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Finite-size effects on self-affine fractal surfaces due to domains

George Palasantzas
Physics Department, Northeastern University, Boston, Massachusetts 02115
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We study the effects on the scaling properties of self-affine fractal surfaces due to domains where a distribution of domain sizes and shapes is simulated through a Gaussian function. Approximate expressions for the roughness spectrum and surface width are confirmed with comparison to surface-width data acquired by means of scanning tunneling microscopy.

I. INTRODUCTION

A wide variety of surfaces and interfaces occurring in nature are well represented by a kind of roughness associated with self-affine fractal scaling as defined by Mandelbrodt in terms of fractional Brownian motion.[1] Examples of physical processes which produce such surfaces include fracture, erosion, and molecular-beam epitaxy, as well as fluid invasion of porous media.[2]

Let us denote by $z(\mathbf{r})$ and $\mathbf{r}=(x,y)$ respectively the vertical and horizontal surface coordinates, with $z(\mathbf{r})$ being a single-valued function of the in-plane positional vector $\mathbf{r}$. Self-affine fractal surfaces exhibit fluctuations in the perpendicular direction which can be characterized in terms of the height-difference correlation $g(\mathbf{R}) = \langle [z(\mathbf{r})-z(\mathbf{r}')]^2 \rangle$, with $z(\mathbf{r})-z(\mathbf{r}')$ assumed a Gaussian random variable whose distribution depends on the relative coordinates $\mathbf{R}=\mathbf{r}'-\mathbf{r}$. The notation $\langle \cdots \rangle$ means an average over all possible choices of the origin, and an ensemble average over all possible surface configurations. If we assume an isotropic surface in the x and y directions and consider

$$g(R) \propto R^{2H} \quad (0<H<1) , \tag{1.1}$$

the associated surface roughness can be attributed to self-affine fractals as defined by Mandelbrodt in terms of fractional Brownian motion.[1] The exponent H characterizes the surface texture and is associated with a local fractal dimension $D=3-H$.[1,3] 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 the following equation:

$$g(\mathbf{R})=2\sigma^2-2C(\mathbf{R}) . \tag{1.2}$$

If $R\rightarrow\infty$, $g(R)\rightarrow\infty$ but $g(R)/R^2\rightarrow 0$ (asymptotically flat surface), which is a rather ideal case because on real surfaces finite-size effects will cause $g(R)$ at large length scales to saturate to the value $2\sigma^2$ [$\sigma=\langle z(\mathbf{0})^2\rangle^{1/2}$ being the saturated value at large length scales of the rms surface width], since a surface with such a power-law roughness does not have a well-defined mean position. This implies the existence of an effective roughness cutoff ξ. The length scale ξ is called the in-plane correlation length. Therefore, $g(R)$ for real self-affine surfaces has the following behavior[4]

$$g(R) \propto R^{2H} \quad (R \ll \xi) , \tag{1.3a}$$

$$g(R)=2\sigma^2 \quad (R \gg \xi) . \tag{1.3b}$$

Furthermore, ξ represents an intrinsic length scale, which together with H controls how far a point can move on a surface before losing memory of the initial value of its z coordinate.

In general, finite-size effects impose effective cutoffs on the long-wavelength surface fluctuations which cause the surface roughness to be bounded. The aim of this work is to correlate known information about various aspects of finite-size effects observed on rough surfaces under a general quantitative scheme. Its motivation lies in a recent study concerning iron-film surfaces, where surface-width measurements reveal scaling behavior which can be attributed to the presence of domains.[5] If the surface consists of a distribution of domain sizes and shapes, the various surface quantities in general have to be averaged over the domain distribution, which will affect their scaling properties in a manner that is determined mainly by the competition between the in-plane correlation length ξ, and some average domain size ζ. More specifically, if $\zeta \gg \xi$ the domain effect can be completely neglected, whereas for $\zeta \le \xi$ the average over the domain distribution will have a significant effect on the scaling properties.

