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Modified Critical Correlations Close to Modulated and Rough Surfaces

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Correlation functions are sensitive to the presence of a boundary. Surface modulations give rise to modified near surface correlations, which can be measured by scattering probes. To determine these correlations, we develop a perturbative calculation in deformations in height from a flat surface. The results, combined with a renormalization group around four dimensions, are also used to predict critical behavior near a self-affine rough surface. We find that a large enough roughness exponent can modify surface critical behavior.

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Bulk properties, such as magnetization, as well as correlation functions are modified on approaching a surface. In particular, critical behavior near surfaces or defects, which is quite different from the bulk, has been extensively studied theoretically [1–4], by experiments [5–7], and in simulations [8]. Along with this development, the method of grazing incidence of x rays and neutrons [3] has become a standard tool of probing critical behavior near surfaces and interfaces [5–7].

Most theoretical studies have been restricted to flat surfaces. However, real surfaces mostly deviate from this idealized picture. Possible deviations can be divided into two classes: (i) advanced experimental methods, e.g., lithographic preparation, allow one to endow surfaces with specific, regular geometrical patterns down to the nanometer scale, with important applications in technology and material science [9]; (ii) surfaces or interfaces can be naturally rough, e.g., due to growth, fracture, or erosion. Most common are self-affine rough surfaces, in which the root mean square height fluctuations on a length scale L grow as L^ζ , where $\zeta < 1$ is the so-called roughness exponent. Self-affine scaling is predicted by many numerical and analytical models of surface growth [10], and is also observed in a number of experiments [11].

In this work we show that the shape of the surface has a distinct influence on the properties of an adjacent medium with long-range correlations. This is demonstrated for two-point correlation functions near a critical point of the medium, for both cases (i) and (ii) outlined above. The diffuse scattering of x rays and neutrons at grazing incidence due to such correlations appears in addition to what would have been observed if the surface were separating two homogeneous media [12]. The modified correlations may thus provide an additional and indirect means of characterizing the surface profile. This may be of value when other techniques are not possible, as in the case of the interior surface of a glass, or an internal crack, whereas scattering from a critical fluid or binary alloy coating the surface may be feasible.

In order to study the effects of the surface shape, we develop a perturbative expansion of two-point correlation functions in the deformations of the height profile. Ini-

tially for a Gaussian field, the calculations are carried out to second order. Already at the first order, the two-point correlation functions track the profile from the substrate, with a modulation that decreases with the distance of the two points from the surface. This leads to explicit predictions for the structure factor, as a function of the lateral wave vector transfer, for a modulated surface [see Eqs. (7) and (8) below]. For example, for a sine modulation with wavelength $2\pi/k$ along one direction, the incident wave vector is scattered by k in that direction, with amplitude proportional to the modulation in height.

For self-affine rough surfaces, second order calculations are necessary, as the first order results vanish on average. For a massless Gaussian field we find the expected result that self-affine roughness leads to subleading corrections to the decay of two-point correlation functions, which at a scale r are smaller by a factor of $r^{-2(1-\zeta)}$ than the leading contribution coming from a flat surface. Typical critical systems, however, are described by a non-Gaussian (interacting) field theory. In this case, the correlations are calculated perturbatively in the strength of the interaction, and the results interpreted with the aid of the renormalization group (RG) in $4 - \epsilon$ dimensions [see Eqs. (11)–(14) below]. We find that the subleading corrections now fall off with a slower power as compared to the Gaussian case and, surprisingly, for a sufficiently large ζ even *dominate*, giving rise to novel surface critical behavior.

Fluctuations in the critical system located above and bounded by the surface will be described by an n -component field $\Phi(\underline{r}) = [\Phi_1(\underline{r}), \dots, \Phi_n(\underline{r})]$, where \underline{r} is assumed to be d dimensional. For example, $n = 1$ can represent an Ising magnet or binary alloy, while $n = 2$ may describe a superfluid. The fluctuations are described by the statistical Boltzmann weight $e^{-\beta\mathcal{H}}$, with

$$\beta\mathcal{H}\{\Phi\} = \int d^d r \left\{ \frac{1}{2} (\nabla\Phi)^2 + \frac{\tau_0}{2} \Phi^2 + \frac{u_0}{4!} (\Phi^2)^2 \right\}, \quad (1)$$

where u_0 is the strength of the interaction, set to zero in the Gaussian theory, and $\tau_0 \sim T - T_c$. The above expression must be supplemented by a boundary condition on the surface. We choose the Dirichlet boundary condition $\Phi = 0$,

which represents the so-called ordinary surface universality class, appropriate to magnets, binary alloys, and for ^4He near the normal to superfluid transition point [1,2,13].

