were measured with a photodiode and with a NaI scintillation counter.[8] The diffuse scattering was detected in a slit geometry, integrating over the direction ϕ_1 parallel to the surface of the sample and perpendicular to the plane of incidence (Fig. 1). Two detection modes were applied:

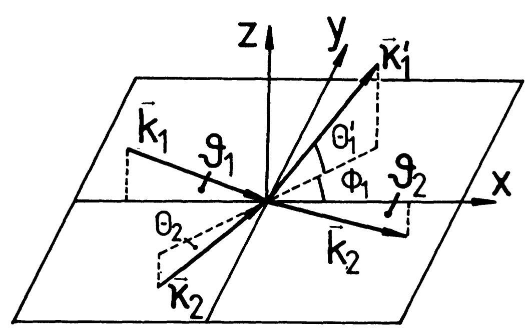

FIG. 1. X rays with wave vector $\mathbf{k}$ are scattered by a rough surface in the direction κ_1'. The pairs of wave vectors $\mathbf{k}_1,\mathbf{k}_2$ and κ_1',κ_2 are related to one another by Snell's law.

one with a fixed angle of incidence θ_1 and varying angle of scattering Θ_1', the other with fixed detector position θ_D and varying sample orientation keeping $\theta_1+\Theta_1'=\theta_D$ constant. The samples used in the present investigation were 4- and 6-in. silicon wafers covered with thin layers of aluminum: 6000 Å of *Al*Si with 1-at.% silicon deposited at 200 °C by sputtering and 4680 Å of *Al*Cu with 1.8-at.% copper deposited at 200 °C by evaporation in high vacuum. Figure 2 shows the reflected and the diffuse scattering at $\theta_1=0.5°$ from an *Al*Cu layer at 8000 eV photon energy. The critical angle of total external reflection is at 0.24°. The specularly reflected intensity shows up at $\Theta_1'=0.5°$. At the angle of total reflection $\Theta_1'=\theta_{1c}$ the diffuse scattering shows a peak. It is called the Yoneda peak and is a typical feature of the diffusely scattered intensity. In the following, we will briefly outline the theory which will enable us to describe quantitatively the experimental results and to extract the parameters which characterize the rough surface.

We consider a half infinite medium at $z<0$ with an index of refraction $n=1-\delta-i\beta$ and a rough interface which is described by a height-height correlation function.

$$C(\tau)\equiv\langle z(0)z(\tau)\rangle/\sigma_D^2=\exp\left[-\left(\frac{\tau}{\xi_D}\right)^{2h_D}\right]. \quad (1)$$

τ is the distance between two points in the x,y plane for which the height-height correlation is considered. Although on the right-hand side of Eq. (1) a special class of functions for the correlation has been chosen, this class is quite general in the sense that it describes smooth as well as jagged surfaces according to the choice of the exponent h_D. As discussed by Mandelbrot[9] h_D takes values between 0 and 1. Values h_D near 0 characterize very jagged surfaces. Values h_D near 1 describe smooth hills and valleys. For $h_D=1$ the correlation decays like a Gaussian and for $h_D=0.5$ as an exponential. σ_D is the rms roughness and the correlation length ξ_D is the scale on which

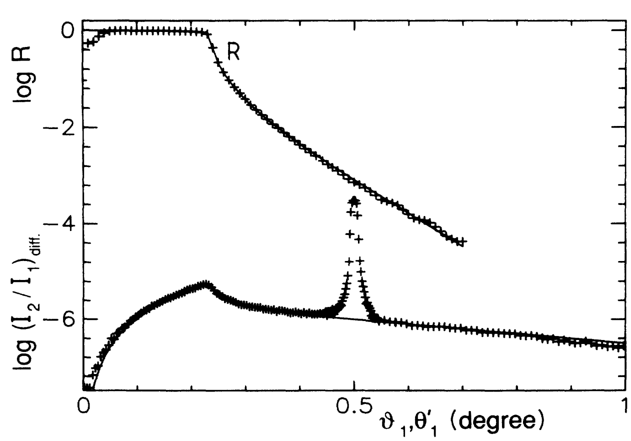

FIG. 2. Diffuse scattering $(I_2/I_1)_{\rm diff}$ and reflectivity R of 8-keV photons from a 468-nm *Al*Cu layer on a 4-in. silicon wafer. The angle of incidence θ_1 for the diffuse scattering is 0.5°. The detector opening is 1.52×10^{-3} for the reflectivity and 1.76×10^{-4} for the diffuse scattering. The fit of the data with Eqs. (2) and (4) is given by the solid lines. The logarithm is to the base 10.

