

Characterization of self-affinity in the global regime

Alexander V. Neimark*

*Institute of Physical Chemistry, Russian Academy of Sciences, Moscow
and Laboratoire de Physique Théorique et Mathématique, Centre National de la Recherche Scientifique,
Chasseneuil du Poitou, France*

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Methods for characterization of self-affine surfaces and measurements of their roughness exponents H are developed. It is shown that for smoothed surfaces, which underwent particular coarse graining or averaging of the small-scale fluctuations, the excess surface area S_{ex} and the mean square root radius of curvature a_c are related by two distinct asymptotic power laws if a_c is well below or well above a certain crossover scale a_{cr} . In the local regime of self-affinity, when $a_c \ll a_{cr}$, $S_{ex} \propto (a_c/a_{cr})^{-(1-H)}$. In the global regime of self-affinity, when $a_c \gg a_{cr}$, $S_{ex} \propto (a_c/a_{cr})^{-2(1-H)/(2-H)}$. The former scaling relationship is consistent with the well known definition of local fractal dimensions $d_{loc} = d_{top} + 1 - H$. The latter scaling relationship offers alternatives for characterization of self-affinity over large scales by means of excess dimensions defined as $d_{ex} = d_{top} + 2(1-H)/(2-H)$ and can be used for determination of roughness exponents from the measurements provided in the global regime. The thermodynamic method of fractal analysis, proposed earlier for self-similar surfaces (A.V. Neimark, Pis'ma Zh. Eksp. Teor. Fiz. **51**, 535 (1990) [JETP Lett. **51**, 607 (1990)]; Physica A **191**, 258 (1992)), is extended for self-affine surfaces for determination of fractal dimensions and roughness exponents from adsorption and capillary experimental data.

Conventional methods of fractal analysis are instructive for self-affine surfaces when they are studied over sufficiently small scales well below a certain crossover scale a_{cr} , which is defined as a lateral distance being equal to the corresponding mean square root (MSR) normal fluctuations

$$\langle [z(x+a_{cr})-z(x)]^2 \rangle^{1/2} = a_{cr} . \tag{1}$$

Here x is the lateral coordinate and z is the normal coordinate. The crossover scale a_{cr} separates so-called local (small scales) and global (large scales) regimes of self-affinity. For the definitions of self-affinity and conventional methods of modeling and analysis of self-affine surfaces, see the original papers of Mandelbrot^{1,2} and Voss.³

Box counting and other measurements using rulers of varying size a give nontrivial values for fractal dimensions in the local regime and indicate nonfractal behavior in the global regime

$$d_{loc} = d_{top} + 1 - H , \quad a \ll a_{cr} , \tag{2a}$$

$$d_{glob} = d_{top} , \quad a \gg a_{cr} . \tag{2b}$$

Here d_{top} is the topological dimension of a given surface and H is the roughness (Hurst or Hoelder) exponent, defined from

$$\langle [z(x+\Delta x)-z(x)]^2 \rangle^{1/2} = a_{cr} [\Delta x/a_{cr}]^H . \tag{3}$$

The different images of a self-affine profile in the local and global regimes are shown in Fig. 1 with the example of a trail of fractional Brownian motion. In Fig. 2, the results of fractal analysis of Gaussian surfaces with self-affine fluctuations are displayed. This relationship between the specific area S of smoothed surfaces and the smallest relative mean square height fluctuations that remained after smoothing,

$$G = \langle [z(x+\Delta x_s)-z(x)]^2 \rangle / (\Delta x_s)^2 \\ = \langle (\Delta z)^2 \rangle / (\Delta x_s)^2 = (\Delta x_s/a_{cr})^{-2(1-H)} , \tag{4}$$

highlights that in the local regime, $G \gg 1$ and $\Delta x_s \ll a_{cr}$, the surface area S increases with the decrease of the smoothing scale Δx_s in conformity with the fractal law

$$S \propto G^{1/2} = (\Delta x_s/a_{cr})^{-(1-H)} . \tag{5}$$

(Smoothed surfaces are regarded here as rough surfaces that underwent a particular procedure of eliminating the small-scale fluctuations below a certain scale named as a smoothing scale Δx_s .) There is no available information about scaling of the surface fluctuations in the global regime when $G \ll 1$ and $\Delta x_s \gg a_{cr}$. A similar situation is typical for the other methods of fractal analysis applied to self-affine systems. For recent reviews and discussions on the current state of art, see Refs. 4-9.