In the absence of overhangs and inlets, the surface profile can be described by a single-valued height function $h(\mathbf{x})$, where \mathbf{x} spans a $D = d - 1$ dimensional base plane (see Fig. 1). The Gaussian correlation $\langle \Phi_i(\underline{r}) \Phi_j(\underline{r}') \rangle = \delta_{ij} G(\underline{r}; \underline{r}')$, according to Eq. (1) with $u_0 = 0$, can be calculated using functional integral methods [14,15] and is given by

$$G(\underline{r}; \underline{r}') = G_b(\underline{r}; \underline{r}') - \int d^D x \int d^D y \times G_b[\underline{r}; \mathbf{x}, h(\mathbf{x})] M(\mathbf{x}, \mathbf{y}) G_b[\underline{r}'; \mathbf{y}, h(\mathbf{y})]. \quad (2)$$

Note that we denote d dimensional vectors with underlined letters, and D dimensional vectors with boldface letters. Position vectors \underline{r} are thus decomposed according to $\underline{r} = (\mathbf{r}_{\parallel}, z)$, where \mathbf{r}_{\parallel} comprises the $D = d - 1$ components parallel to the surface and z is the distance from the base plane; G_b is the bulk correlation function of the Gaussian theory, and the kernel $M(\mathbf{x}, \mathbf{y})$ satisfies

$$\int d^D y M(\mathbf{x}, \mathbf{y}) G_b[\mathbf{y}, h(\mathbf{y}); \mathbf{y}', h(\mathbf{y}')] = \delta^D(\mathbf{x} - \mathbf{y}'). \quad (3)$$

While the above results are generally valid, we focus on the behavior of the correlation functions at the bulk critical point, i.e., for $T = T_c$, where correlations are strongest.

The solution for G can be expanded in a series $G_0 + G_1 + G_2 + \dots$ in powers of $h(\mathbf{x})$. The lowest order result, $G_0(\underline{r}; \underline{r}') = G_b(\mathbf{r}_{\parallel}, z; \mathbf{r}'_{\parallel}, z') - G_b(\mathbf{r}_{\parallel}, z; \mathbf{r}'_{\parallel}, -z')$, corresponds to a flat surface. The bulk correlation function $G_b(\underline{r}; \underline{r}')$ decays as $r^{-(d-2+\eta)}$ for large separations $r = |\underline{r} - \underline{r}'|$, where the bulk critical exponent η is given by $\eta = 0$ in the Gaussian theory. In contrast, if both points remain close to the surface, $G_0(\underline{r}; \underline{r}')$ decays as $r^{-(d-2+\eta_{\parallel})}$, where η_{\parallel} is a surface critical exponent given by $\eta_{\parallel} = 2$ in the Gaussian theory [1,2].

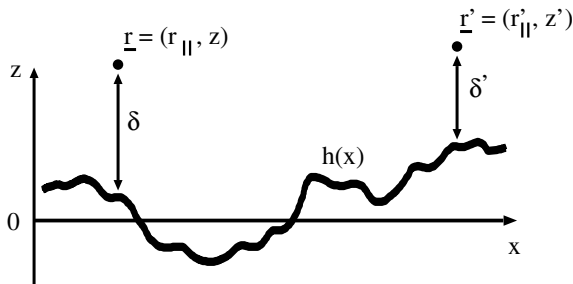


FIG. 1. Position vectors $\underline{r} = (\mathbf{r}_{\parallel}, z)$ and $\underline{r}' = (\mathbf{r}'_{\parallel}, z')$ of the two-point correlation function in the critical system located above and bounded by a deformed surface. The surface profile is described by the height function $h(\mathbf{x})$ and the vertical distances of \underline{r} and \underline{r}' from the surface are given by $\delta = z - h(\mathbf{r}_{\parallel})$ and $\delta' = z' - h(\mathbf{r}'_{\parallel})$, respectively.

