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Roughness spectrum and surface width of self-affine fractal surfaces via the K-correlation model

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Theoretical expressions for the height-height correlation function of self-affine fractal surfaces are discussed in comparison with scanning tunneling microscopy, correlation and surface-width data obtained from rough silver and gold films. Fourier transformations are used to compare with equilibrium phenomena, and lead to a correlation model with an associated roughness spectrum of analytic form.

I. INTRODUCTION

In nature a wide variety of surfaces occurs with roughness well described in terms of self-affine fractal scaling, for example, the nanometer-scale topology of vapor-deposited films, spatial fluctuations of liquid-gas interfaces, and the kilometer-scale structure of mountain terrain.[1] Physical processes which produce such a kind of surface morphology include fracture, erosion, molecular-beam epitaxy, as well as fluid invasion of porous media.[2]

In most of the investigations done so far, a specific form of the height-height correlation function for self-affine fractal surfaces has been used to model x-ray reflectivity and scanning tunneling microscopy (STM), as well as atomic force microscopy (AFM) data,[3-5] which read as $C_S(R)=\sigma^2 e^{-(R/\xi)^{2H}}$ and describe the real height-height correlation in many cases sufficiently well.[6] This is the well-known Kohlrausch-Williams-Watts (KWW) function introduced in 1863 to describe mechanical creep in glassy fibers, and later used by Williams and Watts to describe dielectric relaxation in polymers,[7] as well as lately to fit miscellaneous experimental data including NMR, dynamic light scattering, quasielastic neutron scattering, kinetics reactions, magnetic relaxation, etc.[8] Currently it is well known that this relaxation law can be derived from several different physical or mathematical models based on distributions of relaxation times to complex correlated processes.[9]

However, this relaxation law does not address the question for the height-height correlation in the limit $H\to 0$ which is related with the existence of a lower fractal bound, and is of fundamental interest in account of the equilibrium as well as the nonequilibrium roughening transition; rather it reveals a trivial behavior. Furthermore, the Fourier transform of $C_S(R)$ does not have, in general, an analytic expression, excluding therefore explicit calculation of other relevant surface properties. In addition, it is well known that the computation of the Fourier transform has numerical problems originating from cutoff effects which yield undesirable oscillations, especially when treating real data.

Our purpose in this paper is to investigate the possibility of a surface height-height correlation being described by a form, which has similarities with correlations related to thermal roughening transitions,[10,11] capillary waves in liquids,[3,12] and with nontrivial behavior in the limit $H\to 0$ (logarithmic roughness), as well as with an associated roughness spectrum of analytic form for the whole range of values of the roughness exponent H, $0\le H<1$. The latter favors the analytic calculation of various surface properties with the most important for roughness measurements taken directly by means of STM, the surface width $\sigma(L)$, where L represents a linear length scale on the surface.[13,14] Furthermore, the knowledge of analytic forms for the roughness spectrum is important in a wide variety of roughness studies including, for example, x-ray scattering,[3-6] light scattering,[15] as well as quartz crystal microbalance studies.[16]

II. SURFACE MODEL

Consider a rough surface in terms of a solid-on-solid model (SOS) in which the surface height $z(\mathbf{r})$ is considered a single-valued random function of the in-plane positional vector $\mathbf{r}=(x,y)$. We make the central assumption that the difference $z(\mathbf{r})-z(\mathbf{r}')$ is a random Gaussian variable whose distribution depends on the relative coordinates $(x'-x,y'-y)$, and we write $\langle[z(\mathbf{r})-z(\mathbf{r}')]^2\rangle = g(\mathbf{R})$, $\mathbf{R}=\mathbf{r}'-\mathbf{r}$. The notation $\langle\ \rangle$ means an ensemble over all possible choices of the origin average, and $g(\mathbf{R})$ is the height-difference correlation function. For many isotropic solid surfaces the height-difference correlation can be represented by

$$g(R)=A_0 R^{2H}\quad (0<H<1)\ . \tag{2.1}$$

A_0 is $\propto$ proportionality constant. If $H>1$, at large length scales the interface fluctuations can exceed the system size which is physically impossible. This kind of surface roughness is related to the self-affine surface, defined by Mandelbrodt in terms of fractional Brownian motion.[17] The exponent H is indicative of the surface texture, and is associated with a local fractal dimension $D=3-H$.[9,17] When $R\to\infty$, $g(R)\to\infty$, but $g(R)/R^2\to 0$ since $H<1$ (asymptotically flat surface). This is a rather ideal case because on real surfaces $g(R)$ may saturate to the value $2\sigma^2$ [$\sigma=\langle z(\mathbf{0})^2\rangle^{1/2}$] for many reasons, finite size being one such reason, since a surface with this kind of power-law roughness does not have a

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well-defined mean position which implies the existence of an effective roughness cutoff such that for $R \ll \xi$, $g(R) \propto R^{2H}$.[3,4] Therefore, $g(R)$ for real self-affine surfaces has the following behavior:

$$g(R) \propto R^{2H}, \quad R \ll \xi, \tag{2.2a}$$

$$g(R) = 2\sigma^2, \quad R \gg \xi. \tag{2.2b}$$

The length scale ξ is called the in-plane correlation length which together with H controls how far a point on the surface can move before losing memory of the initial value of its z coordinate. In general, $g(\mathbf{R})$ is related to the height-height correlation function $C(\mathbf{R}) = \langle z(\mathbf{R})z(\mathbf{0})\rangle$ by means of the equation

$$g(\mathbf{R}) = 2\sigma^2 - 2C(\mathbf{R}). \tag{2.3}$$

Equation (2.3) for the correlation function $C_S(R)$ yields $g_s(R) = 2\sigma^2(1 - e^{-(R/\xi)^{2H}})$ which is consistent with the asymptotic behavior in terms of Eqs. (2.2).