In the present study, we shall consider a distribution of domains aligned parallel to each other and with the domain terrace to possess self-affine surface roughness.[6] In addition, following Dutta and Sinha,[7] we shall simulate the effect of a distribution of domain sizes and shapes through the radial Gaussian distribution function $\propto e^{-\pi R^2/\zeta^2}$, with ζ the average domain size. Physically, such a distribution can be understood from the fact that the formation of a domain of size R costs energy which is proportional to the domain area $\sim R^2$, and results in a Boltzmann distribution of domain sizes e^{-aR^2}. This kind of distribution has been widely used in the literature to accommodate finite-size effects in scattering theories, and is inherently associated with the Warren approximation.[4,7,8] But so far its direct applicability to surface quantities which are relevant from the experimental point of view in surface-roughness studies remains undeveloped.

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II. DOMAIN FORMALISM

In our case, finite size manifests itself in two ways, first through the correlation length ξ, and second through the average domain size ζ. We define the height-height correlation function $C(\mathbf{R})$ by[9]

$$C(\mathbf{R})=\frac{1}{A}\int \langle z(\mathbf{P}+\mathbf{R})z(\mathbf{P})\rangle d^2\mathbf{P} , \tag{2.1}$$

and the height Fourier transform by

$$z(\mathbf{k})=\frac{1}{(2\pi)^2}\int z(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}d^2\mathbf{R} . \tag{2.2}$$

The notation $\langle \cdots \rangle$ means an ensemble average, and the constant A represents the macroscopic area of the average smooth surface over which the height-height correlation is calculated for the case where no domains exist. The Wienner-Khinchin theorem yields[10]

$$\langle |z(\mathbf{k})|^2\rangle=\frac{A}{(2\pi)^6}\int C(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}d^2\mathbf{R} . \tag{2.3}$$

If the surface consists of a distribution of domain sizes and shapes, we can proceed under a similar framework to that used by Dutta and Sinha to derive an expression for the scattering structure factor of a finite two-dimensional lattice, as is outlined briefly below.[7] Thus, we write for the roughness spectrum according to Eq. (2.3)

$$\langle |z(\mathbf{k})|^2\rangle_d\sim\int_\zeta C(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}d^2\mathbf{R} , \tag{2.4}$$

where the integration is confined over an area of size $\sim\zeta^2$, and we Fourier-transform $C(\mathbf{R})$ in terms of a wave vector $\mathbf{q}$. If we make the Warren approximation in the continuum limit for the integral $\int_\zeta e^{-i(\mathbf{k}-\mathbf{q})\cdot\mathbf{R}}d^2\mathbf{R}$ over a finite area to the order of $\sim\zeta^2$ (Refs. 7,8)

$$\int_\zeta e^{-i(\mathbf{k}-\mathbf{q})\cdot\mathbf{R}}d^2\mathbf{R}\sim e^{-(\mathbf{k}-\mathbf{q})^2\zeta^2/4\pi} , \tag{2.5}$$

we obtain, after Fourier-transforming back to the real-space integral,

$$\langle |z(\mathbf{k})|^2\rangle_d\sim\int C(\mathbf{R})e^{-\pi R^2/\zeta^2}e^{-i\mathbf{k}\cdot\mathbf{R}}d^2\mathbf{R} , \tag{2.6}$$

where the integration can be extended over the whole x-y plane due to the presence of the Gaussian term, which expresses the average over the domain distribution of sizes and shapes. This result cannot in fact be obtained if the finite integral is performed over a single domain of specified shape and size; rather, the previous approximation takes into account the smearing of the roughness spectra due to the superposition from many domains.[7,11] Assuming the surface to be isotropic in the x and y directions, angular integration in Eq. (2.6) finally yields

$$\langle |z(\mathbf{k})|^2\rangle_d=\frac{A}{(2\pi)^5}\int_0^\infty C(R)e^{-\pi R^2/\zeta^2}RJ_0(kR)dR . \tag{2.7}$$

The knee regime of the roughness spectrum, Eq. (2.7), if $\zeta<\xi$ corresponds to a length scale ($kR\sim2\pi$) to the order of ξ. If $\zeta\geq\xi$, with ζ and ξ of the same order of magnitude, the knee regime determines mainly the domain size ζ; for $\zeta\gg\xi$ the corresponding length scale is approximately $\sim4\xi$ (Fig. 1).

