Bragg diffraction peaks in x-ray diffuse scattering from multilayers with rough interfaces

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The peaks in x-ray diffuse scattering appearing due to diffraction of incident or scattered waves on a periodically layered structure are sensitive to the perfection of multilayers. These peaks are investigated analytically and compared with numerical calculations. It is shown that, if the roughness of periodic interfaces is not correlated, the peaks follow the intensity of the x-ray standing wave on the interfaces. Interference effects due to correlated interfacial roughness can change the sense of the peak (sequence of maximum and minimum). The factors controlling the peak sense are derived and applied to explain the results of numerical calculations.

X-ray diffuse scattering has proved to be an informative nondestructive tool for the studies of roughness of surfaces and interfaces.[1–11] An essential feature of diffuse scattering from multilayer systems is the sensitivity to correlations between the roughnesses of interfaces. This effect is especially pronounced in periodic multilayers: when the roughness is correlated, diffuse intensity is concentrated on equidistantly spaced sheets in reciprocal space. All experimental studies report that the roughness of different interfaces is correlated, at least partially.

Additional singularities in the diffuse scattering are observed when either the incident or the scattered beam occurs in Bragg diffraction conditions due to the periodicity of the multilayer. It has been claimed, without proof, that the intensity distribution in Bragg singularities follows the standing-wave intensity at interfaces.[2,11] The aim of this paper is to investigate the formation of these singularities and their sensitivity to interlayer roughness correlations by means of analytical estimations compared with numerical calculations.

The x-ray diffuse scattering is due to random deviations $\delta\chi(\boldsymbol{r})$ of the polarizability of the scattered object from its mean distribution $\langle\chi(\boldsymbol{r})\rangle$. The most effective way for its calculation, in the lowest order over the perturbation $\delta\chi(\boldsymbol{r})$, is to apply the reciprocity theorem (see, e.g., Refs. 1 and 8). The amplitude of diffuse scattering can be represented as

$$f = (k^2/4\pi)\int \boldsymbol{E}^{\rm out}(\boldsymbol{r})\delta\chi(\boldsymbol{r})\boldsymbol{E}^{\rm in}(\boldsymbol{r})dV\ , \qquad (1)$$

where $\boldsymbol{E}^{\rm in}(\boldsymbol{r})$ and $\boldsymbol{E}^{\rm out}(\boldsymbol{r})$ are the wave fields in the object produced by x-ray plane waves incoming in the incidence direction and from the observation point, and k is the modulus of the wave vector in vacuum. Approximation (1) is commonly referred to as the distorted wave Born approximation (DWBA). Far from the regions of diffraction or reflection the waves $\boldsymbol{E}^{\rm in}$ and $\boldsymbol{E}^{\rm out}$ tend to plane waves with wave vectors inside the medium $\boldsymbol{K}^{\rm in}$ and $\boldsymbol{K}^{\rm out}$, which differ from corresponding vacuum waves $\boldsymbol{k}^{\rm in}$ and $-\boldsymbol{k}^{\rm out}$ due to refraction. The scattering amplitude (1) reduces to the refraction-corrected first Born approximation, $f = (k^2/4\pi)\delta\chi(\boldsymbol{Q})$, where $\boldsymbol{Q} = -(\boldsymbol{K}^{\rm in} + \boldsymbol{K}^{\rm out})$.

When the object possesses periodicity with a period D and either incident or scattered waves occur in the Bragg diffraction conditions, the corresponding wave is no longer a plane wave and the Born approximation is not valid. The diffracted wave has to be taken into account. The diffraction process close to an mth-order Bragg peak with the diffraction vector $H = 2\pi m/D$ can be considered as a scattering on the corresponding Fourier component χ_H of polarizability of the periodic system. Dynamical diffraction effects are governed by the ratio of the extinction length $\Lambda_H = 2\sin\theta/(k|\chi_H|)$ to the thickness $L = ND$ of a multilayer. In most practical cases, the extinction length is larger than the multilayer thickness for $m > 1$. Then the kinematical approximation can be applied to determine the wave fields $\boldsymbol{E}^{\rm in}$ and $\boldsymbol{E}^{\rm out}$.