In Fig. 1, we illustrate the applicability of the correlation function $C_S(R)$ with a fit to correlation data from a silver (Ag) film of thickness ~ 100.0 nm, deposited on a polished quartz crystal held at 106 K by means of thermal evaporation. The deposition rate was 0.3 Å/sec, and the film thickness was monitored by means of a quartz crystal microbalance (QCM).[18] The sample was left to be annealed until it reached room temperature after deposition. The system base pressure was $\sim 1.0 \times 10^{-8}$ Torr, and during the deposition $\sim 5 \times 10^{-7}$ Torr. The STM measurements have been performed under dry N_2 gas atmosphere, and four files of correlation data recorded at different locations on the surface have been averaged. The scan size was 500.0 nm, with 400 points per line scan. The power-law fit permits direct measurement of H uniquely which gives $H = 0.12 \pm 0.05$. The correlation length is estimated from the value $C(R)\sigma^{-2}$, assuming $R = \xi$, which is e^{-1} for $C_S(R)$. The resulting correlation length, $\xi_s = 12.3 \pm 0.5$ nm (↑) is close to the regime where the correlation turns significantly away from the linear behavior, showing therefore its real significance for the corresponding surface morphology, as well as comparing favorably with cluster sizes obtained from the STM images.

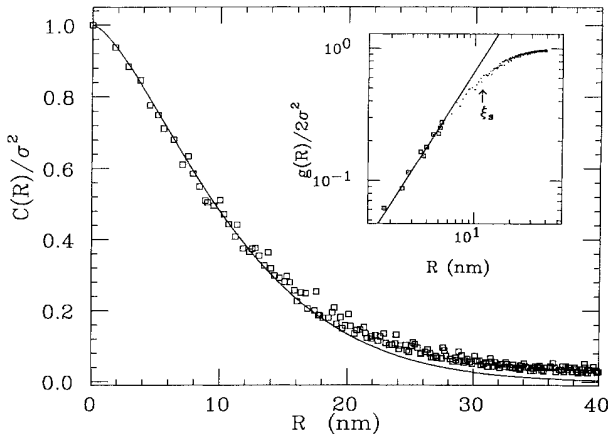

FIG. 1. Fit of $C_S(R)/\sigma^2$ to correlation data for the silver (Ag) film (106 K) of thickness 100.0 nm, with $\xi_s = 12.3$ nm and $H = 0.12$. The inset depicts a power-law fit for $g(R)$ in order to determine H, which yields $H = 0.12 \pm 0.05$. The arrow indicates the position of the correlation length ξ_s and the squares the regime of length scales at which the power-law fit was performed.

III. ROUGHNESS SPECTRUM

Let us denote the average macroscopic sample surface by A. If we define the correlation function by

$$C(\mathbf{R}) = \frac{1}{A}\int \langle z(\mathbf{P}+\mathbf{R})z(\mathbf{P})\rangle d^2P \tag{3.1}$$

and the Fourier transform of $z(\mathbf{R})$ by

$$z(\mathbf{k}) = \frac{1}{(2\pi)^2}\int z(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}d^2R. \tag{3.2}$$

The Wiener-Khinchin theorem yields[19]

$$\langle |z(\mathbf{k})|^2\rangle = \frac{A}{(2\pi)^6}\int C(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}d^2R. \tag{3.3}$$

Since $C(\mathbf{R})$ is assumed to be isotropic, angular integration yields

$$\langle |z(\mathbf{k})|^2\rangle = \frac{A}{(2\pi)^5}\int_0^\infty RC(R)J_0(kR)d^2R. \tag{3.4}$$

Figure 2 shows roughness spectra for small and large values of H in order to be pointed out also in k-space similarities and differences in behavior of the associated correlation functions. The observed linear regime in Fig. 2 for $k\xi \gg 1$, corresponds to a power law $\sim k^{-2(1+H)}$ which permits the determination of the roughness exponent H from Fourier profile analysis.[20] Another interesting property of the roughness spectra is the location of the *knee* regime with respect to the in-plane correlation length ξ (see Fig. 2). An estimation of ξ directly from the location of the *knee* regime, *as has been some-*

