Dynamics of the Roughening Transition

S. T. Chui
Department of Physics, State University of New York at Albany, Albany, New York 12222

and

J. D. Weeks
Bell Laboratories, Murray Hill, New Jersey 07974

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We use the renormalization-group method of Kosterlitz to study the dynamics of a purely relaxational model for the roughening transition. Implications of our results for the behavior of spatial and temporal correlation functions of a crystal-vapor interface and for the crystal growth rate are discussed.

The "roughening transition" from a localized to a delocalized interface in three-dimensional (3D) Ising and related lattice models has recently aroused considerable interest, both because of its practical importance in understanding the process of crystal growth and because of a more fundamental interest in the nature of the transition.[1,2] In this Letter we apply the renormalization-group method of Kosterlitz[3] and José *et al.*[4] to study the dynamics of the roughening transition. That such an approach would be useful was suggested by earlier work of the present authors[5] which first pointed out the equivalence for the static behavior of the roughening transition and the phase transition in the 2D Coulomb gas, to which Kosterlitz originally applied his method.

Our results confirm the expectations of crystal growth theorists[1] for the existence of linear (nonactivated) growth above the roughening temperature T_R with sublinear (nucleated) growth below, and relate this change in growth mechanism directly to the change in the equilibrium structure of the interface at T_R. We predict a qualitative change at T_R in the character of spatial and temporal correlations between different parts of the interface. Although we concentrate herein on the crystal growth and interfacial interpretations of our results, we believe the same procedure can be applied to describe the dynamics of a model for 2D nematic crystals studied by the Gennes,[6] and to the purely relaxational limit (no coupling to a conserved z component of the spin) of the dynamics of the planar XY model studied by Nelson and Fisher.[7] Another possible application, which we discuss later, is to the motion of domain boundaries (or solitons) in 1D crystals when interchain couplings as well as the commensurate-incommensurate energy are present.[8]

We model the interface between a crystal and its vapor using a 2D square array of columns of height h_j. The Hamiltonian which we study is

$$H=\frac{J}{2}\sum_{j,\delta}(h_j-h_{j+\delta})^2+Jg^2\sum_j h_j^2-\sum_j \Delta\mu_j h_j-2y_0J\sum_j \cos(2\pi h_j). \tag{1}$$

The first term gives the interaction energy between a column at site j and its nearest neighbors at sites $j+\delta$, the second gives the interaction with a dimensionless "stabilizing field" g^2, and the third gives the effect of "applied fields" $\Delta\mu_j$. The fourth term, parametrized by the dimensionless quantity y_0, is a *weighting function*[5] which favors integer values of the h_j. The form of the weighting function in Eq. (1) is chosen for convenience; as pointed out in Ref. 5, any periodic weighting function should give the same critical behavior. [Equation (1) is also essentially identical with the small-y_0 limit of the generalized Villain model discussed by José *et al.*[4] in their analysis of the planar XY model. Note, however, that temperatures are inversely related in the roughening and XY descriptions.]

Following the ideas of Halperin, Hohenberg, and Ma[9] and Hohenberg and Halperin[10] we shall assume that the long-wavelength low-frequency behavior of the system at the roughening (critical) point can be described by the simplest possible dynamical model which reduces to the correct static limit and which is consistent with the proper set of conservation laws for the system. Here we have the simplest case ("model A") since $\langle h\rangle$ is not conserved in either crystal-growth or relaxational XY model applications, and thus we study the following dynamical equation[10]

$$\begin{aligned}\partial h_j/\partial t &= -(\Gamma/T)(\delta H/\delta h_j)+\eta_j \\ &= -\Gamma K^{-1}\textstyle\sum_\delta (h_j - h_{j+\delta}) - \Gamma K^{-1}g^2 h_j + \Gamma(\Delta\mu_j/T) - 2\pi K^{-1}\Gamma y_0 \sin 2\pi h_j + \eta_j.\end{aligned} \tag{2}$$

Here $K^{-1} \equiv 2J/T$. We allow the applied fields $\Delta\mu_j$ to be time dependent and assume that the system starts from equilibrium at $t=-\infty$. The Gaussian fluctuating noises $\eta_j(t)$ satisfy

$$\langle \eta_j(t)\rangle = 0, \quad \langle \eta_j(t)\eta_{j'}(t')\rangle = 2\Gamma\delta_{jj'}\delta(t-t'). \tag{3}$$

In crystal growth applications the parameter Γ can be identified with the equilibrium (kink-site) evaporation rate and $\Delta\mu$ with the chemical-potential driving force.[11]

If $y_0=0$, Eq. (2) can be solved exactly by Fourier-transform techniques in terms of an unperturbed Green's function, which in the long-wavelength limit has the form

$$G(q,\omega) = [K^{-1}(q^2+g^2) - i\omega/\Gamma]^{-1}. \tag{4}$$

In the limit $g^2 \to 0^+$, which we consider hereafter, G is the Green's function for 2D diffusion.[6]