the surface shows lateral correlation. A plane wave $\exp(i\mathbf{k}_1\cdot\mathbf{r})$ with wavelength $2\pi/k_1$ may fall in the x,z plane on the interface (Fig. 1). If the surface were ideally smooth we would expect a reflected and a refracted beam according to the Fresnel theory. Roughness reduces the reflected and increases the transmitted amplitude exponentially according to Refs. 3 and 7.

$$r_r(k_1)=\frac{\theta_1-\theta_2}{\theta_2+\theta_2}\exp[-2k_1^2\sigma_1^2\theta_1\theta_2]\,, \quad (2)$$

$$t_r(k_1)=\frac{2\theta_1}{\theta_1+\theta_2}\exp[+\tfrac{1}{2}k_1^2\sigma_1^2(\theta_1-\theta_2)^2]\,. \quad (3)$$

The preexponential factors are the Fresnel amplitudes in the small angle limit, appropriate for hard x rays. In this regime, s and p polarization of the light give the same result. Note the factor $\theta_1\theta_2$ in the damping factor of $r_r(k_1)$ which is different from the usual Rayleigh damping containing a factor θ_1^2 instead. In the hard x-ray range the version with $\theta_1\theta_2$ describes the experimental results whereas that with θ_1^2 does not.[7] The diffuse scattering produced by surface roughness has been described by Sinha *et al.* in the distorted-wave Born approximation (DWBA) and is given by the Eqs. (4.41) and (4.42) in their paper. Because we have used in our experiments a detector with a slit wide open parallel to the surface (the y direction in Fig. 1) the corresponding integration gives for the diffuse scattering

$$(I_2/I_1)_{\rm diff}=\frac{k_1^3}{8\pi^2\theta_1}|1-n^2|^2|t_r(\theta_1)t_r(\Theta_1')|^2S(q_z^t)\delta\Theta_1' \quad (4)$$

with

$$S(q_z^t)=|q_z^t|^{-2}\exp\left[-\frac{\sigma_1^2}{2}\{(q_z^t)^2+(q_z^{t*})^2\}\right]$$
$$\times\int_0^\infty d\tau\cos(q_r\tau)[\exp\{|q_z^t|^2\sigma^2C(\tau)\}-1]\,,$$
$$q_r=\tfrac{1}{2}k_1(\theta_1^2-\Theta_1'^2),\ q_z^t=nk_1(\theta_2+\Theta_2)\,.$$

The opening of our slit detector defines $\delta\Theta_1'=75\ \mu$m/425 mm $=1.76\times10^{-4}$. In the limit of $q_z^t\sigma\ll1$ the structure factor $S(\theta_1,\Theta_1')$ reduces to the Fourier transform of the height-height correlation function of the surface. It turned out that in our data analysis we have to use the full expression for $S(q_z^t)$ as given in Eq. (4).

Due to the standing-wave field which builds up in front of the reflecting surface the transmitted amplitudes $t(\theta_1)$ and $t(\Theta_1')$ have a maximum at the critical angle θ_{1c}. This is the reason for the Yoneda peak observed at θ_{1c} in Fig. 2. The product of the two transmitted amplitudes $t(\theta_1)t(\Theta_1')$ in Eq. (4) is a consequence of the invariance for reversal of the light path, i.e., for exchanging the light source and the detector. In the DWBA given by Sinha *et al.* the transmitted amplitudes $t(\theta_1)$ and $t(\Theta_1')$ are those of the ideally smooth surface, i.e., those without the exponential modifications given in Eq. (3). We cannot explain our data quantitatively in this approximation. If, on the other hand, we replace the transmitted amplitudes for the smooth surfaces by the transmitted amplitudes $t_r(\theta_1)$ and $t_r(\Theta_1')$ for the rough surface, as given by Eq. (3), the data can be explained in a satisfactory way by Eq. (4), at