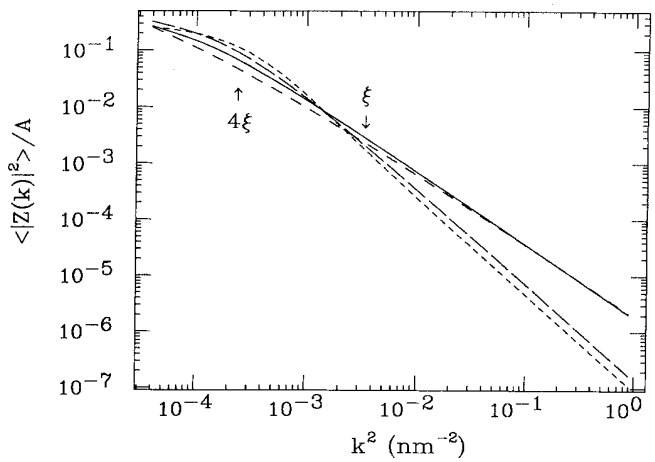

FIG. 2. Roughness spectra are depicted for $\langle |z(\mathbf{k})|^2\rangle_{sf}/A$, $H = 0.3$ (solid line), and $H = 0.7$ (short-dashed line), as well as for $\langle |z(\mathbf{k})|^2\rangle_S/A$ related to $C_S(R)$; $H = 0.3$ (dashed line), $H = 0.7$ (long-dashed line). During the calculation we used; $\xi = 100.0$ nm, and $\sigma = 0.7$ nm. The arrows indicate the corresponding positions of ξ as well as 4ξ in k space in order to point out their relation with respect to the knee regime.

times assumed, can be misleading since the corresponding length scale is approximately 4ξ which is expected since the *knee* regime corresponds to the point where the associated correlation will start having an insignificant effect, $R \geq 4\xi$.

The Fourier transform of $C_S(R)=\sigma^2 e^{-(R/\xi)^{2H}}$ in two-dimensions for values of the roughness exponent H in the interval $0<H<1$ has an analytic form only for $H=0.5$,

$$\langle |z(\mathbf{k})|^2 \rangle = \frac{A}{(2\pi)^5} \frac{\sigma^2\xi^2}{(1+k^2\xi^2)^{3/2}} , \tag{3.5}$$

from which it can be observed that the roughness exponent $H=0.5$ enters the roughness spectrum as a power of $1+k^2\xi^2$ and not of k^2 individually. On the other hand, in temporal relaxation phenomena a similar empirical expression in the frequency domain has been introduced by Haviliak and Negami (HN) to describe the behavior of glass-forming liquids, $\phi(\omega)\sim[1+(i\omega\tau_0)^c]^{-\gamma}$ with the parameters c and γ ranging from 0 to 1.[21] Because of the nonanalytic expression for the Fourier transform of the KWW function $\sim e^{-(\tau/\tau_0)^H}$, as well as its accurate description of real data, Alvarez *et al.* established a relationship among the parameters of these two models which is not an analytic one since these two functions are not exactly Fourier transforms of each other.[22]

However, for two-dimensional spectra the previous observation from Eq. (3.5) related to the k dependence of $\langle |z(\mathbf{k})|^2 \rangle$ signals already the possibility of the existence of a wider family of roughness spectra with an analytic form similar to the HN function, as well as close to those of the $C_S(R)$ function which can describe the real correlation significantly well (Fig. 1). In addition, comparison with relevant equilibrium as well as nonequilibrium cases in the limit $H\rightarrow 0$ will support such a conjecture. Therefore, we turn to a more intuitive method that captures the essence of exact calculations as far as critical behavior is concerned, and leads also to a calculation of the correlation functions.

IV. *K*-CORRELATION FUNCTIONS

From equilibrium phenomena theory a simple form of the free-energy functional evaluated in terms of long-wavelength Fourier components, which can describe the qualitative features of the thermal roughening transition for vicinal or stepped surfaces, was introduced by Jorge *et al.*,[23] namely, $F=\frac{1}{2}\sum_k [nk_y^2+n'k_x^2]|u(k)|^2+U\sum_{my}[1-\cos 2\pi u_m(y)]$. Replacing the cosine by its second-order Taylor expansion, we obtain a harmonic free energy which easily yields an average value below the transition temperature,

$$\langle |u(\mathbf{k})|^2 \rangle \sim \frac{1}{1+ak_y^2+bk_x^2} \tag{4.1}$$

with an associated correlation function $\langle [u_m(y)-u_0(0)]^2 \rangle \propto A-BK_0(p/\xi)\propto \ln(p)$ for $p \ll \xi$; $p^2=(n'/n)y^2+(n/n')m^2$.

The previous formulation captures the qualitative features of a more generalized model for the roughening problem described by Villain, Grempel, and Lapujoulade[10] which can reduce to the SOS model studied by Chui and Weeks under the correspondence $u_m(y)\rightarrow z(\mathbf{R})$ in a discrete version and is known to have a roughening transition.[11] Furthermore during the surface growth under nonequilibrium conditions highly anisotropic phenomena occur, reflected in the stationary case through the roughness exponent H in terms of a power-law roughness $\propto R^{2H}$ of which a limiting case can be considered the logarithmic roughness at $H\rightarrow 0$; $\lim_{H\rightarrow 0}(1/2H)[R^{2H}-1] \rightarrow \ln(R)$. On the other hand, the limiting behavior of the power-law roughness at $H\rightarrow 0$ has its own counterpart, the nonequilibrium analog of the roughening transition which has been predicted to occur at temperatures close to where the equilibrium case is expected as well as with correlation $g(R)\propto \ln(R)$ in the stationary phase.[24]