The goal of this paper is to propose alternative methods for characterization of self-affine surfaces in the global regime. This problem is important and topical in view of possible practical applications of self-affine models to real surfaces, which may most likely display self-affine properties over relatively large scales in the global regime, for example, vapor-deposited films, fractures, etc.

Let us consider the excess area of smoothed surfaces with respect to the unit area of a reference plane defined as

$$S_{ex} = \frac{1}{L^{d_{top}}} \int \left[\sqrt{1+(\nabla z)^2} - 1 \right] d^{d_{top}} \mathbf{x} . \tag{6}$$

The introduction of the excess quantities is reasonable in view of further applications of Gibbs' thermodynamics of interface phenomena, which deals with the excess extensive parameters, to the description of capillary phenomena on rough surfaces. For smoothed surfaces, S_{ex} de-

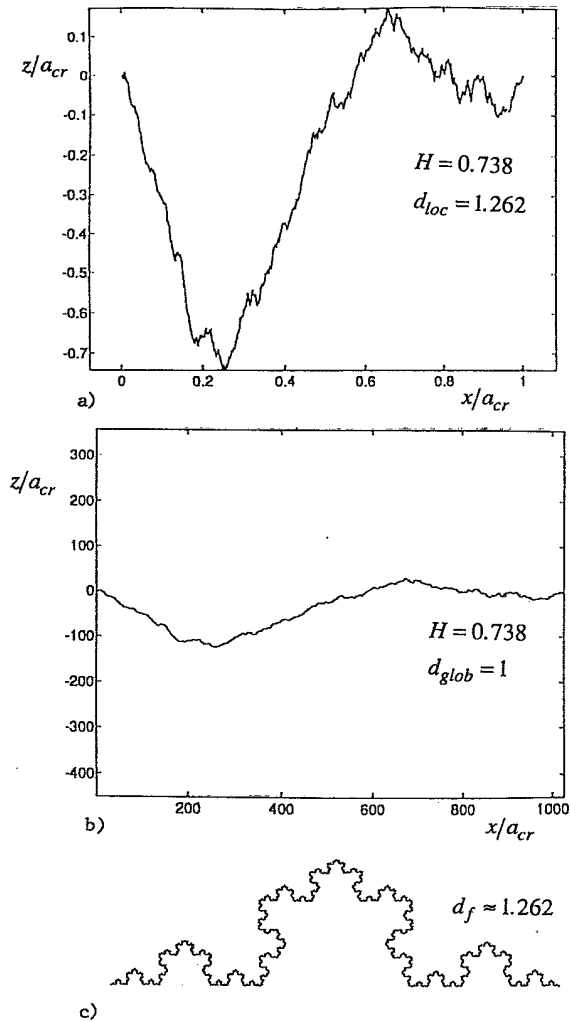


FIG. 1. Typical self-affine profile (a trail of FBM) in the local (a) and global (b) regimes. The normal z and lateral x coordinates are reduced to the crossover scale a_{cr} . The value of the roughness exponential is chosen specially equal to $H=0.738$ in order to get the value of the local fractal dimension being equal to the fractal dimension $d_f \approx 1.262$ of self-similar triadic Koch curve (c), which is presented for comparison.

depends on the smoothing scale Δx_s , and can be calculated explicitly for a given distribution function $P(\Delta z, \Delta x)$ of height differences Δz within lateral distance Δx .

