

Surface roughness of sputtered ZrO_2 films studied by atomic force microscopy and spectroscopic light scattering

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ZrO_2 films were prepared by reactive sputtering. Elastic light scattering was used to determine the cross correlation of the substrate and film interface roughness. Surface profiles were measured with atomic-force microscopy. The power spectral density functions could be fitted by the K -correlation model, suggesting self-affine fractal surfaces. The roughness of the film front surfaces was of the same order of magnitude as the substrate roughness. We have derived a replication factor from experimental data that gives information on the evolution of the contribution of the substrate roughness. [S1063-651X(96)10810-2]

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INTRODUCTION

Thin films are used in many applications and there are numerous methods of producing thin films with specific properties. The surface roughness of thin films is of interest in various applications. Often, one wants to minimize surface roughness, e.g. in optical components [1], while other applications favor rough or porous surfaces, e.g. thin films for electrochromic applications [2]. In this context, the description of surface roughness and its implications for various physical properties is a subject of intense and growing interest. Theoretical studies have been concerned with the scaling of surface roughness as a function of film thickness and lateral length scale [3,4]. Frequently thin film surfaces are found to be self-affine fractals over a considerable length scale. Experimental studies have been carried out on films produced by a variety of methods such as evaporation, sputter deposition, molecular-beam epitaxy, and chemical-vapor deposition [3,5].

The influence from substrate roughness on the growth of thin films has not been studied very much. However, simple approximations for the power-spectral density function of resulting film-front surface roughness do exist [6,7]. A more detailed understanding of the interaction between substrate roughness, smoothing, and roughening is vital in many cases. One example is thin films produced by sputtering, which frequently exhibit surface roughness of the same order of magnitude as conventional glass substrates. Sputter-deposited films are used in many applications, but the scaling of surface roughness has not been extensively studied.

There are several methods of characterization of surface roughness. Most methods are based either on the measurement of a surface profile or on the measurement of radiation scattered by the surface. In the former category are stylus profilers [8] and scanning-force microscopes [9], e.g. the atomic-force microscope (AFM) [10]. In the latter are methods based on x-ray [11] and optical scattering [12]. The measurement of optical scattering from transparent films can give information on the roughness of the two interfaces of a film and the statistical cross correlation, which is important when the influence of the substrate roughness on the surface roughness of the film is to be studied.

In this paper, we study the surface roughness of ZrO_2 thin films sputter-deposited onto glass substrates. We combine AFM and light scattering in order to study the evolution of surface roughness with film thickness and the influence of substrate roughness. We use AFM to characterize the roughness of the film-front surfaces and the substrate, and optical scattering to obtain information on the cross correlation between the film interfaces. We also combine the two methods in order to obtain a replication factor that describes the evolution of the substrate-roughness contribution to the resulting thin film roughness. This replication factor gives important information on the film-growth process.

THEORY

A rough surface can be described by the power-spectral density function (PSD), $g(\mathbf{K})$, which is the square of the Fourier transform of the surface profile $z(\mathbf{r})$ [13]:

$$g(\mathbf{K}) = \lim_{A \rightarrow \infty} \frac{1}{A} \left| \int_A z(\mathbf{r}) \exp(i\mathbf{K} \cdot \mathbf{r}) d\mathbf{r} \right|^2. \quad (1)$$

\mathbf{K} is the spatial wave vector, A an area of integration, and $\mathbf{r}=(x,y)$ the surface vector.

A surface can also be described by an autocorrelation function. The PSD function and the autocorrelation function form a Fourier transform pair. A thin film has two interfaces, each one of which can be described by a PSD function. The statistical correlation between the interfaces requires a third PSD function. The PSD functions describing the roughness of a thin film are [13]

$$g_{nm}(\mathbf{K}) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G_{nm}(\boldsymbol{\tau}) \exp(i\mathbf{K} \cdot \boldsymbol{\tau}) d\boldsymbol{\tau}, \quad (2)$$

where, G_{nm} is the correlation function, defined as

$$G_{nm}(\boldsymbol{\tau}) = \lim_{A \rightarrow \infty} \frac{1}{A} \int_A \int_A z_n(\mathbf{r}) z_m(\mathbf{r} + \boldsymbol{\tau}) d\mathbf{r}. \quad (3)$$

If $n=m$, g_{nm} is the PSD function of interface n . If $n \neq m$, g_{nm} is the cross PSD function, describing the statistical correlation between interfaces n and m .

