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A STUDY OF (1 + 1)-DIMENSIONAL HEIGHT-HEIGHT CORRELATION FUNCTIONS FOR SELF-AFFINE FRACTAL MORPHOLOGIES

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We study analytic forms in Fourier space of one-dimensional height-height correlation functions for self-affine rough surfaces. Comparisons with complex systems suggest three alternative models. However, only the model $C_1(k) \approx (1 + a |k|^2)^{-(1+2n)/2}$ permits analytic calculation of important surface roughness quantities (i.e. surface width) for roughness exponents in range $0 \leq H \leq 1$. Furthermore, the implications of the results to experimental roughness studies by means of STM-AFM are discussed. Copyright © 1996 Published by Elsevier Science Ltd

In the context of relaxation phenomena, a specific correlation function has been used widely to describe the complex nature of various physical processes. This the well known stretched exponential function $C_0(x) = C_s(0) e^{-x^H}$ ($0 < H \leq 1$). It was introduced in 1863 for the description of mechanical creep in glassy fibers, as well as later to describe dielectric relaxation in polymers [1]. Lately, it was used to fit miscellaneous experimental data including NMR, dynamic light scattering, quasi electric neutron scattering, kinetic reactions, magnetic relaxation, etc. [2], as well as to describe height-height correlation functions for self-affine fractal surfaces [3–5].

The knowledge of one-dimensional height-height correlation functions is required in real and/or Fourier space in a wide spectrum of studies which involve random rough surfaces. These studies include X-ray scattering investigations of surface/interface roughness in single and/or multilayer films [3, 4], surface sound wave studies where the direct knowledge of the Fourier transform $C(k)$ of one-dimensional height-height correlation function is required [6], and roughness studies by means of Scanning Tunnelling Microscopy (STM) as well as Atomic Force Microscopy (AFM) [7, 8].

We will denote by $z(x)$ the surface height profile function which is assumed a random variable with zero mean $\langle z(x) \rangle = 0$ over a segment of macroscopic size $L$. The Fourier transform $z(k)$ and the height-height correlation function $C(x)$ are defined respectively by $z(k) = \int \frac{(1/2\pi)}{L} z(x) e^{-ikx} dx$ and $C(x) = \int \frac{(1/L)}{2\pi} \langle z(x + x')z(x') \rangle dx'$, and their combination yields the roughness spectrum $\langle |z(k)|^2 \rangle$ [13]; $\langle |z(k)|^2 \rangle = \int \frac{(1/L)}{2\pi} C(k)$ with $C(k) = \int C(x) e^{-ikx} dx$.

For self-affine fractal surfaces the height-height correlation function $C(x)$ has the scaling behaviour [14] $C(x) \sim \sigma^2 - D(x)^{2H}$ if $x \ll \xi$, and $C(x) = 0$ if $x \gg \xi$, $D(\approx \sigma^2/\xi^{2H})$ is a constant, $\xi$ the in-plane roughness correlation length and $\sigma = \langle |z(x)|^2 \rangle^{1/2}$ the saturated r.m.s. surface roughness. Thus, the Fourier transform $C(k) = F\{C(x)\}$ for self-affine fractals has the scaling behaviour $C(k) \sim k^{-1-2H}$ if $k\xi \gg 1$, and $C(k) \sim \text{const}$ if
Fig. 1. Log–log plots of the one-dimensional roughness spectra $C_1(k)/\sigma^2$ vs $k$ for $\xi = 100$ nm, $a_0 = 0.3$ nm and roughness exponents $H$: $H = 0.3$ (dashes), $H = 0.5$ (solid), $H = 0.8$ (dots). The inset shows the change of the parameter $a = a(H, \xi)$ as a function of $\xi/a_0$ for $H = 0$ (squares), $H = 0.3$ (circles), $H = 0.5$ (up-triangles), and $H = 0.8$ (down-triangles).

$k \xi \ll 1$. The intermediate behaviour at $k \xi \sim 1$ will be based upon suggestions from previous studies in complex systems. The height-difference correlation function $g(x)$ is given by $g(x) = 2a^2 - 2C(x)$. The roughness exponent $H$ characterise the degree of surface irregularity, and has values in the range $0 < H < 1$. Small values of $H (H \to 0)$ correspond to highly irregular surfaces, and large values $(H \to 1)$ to surfaces with a smooth hill-valley structure [9, 14, 15].