Therefore following the previous reasoning and comparing Eqs. (3.5) and (4.1), it is plausible to examine the possibility of a more general class of height-height correlation functions for self-affine fractals with an associated roughness spectrum depending mainly on H as a power of $(1+k^2\xi^2)$ of the following form:

$$\langle |z(\mathbf{k})|^2 \rangle_a = \frac{A}{(2\pi)^5} \frac{\sigma^2\xi^2}{(1+ak^2\xi^2)^{1+H}} , \tag{4.2}$$

which distinguishes itself from the equilibrium analog equation (4.1), through the exponent H. The parameter a in the denominator of Eq. (4.2) has been introduced in order to accommodate values of H other than 0.5. Moreover, an examination of the consequences and implications of such an assumption has to be pursued as far as consistency with the self-affine nature [$g(R)\propto R^{2H}$; $0<H<1$] and logarithmic behavior [$g(R)\propto \ln(R)$; $H\rightarrow 0$] is concerned.

Inverse Fourier transformation of $\langle |z(\mathbf{k})|^2 \rangle_a$ in Eq. (4.2) yields

$$C_a(R)=\sigma^2\xi^2 \int_0^{k_c} \frac{kJ_0(kR)}{(1+ak^2\xi^2)^{1+H}} dk . \tag{4.3}$$

The lower limit of integration in Eq. (4.3) is to the order of $A^{-1/2}$ which for A of macroscopic size can be completely neglected. $k_c=\pi/a_0$ represents an upper cutoff since at an atomic level we do not expect any fractal behavior. For comparison with a lattice model a_0 is identified with the lattice spacing. During the following calculations the limit $H\rightarrow 0$ will be considered in terms of the scheme

$$\lim_{H\rightarrow 0} \frac{1}{H}[x^H-1]\rightarrow \ln(x) . \tag{4.4}$$

In the continuum limit $g(R)$ is related to the roughness spectrum by the following equation:

$$g(\mathbf{R})=2\frac{(2\pi)^4}{A}\int \langle |z(\mathbf{k})|^2 \rangle [1-e^{i\mathbf{k}\cdot\mathbf{R}}]d^2\mathbf{k} \tag{4.5}$$

which, according to Eq. (2.3), imposes the following normalization condition for $\langle |z(\mathbf{k})|^2 \rangle_a$:

$$\frac{(2\pi)^4}{A}\int_{0<k<k_c} \langle |z(\mathbf{k})|^2 \rangle_a d^2\mathbf{k}=\sigma^2 . \tag{4.6}$$

Equation (4.6) permits the determination of the parame-

ter a in a consistent manner for the whole regime of values of the roughness exponent H.

Integration of Eq. (4.) for $0<H<1$ yields

$$a_{\rm sf}=\frac{1}{2H}[1-(1+a_{\rm sf}k_c^2\xi^2)^{-H}] \tag{4.7}$$

which in the limit $H\to 0$ according to Eq. (4.4) takes the form

$$a_{\rm lg}=\tfrac{1}{2}\ln[1+a_{\rm lg}k_c^2\xi^2]\ , \tag{4.8}$$

depicting at the same time the significance of a lower fractal bound or discreteness effect in order for the logarithmic behavior to be recovered from a power law. The notation sf means self-affine fractal and lg logarithmic roughness. Equations (4.7) and (4.8) define in general the parameter $a=a(\xi/a_0,H)$ for the whole range of values of the roughness exponent H, $0\le H<1$.

In the continuum limit, since $a_0\ll\xi$ the integration in Eq. (4.3) can be extended to ∞ which yields in terms of the integral identity,

$$\int_0^\infty k^{1+v}J_v(kr)(k^2+x^2)^{-1-p}dk=\frac{r^p x^{v-p}}{2^p\Gamma(p+1)}K_{v-p}(xr) \tag{4.9}$$

valid for $-1<R(v)<2R(p)+\frac{3}{2}$,[25]

$$C_a(R)=\frac{\sigma^2}{a\Gamma(1+H)}\left[\frac{R}{2\xi\sqrt{a}}\right]^H K_H\left[\frac{R}{\xi\sqrt{a}}\right]\ . \tag{4.10}$$

The functional $K_H(x)$ means the second kind of Bessel function of H order from which the characterization K correlations comes.[25]

A. Power-law roughness correlation functions

In Eq. (4.7) for $H>0$ and $a_0\ll\xi$, neglecting discreteness effects we obtain $a_{\rm sf}\approx 1/2H$. With a substitution in the small-length scale ($R\ll\xi$) expansion of $g_a(R)=2\sigma^2-2C_a(R)$ in terms of Eq. (4.10),

$$g(R)_{\rm sf}\approx 2\sigma^2\left[1-\frac{1}{2Ha_{\rm sf}}\right]+\frac{\sigma^2\Gamma(1-H)}{a_{\rm sf}H\Gamma(1+H)}\left[\frac{R}{2\xi\sqrt{a_{\rm sf}}}\right]^{2H} \tag{4.11}$$

we obtain, finally,

$$g(R)_{\rm sf}\approx\frac{2\sigma^2\Gamma(1-H)}{\Gamma(1+H)}\left[\frac{R}{2\xi\sqrt{a_{\rm sf}}}\right]^{2H}\ , \tag{4.12}$$