The first order result is given by

$$G_1(\underline{r}; \underline{r}') = - \int d^D x \Delta(\mathbf{r}_{\parallel} - \mathbf{x}, z) h(\mathbf{x}) \Delta(\mathbf{r}'_{\parallel} - \mathbf{x}, z'), \quad (4)$$

where $\Delta(\mathbf{x}, z) = \int \frac{d^D p}{(2\pi)^D} e^{i\mathbf{p}\cdot\mathbf{x}} e^{-pz}$, with $p = |\mathbf{p}|$, has the form of a representation of the delta function $\delta^D(\mathbf{x})$, i.e., $\int d^D x \Delta(\mathbf{x}, z) = 1$ and $\lim_{z \rightarrow 0} \Delta(\mathbf{x}, z) = \delta^D(\mathbf{x})$. The first order result G_1 tracks the profile $h(\mathbf{x})$ of the surface. For example, for $r = |\underline{r} - \underline{r}'| \rightarrow \infty$ with z and z' fixed, the results for G_0 and G_1 imply the behavior

$$G(\underline{r}; \underline{r}') \sim [1 - A(\underline{r}) - A(\underline{r}')] r^{-(d-2+\eta_{\parallel})}, \quad (5)$$

up to terms of order $(h/z)^2$ and $(h/z')^2$. Thus, the leading power law is the same as for a flat surface, but the amplitude is modulated by the surface deformations in the vicinity of \mathbf{r}_{\parallel} and \mathbf{r}'_{\parallel} by [16]

$$A(\underline{r}) = \frac{\eta_{\parallel} - \eta}{2} \int d^D x \frac{h(\mathbf{x})}{z} \Delta(\mathbf{x} - \mathbf{r}_{\parallel}, z). \quad (6)$$

Already the results at first order indirectly characterize the surface in scattering experiments. To demonstrate this, we introduce the Fourier transform of the height profile as $h(\mathbf{x}) = \int \frac{d^D k}{(2\pi)^D} e^{i\mathbf{k}\cdot\mathbf{x}} \hat{h}(\mathbf{k})$, with $\hat{h}(-\mathbf{k}) = \hat{h}(\mathbf{k})^*$, and accordingly the lateral structure factor $S(\mathbf{p}, z; \mathbf{p}', z')$ corresponding to $G(\underline{r}, \underline{r}')$ (where the parallel component \mathbf{r}_{\parallel} is transformed to the lateral wave vector \mathbf{p}). The first order result in Eq. (4) then gives

$$S_1 = -e^{-pz} e^{-p'z'} \hat{h}(\mathbf{p} + \mathbf{p}'). \quad (7)$$

For example, for a sine modulation with wavelength $2\pi/k$ along, say, the x axis, this implies that the incident wave vector component p_x is scattered to $p'_x = p_x \pm k$ while the other components of \mathbf{p} remain unchanged.

A similar behavior occurs for the contribution to S at second order in h , which is given by

$$S_2 = -e^{-pz} e^{-p'z'} \int \frac{d^D k}{(2\pi)^D} |\mathbf{p} - \mathbf{k}| \times \hat{h}(\mathbf{k}) \hat{h}(\mathbf{p} + \mathbf{p}' - \mathbf{k}). \quad (8)$$

For the sine modulation, this implies that p_x is scattered by $2k, 0, -2k$. In a scattering experiment with grazing incidence, the length scale perpendicular to the surface is set by the depth b the evanescent wave penetrates the sample, giving rise to diffuse scattering and thereby probing the critical correlations close to the surface [3]. Since this diffuse scattering appears in addition to the contribution already present away from criticality [12], it can in principle be separated out by tuning the temperature deviation $T - T_c$. We assume that b is much larger than the height of the deformations. In this case, the above expansion in the deformations results in an expansion in powers of $h/b \ll 1$ for the elastic scattering cross section, which

allows one to distinguish the corresponding contributions via their intensities.