Let us assume, for the sake of definiteness, that Bragg diffraction conditions are satisfied for the incident wave. Its amplitude is $E^{\rm in}(\boldsymbol{r}) = E_0(\boldsymbol{r})\exp(i\boldsymbol{K}_B\boldsymbol{r}) + E_H(\boldsymbol{r})\exp[i(\boldsymbol{K}_B + \boldsymbol{H})\boldsymbol{r}]$, where $\boldsymbol{H}$ is the actual diffraction vector and $\boldsymbol{K}_B$ is the wave vector in crystal satisfying the Bragg condition. In the kinematical approximation, the amplitude of the transmitted wave $E_0(\boldsymbol{r})$ does not change in the direction of its propagation, $\partial E_0(\boldsymbol{r})/\partial s_0 = 0$, where s_0 is the coordinate along $\boldsymbol{K}_B$. Applying the boundary condition at the multilayer surface $E_0|_{z=0} = \exp[i(\boldsymbol{K} - \boldsymbol{K}_B)\boldsymbol{r}]$, one has $E_0 = \exp(-2i\kappa s_H\sin\theta)$, where θ is the Bragg angle, $\kappa = (\boldsymbol{K} - \boldsymbol{K}_B)_z$ is the z component of the wave vector deviation from the Bragg condition, and s_H is the coordinate along $\boldsymbol{K}_B + \boldsymbol{H}$. The kinematical approximation implies that the amplitude of the diffracted wave E_H varies in the direction of its propagation due to scattering from the transmitted wave, $\partial E_H(\boldsymbol{r})/\partial s_H = -i(k\chi_H/2)E_0$. The integration of this equation gives

$$\begin{aligned} E_H &= i\frac{k\chi_H}{2}\int E_0 ds_H \\ &= \frac{k\chi_H}{4\kappa\sin\theta}\left\{e^{-2i\kappa s_H\sin\theta} - e^{2i\kappa L}e^{-2i\kappa s_0\sin\theta}\right\}. \end{aligned} \qquad (2)$$

The second term on the right-hand side of Eq. (2) is the integration constant ensuring the boundary condition on the bottom surface of the multilayer $E_H|_{z=L} = 0$. Proceeding to the rectangular coordinates along the surface and a normal to it, one has

$$E^{\rm in}(\boldsymbol{r}) = \exp(i\boldsymbol{K}^{\rm in}\boldsymbol{r}) + \frac{k\chi_H}{4\kappa\sin\theta}\{1 - e^{2i\kappa(L-z)}\} \times \exp[i(\boldsymbol{K}^{\rm in} + \boldsymbol{H})\boldsymbol{r}] \ . \tag{3}$$

Two terms in the curley brackets of Eq. (3) are due to direct diffraction at point $\boldsymbol{r}$ and reflection from the bottom surface of the slab, respectively. Substituting (3) into (1), we get

$$\frac{d\sigma}{d\Omega} = \langle |f|^2\rangle = \left(\frac{k^2}{4\pi}\right)^2 \Big\{\langle\delta\chi(\boldsymbol{Q})\delta\chi^*(\boldsymbol{Q})\rangle + {\rm Re}\frac{k\chi_H}{2\kappa\sin\theta}[\langle\delta\chi(\boldsymbol{Q}-\boldsymbol{H})\delta\chi^*(\boldsymbol{Q})\rangle - \exp(2i\kappa L)\langle\delta\chi(\boldsymbol{Q}-\boldsymbol{H}+2\boldsymbol{\kappa})\delta\chi^*(\boldsymbol{Q})\rangle]\Big\} \ . \tag{4}$$

Here the angular brackets $\langle\cdots\rangle$ denote averaging over fluctuations of polarizability, $\boldsymbol{\kappa}$ is the vector of the length κ directed normally to the interfaces, and $\delta\chi(\boldsymbol{Q})$ is the Fourier transform of the polarizability fluctuations. The first term in Eq. (4) corresponds to the Born approximation and two other terms are due to direct diffraction of the incident wave and the wave scattered from the bottom surface of the slab, respectively. Comparing them with the result of the Born approximation, one can see that they involve correlations between fluctuations of polarizability on two different wave vectors and thus, in principle, contain more information about correlations.