which confirms consistency with the self-affine nature in the continuum limit and $0<H<1$ as is defined in terms of Eqs. (2.2). Equation (4.10) for $a_{\rm sf}\approx 1/2H$ yields

$$C(R)_{\rm sf}=\frac{2H\sigma^2}{\Gamma(1+H)}\left[\frac{R\sqrt{2H}}{2\xi}\right]^H K_H\left[\frac{R\sqrt{2H}}{\xi}\right] \tag{4.13}$$

which for $H=0.5$ is equivalent to $C_S(R)$. In addition, $C(R)_{\rm sf}$ reveals an inversion of its decay rate as a function of H at a length scale $R\sim 2.5\xi$, in a manner similar to

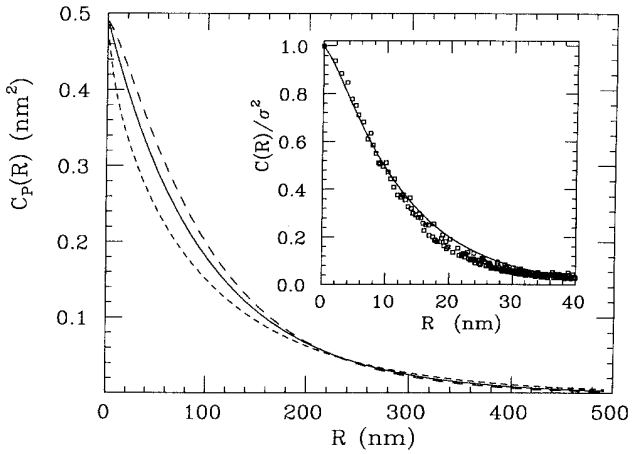

FIG. 3. Schematics for $C(R)_{\rm sf}$ correlation function, $\sigma=0.7$ nm; $\xi=100.0$ nm, $H=0.3$ (short-dashed line), $H=0.5$ (solid line), $H=0.7$ (long-dashed line). The inset shows a fit of $C(R)_{\rm sf}/\sigma^2$ (solid line) to correlation data (squares) of the cold silver (Ag) film (106 K), with $H=0.69$, and correlation length $\xi_{\rm sf}=19.1$ nm.

$C_S(R)$ for which the inversion occurs at $R=\xi$ (Fig. 3). The inset in Fig. 3 depicts a fit of $C(R)_{\rm sf}$ to the previous cold silver film (106 K) correlation data with $H=0.65$, and $\xi_{\rm sf}=11.6$ nm, which shows clearly that the $C(R)_{\rm sf}$ model is a realistic one since the fit parameters are in between the limits of H and ξ obtained previously, and with an associated roughness spectrum given by

$$\langle|z(\mathbf{k})|^2\rangle_{\rm sf}=\frac{A}{(2\pi)^5}\frac{\sigma^2\xi^2}{\left[1+\dfrac{k^2\xi^2}{2H}\right]^{1+H}}\ . \tag{4.14}$$

However, it is noteworthy to point out that the approximation $a_{\rm sf}\approx 1/2H$ as the ratio ξ/a_0 decreases is more consistent with higher values of H. If we go beyond this approximation, we can estimate the length scale $R_{\rm sf}^0$ for which the continuum limit breaks down as $\propto$ function of H, ξ, and a_0. Since from Eq. (4.7) $a_{\rm sf}\le 1/2H$, if we consider Eq. (4.11), $R_{\rm sf}^0$ can be defined from the condition $g_{\rm sf}(R)=0$ which yields $R_{\rm sf}^0\approx 2\xi\sqrt{a_{\rm sf}}(1-2Ha_{\rm sf})^{1/2H}$.

B. Logarithmic roughness correlation functions

Therefore, if we consider the existence of a lower fractal bound logarithmic roughness follows naturally from the power-law roughness in the limit $H\to 0$, with the corresponding correlation function in the continuum limit,

$$C(R)_{\rm lg}=\frac{\sigma^2}{a_{\rm lg}}K_0\left[\frac{R}{\xi\sqrt{a_{\rm lg}}}\right] \tag{4.15}$$

and an associated roughness spectrum given by

$$\langle|z(\mathbf{k})|^2\rangle_{\rm lg}=\frac{A}{(2\pi)^5}\frac{\sigma^2\xi^2}{1+a_{\rm lg}k^2\xi^2}\ . \tag{4.16}$$

Equation (4.15) for $R\ll\xi$ yields $g(R)_{\rm lg}\approx 2\sigma^2+2(\sigma^2/a_{\rm lg})\ln(R/2\xi\sqrt{a_{\rm lg}})$ which alterna-

tively can be from Eq. (4.11) in the limit $H \to 0$ in account of Eq. (4.4). The corresponding lower limit for the validity of the continuum approximation is given in this case by $R^0_{lg} \approx 2\xi\sqrt{a_{lg}}e^{-a_{lg}}$.

