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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 $C_1(k) \propto (1 + a|k|\xi)^{-(1+2H)}$ 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 $C_s(x) = C_s(0)\,e^{-(x/\xi)^{2H}}$ $(0 < H \leq 1)$. 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].

Despite the simple analytic form of $C_s(x)$, its Fourier transform (except $H = 0.5, 1$) is not known analytically and has a rather trivial behaviour in the limit $H \to 0$; $C_s(x) = C_s(0)/e$ [9, 10]. Because of the non analytic Fourier transform of $C_s(x)$ and its accurate description in many cases of real data in relaxation phenomena, Alvarez *et al.* [11] attempted to connect its characteristic parameters (H, ξ) with those of the Havriliak–Negami (HN) function in Fourier space, $\Phi(k) \propto [1 + i(ck)^a]^{-b}$ $(0 < a, b < 1)$, which has been used widely in glass-forming-liquid studies [12].

In our study, we shall examine directly in Fourier space height–height correlation functions $C(k)$ for one-dimensional self-affine rough profiles due to its necessity in various physical systems and surface/interface roughness studies [3–8]. Furthermore, extensive calculation of other roughness quantities will be performed, with emphasis on the surface width $\sigma(x)$ (as a function of the lateral length scale x) because of its direct measurement in roughness studies by means of STM and 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 $z(x)$ and the height–height correlation function $C(x)$ are defined respectively by $z(k) = (1/2\pi)\int z(x)\,e^{-ikx}\,dx$ and $C(x) = (1/L)\int\langle z(x+x')z(x')\rangle\,dx'$, and their combination yields the roughness spectrum $\langle|z(k)|^2\rangle$ [13]; $\langle|z(k)|^2\rangle = [L/(2\pi)^2]C(k)$ 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] $C(x) \sim \sigma^2 - Dx^{2H}$ if $x \ll \xi$, and $C(x) = 0$ if $x \gg \xi$. $D(\approx\sigma^2/\xi^{2H})$ is a constant, ξ 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 $C(k) \sim$ const. if