Another useful quantity is the cross-correlation coefficient [14] g_{12}/g_{22} . If $g_{12}/g_{22}=1$, interface 2 is perfectly correlated to interface 1; if $g_{12}/g_{22}=0$, the interfaces are completely uncorrelated.

The front surface of a thin film, grown on a rough substrate, depends on the replication of substrate roughness and the additive roughness [6,7]. If a film is deposited onto a rough substrate, with roughness profile $z_1(\mathbf{r})$, two statistically independent processes can take place: replication of the substrate roughness and additive roughness, i.e.,

$$z_2(\mathbf{r}) = a(\mathbf{r}) * z_1(\mathbf{r}) + z_\mu(\mathbf{r}) \quad (4)$$

Here, $z_2(\mathbf{r})$ is the profile of the front surface of the film. The replication is described by the convolution (*) of the substrate roughness $z_1(\mathbf{r})$, with $a(\mathbf{r})$. The additive roughness $z_\mu(\mathbf{r})$ is independent of $z_1(\mathbf{r})$.

The PSD function of the front surface, g_{22} , is given by [6,7]

$$g_{22}(\mathbf{K}) = g_a(\mathbf{K})g_{11}(\mathbf{K}) + g_{\mu\mu}(\mathbf{K}), \quad (5)$$

where g_{11} is the PSD function of the substrate and $g_{\mu\mu}$ that of the additive roughness. The cross PSD function is given by

$$g_{12}(\mathbf{K}) = \mathcal{F}\{a(\mathbf{r})\}g_{11}(\mathbf{K}), \quad (6)$$

where the replication factor $\mathcal{F}\{a(\mathbf{r})\}$ is the Fourier transform of $a(\mathbf{r})$. Furthermore,

$$g_a(\mathbf{K}) = |\mathcal{F}\{a(\mathbf{r})\}|^2. \quad (7)$$

EXPERIMENT

A. Sample preparation

The ZrO_2 films were produced using a dc magnetron sputtering system [15]. The substrate was Corning borosilicate glass no. 7059. The films were made by reactive sputtering with a Zr target. The base pressure of the deposition chamber was below 10^{-5} Pa. During deposition, the total pressure was held at 1.0 Pa, with an argon and an oxygen flow of 100 sccm each. The plasma current was 6.0 A and the target potential was 290 V. No substrate heating was used. The distance between the target and the substrate was 10 cm. The mean free path of the atoms in the plasma is approximately 1 cm at 1.0 Pa. Since the mean free path is much shorter than the target-substrate distance, the atoms hit the substrate with a uniform distribution of incidence angles.

B. Optical characterization

The optical characterization was made by using a spectroscopic total integrated scattering instrument [16]. The total reflectance and transmittance spectra were measured in the wavelength range $0.4 < \lambda < 1.0 \mu\text{m}$. Diffuse reflectance and transmittance spectra were measured in the same wavelength range in the angle intervals, 2.5° – 10° , 10° – 20° , 20° – 40° , 40° – 70° , and 2.5° – 70° .

The film thickness and dielectric function were determined from the total spectra by using the method of Manificier, Gasiot, and Fillard [17]. The data were in good agreement with those reported by Andersson, Veszelei, and Roos [18]. The thickness d were 0.11, 0.18, 0.21, 0.34, 0.41, and $0.66 \mu\text{m}$.

C. Atomic-force microscopy

Surface profiles were obtained with a Nanoscope III atomic-force microscope. The cantilever, made of etched silicon, had a tip with a radius of 10 nm and an apex angle of 35° . The contact force was approximately 10^{-7} N and scans were taken over areas of $1 \times 1 \mu\text{m}$, $5 \times 5 \mu\text{m}$ and $50 \times 50 \mu\text{m}$, with a resolution of 256×256 pixels.

RESULTS

Figure 1 shows the surface profiles for the scan sizes $1 \times 1 \mu\text{m}$, $5 \times 5 \mu\text{m}$, and $50 \times 50 \mu\text{m}$ as obtained with the AFM for one of the samples ($d=0.34 \mu\text{m}$). The surface seems to be dominated by roughness at the length scale of a few tenths of a μm up to $10 \mu\text{m}$. The surface profiles of the other samples had a similar appearance.

The PSD functions, calculated from the surface profiles of the AFM measurements, are shown in Fig. 2. The PSD function of each sample was thus calculated from three surface profiles, with different scan area. The use of different scan areas gives PSD functions over a larger K interval. The arithmetic average was taken where PSD functions from two scan sizes overlapped. The $1 \times 1 \mu\text{m}$ scans were slightly disturbed by tip artifacts [19] which manifest themselves in the bending of the PSD functions at $K > 10^2 \mu\text{m}^{-1}$.