It is interesting at this point to examine the conditions under which Eq. (4.8) has a solution, as well as the relation of such a solution to the universal amplitude $2/\pi^2$ [$g(R) \approx (2/\pi^2)\ln(R/a_0)$], observed in equilibrium Kosterlitz-Thouless phase transition.[26] Equation (4.8) can be rewritten in the form $e^{2a} = 1 + ak_c^2\xi^2$, and if we denote by $f = e^{2a}$ and $X = 1 + ak_c^2\xi^2$, the necessary and sufficient condition for a nontrivial ($a_{lg} > 0$) solution to exist reads as $(df/da)_0 < (dX/da)_0$. This condition is satisfied since for $\xi \gg a_0$ we obtain $(df/da)_0 = 2 \ll (dX/da)_0 = (\pi\xi/a_0)^2$. Figure 4 shows a numerical solution of Eq. (4.8) where a saturated behavior of a_{lg} at large correlation lengths is observed. The latter is expected since $(da_{lg}/d\xi) = 2a_{lg}k_c^2\xi/(1 + 2a_{lg}k_c^2\xi^2) \sim 1/\xi \ll 1$ for $\xi \gg a_0$. The crossover with the value $2/\pi^2$ occurs for correlation lengths $\xi \sim 10^3 a_0$ in an increasing manner as a_0 becomes larger (solid line, Gig. 4). This behavior is in remarkable agreement with equilibrium cases,[10] where the corresponding prefactor approaches the value $2/\pi^2$ for temperatures near to the transition temperature, $T \leq T_c$, with the correlation length diverging as $\ln(\xi) \approx |(T - T_c)/T_c|^{1/2}$. As can be observed in the inset of Fig. 4, the two curves collapse into each other which is expected since a_{lg} is determined by the ratio ξ/a_0 in Eq. (4.8). The reason we compare only the prefactor $2/a_{lg}$ with the universal value $2/\pi^2$ (excluding σ^2) is related to the fact that a measure of the height-height density fluctuations in the continuum limit is provided by the relative interface fluctuation, $g_{a_{lg}}(R)/\sigma^2$ with the prefactor $2/a_{lg}$ characterizing its strength. In various models describing the irreversible growth of surfaces under nonequilibrium conditions, the observed logarithmic behavior occurs in some cases with a prefactor having the universal value $2/\pi^2$,[24] as well as with the nonuniversal value in some other cases.[27] In our case, the transition from power-law to logarithmic roughness in the stationary phase occurs with a prefactor which has a nonuniversal character, however, it approaches the universal constant $2/\pi^2$ as the correlation length increases with the crossover value to the order of $10^3 a_0$. Therefore the logarithmic behavior as a limiting case of the power-law roughness bears characteristics close to those observed in corresponding equilibrium as well as nonequilibrium systems,[10,11,24,27] depicting in that way the relevance of our arguments.

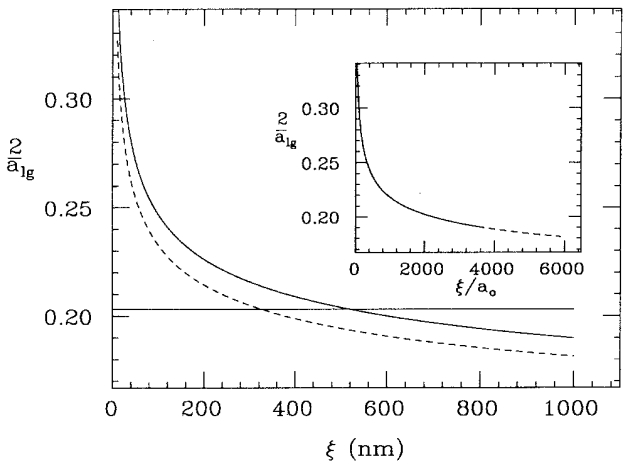

FIG. 4. Numerical solution of Eq. (4.8) where the behavior of $2/a_{lg}$ is depicted as a function of ξ for $a_0 = 0.17$ nm (short-dashed line), and $a_0 = 0.27$ nm (solid line). The solid line parallel to the x axis corresponds to the universal value $2/\pi^2$. The inset depicts the collapse toward each other of the previous two curves in agreement with the fact that a_{lg} is determined by the ratio ξ/a_0.

In the case of liquid surfaces, or purely two-dimensional systems, it is well known that if we ignore finite-size effects, the capillary-wave fluctuations cause $g(R)$ to follow logarithmic behavior,[3,12] $g(R) \sim \ln(R)$. For liquids finite-size effects due to gravity or finite depth cut off the long-wavelength surface modes causing $g(R)$ to be given by (gravitational cutoff) $g(R) \sim 2\sigma^2 - BK_0(R)^3$, comparing directly to $g(R)_{lg} \sim 2\sigma^2 - B'K_0(R)$. In general for $H \to 0$, $D = 3 - H \to 3$, which is a rather subtle situation since a three-dimensional object can be either a fractal or a volume. For the case of a fractal, Eq. (4.15) represents a plausible candidate.