For Gaussian surfaces, with

$$P(\Delta z, \Delta x) = \frac{1}{\sqrt{2\pi g(\Delta x)}} \exp \left\{ -\frac{\Delta z^2}{2g(\Delta x)} \right\}, \quad (7)$$

where $g(\Delta x) = \langle \Delta z^2 \rangle$, the dependence S_{ex} versus G [Eq. (4)] is presented in Fig. 3. Here we see two distinct power-law asymptotics. In the local regime, $G \gg 1$ and $\Delta x_s \ll a_{cr}$, as one might expect, the well-known asymptotic (5) is observed. In the global regime, $G \ll 1$ and $\Delta x_s \gg a_{cr}$, the asymptotic law is as follows:

$$S_{ex} \propto G = (\Delta x_s / a_{cr})^{-2(1-H)}. \quad (8)$$

Looking at the scaling relationship (8), it is tempting to introduce the exponent $\beta_{ex} = d_{top} + 2(1-H)$ as a new

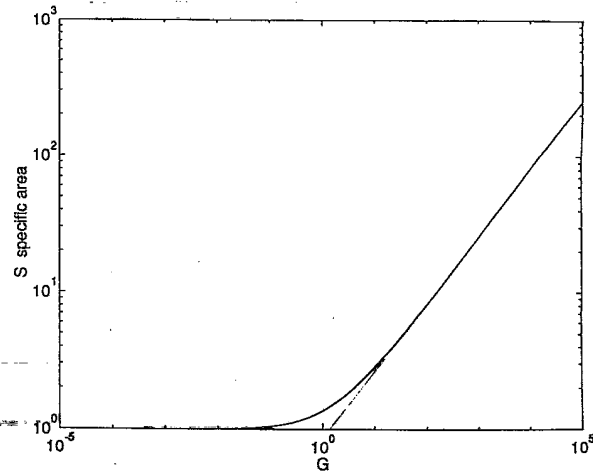


FIG. 2. Specific area S of smoothed surfaces vs relative mean square height fluctuations, G [Eq. (14)]. The scaling asymptotic [Eq. (5)] is observed only in the local regime ($G \gg 1$).

fractal dimension in the global regime. However, it is worth noting that β_{ex} cannot serve as a real fractal dimension for several reasons: (i) the excess area is not the area of any real surface; (ii) the smoothing parameter Δx_s does not play the role of a ruler and (iii) β_{ex} varies between 2 and 4 and exceeds 3 as $H < \frac{1}{2}$.

From the point of view of some experimental measurements, it is more instructive, and it will be shown below why, to use the MSR radius of curvature of smoothed surfaces a_c instead of Δx_s . The MSR radius of curvature of a surface is defined as

$$a_c = \langle K(\mathbf{x})^2 \rangle^{-1/2}. \quad (9)$$

Here $K(\mathbf{x})$ is the surface curvature

$$K(\mathbf{x}) = \frac{1}{2} \frac{\nabla^2 z}{[1 + (\nabla z)^2]^{3/2}}. \quad (10)$$

For smoothed surfaces, a_c depends on the lateral

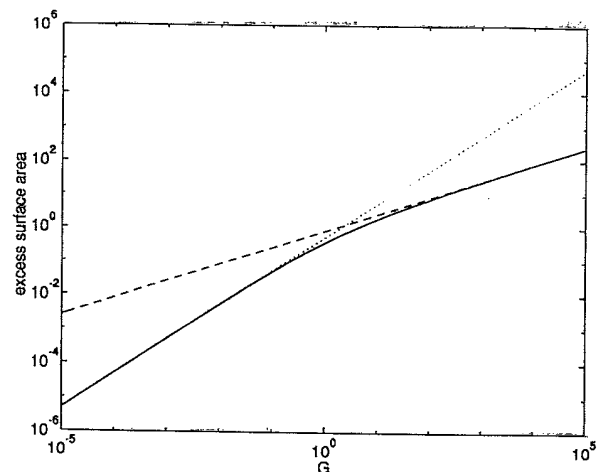


FIG. 3. Excess area S_{ex} of smoothed surfaces vs relative mean square height fluctuations G [Eq. (4)]. Two distinct scaling asymptotes (5) and (8) are observed, respectively, in the local ($G \gg 1$) and in the global ($G \ll 1$) regimes.