We will perform the construction of self-affine height–height correlations in Fourier space, since it is of primary importance to investigate analytic forms for $C(k)$. Thus, we will suggest and investigate the following three models in Fourier space

$$C_1(x) = \frac{\sigma^2 \xi}{(1 + a |k|^{1+2H})}; \quad a = \frac{1}{H} [1 - (1 + aV_c)^{-2H}],$$

$$C_2(x) = \frac{(\sigma^2 \xi/g)}{1 + (|k|^{1+2H})}; \quad g = 2 \int_0^{V_c} (1 + \nu^{1+2H})^{-1} d\nu,$$

where $V_c = k_c \xi$. The parameters $\{a, y, g\}$ are calculated from the normalisation condition $2 \int C_i(k) \, dk = \sigma^2 (0 < k < k_i, i = 1, 2, 3)$ with $k_i = \pi/a_0$, and $a_0$ the atomic spacing. The existence of the finite bound $k_c$ is related with the fact that any notion of continuum treatment at length scales lower than $a_0$ becomes meaningless. Expressions in the limit $H \to 0$ can be obtained from those at $H > 0$, if we consider the identity $H \to 0: (1/H)\ln(1 + uV_c) \to \ln(u)$ [9, 10]. Thus, we have $a = 2 \ln(1 + aV_c)$ for $H = 0$.

In equation (1) for $V_c \gg 1$ and $H > 0$, we obtain $a \sim 1/H$ (inset of Fig. 1). The model $C_1(k)$ has already been used for the calculation of eigenwave spectrums [6].

It originates from similar studies of two-dimensional self-affine correlation models of the form $C(k) \sim (1 + a k^2 \xi^2)^{-1-H}$ [10]. However, $C_1(k)$ does not
reproduce the behaviour of the Fourier transform of \( C_r(x) \) at \( H = 0.5 \) or \( C_r(k, H = 0.5) \) suggests the generalisation for \( H \neq 0.5 \) that is given by \( C_r(k) \). In equation (2) for \( H = 0 \), we have \( g(H = 0) = 2 \ln(1 + V_c) \). While for \( 0 < H < 1 \) and \( V_c \gg 1 \), the integral identity \( \int^{v_{\max}}_{v_{\min}} [1 + \nu^2]^{-1} d\nu = \pi/\sin(\pi/2) \) yields \( g(H > 0) \approx 2\pi(1 + 2H) \sin(\pi(1 + 2H))^{-1} - 1/2H \). Finally, the correlation function \( C_r(k) \) is suggested according to the form of \( \Phi(k) \) [12] whose connection with the \( C_r(x) \) was established in the past [11]. The constraint \( u_f = 1 + 2H \) is imposed the requirement that at large \( k(k\xi \gg 1) \); \( C_r(k) \propto k^{-1 - 2H} \).

In former studies, a model similar to \( C_{\alpha\epsilon}(k) \propto \sigma^2\eta^2(l + \alpha\kappa^2\eta^2)^{-1 + 2H/2} \) was introduced by Church and Takacs [16]. This is based again on the two-dimensional analogue of \( C_r(k) \propto (1 + \alpha\kappa^2\eta^2)^{-1 - H} \) [10], and \#\# \( C_{\alpha\epsilon}(k) \#\# \) is obtained from the normalisation of \( C_{\alpha\epsilon}(k) \), however, only in an integral form: \( 2 \int [1 + \alpha^2\eta^2]^{-1 - H} d\eta = 1 \) \( (0 < \nu < V_c) \).

Direct measurements of one-dimensional integrated \( C(k) \) spectra have been performed in roughness studies by Mitchell and Bonnell [7]. Besides the roughness exponent \( H \) that we obtain in a \( \ln C(k) \) vs \( \ln(k) \) plot (linear regime with slope \(-1 - 2H\)), the knee regime \( \sim 2\pi/\xi \) (or \( X_{\text{corr}} \sim 4\xi \) on a surface width \( \ln[\sigma(x)] \) vs \( \ln(x) \) plot as in Fig. 2) where a turning occurs from the linear behaviour with slope \(-1 + 2H\) to a saturated regime \(-\ln(\sigma^2\eta^2)\) (plateau, see Fig. 1) is important experimentally. This is because it provides a sufficient means to determine the correlation length \( \xi \#\# \), and its dynamic evolution with film thickness in growth studies [10, 17, 18].