V. SURFACE WIDTH

A characteristic and important surface quantity that represents a measure of the correlations along the direction of surface growth is the surface width. In the stationary phase the surface width, $\sigma(L)$, scales with the linear size L on the surface for self-affine fractals as follows. $\sigma(L) \sim L^H$ for $L \ll \xi$ and $\sigma(L) \to \sigma$ for $L \gg \xi$.[14] Measurement of $\sigma(L)$ in various physical systems has been performed in order to explore the scaling properties of the involved surface morphology,[22,28] and stands as a suitable technique for roughness characterization by means of STM especially for surfaces with large-scale roughness, $\xi \geq 100.0$ nm (Fig. 4). For the case of surfaces with a shorter roughness scale $\xi \leq 100.0$ nm, the appropriate alternative is a correlation function measurement (Figs. 1 and 6, details will be given elsewhere). The surface width for a surface section of linear size L in the continuum limit is given by [29]

$$\sigma^2(L) = \int_{2\pi/L < k,k' < k_c} \langle z(\mathbf{k})z(\mathbf{k}')\rangle d^2\mathbf{k}\, d^2\mathbf{k}' \,. \quad (5.1)$$

Since the surfaces we consider are stationary stochastic processes,

$$\langle z(\mathbf{k})z(\mathbf{k}')\rangle = \frac{(2\pi)^4}{A}\delta(\mathbf{k}+\mathbf{k}')\langle |z(\mathbf{k})|^2\rangle \,. \quad (5.2)$$

Substitution in Eq. (5.1) from Eqs. (5.2) and (4.2), carrying out the integration yields for $0 < H < 1$ and $k_l = 2\pi/L$

$$\sigma^2(L)_{sf} = \frac{\sigma^2}{2Ha_{sf}}[(1 + a_{sf}k_l^2\xi^2)^{-H} - (1 + a_{sf}k_c^2\xi^2)^{-H}] \quad (5.3)$$

which for $a_0 \ll \xi$ and $a_{sf} \approx 1/2H$ is simplified to have the form $\sigma^2(L)_{sf} \approx \sigma^2(1 + k_l^2\xi^2/2H)^{-H}$. Equation (5.3) in the

limit $H \rightarrow 0$ with Eq. (4.2) yields

$$\sigma^2(L)_{\lg} = \frac{\sigma^2}{2a_{\lg}} \ln \left[\frac{1+a_{\lg}k_c^2\xi^2}{1+a_{\lg}k_l^2\xi^2} \right] . \quad (5.4)$$

For $L \ll \xi$, Eqs. (5.3) and (5.4) yield $\sigma(L)_{sf} \approx (2\pi)^{-H} a_{sf}^{-(1+H)/2} \sigma (L/\xi)^H$ and $\sigma(L)_{\lg} \approx (\sigma/\sqrt{a_{\lg}}) \ln^{1/2}(L/2a_0)$, respectively. In the opposite limit $\sigma(L) \rightarrow \sigma$ in both cases, as can easily be verified by the defining equations for the parameter a.

The *knee* regime for $\sigma(L)$ corresponds to a length scale approximately 4ξ in agreement with our earlier comment for the location of the roughness spectrum knee regime. The inset in Fig. 5 shows a fit to surface width data from a previous work by Krim *et al.*[13] (Fig. 2 in Ref. 13) for a gold (Au) film with large-scale roughness $\xi \sim 2000.0$ nm, in order to illustrate the applicability of Eq. (5.3). It is noteworthy to point out that in this case because of STM head limitation, surface-width data acquisition for larger scan sizes was impossible. Therefore the only way to estimate σ and ξ easily and accurately (power-law fit gives H) is to proceed through a fit in terms of Eq. (5.3) where the *knee* regime ~ 8000.0 nm signals qualitatively an estimation of ξ as we explained previously.

Furthermore, we illustrate the relation of the surface-width *knee* regime with respect to ξ directly with comparison of correlation and surface-width data obtained from a rough silver film ~ 80.0 nm thick, Fig. 6. The experimental conditions during film preparation and measurement, were similar to those of the low-temperature (106 K, Fig. 1) film except that the sample was held at room temperature ~ 300 K during deposition. Five files of correlation data acquired from STM images with 400 points per line scan and scan size 500.0 nm have been averaged. The inset depicts a fit to surface-width data of the same film by means of Eq. (5.3) with $H=0.70$, $\sigma=2.08$ nm, and $\xi_{sf}=24.0$ nm (↑). Each point of the surface-width data represents an average rms roughness of 5–10 images after planefit was performed in order to remove mainly the sample tilt. The value of the correlation length is approximately to $\frac{1}{4}$ of the *knee* regime length scale ~ 80–90 nm. The correlation length estimated directly from the regime where the correlation data in Fig. 6 deviate significantly from the linear behavior is roughly $\sim 20.0 \pm 5.0$ nm, $\sigma = 2.1$ nm which was calculated as the average rms roughness from the corresponding images used to acquire the correlation data, as well as the value of H in terms of a power-law fit; $H = 0.68 \pm 0.02$. It is worthwhile to point out that the slight oscillatory behavior of the correlation data in the regime of length scales 30–50 nm is reflected also on the surface width for the same approximate range of length scales as a steppedlike feature. Therefore, the comparison between correlation and surface-width data according to the specified limits of the surface parameters H and ξ confirm experimentally the theoretical prediction for the *knee* regime of the roughness spectrum and surface width, as well as depict the relevance of the corresponding formalism.