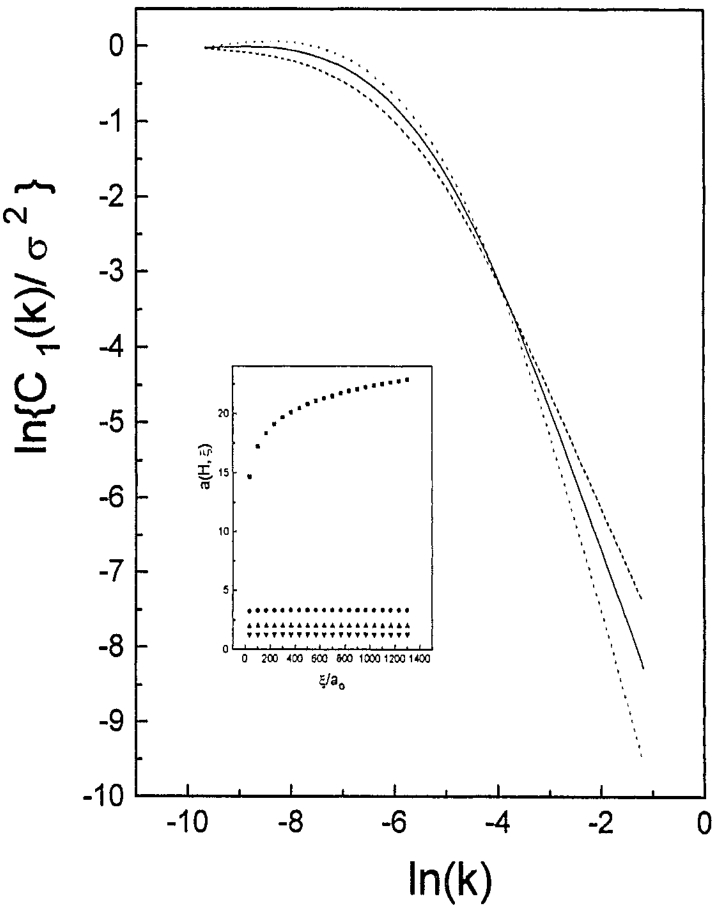

Fig. 1. Log–log plots of the one-dimensional roughness spectra $C_1(k)/\sigma^2$ vs k for $\xi = 100\,\mathrm{nm}$, $a_0 = 0.3\,\mathrm{nm}$ and roughness exponents H: $H = 0.3$ (dashes), $H = 0.5$ (solid), $H = 0.8$ (dots). The inset shows the change of the parameter $a = a(H, \xi)$ as a function of ξ/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 $g(x) = 2\sigma^2 - 2C(x)$. The roughness exponent H characterise the degree of surface irregularity, and has values in the range $0 < H < 1$. Small values of H ($H \sim 0$) correspond to highly irregular surfaces, and large values ($H \sim 1$) 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

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

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

$$C_2(x) = \frac{(\sigma^2 \xi / y)}{[1 + (|k|\xi)^f]^{-u}}; \quad y = 2\int_0^{V_c} (1 + \nu^f)^{-u}\,\mathrm{d}\nu \quad (uf = 1 + 2H), \tag{3}$$

where $V_c = k_c \xi$. The parameters $\{a, y, g\}$ are calculated from the normalisation condition $2\int C_i(k)\,\mathrm{d}k = \sigma^2$ ($0 < k < k_c$, $i = 1, 2, 3$) with $k_c = \pi/a_0$, 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 $H \to 0$: $(1/H)[x^H - 1] \to \ln(x)$ [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 $a \approx 1/H$ (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 $C(k) \sim (1 + ak^2\xi^2)^{-1-H}$ [10]. However, $C_1(k)$ does not reproduce the behaviour of the Fourier transform of $C_s(x)$ at $H = 0.5$ or $C_s(k, H = 0.5) \sim (1 + k^2\xi^2)^{-1}$.

On the other hand, $C_s(k, H = 0.5)$ suggests the generalisation for $H \neq 0.5$ that is given by $C_2(k)$. 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 $\int v^{c-1}[1 + v^a]^{-1}\,dv = \pi/[a\sin(\pi c/a)]$ $(0 < v < +\infty,\ 0 < c < a)$ yields $g(H > 0) \approx 2\pi[(1 + 2H)\sin(\pi/1 + 2H)]^{-1} - (1/H)[k_c\xi]^{-2H}$. Finally, the correlation function $C_3(k)$ is suggested according to the form of the HN-function or $\Phi(k) \sim [1 + i(ck)^a]^{-b}$ [12] whose connection with the $C_s(x)$ was established in the past [11]. The constraint $uf = 1 + 2H$ is imposed the requirement that at large k $(k\xi \gg 1)$; $C_3(k) \sim k^{-1-2H}$.

In former studies, a model similar to $C_{ct}(k) = \sigma^2\xi/(1 + a_{ct}k^2\xi^2)^{-(1+2H)/2}$ was introduced by Church and Takacs [16]. This is based again on the two-dimensional analogue of $C(k) \sim (1 + ak^2\xi^2)^{-1-H}$ [10], and "a_{ct}" is obtained from the normalisation of $C_{ct}(k)$, however, only in an integral form: $2\int(1 + av^2)^{-(1+H)/2}\,dv = 1$ $(0 < v < V_c)$.

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 $\sim 2\pi/4\xi$ (or $X_{knee} \sim 4\xi$ 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 $\sim\ln(\sigma^2\xi)$ (plateu, see Fig. 1) is important experimentally. This is because it provides a sufficient means to determine the correlation length ξ, 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]

$$\sigma^2(x) = 2\iint_{kx<k,k'<kc} \langle z(k)z(k')\rangle\,dk\,dk'; \tag{4}$$

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

where $k_x = 2\pi/x$, 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

$$\sigma_1^2(x) = \frac{\sigma^2}{aH}[(1 + ak_x\xi)^{-2H} - (1 + ak_c\xi)^{-2H}] \quad (0 < H < 1), \tag{5}$$

$$\sigma_1^2(x) = \frac{2\sigma^2}{a}\ln\left[\frac{1 + ak_c\xi}{1 + ak_x\xi}\right] \quad (H = 0). \tag{6}$$