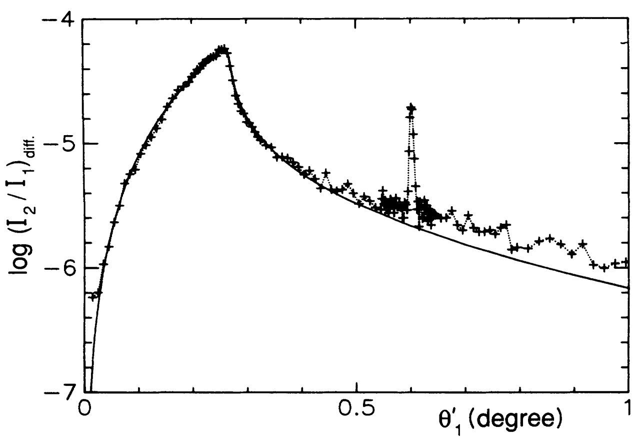

FIG. 3. Diffuse scattering $\log_{10}(I_2/I_2)_{\rm diff}$ of 7000-eV photons from a 600-nm *Al*Si layer on a 6-in. silicon wafer. The angle of incidence θ_i is 0.6°. The fit of the data with Eq. (4) is given by the solid line.

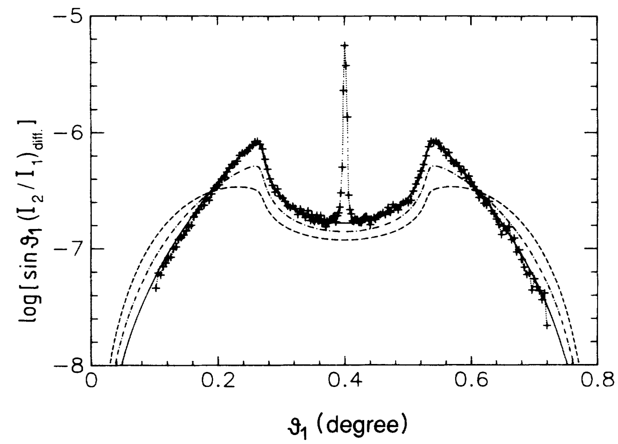

FIG. 4. Diffuse scattering $\log_{10}(I_2/I_1)_{\rm diff}$ of 7000-eV photons from the sample in Fig. 3. The detector forms with the incident beam an angle of 0.8°. The sample is rotated. The fit of the data with Eq. (4) is given as a solid line. The fits with the smooth transmitted amplitude as proposed by Sinha *et al.* (-·-·) and with the Rayleigh transmitted amplitude (---) do not describe the experimental results.

least for degrees of roughness which are not too strong. Figure 2 shows a fit of the experimental data with Eq. (4). The fit gives $\sigma_D = 17 \pm 2$ Å, $\xi_D = 1100 \pm 50$ Å, and $h_D = 0.7 \pm 0.1$. Also shown is a fit of the reflectivity according to Eq. (2). It gives $\sigma_R = 19 \pm 1$ Å in excellent agreement with the value found from diffuse scattering. A value for h_D of about 0.7 seems to be typical for many natural phenomena as observed by Hurst.[10] A detailed analysis of the data has shown that the small-angle data determine the value for σ_D, the high angle determines the exponent h_D, whereas the absolute height of the diffuse scattering determines the correlation length ξ_D. A second example of diffuse scattering is given in Fig. 3. The sample was the *Al*Si layer mentioned above. The reflectivity gives an rms roughness of $\sigma_R = 41 \pm 1$ Å, whereas the diffuse scattering gives $\sigma_D = 43 \pm 2$ Å, and $\xi_D = 4000 \pm 50$ Å. Again the rms roughnesses agree very well. It is noteworthy that the simulation of the data with our model [Eq. (4)] starts to fail at angles above 0.4°. This is due to breakdown of the first Born approximation. In other words, an rms roughness of 42 Å can no longer be treated as a small perturbation. Because the exponent h_D is mainly determined by the high angle data, its value can only be determined by this experiment to be about 1. Figure 4 shows a scan in the other detection mode, mentioned above, in which the detector is fixed and the sample is rotated. The sample is again the *Al*Si layer used in Fig. 3. The data are multiplied by $\sin\theta_i$ accounting for the change in the illuminated sample area. The symmetry in the curves about the angle of specular reflection expresses the reciprocity theorem of invariance against reversal of the optical path. The symmetry implies that the roughness is homogeneous over the illuminated sample area. The values for σ and ξ are identical to within the error with the parameters obtained in the other detection mode. Figure 4 shows, in addition, a fit of the data with the smooth transmission amplitudes as proposed by Sinha *et al.* and with the rough transmission amplitudes according to the Rayleigh description of roughness [which has a negative sign in the exponent of the exponential in Eq. (3)]. Obviously both models cannot describe the data quantitatively, whereas out extension [Eq. (4)] of the theory of Sinha *et al.* describes the experimental data in the whole angular range in a very satisfactory manner.