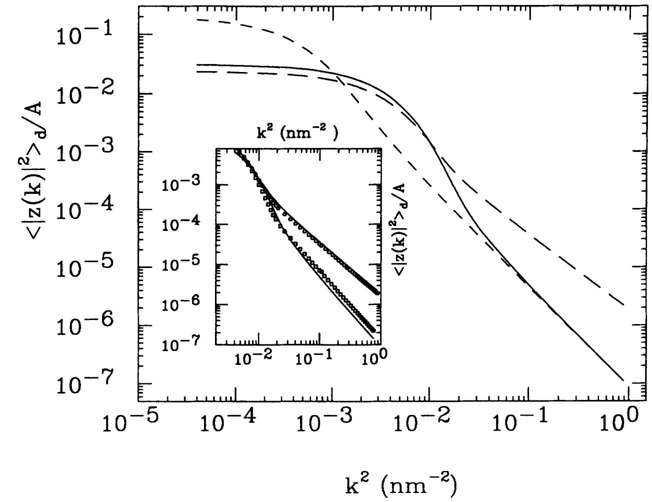

FIG. 1. Schematics of $\langle |z(\mathbf{k})|^2\rangle_d/A$ calculated by means of Eq. (2.7) with $\sigma=0.7$ nm and $\xi=100.0$ nm. For $H=0.7$, $\zeta=70.0$ nm: solid line; $\zeta=300.0$ nm: dashed line. For $H=0.3$, $\zeta=70.0$ nm: long-dashed line. The inset depicts a comparison of Eq. (2.8) with calculations performed in terms of Eq. (2.7) for the correlation function $C_S(R)$. For $H=0.3$, Eq. (2.7): solid line; Eq. (2.8): circles; and for $H=0.7$, Eq. (2.7): solid line; Eq. (2.8): squares.

Equation (2.7) for $\zeta\ll\xi$ after expansion of $C(R)$ according to Eqs. (1.2) and (1.3), $C(R)\approx\sigma^2-DR^{2H}$, yields $\langle |z(\mathbf{k})|^2\rangle_d\approx[A/(2\pi)^6]\sigma^2\zeta^2e^{-k^2\zeta^2/4\pi}+o(\zeta^2/\xi^2)$. In the opposite asymptotic limit $\zeta\gg\xi$, Eq. (2.7) after expansion of $e^{-\pi R^2/\zeta^2}(\approx1-\pi R^2/\zeta^2)$ yields $\langle |z(\mathbf{k})|^2\rangle_d\approx\langle |z(\mathbf{k})|^2\rangle+o(\xi^2/\zeta^2)$.[12] Therefore, the previous limiting cases suggest a crossover form for $\langle |z(\mathbf{k})|^2\rangle_d$ whenever ξ and ζ are of the same order of magnitude,

$$\langle |z(\mathbf{k})|^2\rangle_d\approx\langle |z(\mathbf{k})|^2\rangle+\frac{A}{(2\pi)^6}\frac{\sigma^2\pi\xi^2\zeta^2}{(\pi\xi^2+\zeta^2)}e^{-k^2\zeta^2/4\pi} . \tag{2.8}$$

