Roughening transition on an imperfect surface

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We consider a roughening phase transition on a surface whose imperfection can be described as the presence of random surface tension, as well as of random slopes. It is shown that only the latter can alter the character of the critical behavior. In the case of sufficiently strong correlations (defects of the random field), the system remains rough at all temperatures. If the correlation function falls off more rapidly than $1/r^2$, the phase transition has the same character as for uncorrelated random slopes. The latter case is analyzed by the renormalization-group method. It is shown that a universal relation exists between the transition temperature, the surface tension, and the variance of the random slopes. On the other hand, the relation between the transition temperature and the surface tension becomes non-universal. It is shown that the variance of the random slopes has a limiting value above which the surface is rough at all temperatures. A second low-temperature transition into a rough phase is possible in a certain variance range. Quantum effects, which possibly play an important role at low temperatures, are not taken into account.

Experimental research into the roughening phase transition is still in its initial stage. This transition was directly observed only in a few cases: for surface solid-helium crystals[1–4] and for certain metals.[5–7] In a number of organic crystals there were observed evidences of this transition, such as vanishing of the faceting[8] and a change in the morphology of growing crystals.[9] From the viewpoint of further study of the roughening transition, considerable interest attaches to the character of this transition on an imperfect crystal surface. Note that, despite the large number of theoretical papers dealing with the roughening transition (see, e.g., Refs. 4 and 10 and the literature cited therein), the influence of defects on this transition has, to our knowledge, hardly been discussed. There are, to be sure, a number of papers dealing with related systems, viz., two-dimensional isotropic magnets with various defects (see Ref. 11 and the literature therein) and solid films with impurities.[12] It is natural to use the results of thes studies to examine the influence of defects on the roughening transition. We shall show below, however, that the last question must be analyzed separately. The present paper is devoted to an investigation of this kind.

HAMILTONIAN OF A SYSTEM WITH DEFECTS

We shall study the roughening transition using hereafter the so-called discrete Gaussian model (see, e.g., Ref. 10), but it is more convenient at first to start from a continuous Gaussian model corresponding to a rough surface. The Hamiltonian is then of the usual form

$$H=\frac{1}{2}\int \alpha(\nabla f)^2\, d^2r. \tag{1}$$

Here α is the surface-tension coefficient, and f the deviation of the surface from a certain reference plane. If the surface is imperfect, it is necessary to separate the "frozen" part of the displacement from those displacements which are due to thermal fluctuations. As a result we have

$$H=\frac{1}{2}\int (\alpha+\delta\alpha(\mathbf{r}))\,(\nabla\psi+\mathbf{p}(\mathbf{r}))^2\, d^2r, \tag{2}$$

where $\delta\alpha(\mathbf{r})$ and $\mathbf{p}(\mathbf{r})$ are random functions. The function $\mathbf{p}(\mathbf{r})$ can be called a random slope. The statistical properties of the random surface tension $\delta\alpha(\mathbf{r})$ and of the random slope $\mathbf{p}(\mathbf{r})$ depend on the type and placement of the crystal defects. We discuss below several possible cases. Since $\alpha(\mathbf{r})$ is perfectly analogous to the "random temperature," the results obtained for defects of the "random temperature" type in two-dimensional spin systems[11] remain in force. It is shown in Ref. 11 that for a correlation function

$$\langle\delta\alpha(\mathbf{r})\delta\alpha(\mathbf{r}')\rangle\sim\delta(\mathbf{r}-\mathbf{r}'),$$

defects of this type are immaterial. In other words, all the universal relations of the theory of a phase transition in an ideal crystal remain in force also for crystals with defects of this kind. Using the so-called Harris criterion,[13] we can show that for a roughening transition, when the correlation length diverges more rapidly than any power of the reduced temperature, this conclusion remains valid also for any power-law decrease of the correlation function $\langle\delta\alpha(\mathbf{r})\delta\alpha(\mathbf{r}')\rangle$. Note that an analysis carried out for random surface tension is valid for all terms of even powers in ψ with random coefficients.