As we shall see in the following section, the \( C_1(k) \) model will also be preferable to investigate more, since it allows analytic calculation of surface quantities, i.e. surface width, which are of crucial importance in roughness studies [8]. In the continuum limit, the surface width (r.m.s.-roughness) of one-dimensional rough profile over a lateral size \( x \) is given by [19]

\[
\sigma^2(x) = \int_{k < k_c} \int_{k' < k_c} \langle z(k)z(k') \rangle dk dk';
\]

(4)

\[
\langle z(k)z(k') \rangle = \delta(k + k')C(k),
\]

where \( k_c = 2\pi/x \), and \( \langle z(k)z(k') \rangle = \delta(k + k')C(k) \) since the surfaces we consider here are assumed statistically stationary up to second order (translation invariance).

Analytic calculation of \( \sigma(x) \) can be performed in terms of the correlation function \( C_1(k) \) for all values of the roughness exponent \( H \) in the range \( 0 \leq H \leq 1 \).

After substitution in equation (4) of \( C_1(k) \), we obtain

\[
\sigma^2(x) = \frac{\sigma^2}{aH} [(1 + ak_\xi^2)^{-2H} - (1 + ak_\xi^2)^{-2H}]
\]

(0 < H < 1).

(5)

\[
\sigma^2(x) = \frac{2\sigma^2}{a} \ln \left[ \frac{1 + ak_\xi^2}{1 + ak_\xi^2} \right] \quad (H = 0).
\]

(6)

The asymptotic behaviour of equations (5) and (6) is given by \( \sigma_1(x) = [\sigma^2/(H_a)^2(2\pi\xi^2)^2]^{1/2} \) \( (0 < H < 1) \) and \( \sigma_1(x) = [\sigma^2/(a^2)] \ln(\sigma/a_0)^{1/2} \) \( (H = 0) \) for \( a_0 \ll x \ll \xi \), and \( \sigma_1(x) = \sigma \) if \( x \gg \xi \). The asymptotic behaviour for \( \sigma(x) \) obeys the general scaling behaviour attributed to \( \sigma(x) \) for self-affine fractal roughness or \( \sigma(x) \sim x^H \) for \( x \ll \xi \), and \( \sigma(x) = \sigma \) for \( x \gg \xi \) [14]. Figure 2 depicts plots of \( \sigma_1(x) \) vs \( x \) where the knee regime occurs at \( X_{\text{corr}} \sim 4\xi \).

Measurements of \( \sigma(x) \) vs \( x \) have been performed from Salvarezza et al., Vazquez et al., Herrasti et al. for Au-films by means of STM, and by Tong et al. on CuCl/CaF$_2$-films by means of AFM [8], in an effort to measure the roughness exponent \( H \). Our schematics in Fig. 2 for \( \sigma_1(x) \) compare significantly well to the surface width measurements of Tong et al. on CuCl/CaF$_2$ films (Fig. 3 in [8]). They compare also with part of the measurements by Herrasti et al. for Au-films (i.e. Fig. 6 and Fig. 11 in [8]), and Vazquez et al. for Au-films (Fig. 3, Surf. Sci., in [8]). The inset of Fig. 2 is plotted according to parameters observed in Fig. 3 (Surf. Sci.) of Vazquez et al. \( (\sigma = 4\text{nm}, \xi = 38.9\text{nm}, H = 0.83) \) [8] in terms of equation (5).

According to the scaling theory approach, during film growth [20], the normal roughness \( \sigma \) and the in-plane correlation length \( \xi \) evolve with film thickness \( h \) as \( \sigma \sim h^b \) and \( \xi \sim h^{bH} \) (\( b \) and \( z = H/b \) are respectively the growth and the dynamic exponent), as well as the surface width scales as \( \sigma(x, h) = x^b F(h/x^b) \) (\( F(y) \sim y^y \) if \( y \gg 1 \), and \( F(y) \sim \text{const.} \) if \( y \ll 1 \) with lateral size \( x \) and film time evolution \( -h \). The scaling properties of \( \sigma(x, h) \) during growth on one-dimensional substrates are fundamentally important, since they give strong physical insight in complex continuum growth models (i.e. KPZ-equation [20–23]) where exact knowledge of scaling exponents is feasible only in (1 + 1)-dimensions. Equation (5) for \( x \) and \( \xi \gg a_0 \) yields \( \sigma_1(x) = \sigma H [(x + 2\pi\xi^2)/H]^H \), where after substitution of the scaling relations \( \sigma = h^b \) and \( \xi = h^{bH} \) we obtain for \( \sigma_1(x, h) = x^b A_1 Y^{(b - 1)/2} \) with \( Y = h^{bH}/h^{bH} \), \( A_1 = v \) and \( A_2 = 2\sigma_0/H \). \( \sigma_1(x, h) \) is similar in form with the one that was developed for the two-dimensional case [24].