The polarizability fluctuations are due to variations in positions of the interfaces and can be represented as a sum over the interfaces, $\delta\chi(\boldsymbol{Q}) = \sum_j \delta\chi_j(\boldsymbol{Q})$. Then the correlations of polarizability fluctuations on the interfaces can be written, for a pair of the wave vectors $\boldsymbol{Q}_1$ and $\boldsymbol{Q}_2$ entering Eq. (4), as $\langle\delta\chi_j(\boldsymbol{Q}_1)\delta\chi_k^*(\boldsymbol{Q}_2)\rangle = \Delta\chi_j\Delta\chi_k e^{-i(Q_{1z}z_j - Q_{2z}z_k)}C_{jk}$. Here $C_{jk}(Q_{1z}, Q_{2z}, \boldsymbol{q}_\perp)$ is a correlation function, the explicit form of which depends on the model of roughness and can be found elsewhere,[7–9] and $\Delta\chi_j$ is the difference between the polarizabilities of two media forming the interface.

Consider a slab of N periods of thickness $D = d_1 + d_2$, each period consisting of two layers with thicknesses d_1 and d_2 and polarizabilities χ_1 and χ_2, respectively. The Fourier component of polarizability of the slab is $\chi_H = 4/(HD)\Delta\chi\exp(-iHd_1/2)\sin(Hd_1/2)$, where $\Delta\chi = \chi_2 - \chi_1$. The phase factor $\exp(-iHd_1/2)$ arises due to the distance $z = d_1/2$ of the symmetry plane of multilayer "unit cell" from the origin $z = 0$ chosen at the multilayer surface.

If roughness of different interfaces is not correlated, one can set $C_{jk} = C\delta_{jk}$. Then one arrives at

$$\frac{d\sigma}{d\Omega} \sim NC\,[1 + g\xi S_u\Psi(\kappa D)] \ , \tag{5}$$

where $g = 4k^2\Delta\chi/H^2$, parameter ξ is a factor $\simeq 1$, and

$$S_u = \sin(Hd_1) \tag{6}$$

is a factor determining the sign and magnitude of the Bragg diffraction effect. The function $\Psi(x) = x^{-1}\left[1 - \cos\left((N+1)x\right)\sin(Nx)/(N\sin x)\right]$, antisymmetric with respect to its argument, possesses dispersionlike behavior with a minimum and a maximum.

Let us calculate, for comparison, the total intensity of the wave field $E^{\rm in}(\boldsymbol{r})$ on the interfaces $\boldsymbol{r}_j = (\boldsymbol{\rho}, z_j)$: $I = \sum_j |E^{\rm in}(\boldsymbol{\rho}, z_j)|^2 = 2N\,[1 + gS_u\Psi(\kappa D)]$. Thus, the diffuse scattering on multilayers with uncorrelated roughness follows the intensity of an x-ray standing wave on interfaces, differing from it only by a slowly varying factor $\xi \sim 1$. The dispersionlike curve of $\Psi(x)$ is typical of the standing wave.

Let us proceed now to the opposite limiting case of completely correlated roughness and set $C_{jk} = C$. Then we obtain

$$\frac{d\sigma}{d\Omega} \sim C\Phi^2(Q_z)\left[\sin^2(Q_zd_1/2) + g\xi S_c\Psi_c(\kappa D)\right] \ , \tag{7}$$

where $\Phi(Q_z) = \sin(Q_zND/2)/\sin(Q_zD/2)$, and

$$S_c = \sin(Hd_1/2)\sin\left[(Q_z - H)d_1/2\right]\sin(Q_zd_1/2) \ . \tag{8}$$

In a general case the function $\Psi_c(x)$ differs from $\Psi(x)$. However, for scans along a resonant diffuse scattering (RDS) maximum, where Q_z is equal to one of the Bragg diffraction vectors $Q_z = G = 2\pi n/D$, the function $\Psi_c(x)$ is reduced to $\Psi(x)$. Then the contrast of diffuse scattering peaks due to completely correlated roughness differs from that due to uncorrelated roughness by substitution of S_c for S_u only.