The asymptotic behaviour of equations (5) and (6) is given by $\sigma_1(x) = [\sigma^2/(Ha)^{1/2}(2a\pi\xi)^H]x^H$ $(0 < H < 1)$ and $\sigma_1(x) = [\sigma^2/a^{1/2}][\ln(x/a_0)]^{1/2}$ $(H = 0)$ 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 $X_{knee} \sim 4\xi$.

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.* ($\sigma \approx 4$ nm, $\xi = 38.9$ nm, $H = 0.83$) [8] in terms of equation (5).

According to the scaling theory approach, during film growth [20], the normal roughness σ and the in-plane correlation length ξ evolve with film thickness h as $\sigma \sim h^b$ and $\xi \sim h^{b/H}$ (b and $z = H/b$ are respectively the growth and the dynamic exponent), as well as the surface width scales as $\sigma(x, h) = x^H F(h/x^{b/H})$ ($F(y) \sim y^b$ if $y \gg 1$, and $F(y) \sim$ const. if $y \ll 1$) with lateral size x and film time evolution $\sim h$. 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 $\sigma_1(x) = \sigma x^H[x + (2\pi\xi)/H]^{-H}$, where after substitution of the scaling relations $\sigma = vh^b$ and $\xi = qh^{b/H}$ we obtain for $\sigma_1(x, h) = x^H[A_1Y^b/(1 + A_2Y^{b/H})^H]$ with $Y = h/x^{b/H}$, $A_1 = v$ and $A_2 = 2\pi q/H$. $\sigma_1(x, h)$ 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 $C_1(x) \approx \sigma^2 - Dx^{2H}$