This form captures the essence of exact calculations in terms of Eq. (2.7) for the regime of spatial wave vectors $k>2\pi/\max(\zeta,\xi)$ (Fig. 1, inset). In fact, the Gaussian prefactor in Eq. (2.8) is an exact result for the case of the Gaussian correlation function $C(R)=\sigma^2e^{-R^2/\xi^2}$. In this case, Eq. (2.7) yields $\langle |z(\mathbf{k})|^2\rangle_d\sim\xi^2\zeta^2/(\pi\xi^2+\zeta^2)e^{-k^2\xi^2\zeta^2/4(\pi\xi^2+\zeta^2)}$. In addition, we shall consider in Eq. (2.8) for $\langle |z(\mathbf{k})|^2\rangle$ the analytic form

$$\langle |z(\mathbf{k})|^2\rangle=\frac{A}{(2\pi)^5}\frac{\sigma^2\xi^2}{(1+k^2\xi^2/2H)^{1+H}} , \tag{2.9}$$

which is associated with the self-affine correlation function $C(R)\sim R^HK_H(R)$.[13]

However, in order to gauge the effect of any particular choice of the correlation function $C(R)$ and to justify the adequacy of Eq. (2.8) in the regime of spatial wave vectors $k>2\pi/\max(\zeta,\xi)$, we performed comparisons between Eq. (2.8) and numerical calculations of $\langle |z(\mathbf{k})|^2\rangle_d$ in terms of Eq. (2.7) for the correlation function $C_S(R)=\sigma^2e^{-(R/\xi)^{2H}}$. This correlation function has been used widely in the analysis of diffuse x-ray reflectivity data,[4,14] and its associated roughness spectrum coincides with that of Eq. (2.9) for $H=0.5$.[13,14] Comparison shows that Eq. (2.8) is in better agreement with the direct calculation by means of Eq. (2.7) for small values of H for the whole regime of spatial wave vectors $k > 2\pi/\max(\zeta,\xi)$. However, Eq. (2.8) depicts the correct physical behavior rather well, taking also into account the fact that we considered for the roughness spectrum $\langle |z(\mathbf{k})|^2 \rangle$ (no domains), a specific model which will cause additional deviations from Eq. (2.7). Furthermore, Eq. (2.8) contains no adjustable parameters and its simple analytic form permits explicit calculation of the surface width, which is an experimentally important quantity in surface-roughness studies.[5,13]

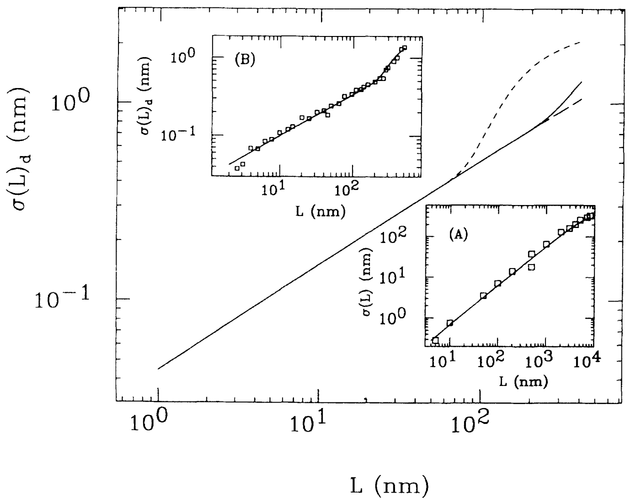

FIG. 2. Schematics for the surface width in terms of Eq. (3.3) with $\sigma=1.7$ nm, $\xi=400.0$ nm. $\zeta=100.0$ nm: short-dashed line, $\zeta=300.0$ nm: solid line, and $\zeta=500.0$ nm: long-dashed line. The inset (A) shows the fit to surface-width data (squares) from a gold film when no domains exist with $H=0.96\pm0.02$, $\sigma=450.0$ nm, and $\xi=2000.0$ nm. The inset (B) shows the fit to surface-width data (squares) from an iron film after ion bombardment, with $H=0.53\pm0.02$, $\xi=650.0$ nm, and $\zeta=260.0$ nm. The value of the roughness exponent H has been determined in both cases in terms of a power-law fit.