Bragg diffraction of the incident wave on a periodic multilayer structure gives rise to a standing wave illuminating rough interfaces.[2,11,12] If roughness of the interfaces is not correlated, the diffraction peaks in diffuse scattering follow the standing-wave distribution, the sequence of the maximum and the minimum being governed by the sign of the factor S_u. Correlations between roughness of the interfaces gives rise to the interference of diffusely scattered waves, which can reverse the sequence of the maximum and the minimum, depending of the sign of the factor S_c for given reciprocal-lattice vectors G and H. It is worth noting that the shape of the Bragg diffraction peaks can also be sensitive to an asymmetry between B/A and A/B interfaces in periodic multilayers.[12]

It can be shown that the qualitative features of Bragg peaks are not sensitive to minor deviations from completely correlated roughness. Let us assume a slow variation of C_{jk} with j and k, so that for two interfaces belonging to one and the same period of multilayer $C_{jk} \approx C_{j+1,k}$ and we can proceed from summation over interfaces to summation over periods. That gives the expression for $d\sigma/d\Omega$ where the sign of the Bragg peak is still controlled by S_c. Therefore, a qualitative transition from one extreme to the opposite one can be expected in case of a fast decrease in correlations with a distance between interfaces, when the assumptions of slow variations of C_{jk} within one period are not valid. This conclusion is confirmed by numerical calculations below.

To proceed to the numerical tests of our analytical estimations, we have to choose an explicit form of the correlation function C_{jk}. A commonly used approximation is to treat a rough interface as a sharp boundary between two media with different polarizabilities, randomly displaced by $u_j(\boldsymbol{\rho})$ from its mean position z_j. Then the correlation functions C_{jk} can be expressed[7–9] in terms of the rms displacements of interfaces, $\sigma_j^2 = \langle u_j^2(\boldsymbol{\rho})\rangle$, and the displacement-displacement correlation functions $\mathcal{K}_{jk}(\boldsymbol{\rho}) = \langle u_j(0)u_k(\boldsymbol{\rho})\rangle$. For $j = k$ the correlation functions are commonly taken, after Ref. 1, as $\mathcal{K}_{jj}(\rho) = \sigma_j^2 \exp[-(\rho/\xi_j)^{2h}]$, where ξ_j is the lateral correlation length of roughness at the jth interface, and $3-h$ is a fractal dimension. For $j \neq k$ there is a variety of suggestions, assuming either nonaccumulated[3,4] or accumulated[5,7] roughness. In the first case, the correlations between interfaces are supposed to depend on the distance between them. For example, in the model by Ming *et al.*[3] the correlation function has the form

$$\mathcal{K}_{jk}(\boldsymbol{\rho}) = \sqrt{\mathcal{K}_{jj}(\boldsymbol{\rho})\mathcal{K}_{kk}(\boldsymbol{\rho})}\, \exp(-|z_j - z_k|/\xi_{\text{vert}}), \tag{9}$$

where ξ_{vert} is a vertical correlation length of roughness. Usually, one takes all $\mathcal{K}_{jj}$ equal to each other, $\mathcal{K}_{jj} = \mathcal{K}$. The correlations of roughness of different interfaces can vary between the limits of uncorrelated interface roughness, $\mathcal{K}_{jk}(\boldsymbol{\rho}) = \mathcal{K}(\boldsymbol{\rho})\delta_{jk}$, at $\xi_{\text{vert}} \to 0$, and completely correlated (replicated) roughness, $\mathcal{K}_{jk}(\boldsymbol{\rho}) = \mathcal{K}(\boldsymbol{\rho})$, at $\xi_{\text{vert}} \to \infty$.

Spiller, Stearns, and Krumrey[5] took into consideration the fact that interfaces are formed successively one after another, starting with the substrate and with the layers repeating long-wavelength modulations in the interface positions, whereas the short-wavelength roughness of an interface is smeared out and appears on the next interface independently. Then the Fourier transforms of displacements of subsequent interfaces are related by the recurrent formula[5] $u_j(\boldsymbol{f}) = h_j(\boldsymbol{f}) + a_j(\boldsymbol{f})u_{j+1}(\boldsymbol{f})$, where $\boldsymbol{f}$ is a two-dimensional wave vector in the interface plane, and h_j are random displacements acquired at the jth interface ($\langle h_j h_k\rangle = 0$ for $j \neq k$). The functions a_j govern the roughness transfer; they were taken[5] on the assumption of diffusionlike roughness propagation as $a_j(\boldsymbol{f}) = \exp[-\nu_j(z_{j+1} - z_j)f^2]$, where ν_j are relaxation parameters. A spatial frequency dependence of the number of interfaces involved in correlations has been observed recently.[4,10]