The cross-correlation coefficient g_{12}/g_{22} was obtained from the diffuse optical spectra by using a method [20] based on the differences in the spectral behavior of the scattering in different angle intervals. The spectral behavior of diffuse spectra from a thin film strongly depends on the roughness cross correlation between the interfaces [21], and roughness on different length scales causes scattering in different angles. In Fig. 3, g_{12}/g_{22} versus K is shown for the ZrO_2 films. There is a clear tendency for the correlation to decrease with d and, in particular, the shift from correlated to uncorrelated occurs at lower K . Note that for the thickest film ($d=0.66 \mu\text{m}$) no correlation could be found on the length scales that the method is sensitive to.

The shape of the PSD function is approximately characterized by the K -correlation model [22,23]. The analytical expression of the K -correlation model for a two-dimensional surface is

$$g(\mathbf{K}) = \frac{A}{[1 + (BK)^2]^{(C+1)/2}}. \quad (8)$$

C determines the slope at high K values, B the position of the ‘‘knee,’’ and A the value at small K . The K -correlation model is useful, since it describes self-affine fractal surfaces with a crossover region. The fractal dimension is given by $D_f = (7 - C)/2$. B is equal to the correlation length and the crossover length $L_c = 4B$ [23]. PSD functions given by Eq. (8) were fitted to the PSD functions determined from AFM profiles for all the samples. The self-affinity of the surfaces

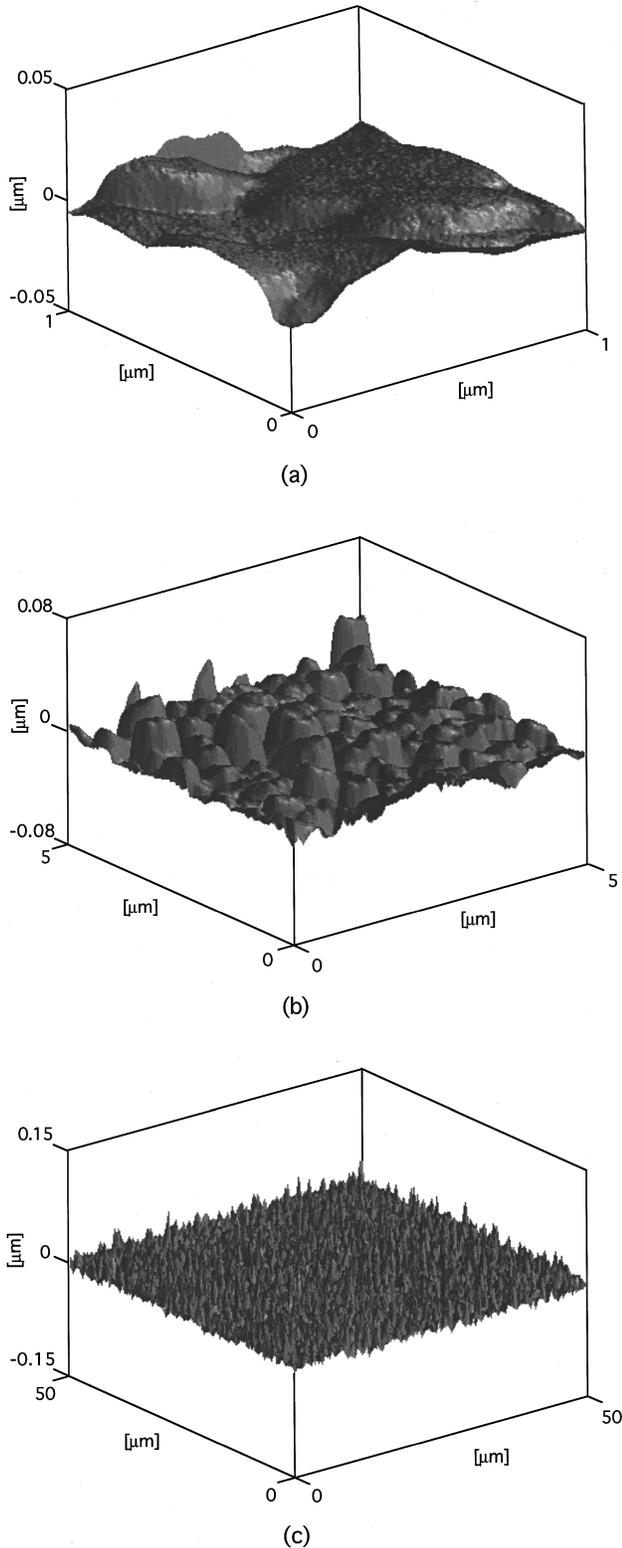


FIG. 1. Surface profiles of a ZrO₂ film (thickness $d=0.34 \mu\text{m}$) at scan sizes of (a) $1 \times 1 \mu\text{m}$, (b) $5 \times 5 \mu\text{m}$, and (c) $50 \times 50 \mu\text{m}$, as obtained from the AFM.