For nonzero y_0, Eq. (2) can be rewritten

$$h(s,t) = \int_{-\infty}^{\infty} ds' \int_{-\infty}^{\infty} dt'\, G(s-s', t-t')[\Delta\mu(s't')/T + \eta(s't')/\Gamma - 2\pi K^{-1} y_0 \sin 2\pi h(s't')]. \tag{5}$$

Here s is a dimensionless 2D lattice vector (the unit of length being the lattice spacing) locating the center of a column. We have taken the limit of an infinite system and replaced sums by integrals. Expanding the solution of Eq. (5) in powers of $\Delta\mu/T$

$$h(s,t) = h_0(s,t) + \int ds' \int dt'\, h_1(st, s't')\Delta\mu(s't')/T + O((\Delta\mu/T)^2) \tag{6}$$

the linear response function $\chi(q,\omega)$ is given by the ensemble average over the noise

$$\chi(q,\omega) = \langle h_1(q,\omega)\rangle \tag{7}$$

and using Eqs. (4)–(6), the unperturbed ($y_0=0$) response function explicitly is

$$\chi_0(q,\omega) = G(q,\omega) = [K^{-1}(q^2+g^2) - i\omega/\Gamma]^{-1}. \tag{8}$$

The effect of a nonzero y_0 is conveniently expressed in terms of a self energy $\Sigma(q,\omega)$, defined as

$$\chi^{-1}(q,\omega) = \chi_0^{-1}(q,\omega) + \Sigma(q,\omega). \tag{9}$$

Substituting Eq. (6) into Eq. (5) we find after some manipulation a formally exact expression for Σ given by

$$\Sigma(q,\omega) = \frac{4\pi^2 y_0 K^{-1} F\{\langle \cos[2\pi h_0(st)]\, h_1(st, s't')\rangle\}}{\langle h_1(q,\omega)\rangle}, \tag{10}$$

where $F\{\}$ denotes a Fourier transform in space and time.

The behavior of Σ in the limit of very low temperatures is easy to analyze. The equilibrium fluctuations of h_0 are very small at low temperatures and the weighting function localizes the interface very near $h_0=0$. Linearizing the sine term in Eq. (2) then gives a constant value for Σ of

$$\Sigma(q,\omega) \cong 4\pi^2 y_0 K^{-1}. \tag{11}$$

Thus there is a finite response even in the $q,\omega \to 0$ limits. The analysis we give below shows that this gap vanishes for all $T \geq T_R$ and that the response can then be related to the divergent response function $\chi_0(q,\omega)$.

To see this, consider an expansion of $\chi^{-1}(q,\omega)$ in powers of y_0. We assume y_0 is less than unity. The recursion relations we derive from this expansion drive y_0 to zero for all $T \geq T_R$, thus justifying the expansion in this region. The motivation here is exactly the same as for the expansion of the static correlations used in the work of José *et al.*[4] and Nelson and Kosterlitz[12] for the *XY* model and the 2D Coulomb gas.

Expanding h_0, h_1, and Σ in powers of y_0, we find, using Eqs. (5)–(10), after some straightforward but tedious algebra, that Eq. (9) can be written to lowest order in q and ω as

$$\chi^{-1}(q,\omega)=[K^{-1}+\pi^3K^{-2}y^2\int_1^\infty ds\, s^{3-2\pi K}]q^2-i\omega[\Gamma^{-1}+\Gamma^{-1}\pi^4y^2/(\pi K-1)\int_1^\infty ds\, s^{3-2\pi K}]+O(y^4), \tag{12}$$

where $y\equiv y_0\exp(-Kc)$ and c is a constant approximately equal to $\frac{1}{2}\pi^2$.

Following the methods of Refs. 4 and 12, we find on integrating over length scales between $s=1$ and $s=e^l$ that Eq. (12) can be rewritten in exactly the same functional form with K, y, and Γ replaced by $K(l)$, $y(l)$, and $\Gamma(l)$. These quantities obey differential recursion relations

$$dK(l)/dl=-\pi^3y^2(l), \tag{13}$$

$$\tfrac{1}{2}dy^2(l)/dl=-[\pi K(l)-2]y^2(l), \tag{14}$$

$$d\ln\Gamma(l)/dl=-\pi^4y^2(l)/[\pi K(l)-1], \tag{15}$$

subject to the boundary conditions $K(l=0)=K$, etc. The roughening temperature is identified as the lowest temperature for which the recursion relations drive $y(l)$ to zero as $l\rightarrow\infty$; $y(l=\infty)=0$ holds for all $T\geq T_R$. In agreement with the assumptions of Ref. 7, the only effect of a nonzero initial value of y_0 for $T\geq T_R$ is to renormalize the coefficients K and Γ in the unperturbed response function χ_0 in Eq. (8).