Here we derive the correlation function of displacements $\mathcal{K}_{jk}(\boldsymbol{\rho})$ for this model on the assumption that the relaxation parameters are equal to each other, $\nu_j = \nu$. Starting from the substrate roughness $u_N \equiv h_N$ and iteratively expressing u_j's, we arrive at

$$\mathcal{K}_{jk}(\boldsymbol{\rho}) = \sum_{n=\max(j,k)}^{N} \int d^2 f\, \langle h_n^2(\boldsymbol{f})\rangle \times \exp[-\nu(2z_n - z_j - z_k)f^2] \exp(i\boldsymbol{f}\boldsymbol{\rho})\ . \tag{10}$$

The limit $\nu \to 0$, implying complete roughness transfer at all spatial frequencies and the assumption of similar acquired roughness at all interfaces, gives rise to the correlation function applied in Ref. 7. We consider the more general case $\nu \neq 0$. As noted in Ref. 6, the major part of the diffuse scattering pattern has a weak dependence on the fractal dimensionality h in this function. Taking $h = 1$ (Gaussian distribution) we obtain

$$\mathcal{K}_{jk}(\boldsymbol{\rho}) = \sum_{n=\max(j,k)}^{N} \sigma_n^2 \frac{\xi_n^2}{\xi_n^2 + p_n^2} \exp\left[-\frac{\boldsymbol{\rho}^2}{\xi_n^2 + p_n^2}\right]\ , \tag{11}$$

where $p_n^2 = 4\nu(2z_n - z_j - z_k)$. In the limit $\nu \to \infty$ one has $\mathcal{K}_{j\neq k}(\boldsymbol{\rho}) = 0$, i.e., the absence of vertical correlations. In the opposite limit $\nu \to 0$ the roughness is completely transferred and accumulated. Taking all ξ_n equal to each other, $\xi_n = \xi$, one can introduce, for convenience of comparing with the model of nonaccumulated roughness, the vertical correlation length $\xi_{\text{vert}} = \xi^2/\nu$.

Figure 1 presents the distributions of diffuse scattering intensity for different values of ξ_{vert} in (9). The most pronounced feature due to the correlations of roughness of interfaces is the concentration of the intensity on equidistantly spaced RDS sheets.[2–4,7,9,10] These sheets are indicated in the figure by arrows with the mark "1." The diffuse scattering at all ξ_{vert} possesses, in addition, the singularities along the lines where either the incident or the scattered wave occurs in the Bragg diffraction condi-

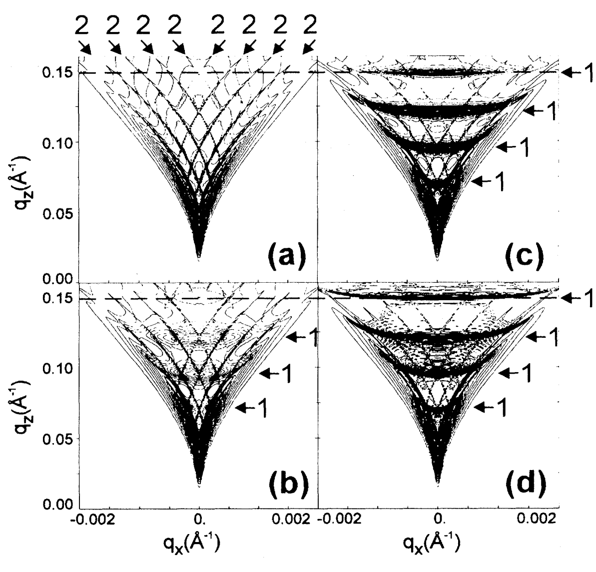

FIG. 1. Formation of resonant sheets in x-ray diffuse scattering from periodic multilayers with an increase in the vertical correlation length of interface roughness. The computations are for the superlattice consisting of 20 periods of (95 Å GaAs/125 Å AlAs) on a GaAs substrate; $\lambda = 1.5$ Å; the parameters of interfacial roughness are $\sigma = 8.6$ Å, $h = 1$, and $\xi = 2000$ Å. The vertical correlation length is $\xi_{\text{vert}} = 0$ (a), 200 Å (b), 1000 Å (c), and ∞ (d), respectively. Arrows 1 and 2 mark the resonance sheets and the traces of Bragg singularities. The dashed lines show the place of sections presented in Fig. 2.