In order to see how the parameters σ, ξ, and h obtained by x-ray scattering compare with those obtained by other techniques not based on scattering we have probed both samples used in this investigation with a scanning force microscope. A vibrating tungsten cantilever ending in a tip with 100-nm radius of curvature was scanned over several areas 5×5 μm^2 in size of the sample in 128 lines. The distance between sample and tip was kept constant at about 200 Å. We have calculated the height distribution dN/dZ and the autocorrelation function $C(\tau)$. They are shown in Fig. 5 for the *Al*Si sample. The correlation is Gaussian ($h_F = 1.00 \pm 0.02$) with a correlation length

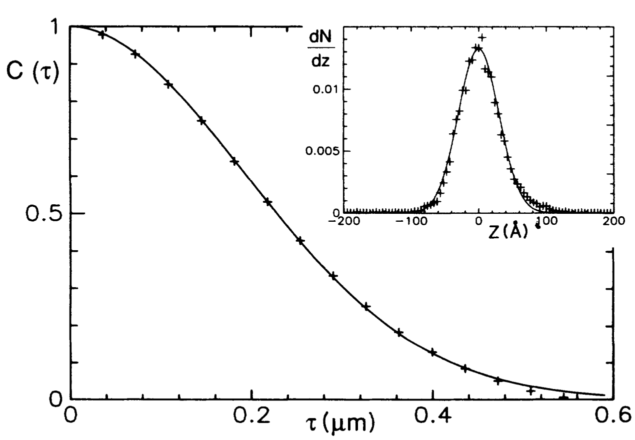

FIG. 5. Height-height correlation function $C(\tau)$ and height distribution dN/dz for the sample from Figs. 3 and 4 measured by scanning force microscopy. The data are best described by Gaussian distributions (solid lines).

$\xi_F = 2800 \pm 110$ Å. dN/dz is also Gaussian with $\sigma_F = 32 \pm 1$ Å. The values for σ deduced from force microscopy and from x-ray techniques differ substantially. The force microscopy value is smaller. This is due to the finite size of the *W* tip which cannot enter into deep crevices. This effect is especially pronounced in surfaces with a correlation length shorter than the radius of curvature of the tip. Therefore the force microscopy feigns surfaces smoother than they are in reality. This conclusion is supported by the data obtained on the *Al*Cu sample (Fig. 2). Here, the force microscopy data are $\sigma_F = 4 \pm 0.5$ Å, $\xi_F = 1200 \pm 170$ Å, and $h_F = 0.7 \pm 0.06$, compared to the x-ray data $\sigma_D = 17 \pm 2$ Å, $\xi_D = 1100 \pm 50$ Å, and $h_D = 0.7 \pm 0.1$.

To conclude, we have improved the description of diffuse x-ray scattering from rough surfaces, given by Sinha *et al.* and are now able for the first time to quantitatively describe the intensity scattered by rough solid surfaces. Our description replaces the transmission amplitudes for the smooth surface, used by Sinha, by those for the rough surface as given by Nevot and Croce, where roughness destroys the fixed phase relationship between the incoming, reflected, and transmitted amplitudes. It is noteworthy that our description of the diffuse scattering [Eq. (4)] fulfills the reciprocity theorem (which is confirmed by our experimental data). This is in contrast to a recent model given by Pynn,[11] who uses the transmission of the incoming wave through the smooth surface and the transmission of the scattered wave through the rough surface. In contrast, our approach gives a consistent description for the influence of the roughness on the reflected, transmitted, and diffusively scattered intensity. The description of the transmissivity through rough surfaces presented here is also relevant for x-ray diffraction and depth profiling by x-ray fluorescence at grazing incidence from samples with rough surfaces.

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