The second order results are particularly useful when dealing with rough surfaces, where the quench averaged first order corrections vanish. In particular, we shall assume that the rough surface is described by a height function $h(\mathbf{x})$ with $\overline{h(\mathbf{x})} = 0$, and

$$\begin{aligned} \overline{[h(\mathbf{x}) - h(\mathbf{y})]^2} &= \omega^{2-2\zeta} |\mathbf{x} - \mathbf{y}|^2 \\ &\times \int \frac{d^D p}{(2\pi)^D} e^{i\mathbf{p}\cdot(\mathbf{x}-\mathbf{y})} p^{-D+2-2\zeta} e^{-p\lambda}, \end{aligned} \quad (9)$$

where the overbar denotes averaging over different surface profiles. While at large separations the above correlations grow as $|\mathbf{x} - \mathbf{y}|^{2\zeta}$, we have also introduced a cutoff length λ to regulate the behavior of the surface at short distances, and an overall amplitude length ω .

A characteristic feature of self-affine roughness is statistical translational invariance, since the rhs of Eq. (9) depends on the distance $|\mathbf{x} - \mathbf{y}|$ only. This implies that the averaged structure factor \overline{S} is proportional to $\delta^D(\mathbf{p} + \mathbf{p}')$, and depends on z , z' , and $p = |\mathbf{p}|$ only. In order to maintain translational invariance, it is convenient to express the results for the correlation functions in terms of the local distance $\delta = z - h(\mathbf{r}_{\parallel})$ from the surface rather than z (see Fig. 1). The two-point correlation function must now vanish as δ or δ' go to zero. In terms of these coordinates, the leading power law behavior of correlations is the same as for a flat surface, but the corresponding amplitude depends on the roughness and is modified by a factor of $[1 - \alpha(\omega/\lambda)^{2(1-\zeta)}]$ as compared to a flat surface, where $\alpha > 0$ is a number of order unity. The subleading correction of order $\overline{h^2}$ decays with the separation $r = |\underline{r} - \underline{r}'|$ with an additional factor of $r^{-2(1-\zeta)}$ compared to the leading term.

For the interacting field theory, governed by Eq. (1) with $u_0 \neq 0$, standard perturbation theory can be applied to get the correlation function

$$\begin{aligned} \langle \Phi_i(\underline{r}) \Phi_i(\underline{r}') \rangle &= G(\underline{r}; \underline{r}') - \frac{n+2}{3} \frac{u_0}{2} \int d^d R \\ &\times G(\underline{r}; \underline{R}) G(\underline{R}; \underline{R}) G(\underline{R}; \underline{r}') + \mathcal{O}(u_0^2). \end{aligned} \quad (10)$$

For a flat surface, the one-loop addition in u_0 can be regularized and renormalized by minimal subtraction of poles in $\varepsilon = 4 - d$, leading to logarithmic contributions in the separation $r = |\underline{r} - \underline{r}'|$. This perturbative result can then be improved by RG, resulting in power laws in r with corresponding surface critical exponents [1,2]. For a self-affine rough surface, the expansion of G to second order in $h(\mathbf{x})$ is substituted in the above equation, and the quench average is obtained using Eq. (9). The one-loop correction then gives rise to six diagrams of order $u_0 \overline{h^2}$. Similarly as for a flat surface, the new diagrams in the

quench average at order of $u_0 \overline{h^2}$ produce logarithmic contributions in r , which can again be recast as power laws. The final results for the two-point correlation function at criticality are summarized below.

Perpendicular correlations are obtained when \underline{r} moves into the bulk while \underline{r}' remains close to the surface, which implies $r = |\underline{r} - \underline{r}'| \rightarrow \infty$ while δ' is fixed (see Fig. 1). In this case the correlations decay as

$$\overline{\langle \Phi_i(\underline{r}) \Phi_i(\underline{r}') \rangle} \sim \frac{1}{r^{d-2+\eta_{\perp}}} + \frac{a}{r^{d-2+\tilde{\eta}_{\perp}}}, \quad (11)$$

where the first term corresponds to a flat surface with $\eta_{\perp} = 1 - \frac{1}{2} \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2)$. The second term describes the effect of self-affine roughness, with an amplitude a depending on ω , λ , and ζ , and the new universal exponent

$$\tilde{\eta}_{\perp} = (2 - 2\zeta) + 1 - 2 \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2). \quad (12)$$