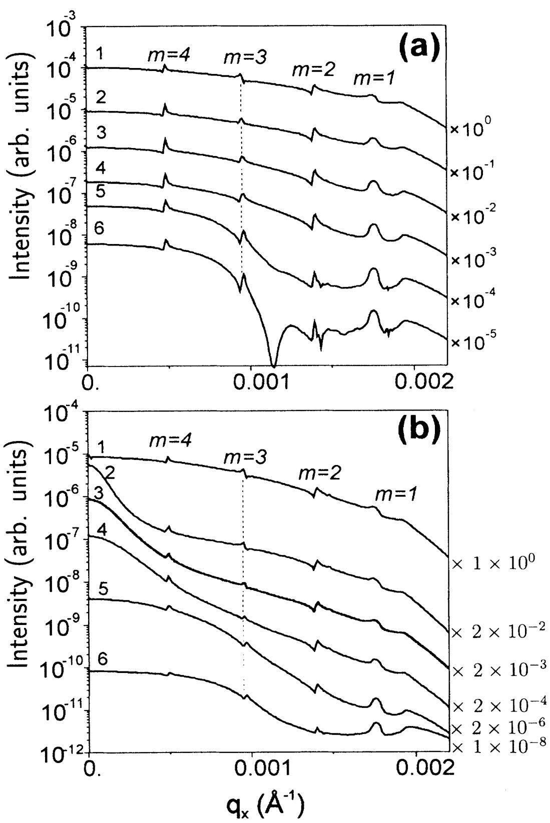

FIG. 2. Transformation of Bragg peaks in the section q_z =const along the fifth resonance sheet with an increase in the vertical correlation length of interface roughness for models of nonaccumulated (a) and accumulated (b) roughness. Lines 1–6 correspond to $\xi_{\rm vert}$ = 0, 200, 500, 1000, 10 000, and ∞ Å, respectively. The vertical dotted line follows the transformation of the third-order Bragg peak. The parameters of calculations are the same as in Fig. 1 with the exception of acquired rms roughness in (b): $\sigma_{n<N} = 1.6$ Å and $\sigma_N = 8.6$ Å.

tion. These lines form a regular mesh on the isointensity maps (see arrows with the mark "2"). In the case of uncorrelated roughness, the singularities on the lines are completely characterized by the order m of the diffraction vector $H = 2\pi m/D$ and consist of a maximum and a minimum of intensity, whose sequence is controlled by the factor S_u, Eq. (6). In the case of correlated roughness, the sense of the Bragg diffraction peaks is controlled by another factor, S_c, Eq. (8), and depends also on the order n of the RDS sheet ($Q_z = 2\pi n/D$). For the value $d_1/D = 0.43$ used in the calculations and for m=3, $n = 5$, the two factors, $S_u = 0.97$ and $S_c = -0.15$, have opposite signs. Then the third-order peak on the fifth RDS is expected to possess *opposite contrasts* in cases of correlated and uncorrelated roughness. This behavior is confirmed by the lines 1 and 6 in Fig. 2(a).

Figure 2 also illustrates the process of the Bragg singularity transformation with increasing vertical correlation for the two models of vertical roughness correlation. Figure 2(a) shows that the contrast becomes already inverted at minor vertical correlations, the parameter $\xi_{\rm vert}$ = 200 Å being less than one multilayer period, thus confirming the estimations given above. Comparison of Figs. 2(a) and 2(b) demonstrates that, with increasing the vertical correlation length, the diffuse intensity in Spiller's model increases first for small q_x, due to prior transfer of the long-wavelength interface roughness. It can also be noted that the contrast of the third Bragg peak inverts later than that in the nonaccumulated roughness model, although this peak changes its sense at a rather small vertical correlation length as well ($\xi_{\rm vert}$ is less than five periods).

Thus, for completely uncorrelated roughness of interfaces, Bragg singularities follow the intensity of the x-ray standing wave at interfaces. The contrast of the peaks can invert due to minor correlations between roughness of different interfaces, which are insufficient for formation of the resonance diffuse scattering sheets. The factors controlling the contrast of these singularities have been found analytically and confirmed by numerical calculations. The inversion of the Bragg peak sense might be helpful in studies of minor correlations between rough interfaces.

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