We turn now to the study of the defects that produce the random slope. It will be shown below that they can in principle also eliminate the phase transition. Indeed, the presence of a face corresponds to allowance, in the Hamiltonian, for the fact that the surface displacements (in units of the lattice constant) are whole numbers. Random slopes violate the absolute preference of whole-number displacements and at $\langle\mathbf{p}^2\rangle\sim 1$ one can expect the surface to be rough. The exact conditions for the vanishing of the roughening transition, as will be shown below, are determined not only by the quantity $\langle\mathbf{p}^2\rangle$ but also by the form of the correlation function of the random slopes. What is actually important is only the longitudinal part of the correlation function. The characteristic form of this function for small $\mathbf{k}$ is

$$\langle p_i(\mathbf{k})\,p_l(-\mathbf{k})\rangle=\delta_{il}\begin{cases}\sigma+wk^{a-2}, & a\neq 2,\\ \sigma-w\ln k, & a=2,\end{cases} \tag{3}$$

where a is the exponent in the coordinate dependence of the

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correlation function, and σ and w are positive. We emphasize that when random slopes are considered, we are dealing with a large class of defects. Thus, defects of the random-field type might be taken into account by adding to Eq. (2) the term $-h(\mathbf{r})\psi(\mathbf{r})$, where $h(\mathbf{r})$ is the "random field." However, integration by parts reduces to the same form the term $\mathbf{p}\psi$ contained in (2), where $h = \operatorname{div}\mathbf{p}$. By considering correlation functions of the general form (3) we automatically take into account also such a possibility. Thus, corresponding to a random field is a slope correlation function of the form $1/k^2$.

We introduce now a discreteness in our model. Replacing in (2) the integral by a sum over the lattice sites, and the gradient by a vector discrete difference, we obtain

$$H=\frac{1}{2}\sum_{\mathbf{r}}\alpha(\Delta n(\mathbf{r})+\mathbf{p}(\mathbf{r}))^2. \tag{4}$$

Here n are integers specified at the lattice sites. The partition function for the Hamiltonian (4) can be expressed, using as for an ideal system[10] the Poisson summation formula, in the form

$$Z=\sum_{n(\mathbf{r})=-\infty}^{\infty}\exp\left(-\frac{\tilde{\alpha}}{2}\sum_{\mathbf{r}}(\Delta n(\mathbf{r}))^2-\tilde{\alpha}\sum_{\mathbf{r}}\mathbf{p}\Delta n\right)$$
$$=\int_{-\infty}^{\infty}\prod_{\mathbf{r}}d\varphi\sum_{m(\mathbf{r})=-\infty}^{\infty}\exp\left\{\sum_{\mathbf{r}}\left(-\frac{\tilde{\alpha}}{2}(\Delta\varphi)^2-\tilde{\alpha}\mathbf{p}\Delta\varphi+2\pi i m\varphi\right)\right\}, \tag{5}$$

where $\tilde{\alpha}=\alpha/T$. This transformation is convenient because the discrete variable n is now replaced by the continuous variable φ.

Changing to a Fourier representation in the long-wave approximation, the sum over r in the argument of the exponential is transformed into

$$\sum_{\mathbf{k}}\left[-\frac{\tilde{\alpha}}{2}k^2|\varphi_{\mathbf{k}}|^2+i(2\pi m_{-\mathbf{k}}-\tilde{\alpha}\mathbf{k}\mathbf{p}_{-\mathbf{k}})\varphi_{\mathbf{k}}\right]. \tag{6}$$

Here $\mathbf{k}$ is the wave vector and $\varphi_{\mathbf{k}}$, $P_{\mathbf{k}}$, and $m_{\mathbf{k}}$ are the Fourier components of the corresponding quantities. We can now integrate with respect to $\varphi_{\mathbf{k}}$ and $\varphi_{\mathbf{k}}^*$. Accurate to an insignificant pre-exponential factor, this integration is equivalent to finding the minimum of the expression under the summation sign in (6) and substitution of this minimum in the exponential. This operation changes the argument of the exponential into an expression whose integral form is

$$\tilde{H}=\int\frac{d^2k}{(2\pi)^2}\left(2\pi^2J\frac{m_{\mathbf{k}}m_{-\mathbf{k}}}{k^2}+\frac{2\pi}{k^2}(\mathbf{k}\mathbf{p}_{\mathbf{k}})m_{-\mathbf{k}}\right), \tag{7}$$