cannot be regarded as definitely proven due to the noisy experimental PSD functions (cf. Fig. 2). However, we obtain an effective D_f from the fit to the K -correlation model, suggesting that a self-affine model may at least be a good approximation.

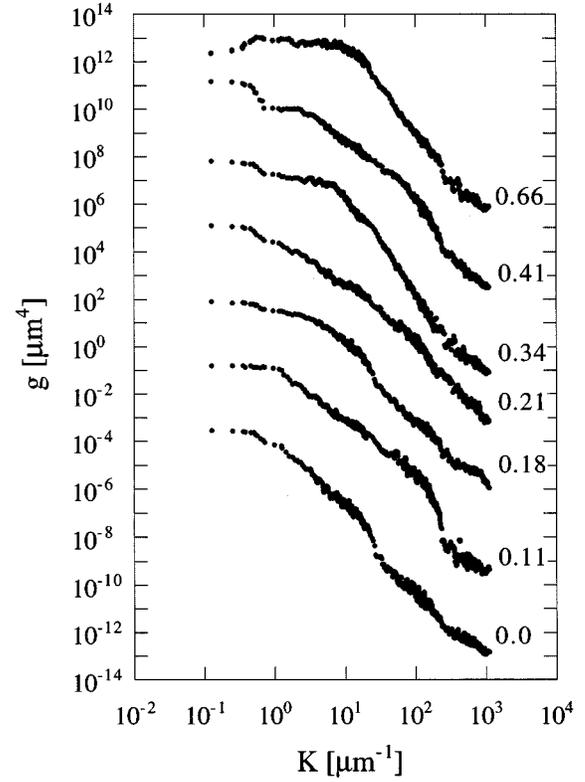


FIG. 2. PSD functions calculated from AFM surface profiles. The PSD functions of the uncoated glass and of films with increasing thickness (in μm) are shown. The curves have been displaced with a multiplicative factor of 10^3 .

The bandwidth-limited rms roughness δ was calculated by integrating the fitted PSD functions:

$$\begin{aligned} \delta &= \left(\frac{1}{(2\pi)^2} \int \int_{K_A} g(\mathbf{K}) d\mathbf{K} \right)^{1/2} \\ &= \left(\frac{1}{2\pi} \int_{K_{\min}}^{K_{\max}} g(K) K dK \right)^{1/2}. \end{aligned} \quad (9)$$

Here, K_A is the integration area in the K plane, determined by the bandwidth limits of the AFM measurement. The last equality is valid for isotropic rough surfaces, where K_{\min} and K_{\max} are the maximum and minimum K values, respectively. Figure 4 shows δ versus d . These δ values are larger than those of the separate AFM profiles due to their larger bandwidths. There is a weak tendency for δ to increase with d . The rms roughness δ is not much higher for the films than for the glass substrate. The assumption of a negligible substrate roughness cannot be made in our case.

D_f versus d is shown in Fig. 5; D_f is between 2 and 2.5 for all samples. There is a tendency for D_f to decrease with increasing d , but scatter in the data and the errors in the determination are too great to verify this. The data from the $1 \times 1\text{-}\mu\text{m}$ scans were disturbed by tip artifacts. If these data were not included in the fit to Eq. (8) giving the determination of D_f , slightly different values were obtained. The errors in D_f were estimated from these differences.