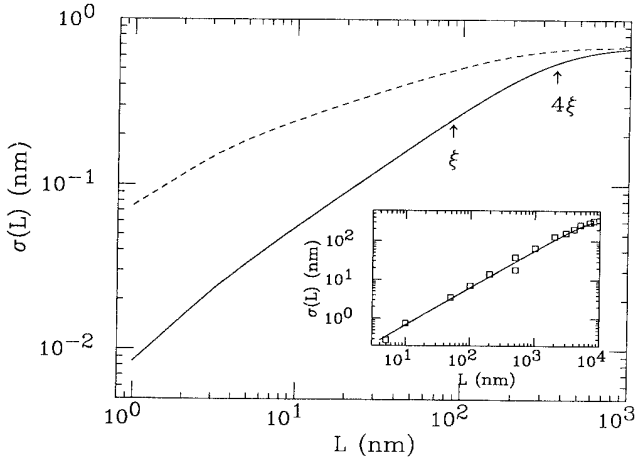

FIG. 5. Schematics for the surface width, Eq. (5.3) with $a_{sf} \approx 1/2H$, as a function of H, $a_0 = 0.27$ nm, $\xi = 100.0$ nm, and $\sigma = 0.7$ nm; $H = 0.3$ (short-dashed line), and $H = 0.7$ (solid line). The arrows indicate the positions of ξ and 4ξ in order to point out their relation with respect to the knee regime. The inset depicts a fit of the gold (Au) surface width data with parameters $H = 0.96$ (determined by power-law fit), $\xi = 2000.0$ nm, and $\sigma = 450.0$ nm.

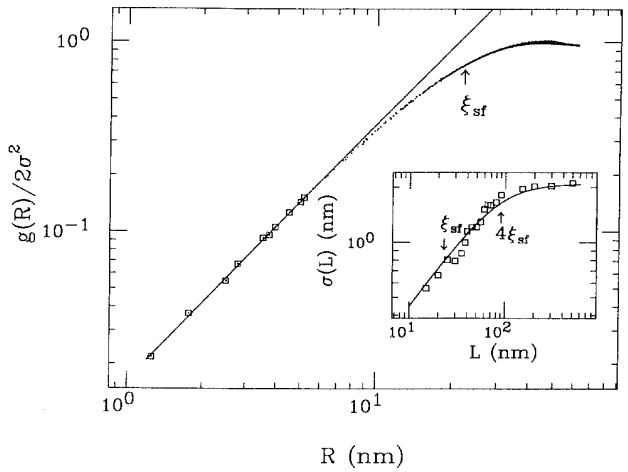

FIG. 6. Power-law fit for the room-temperature silver (Ag) film of thickness 80.0 nm in terms of $g(R)/2\sigma^2$ which gives $H = 0.68 \pm 0.02$ (squares indicate the regime of length scales for the power-law fit). The inset depicts a fit to surface width data of the same film in terms of Eq. (5.3) with $H = 0.70$, $\sigma = 2.08$ nm, and $\xi = 24.0$ nm. The arrows indicate the positions of ξ on the correlation data as well as on the surface width data.

VI. CONCLUSIONS AND DISCUSSION

In conclusion, we described a class of height-height correlation functions for self-affine fractals with the correct limiting behavior at $H \rightarrow 0$, and associated roughness spectrum of analytic form which enables straightforward calculation of other relevant surface properties, as well as enlarge the class of height-height correlations for self-affine fractal surfaces which are related to analogous

equilibrium phenomena as far as critical behavior is concerned, under a more general unifying scheme, Eqs. (4.7), (4.8), and (4.10). Although the assumption, Eq. (4.2), on which we based our derivation has an *ad hoc* nature, the qualitative comparison with exactly solvable models from equilibrium phenomena in the limit $H \to 0$ with the nonequilibrium analogs, justifies the relevance of our conjecture. Furthermore, the analytic calculation of the surface width can provide an estimation of surface parameters also for cases where the corresponding length scales cannot be probed in practice due to equipment limitations.

As a general comment on the importance of roughness spectra in indirect methods of surface roughness studies, we point out the following. The diffuse x-ray or neutron scattering differential cross section, $d\sigma(k)/d\Omega$, for incidence angles close to the angle of total external reflection, yields the Fourier transform of $C(R)$ which is given by Eq. (3.3), $d\sigma(k)/d\Omega \sim \langle |z(\mathbf{k})|^2 \rangle$.[3] Furthermore, in light scattering theories on surface roughness studies, the differential scattering cross sections either for scattering or adsorption (surface plasmons) of electromagnetic radiation due to the presence of surface roughness, are directly proportional to the roughness spectrum, Eq. (3.3).[15] Under the same frame in a recent work on QCM studies involving surface roughness, the frequency change is related directly to the roughness spectrum.[16] The previous examples show the evolution of surface roughness in a wide variety of surface studies, and therefore reflect the significance for the knowledge of analytic forms for the roughness spectrum. More specifically, this need is satisfied by the roughness spectra associated with the K-correlation functions, Eq. (4.2), (4.7), (4.8), and (4.10), not only because of its simple analytic form, but also because of the correct asymptotic behavior in the limit $H \to 0$ (logarithmic roughness) as far as critical behavior is concerned.

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