where $J=1/\tilde{\alpha}=T/\alpha$. The first term reduces in the usual fashion to the Hamiltonian of a two-dimensional Coulomb gas[10] (this gives rise to an additional quantity y connected with the self-energy of the charge), while the second has the form of the Hamiltonian of the interaction of Coulomb charges with random dipoles, having imaginary dipole moments. An effective Hamiltonian similar to (7) appears in problems dealing with the influence of random Dzyaloshinskiĭ-Moriya interactions on the critical properties of a two-dimensional isotropic magnet[11] and with the influence of random impurities on the melting of solid films.[12] There is, however, a substantial difference: in the last two cases the second term of (7) is preceded by an additional factor iJ. It will be shown that this influences the results substantially.

ANALYSIS OF RECURRENCE RELATIONS

Using the results of Ref. 11, we can obtain for the case $w=0$ the renormalization-group equation

$$\dot{J}=-4\pi^3J^2y^2, \tag{8a}$$
$$\dot{y}=(2-\pi J-\pi\sigma)y, \tag{8b}$$
$$\dot{\sigma}=-8\pi^3J\sigma y^2. \tag{8c}$$

At $\sigma=0$ Eqs. (8) reduce to the known Kosterlitz-Thouless equations (see, e.g., Ref. 10).

Note that σ is renormalizable, unlike in Refs. 11 and 12. Equations (8) must be solved with initial conditions $\sigma=\sigma_0$, $y_0=\exp(-\pi^2J_0/2)$ (Ref. 10). If the initial point, moving in accordance with (8), lands on the $y=0$ plane, the surface is rough, but if y begins to increase without limit, such a regime corresponds to a smooth surface.[10] It can be seen from (8) that if the point is initially in the plane (y, J) or (y,σ), its trajectory does not leave the respective plane. At $J=0$ ($T=0$) the system (8) reduces to

$$\dot{y}=(2-\pi\sigma)y, \quad \dot{\sigma}=0. \tag{9}$$

Obviously, its trajectories are straight lines parallel to the y axis. Thus, the system undergoes at absolute zero a transition from a smooth state to a rough one at the universal value $\sigma=2/\pi$. It is possible, of course, that allowance for quantum effects will change this result.

It follows from (8a) and (8b) that the following relation holds for any trajectory:

$$\sigma=\text{const}\,J^2. \tag{10}$$

The constant in (10) depends on the initial conditions. Thus, the system trajectories lie on the cylindrical surface (10), and their projection on the (J,σ) plane is a parabola. It can be seen from (8b) that y decreases in the region $\sigma+J>2/\pi$ and increases in the region $\sigma+J<2/\pi$. If the initial point, starting from the region $\sigma+J>2/\pi$, lands on the line $\sigma+J=2\pi$ in the $y=0$ plane, the system is at a phase-transition point. The observable values α_∞ and J_∞ are obtained from the relations

$$\sigma_\infty=(\sigma_0/J_0^2)J_\infty^2, \quad \sigma_\infty+J_\infty=2/\pi, \tag{11}$$

where σ_0 and J_0 are the corresponding initial values. We have hence for J_∞ and σ_∞

$$J_\infty=(J_0^2/2\sigma_0)[(1+8\sigma_0/\pi J_0^2)^{1/2}-1],$$
$$\sigma_\infty=(J_0^2/4\sigma_0)[(1+8\sigma_0/\pi J_0^2)^{1/2}-1]^2. \tag{12}$$

The universal values $J_\infty=2/\pi$ and $\sigma_\infty=2/\pi$ are reached respectively only in a pure system and at absolute zero. However, the sum of the observable values σ_∞ and J_∞ at the phase-transition point remains universal. We note that it is precisely this sum which enters in the factor preceding the logarithm in the correlation function for the surface displacement

$$\langle(\psi(\mathbf{r})-\psi(0))^2\rangle=(1/\pi)(J_\infty+\sigma_\infty)\ln r.$$

Recall that in a defect-free system this factor is equal to T/α_∞. In our case it is necessary to relate J_∞ to the surface