Figure 6 shows L_c versus d . The crossover length L_c is decreasing with increasing d . The crossover length of the

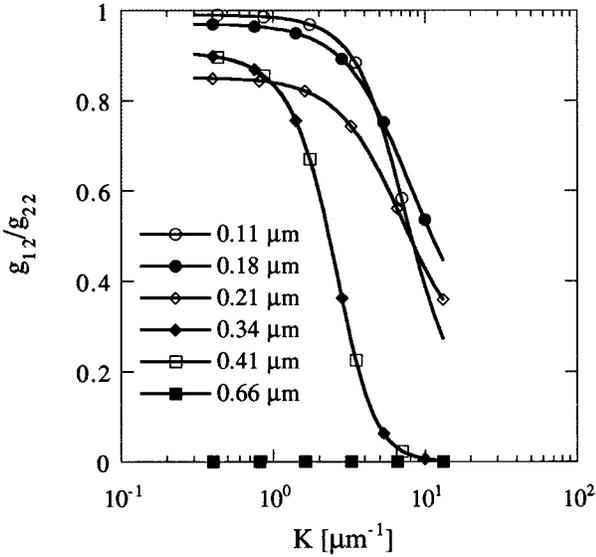


FIG. 3. The cross-correlation coefficient g_{12}/g_{22} vs spatial wave number K for the ZrO_2 films. Note that the curves for $d=0.34$ and $0.41 \mu\text{m}$ cannot be separated, and that for $d=0.66 \mu\text{m}$ no correlation can be seen.

cross-correlation function, denoted L_{12} , is also shown. L_{12} was determined from the curves in Fig. 3, and increases with increasing d .

We have determined the PSD functions of the glass substrate and the film-front surfaces from AFM measurements, and the cross-correlation coefficient g_{12}/g_{22} from light-scattering measurements. Equation (6) can then be rearranged:

$$\mathcal{F}\{a(\mathbf{r})\} = \left(\frac{g_{12}}{g_{22}} \right) \frac{g_{22}}{g_{11}}. \quad (10)$$

The replication factor can be calculated by using the cross-correlation coefficient g_{12}/g_{22} , obtained from the light-scattering experiments, g_{11} from the AFM measurements of the uncoated glass, and g_{22} from the AFM measurements of the film-front surfaces. Figure 7 shows the replication factor versus K for films of different thicknesses. One generally assumes the replication factor to be below one and to de-

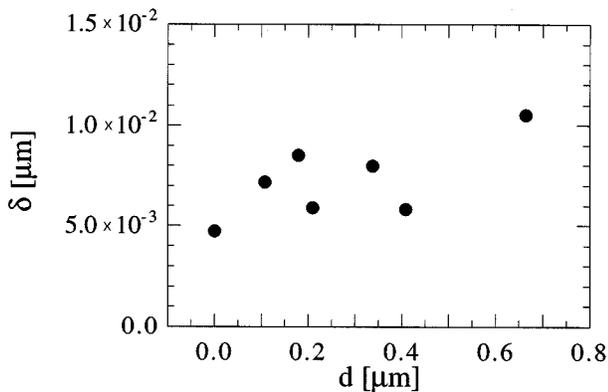


FIG. 4. The rms roughness δ vs film thickness d of the ZrO_2 films.

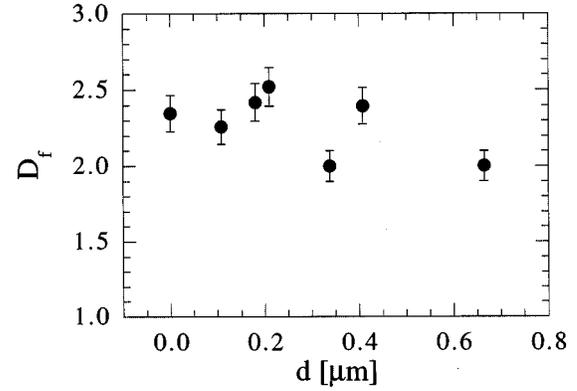


FIG. 5. The effective dimension D_f vs film thickness d for the ZrO_2 films. Estimated error bars are also shown.

crease monotonically with K . For some of our films, $\mathcal{F}\{a(\mathbf{r})\}$ reaches a maximum, higher than one, before it decreases with increasing K . The errors in the determined $\mathcal{F}\{a(\mathbf{r})\}$ are large. We do, however, believe that the maxima in $\mathcal{F}\{a(\mathbf{r})\}$ are significant.