smoothing scale Δx_s , and can be calculated by averaging the square of the differential form (10). The asymptotic in the local regime is obvious,

$$a_c \propto \Delta x_s, \quad a_c \ll a_{cr}. \quad (11)$$

In the global regime, the asymptotic gives

$$a_c \propto \Delta x_s G^{-1/2} = a_{cr} (\Delta x_s / a_{cr})^{2-H}, \quad a_c \gg a_{cr}. \quad (12)$$

The relationship between the excess area and MRS radius of curvature for smoothed surfaces is presented in Fig. 4 for Gaussian self-affine surfaces with different values of roughness exponents $H=0.1, 0.3, 0.5, 0.7,$ and 0.9 . The power-law asymptotics in the local and global regimes are as follows:

$$S_{ex} \propto (a_s / a_{cr})^{-(1-H)}, \quad a_c \ll a_{cr} \quad (13)$$

and

$$S_{ex} \propto (a_s / a_{cr})^{-2(1-H)/(2-H)}, \quad a_c \gg a_{cr}. \quad (14)$$

The exponent $2(1-H)/(2-H)$ varies with H between 0 and 1. Therefore, it is instructive to introduce a new exponent, characterizing self-affinity over large scales, and to name this exponent as excess dimension

$$d_{ex} = d_{top} + 2(1-H)/(2-H). \quad (15)$$

Despite the fact that d_{ex} meets some requirements for surface fractal dimensions— $d_{top} < d_{ex} < d_{top} + 1$ —and links two measured quantities S_{ex} and a_c , I do not use the term fractal dimensions but rather excess dimensions.

Relationship (14) offers new possibilities for measuring roughness exponents. It is well known that in experiments with wetting and nonwetting fluids on rough solid surfaces, the fluid surface can be regarded as a set of menisci of equal mean radius of curvature r_c , which are inscribed into the solid surface.¹⁰ It has been shown¹¹ that for two-dimensional (2D) self-similar substrata the area S_c of these interfaces scales with the mean radius of cur-

vature r_c as

$$S_c \propto r_c^{-(d_{fs}-2)}, \quad (16)$$

where d_{fs} is the surface fractal dimension $2 < d_{fs} < 3$.

The scaling relationship (16) is the basis of thermodynamic methods of fractal analysis, which have been proposed and proofed for self-similar surfaces and porous solids.¹²⁻¹⁴ Now I can extend this method for self-affine surfaces. Actually, interfaces of fluids wetting rough surfaces can be regarded as smoothed surfaces with a natural smoothing scale equal to the mean radius of interface curvature, which is determined by the conditions of thermodynamic equilibrium, namely, by the chemical potential μ of the wetting fluid

$$r_c \propto |\mu|^{-1}. \quad (17)$$

The interface area can be calculated from the experimental adsorption isotherm $N(\mu)$, where N is the amount of wetting fluid adsorbed at a given chemical potential, by means of the following general thermodynamic relationship:

$$S_c(\mu) \propto \int_{N(\mu)}^{N_0} \mu dN. \quad (18)$$

It is worth noting that the thermodynamic quantities used here are the excess quantities with respect to the saturation conditions when the surface is covered by a thick layer of wetting fluid. Thus, S_c is the excess area with respect to the area of the interface of this layer which, in turn, can be regarded as a reference flat surface without loss of accuracy of scaling relationships. That is, for self-affine substrata r_c [Eq. (17)] and S_c [Eq. (18)] are related by the asymptotic power laws similar to (13) in the local regime and (14) in the global regime, namely,