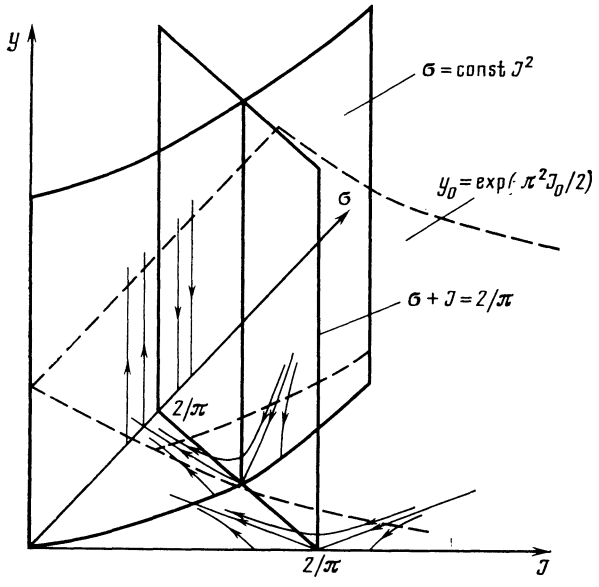

FIG. 1. Phase trajectories of the system (9). The dashed lines mark the positions of initial points, and the arrows the direction of motion.

tension and not the sum $J_\infty + \sigma_\infty$. Indeed, there are two sources of surface fluctuations, thermal motion and the presence of defects. The latter lead to fluctuations even at $T = 0$ and are manifested in elastic scattering of light by a surface. They can be easily singled out when considering a spatiotemporal correlation function that takes at $T \geqslant T_R$ the form $\delta(\omega)\sigma_\infty/k^2 + 2\Gamma/(\Gamma^2 k^4/J_\infty^2 + \omega^2)$, where Γ is a measurable transport coefficient (see, e.g., Ref. 10). The surface-tension coefficient, as the rigidity of the system to changes of its surface area, is related just to the second term.

Linearization, with the aid of Eq. (10), of the system (8) near the point $\sigma_\infty, J_\infty, y = 0$ reduces the system of three equations into a system of two of the Kosterlitz-Thouless form. The only difference is that the slope of the corresponding separatrices does not remain constant but increases continuously from a certain constant value in the $\sigma = 0$ plane to infinity in the $J = 0$ plane. Both equation systems, however, correspond to the same class of critical behavior. Summarizing all the foregoing, we can visualize the trajectories in the following manner (Fig. 1).

In the (y, J) plane we have the usual Kosterlitz-Thouless diagram. On moving along the line $\sigma + J = 2/\pi$ this diagram is deformed: the trajectories lie on the cylindrical surface (10), and the slope of the separatrices increases continuously. In the (y,σ) plane they merge into the straight line $\sigma = 2/\pi$, and we arrive at the system (9). If the initial point is on a separatrix, the system is at a phase-transition point. Since the slopes of the separatrices increases on moving along the line $\sigma + J = 2/\pi$, it is clear that the $\sigma_0(J_0)$ curve that constitutes the geometric locus of the transition points comes ever closer to the line $\sigma + J = 2/\pi$ and they have a common point at $J = 0$, $\sigma = 2/\pi$. It follows from the analysis of the system (8) that $d\sigma_0/dJ_0 \to +\infty$ as $J_0 \to 0$. The $\sigma_0(J_0)$ curve lands on the J axis at $J_0 = J_R$ (at the transition point of the pure system). It can be easily shown that $d\sigma_0/dJ_0 < 0$ as $\sigma_0 \to 0$, $J_0 \to J_R$. The initial growth of $\sigma_0(J_0)$ is stopped by the weakening of the role of the initial conditions: $y_0 = \exp(-\pi^2 J_0/2)$, and subsequently the slope of the curve is close to the slope of the line $\sigma + J = 2/\pi$.