The correlation function in real space is given by \( C_1(x) = 2\int C_1(k) dk \) \( (0 < k < k_c) \), which re-generates in real space the power law behaviour \( C_1(x) = \sigma^2 - Dx^{2H} \)
or \( g_1(x) \approx D x^{2H} \) for \( x \ll \xi \). In fact recently, there was a discussion about violation of the asymptotic power law behaviour \( g(x) \approx D x^{2H} \) or \( C(k) \approx k^{-(1+2H)} \) in the non-self-affine regime \( H \geq 1 \) (more precisely for \( H > 0.9 \)) [26]. These values are observed in MBE growth models with linear diffusion dynamics [27], and diffusion induced instabilities [28]. The \( C_1 \)-model extended in the non-self-affine regime \( H \geq 1 \), however, does not display this type of inconsistency. Figure 3 depicts plots \( g_1(x) \) vs \( x \) for \( H = 0.5, 1, 1.5 \) [29]. The power law behaviour (linear regime) is conserved from Fourier to real space and vice versa for \( 0 < H \leq 1 \).

The semi-log plot in the inset for \( H = 0 \) shows that the \( C_1 \)-model has logarithmic behaviour; \( g(x) - \ln(x) \) at \( x \ll 1 \) [29]. The latter is related with growth model predictions of the non-equilibrium analogue [10, 30], of the equilibrium roughening transition [31]. Finally, we point out that the \( C_1 \)-model does not reverse its decay rate at \( x = \xi \) as \( H \) increases from 0 to 1 (self-affine regime, see Fig. 3). In contrast, this un-natural reversibility is inherent to the \( C_2(x) \) [or \( g_2(x) \)] correlation function as pointed out in earlier studies [4, 32].

In conclusion, we convoluted known information for one-dimensional correlation functions with general concepts of self-affine fractals in order to suggest one-dimensional analytic correlation models \( [C(1,2,3)](k) \) in Fourier space. Our conjectures for the models \( C(1,2,3)(k) \) have an ad-hoc nature. However, comparisons with studies in other complex systems suggest the assumed generalisations for one dimensional self-affine fractal morphologies. We studied models in Fourier space since in a wide variety of roughness studies, the knowledge of the Fourier transform of \( C(x) \) is needed [3–8]. Moreover, analytic calculation of other important roughness quantities (i.e. surface width) which can be directly useful in roughness studies, became feasible especially in terms of the \( C_1(k) \) model [equation (1)]. More precisely, in STM–AFM measurements of the surface width \( \sigma(x) \), the knowledge of analytic forms
of $\sigma(x)$ can be useful to estimate roughness parameters ($\sigma$ and $\xi$) when the appropriate length scales cannot be probed due to system scan head limitations [10].

Finally, as a general comment on the importance of one-dimensional correlation functions $C(k)$, we point out the following. In eigenwave spectrum $[\omega(k)]$ studies of surface sound waves on rough surfaces, the roughness effect is proportional to an integral of the correlation function $C(k)$; $\omega(k) \sim [\int f(k)C(k)dk]^2$ [6]. In X-ray scattering studies, the integrated (in the direction perpendicular to the scattering plane) diffuse cross section $I(k)$ for incidence angles close to the angle of total external reflection, is directly proportional to $C(k)$; $I(k) \sim C(k)$ [3, 25].

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21. The plots \( g(x) \) vs \( x \) of Fig. 3 have been performed for \( \theta_d(H, \xi)/\theta_e = 0 \), where in the inset of Fig. 1 this corresponds to \( 300 \leq \xi/a_0 \leq 350 \).
22. The correlation function \( C_\eta(x) \) decays slower at \( x < \xi \) as \( H \) increases from low values \( H < 0.5 \) to high values \( 0.5 < H < 1 \). The opposite takes place at length scales \( x > \xi \) as a function of \( H \). Such a behaviour is not observed in the \( C_\eta \)-model.