$$S_c \propto r_c^{-(1-H)}, \quad r_c \ll a_{cr} \quad (19)$$

and

$$S_c \propto r_c^{-2(1-H)/(2-H)}, \quad r_c \gg a_{cr}. \quad (20)$$

The relationships (16)–(20) can be used for the interpretation of experimental data obtained by quasiequilibrium adsorption or capillary measurements, such as capillary condensation of vapors, intrusion of nonwetting fluids (e.g., mercury porosimetry), impregnation and drainage of wetting fluids, thermoporosimetry, etc. When the correlation between the excess area [Eq. (18)] and the mean radius of curvature r_c [Eq. (17)] of equilibrium interfaces at varying chemical potential μ follows the power law (16) over a certain scale range, one can argue that the substratum under study may be either a self-similar surface with the surface fractal dimension d_{fs} or a self-affine surface. In the latter case, there are two variants according to the scale range of measurements. If the measurements are carried out in the local regime of self-affinity, d_{fs} should be referred to as the local fractal dimension d_{loc} [Eq. (2c)] and the roughness exponent happens to be equal to

$$H = 3 - d_{fs}. \quad (21)$$

On the contrary, if the measurements are carried out in

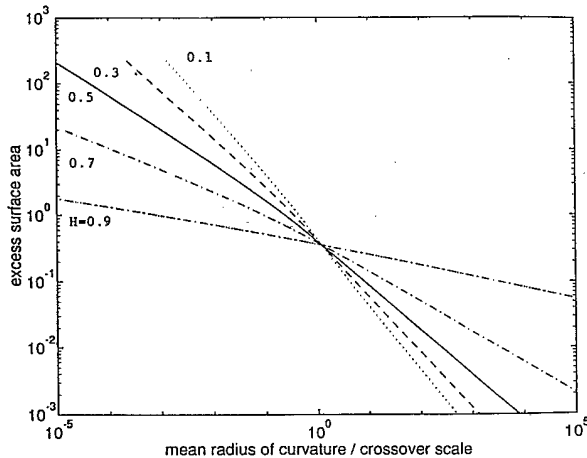


FIG. 4. Excess area S_{ex} vs MRS radius of curvature reduced to the crossover scale, a_c/a_{cr} , for smoothed affine surfaces with different values of roughness exponents $H=0.1, 0.3, 0.5, 0.7,$ and 0.9 . Two distinct scaling asymptotes (13) and (14) are observed, respectively, in the local ($a_c/a_{cr} \ll 1$) and global ($a_c/a_{cr} \gg 1$) regimes.

the global regime of self-affinity, d_{fs} should be referred to as the excess dimension d_{ex} [Eq. (2c)] and the roughness exponent happens to be equal to

$$H = 2(3 - d_{fs}) / (4 - d_{fs}). \quad (22)$$

It is worth noting that in the special case of experimental absorption isotherms of the so-called Halsey type, $N \propto |\mu|^{-1/m}$, the thermodynamic method gives the same results as the theories proposed earlier by (i) de Gennes¹⁵ for self-similar pore structures $d_{fs} = 3 - 1/m$; (ii) Pfeifer and Cole¹⁶ for self-affine surfaces in the local regime $d_{loc} = 3 - 1/m$; and (iii) Kardar and Indekeu¹⁷ for self-affine surfaces in the global regime $H = 2/(1+m)$. In this way, a proper compromise is established in the discussion¹⁶⁻²³ about the interpretation of capillary condensation experiments on rough surfaces triggered by the paper of Pfeifer *et al.*¹⁸

Application of the thermodynamic method to the analysis of the experimental isotherm of N_2 adsorption at 77 K on 1000-Å-thick silver film deposited at 80 K (Ref.

23) testifies that in this particular case the power correlation between S_c and a_c holds in the scale range 10–1000 Å with the formal surface fractal dimension $d_{fs} = 2.78 - 2.79$ calculated from Eq. (16). Suppose a surface under study to be a self-affine surface in the global regime (that is typical for vapor deposited films^{5,6}), the corresponding roughness exponent is estimated from Eq. (22) as $H = 0.35 - 0.36$. This value is close to the value $H \approx 0.39$ predicted by theoretical models of vapor deposition onto 2D substrata without surface diffusion of deposited particles.²⁴⁻²⁷

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*Present address: Department of Chemical Engineering, P.O. Box 208286, Yale University, New Haven, CT 06520-8286.

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