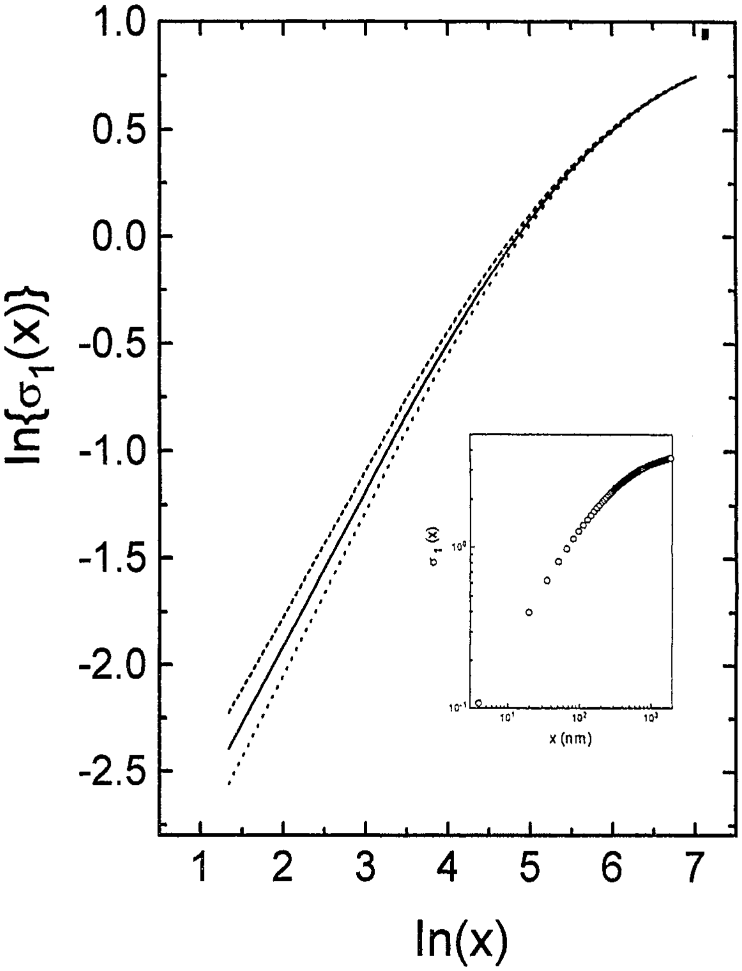

Fig. 2. Log–log plots of the one-dimensional surface width $\sigma_1(x)/\sigma$ vs x for $\xi = 100\,\text{nm}$, $a_0 = 0.3\,\text{nm}$ and roughness exponents H: $H = 0.3$ (dashes), $H = 0.5$ (solid-line), $H = 0.8$ (dots). The inset shows a plot (for comparison purposes) of the surface width for parameter values $\sigma = 4\,\text{nm}$ and $\xi = 39.8\,\text{nm}$ observed in Au-films by Vazquez *et al.* (Fig. 3, *Surf. Sci.* [8]) for $H = 0.83$.

or $g_1(x) \approx Dx^{2H}$ for $x \ll \xi$. In fact recently, there was a discussion about violation of the asymptotic power law behaviour $g(x) \approx Dx^{2H}$ 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; $g(x) \sim \ln(x)$ at $x \ll \xi$ [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 $C_s(x)$ [or $g_s(x)$] 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

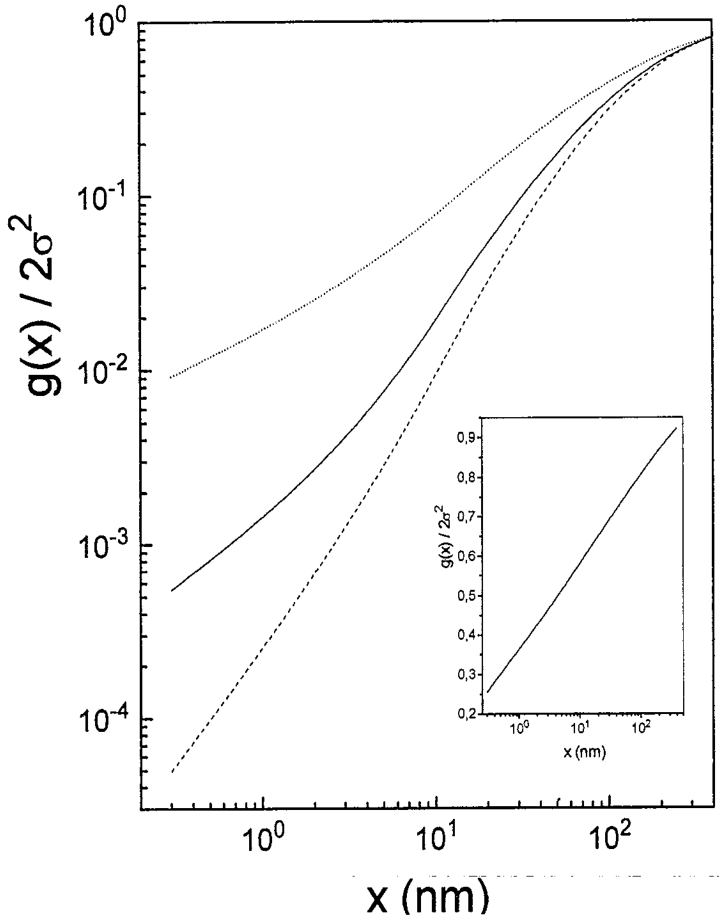

Fig. 3. Log–log plots of the height-difference correlation function $g_1(x) = 2\sigma^2 - 2C_1(x)$ vs x for $\xi = 100$ nm and $H = 0.5$ (dots), $H = 1$ (solid-line) and $H = 1.5$ (dashes). The inset shows a semi-log plot of $g_1(x)$ vs ln (x) for $H = 0$, where the linear behaviour reveal the logarithmic behaviour of the C_1-model.

of $\sigma(x)$ can be useful to estimate roughness parameters (σ and ξ) 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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