III. SURFACE-WIDTH DOMAINS

When the surface height $z(\mathbf{r})$ is single valued as a function of position, its variance or surface width $\sigma(L)$ is well defined and can be analyzed for self-affine fractals according to the form $\sigma(L) \propto L^H$, where H is the roughness exponent and L a linear size on the surface.[15] In the present study, we shall examine the effect of domains on the surface width, which in the continuum limit is given by[16]

$$\sigma^2(L)_d = \int_{k_L < k,k' < k_c} \langle z(\mathbf{k}) z(\mathbf{k}') \rangle_d \, d^2\mathbf{k} \, d^2\mathbf{k}' \,, \qquad (3.1)$$

with $k_L = 2\pi/L$, $k_c = \pi/a_0$, a_0 the atomic spacing, and

$$\langle z(\mathbf{k}) z(\mathbf{k}') \rangle_d = \frac{(2\pi)^4}{A} \delta(\mathbf{k}+\mathbf{k}') \langle |z(\mathbf{k})|^2 \rangle_d \,. \qquad (3.2)$$

Substituting in Eq. (3.1) from Eqs. (2.8), (2.9), (3.2), and carrying out the integration, we obtain

$$\sigma^2(L)_d \approx \sigma^2 \left[1 + \frac{4\pi^2 \xi^2}{2HL^2} \right]^{-H} + \frac{\sigma^2 \pi \xi^2}{\pi \xi^2 + \zeta^2} e^{-\pi \zeta^2 / L^2} \,, \qquad (3.3)$$

since $\xi, \zeta \gg a_0$.

Calculations of $\sigma(L)_d$ versus scan size L in terms of Eq. (3.3), are shown in Fig. 2. From these schematics, the effect of the competition between the characteristic length scales ξ and ζ on the self-affine fractal behavior at large L can be observed. The first term in Eq. (3.3) represents the surface width when there is no domain effect, and its applicability is illustrated in terms of a fit to surface-width data (Fig. 2 in Ref. 5), acquired by means of scanning tunneling microscopy (STM) from a gold film with $H=0.96$, $\xi=2000.0$ nm, and $\sigma=450.0$ nm [Fig. 2, inset (A)]. The Gaussian term will cause the surface width to have a rather sharp rise for linear sizes $L \geq \zeta$ and $\zeta < \xi$. Such a behavior can be seen in Fig. 2, and has been also observed in real surface-width data (Fig. 3 in Ref. 5). These data were acquired by means of STM from rough iron-film surfaces fabricated with ion-beam erosion.[5] The observed sharp increment cannot be explained only by the existence of the effective roughness cutoff ξ. A second length scale is required, which can be identified as a domain effect. Such an explanation can be understood also from the fact that the iron-film surface prior to ion bombardment revealed atomically stepped terraces with typical terrace area of the order of $5-10 \times 10^2$ nm^2. This might favor the creation of domain boundaries during ion bombardment, obscuring therefore the expected power law $\sigma(L) \sim L^H$, $H=0.53\pm0.02$, above 200.0 nm. Furthermore, the picture of domains is supported also by the fact that the surface-width data acquired prior to ion bombardment approach those after erosion at length scales of the order of 200.0 nm, which supports the obscuring of the power-law behavior due to pre-existing roughness, as explained in terms of domain formation. The moderately sharp increment above 200.0 nm, according to previous results, signals a domain size ζ smaller than the correlation length ξ. The fit of the iron-film width data in terms of Eq. (3.3) with $H=0.53$ (obtained from a power-law fit) yields $\sigma=1.7$ nm, $\zeta=260.0$ nm, and $\xi=650.0$ nm [Fig. 2, inset (B)]. The value of the average domain size ζ is significantly close to the regime where the sharp increment of the surface width is observed, supporting therefore the consistency of the corresponding formalism as well as its relevance to experimental studies.

IV. CONCLUSIONS

In conclusion, the aim of this work was to correlate known information regarding finite-size effects due to domains for the particular case of self-affine fractal surfaces. This is accomplished in terms of a simple formalism which can be relevant in experimental studies of surface roughness by means of scanning tunneling microscopy. Direct fit to surface-width data shows the adequacy of this formalism for a certain regime of length scales, which is, however, sufficiently wide to capture the full physical behavior of the fractal system, and to yield its characteristic parameters.

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