The first two equations [(13) and (14)] are essentially identical[13] with those found earlier in the static calculations of Nelson and Kosterlitz[12] and José *et al.*[4] It is interesting that we obtain these equations directly without a duality transformation to a 2D Coulomb gas representation as was done in the previous work. The third equation describes the behavior of the dynamical parameter Γ. Eliminating $y^2(l)$ between Eqs. (13) and (15) and integrating from $l=0$ to $l=\infty$, we have

$$\Gamma(\infty)/\Gamma=[\pi K(\infty)-1]/(\pi K-1). \tag{16}$$

Here $\Gamma(\infty)$ and $K(\infty)$ are the renormalized values of the bare parameters Γ and K. Thus Γ effectively scales with K, whose behavior has already been discussed by Kosterlitz[3] and José *et al.*[4] Eq. (16) shows that the renormalized Γ is reduced from its bare value, but does not vanish along the entire fixed line of critical points which characterizes the roughened phase including the endpoint at T_R. Using the language of Hohenberg and Halperin,[10] the dynamics is thus *conventional.* However, the mutual scaling of K and Γ represents an interesting and somewhat unconventional feature of the model. Since $K(\infty)$ has a square-root cusp as $T\rightarrow T_R$,[3,12] it should be possible to observe a similar anomaly in $\Gamma(\infty)$.

The average growth rate R of a crystal is related to the response to a spatially and temporally uniform driving force when the stabilizing field $g^2=0$. To first order of $\Delta\mu$ it is given by

$$R=\lim_{\omega\rightarrow 0}[-i\omega\chi(q=0,\omega)\Delta\mu/T], \tag{17}$$

$$=\Gamma(\infty)\Delta\mu/T, \quad (T\geq T_R). \tag{18}$$

Thus the theory predicts linear growth at and a above T_R in agreement with conventional theories of crystal growth.[1,11] Below T_R the situation is very different. Here

$$\chi(q,\omega)=[K'(q^2+\xi^{-2})-\omega/\Gamma']^{-1} \tag{19}$$

with a finite correlation length ξ and renormalized coefficients K' and Γ'. Equation (17) then predicts a zero growth rate for $T<T_R$ to first order in $\Delta\mu/T$. This result is consistent with the fact that growth at low temperatures occurs by a nucleation mechanism. Nucleation theory gives the result $R\sim\exp(-c/\Delta\mu)$; so in fact below T_R all terms in a power series about $\Delta\mu=0$ should vanish.[1,11]

Predictions for the equilibrium height-height correlation function can be obtained from the fluctuation-dissipation theorem[10]

$$\langle|h_0(q,\omega)|^2\rangle=(2/\omega)\operatorname{Im}[\chi(q,\omega)], \tag{20}$$

where Im[] denotes the imaginary part. In particular, for $T\geq T_R$ and large s or large t,

$$\langle[h_0(s,t)-h_0(0,0)]^2\rangle$$

$$\cong[K(\infty)/2\pi]\ln[\max\{s^2,4\Gamma(\infty)t/K(\infty)\}], \tag{21}$$

where we have used some results of de Gennes.[6] Thus there are logarithmically diverging correlations in space and time above T_R. Below T_R, Eq. (19) holds and correlations decay exponentially at large s and t. All these results as well as Kosterlitz's prediction for the very rapid divergence of the correlation length as $T\rightarrow T_R$ from below:

$$\xi\sim\exp[c/(T_R-T)^{1/2}], \tag{22}$$

can be checked using presently available computer simulation methods.[14] Light scattering experiments on the interfacial structure of real crystal-vapor systems appear to us very feasible, and would provide a definitive test of the applicability of these ideas to realistic systems.[15]

Another possible application of these results is to the motion of domain boundaries in 1D crystals.[8] Assume there is one domain boundary per chain, whose position in chain j is given by h_j. The term $h_j - h_{j+\delta}$ in Eq. (2) describes the interchain coupling, while the commensurate-incommensurate interaction is described by the term $\sin 2\pi h$. The linear response function then essentially describes the conductivity. Our results thus predict that below T_R the domain boundaries are pinned, yielding an activation-type conductivity, but they become unpinned above T_R, giving metallic conductivity. Of course, a finite amount of energy is required to create a domain boundary and this may limit the temperature dependence of the conductivity. Further, the restriction of one domain boundary per chain is unrealistic; so these results are merely suggestive.

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[13] The only difference arises because the weighting function in Eq. (1) has the quantity J factored out explicitly, while Refs. 4 and 12 did not do this factorization. This has no effect on the critical behavior and Eqs. (13)–(15) are slightly simpler with our factorization.

[14] G. H. Gilmer (private communication) has found fairly good agreement between the predictions of Eq. (22) and the behavior of static correlation functions calculated by the Monte Carlo method. Some of his results are found in Ref. 1. Other Monte Carlo work is found in R. H. Swendsen, Phys. Rev. B 15, 5421 (1977).

[15] See K. A. Jackson and C. E. Miller, J. Crys. Growth 40, 169 (1977), for a recent experiment on the effects of the roughening transition on crystal growth kinetics and morphology.