DISCUSSION

The roughness growth of self-affine fractal surfaces can be described by scaling laws. They involve a growth exponent β and a roughness exponent α [24]. Thus, for a self-affine fractal surface, δ increases with d :

$$\delta \propto d^\beta. \quad (11)$$

L_c increases with d .

$$L_c \propto d^{\beta/\alpha}. \quad (12)$$

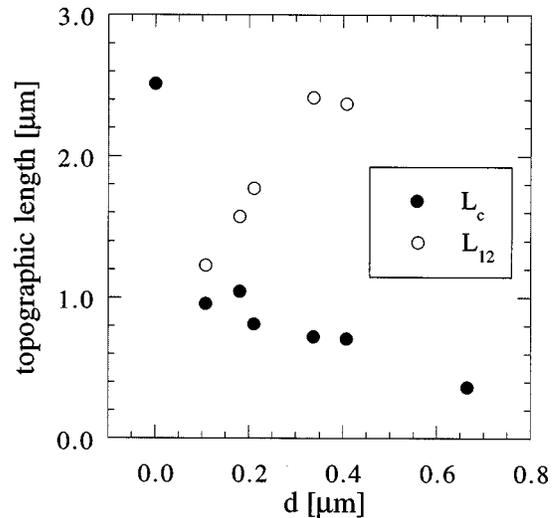


FIG. 6. The crossover lengths of the film-front surface L_c , and of the cross-correlation coefficient L_{12} vs film thickness d .

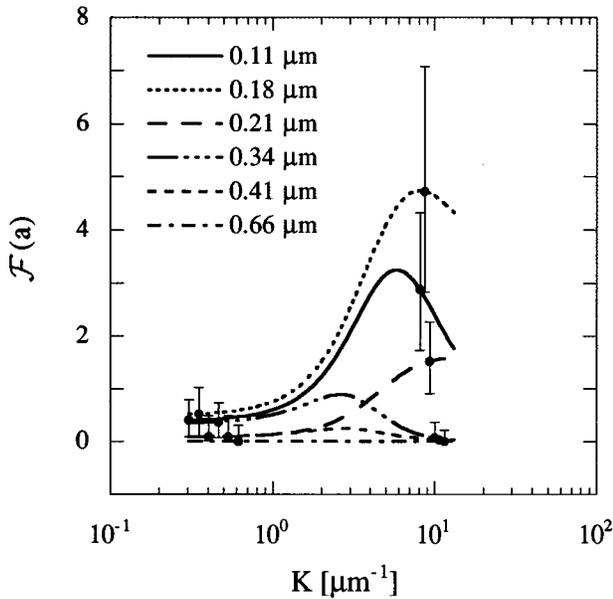


FIG. 7. The replication factor $\mathcal{F}(a)$ vs K as obtained from combining the AFM and light-scattering data. The estimated errors are given as bars at some values of K .

For length scales shorter than L_c (large K), the PSD function is determined by α and is independent of d . α is related to D_f by

$$D_f = 3 - \alpha. \quad (13)$$

In Fig. 4, δ versus d does not seem to obey a power law. The results are not clear due to scatter in the data and that the substrate roughness is not negligible. The crossover length L_c decreases with d (cf. Fig. 6) instead of increasing as in Eq. (12), and D_f is not independent of d (cf. Fig. 5). The surface-roughness growth seems to follow other statistics than the growth of a self-affine fractal surface despite the fact

that the roughness statistics of each surface can be described by the K -correlation model. Furthermore, the replication factor does not decrease monotonically with K as expected. We believe that the reason for this is that the roughness of the film is not negligible in comparison with the substrate roughness and that the interfaces are partially correlated.

We propose that the roughness growth is the sum of three processes.

(I) Additive roughness from the deposition process. This roughness, described by $g_{\mu\mu}$, is statistically independent of the substrate roughness and increases with thickness.

(II) Smoothing effects due to surface diffusion. This effect causes $\mathcal{F}\{a(\mathbf{r})\}$ to decrease with increasing K for a fixed d and to decrease with increasing d for a fixed K .

(III) Shadowing effects, which cause preferential growth of surface roughness on a specific length scale.

For random incidence, shadowing effects have been described by the *grass model* [25,26]. In our case we have random incidence, since the mean free path is much less than the target-to-substrate distance. Shadowing effects are most important in the first stages of the film growth and give the maximum in $\mathcal{F}\{a(\mathbf{r})\}$. Smoothing effects become more important for thicker films. This may be because the temperature increases with deposition time. For the thickest films, the substrate roughness is smoothed out and only the additive roughness remains. Thus, due to the combination of surface diffusion, shadowing effects, and additive roughness, the roughness of our films does not scale in the way generally expected for self-affine rough film-front surfaces.

The important result reported in this paper is that the substrate roughness seems to have significant impact on the roughness of sputtered films and that this impact is different at different length scales and different film thicknesses. A further investigation could include comparison of films grown on rough and smooth substrates and the influence of substrate temperature. A film grown on a rougher substrate is likely to show a higher correlation between the two film interfaces.

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