Consequently, the function $\sigma_0(J_0)$ has a maximum at a

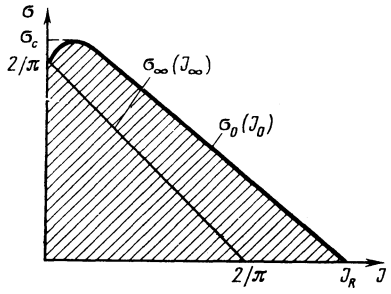

FIG. 2. Phase diagram in the variables σ and J. The shaded region corresponds to the smooth phase.

certain $\sigma_0 = \sigma_c > 2/\pi$. The numerical value of σ_c is universal. The positions of the phase-transition points and of the end points of the trajectories ($\sigma + J = 2/\pi$, $y = 0$) are shown schematically in Fig. 2.

At $\sigma_0 > \sigma_c$ the surface is rough at all temperatures. At $2/\pi < \sigma_0 < \sigma_c$, on moving from the high-temperature region, a transition first takes place into a smooth state, and then at lower temperature back to a rough state.

We examine now how the results change if $w \neq 0$. It is necessary to add to the recurrence relations (8), which now contain also w, an equation for w. Following Ref. 14, we can verify that at small w this relation is

$$\dot{w} = (2-a)w. \tag{13}$$

This shows that at $a > 2$ the fixed point corresponds to $w = 0$, i.e., the critical behavior of the system reduces to the one described above. At $a \leqslant 2$, w becomes substantial and the critical behavior changes. To assess its character we must turn to the perturbation-theory series in y, which was used to obtain the recurrence relations. Recall that this series diverges in the parameter region corresponding to a smooth surface. At $a \leqslant 2$ this series does not diverge. This means that the surface is rough at all temperatures. In particular, it turns out to be rough in the presence of defects of the random-field type.

DISCUSSION OF RESULTS

We comment now on the main results of the paper and discuss the feasibility of verifying them in experiment. Recall that the essential role in the roughening transition is played by defects that produce random slopes. If the correlation function of the random slopes decreases slowly enough with distance (e.g., for defects of the random-field types), there is no roughening transition, and the surface is always rough. A strong correlation of the slopes can take place if the random slopes are produced by dislocation lines that pass through the entire crystal. If the decrease of the correlations is rapid enough, the character of the phase transition is the same as in the case of uncorrelated random slopes. The random slopes become uncorrelated, for example if dislocation dipoles are randomly distributed in the bulk of the crystal. Note that in this case the phase transition takes place also on a highly imperfect surface, all the way to $\langle \mathbf{p}^2 \rangle \sim 1$. Note also that the variance σ_∞ of the random slopes depends on the temperature. Near the phase-transition point it behaves like $1/\tilde{\alpha}_\infty^2$ (Ref. 4) [Eq. (10)], i.e., on the rough-phase side we have the relation

$$\sigma_\infty = \sigma_\infty(T_R)[1 + \text{const}((T - T_R)/T_R)^{1/2}]. \tag{14}$$

This relation can be verified by investigating elastic scattering of light by the surface.

We note also that in our case the universal relation between the surface tension and the transition temperature, which obtains for an ideal surface,[10] no longer holds. Nonetheless, the sum of σ and J, measured at the phase-transition point, is universal. At absolute zero there exists the universal value $\sigma = 2/\pi$ above which, neglecting quantum effects, the surface is always rough. There exists a certain value $\sigma_c > 2/\pi$ above which the surface remains rough at all temperatures. At $2/\pi < \sigma_0 < \sigma_c$, as the temperature is lowered, a transition takes place from a rough to a smooth state, and then from a smooth to a rough one. Similar results for two-dimensional spin systems in solid films were obtained in Refs. 11 and 12.

If the random-slopes correlation function differs from a delta function (but decreases rapidly enough with distance), the form of the correlation function changes substantially near the phase-transition point. Namely, the long-range-correlation amplitude on a smooth surface decreases exponentially rapidly to zero as the phase transition point is approached, and remains equal to zero in the rough phase.

This evolution of the correlation function is naturally reflected in the change of the character of the dependence of the elastic scattering of light on the scattering wave vector.

We note in conclusion that in the case of a boundary between two phases in a solid, or in the case of a domain wall, it is important to take into account the defects that determine the position of the boundary. For a crystal-melt (vapor) interface, however, they can hardly be regarded as frozen. We did not consider such defects.

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