Similarly, when both points remain close to the surface, i.e., both δ and δ' are fixed, correlations fall off as

$$\overline{\langle \Phi_i(\underline{r}) \Phi_i(\underline{r}') \rangle} \sim \frac{1}{r^{d-2+\eta_{\parallel}}} + \frac{a'}{r^{d-2+\tilde{\eta}_{\parallel}}}. \quad (13)$$

In this case the flat surface is governed by $\eta_{\parallel} = 2 - \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2)$, while self-affine roughness gives

$$\tilde{\eta}_{\parallel} = (2 - 2\zeta) + 2 - 4 \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2). \quad (14)$$

The corrections due to roughness now decay with a slower power as compared to the Gaussian case. Indeed, for a sufficiently large roughness exponent ζ , these corrections can even dominate the result for the flat surface. The borderline roughness exponent is $\zeta_{\perp}^* = 1 - \frac{3}{4} \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2)$ for perpendicular, and a different value of $\zeta_{\parallel}^* = 1 - \frac{3}{2} \frac{n+2}{n+8} \varepsilon + \mathcal{O}(\varepsilon^2)$ for parallel correlations. This is a surprising result from a naive point of view since, due to $\zeta < 1$, on larger and larger length scales a self-affine rough surface looks more and more like a flat surface. Note that this effect becomes visible only beyond the Gaussian approximation, which corresponds to $\varepsilon = 0$.

To test the generality of this result, we examined the correlations for a $d = 2$ dimensional XY model below the Kosterlitz-Thouless temperature [17]. Correlations in the spin variables $s(\underline{r}) = e^{i\theta(\underline{r})}$ decay with power laws in this case. We found that the surface correlations fall off with the simple relative factor of $r^{-2(1-\zeta)}$. We attribute this to the Gaussian nature of the fluctuations in the phase angle $\theta(\underline{r})$, which are retained in the asymptotics of correlations for $s(\underline{r})$.

Thermodynamic quantities and correlation functions can be obtained from derivatives of the free energy with respect to magnetic fields. To discuss surface behavior, we introduce distinct fields h_b and h_s in the bulk and close to the surface, respectively. Assuming that our underlying assumption of the validity of an expansion in $h(\mathbf{x})$ holds, the

results for the two-point correlation function are consistent with the following form for the scaling of the leading singular part of the surface free energy per projected area,

$$f_s^{(\text{sing})} = \xi^{-d+1} [g_s(h_b \xi^{y_b}, h_s \xi^{y_s}) + \xi^{-2(1-\zeta)} g_r(h_b \xi^{y_b}, h_s \xi^{\tilde{y}_s})], \quad (15)$$

where $\xi \sim |T - T_c|^{-\nu}$ is the correlation length that diverges at the critical point. The first term in square brackets corresponds to a flat surface with y_b and y_s describing the relevance of bulk and surface fields, respectively [1,2]. The second term gives the effect of surface roughness, with $\xi^{-2(1-\zeta)}$ reflecting the average increase in area. However, to regain the results in Eqs. (11)–(14) we have to use a value of $\tilde{y}_s = 1 + \frac{3n}{2(n+8)}\varepsilon + \mathcal{O}(\varepsilon^2)$ which is different from $y_s = 1 - \frac{3}{n+8}\varepsilon + \mathcal{O}(\varepsilon^2)$. To motivate and justify this difference, we resort to an analogy in which the rough surface is replaced with a collection of edges with a (possibly scale-dependent) distribution of opening angles. Already for a single edge, describing correlations requires a distinct and angle-dependent value of y_e for the magnetic field close to the edge [18,19]. Similarly, results obtained recently for correlations in the vicinity of a fractal surface with fractal dimension d_f [20,21] cannot be obtained using the value of y_s for a flat surface [with ξ^{-d_f} replacing ξ^{-d+1} in Eq. (15) and omitting the second term in square brackets]. Thus \tilde{y}_s can be regarded as inherently related to self-affine geometry. Interestingly, however, it does not depend on the roughness exponent ζ , at least to order ε .

Future extensions of this research could focus on other surface universality classes, the possibility of multifractal correlations, Casimir-type effects, and in particular exact results in two dimensions. However, our results derived here already allow for meaningful tests by simulations and experiments.

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