Preroughening Transitions in Surfaces

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We introduce a new type of phase of crystal surface and interfaces. This disordered flat phase appears intermediate between the familiar flat and rough phases in the presence of short-range interactions of a type common in experiments. The surface remains flat on average although it contains a disordered array of steps. The preroughening transition into the disordered flat phase belongs to a new universality class. Finite-size–scaling calculations for the restricted solid-on-solid model confirm the existence of the disordered flat phase and the preroughening transition.

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Crystal surfaces and interfaces undergo several types of phase transitions. One type is the roughening transition, where thermodynamically excited steps transform a flat surface into a terraced mountainlike landscape. Conventionally[1] this is a Kosterlitz-Thouless (KT) transition. The step free energy vanishes with an essential singularity. In the rough phase, the height-height correlation function diverges logarithmically. Its amplitude, $1/\pi K_g$, measures the roughness of the surface. $K_g = \pi/2$ at the KT transition and decreases monotonically with temperature. Numerous numerical and exact results for solid-on-solid (SOS) models[1-3] and experimental evidence[4] have confirmed this.

Here we show that a novel type of phase appears intermediate between the flat and rough phases in the presence of short-range interactions between the steps. We call this the disordered flat (DOF) phase, because the surface remains flat on average although it contains a disordered array of steps. First the step free energy vanishes at the preroughening transition from the flat into the DOF phase. Next the surface undergoes a KT transition from the DOF phase into the rough phase. Before we present our results for the restricted solid-on-solid (RSOS) model, we discuss the mechanism that stabilizes the DOF phase in general.

A flat surface contains thermodynamically excited terraces where the surface is higher or lower (by one unit). The free energy of a terrace is proportional to the length of its edge (the step length). This determines the size of a typical terrace, since its free energy is of order $k_B T$. The terraces increase with temperature because meander entropy reduces the step free energy. This continues until temperatures at which the terraces start to see each other. The topological rules that govern how steps intertwine and the nature of the short-range interactions between steps become the limiting factors in the increase in entropy and determine the universality class of the transition—a KT transition in the conventional case, when hard-core repulsion between steps dominates.

Most studies considered only nearest-neighbor (nn) interactions. The DOF phase appears when we merely extend the range of the interactions, such that the further-than-nn columns prefer to be at the same height, just as nn columns do. Such interactions must be common in experimental systems. It is important to distinguish between up and down steps, i.e., to acknowledge the direction of the change in height. In Fig. 1 we denote this by arrows along the steps: When one looks in the direction along the arrow, the height to the left of the step is (one unit) lower. The nn interactions contribute only to the step energy. They are blind to the arrows. The further-than-nn interactions can look across two or more steps. They imply a short-range repulsion between steps with parallel arrows, but an attraction (or no interaction, like in the RSOS model discussed below) between steps with opposite arrows. In the extreme case when this interaction is infinitely strong, steps with parallel arrows are forbidden to approach each other closer than the interaction range, while steps with antiparallel arrows can approach each other at will. So the meander entropy is larger in configurations where up and down steps alternate, Fig. 1(a), than in configurations where they form a staircase, Fig. 1(b). At temperatures high enough that

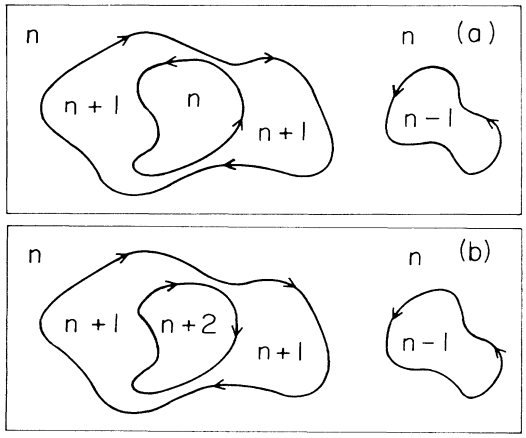

FIG. 1. Surface configuration with (a) nested up-down steps and (b) up-up steps.

the terraces intertwine, the surface prefers a structure where the steps have an up-down-up-down order.

Our results for the RSOS model show that this entropy difference is sufficient to stabilize the DOF phase, with a nonzero up-down-up-down step order parameter. The DOF phase represents a new type of surface reconstruction with randomly rather than periodically arranged steps. Only the arrows have long-range order.

In SOS models, the surface is characterized by integer-valued column-height variables $h(\mathbf{r})$. In the RSOS model, nn columns $\mathbf{r}$ and $\mathbf{r}'$ are allowed to differ by at most one, $h(\mathbf{r})-h(\mathbf{r}')=0,\pm 1$. This restriction retains the effect that we want to study, while it allows high-accuracy numerical calculations. We consider the Hamiltonian

$$H_{\mathrm{RSOS}}=-K\sum_{\langle \mathbf{r},\mathbf{r}'\rangle}\delta(|h(\mathbf{r})-h(\mathbf{r}')|-1)-L\sum_{(\mathbf{r},\mathbf{r}'')}\delta(|h(\mathbf{r})-h(\mathbf{r}'')|-2). \quad (1)$$

$\langle \mathbf{r},\mathbf{r}'\rangle$ denotes nn bonds on a square lattice: $(\mathbf{r},\mathbf{r}'')$ denotes next-nearest-neighbor (nnn) bonds; $\delta(x)=1$ when $x=0$ and vanishes otherwise. Energies are measured in units of $-1/k_BT$. $L>0$ favors equal column heights of second neighbors.

The steps follow the bonds of the dual lattice and form closed loops. To distinguish their up or down direction, we place arrows along these loops, as in Fig. 1. At each intersection of loops the flux of arrows must be equal to zero. K is the energy of a loop element and L favors an alternating arrow order at loop intersections.

To expose the order parameter of the DOF phase, we distinguish between loops and arrows. Associate an Ising spin $s(\mathbf{r})=\pm 1$ to each column. Each Ising Bloch wall forms a closed loop and represents the presence of a step. Next consider the body-centered solid-on-solid (BCSOS) model (the six-vertex model).[2] This is a special SOS model with an arrow (a step) on every bond of its lattice. We rewrite our partition function as

$$Z_{\mathrm{RSOS}}=\sum_{\{s(\mathbf{r})\}}\exp\left[\tfrac{1}{2}K\sum_{\langle \mathbf{r},\mathbf{r}'\rangle}\{s(\mathbf{r})s(\mathbf{r}')+1\}\right]Z_{\mathrm{BCSOS}}(\{s(\mathbf{r})\},L). \quad (2)$$

$s(\mathbf{r})$ represents the parity of the column height at $\mathbf{r}$. The conventional BCSOS model has a rigid lattice. In (2) the annealed Ising-Bloch-wall structure plays the role of BCSOS lattice. The Ising Bloch walls are its bonds and the wall intersections its vertices. This lattice has a two-sublattice structure; the column heights are even on one sublattice and odd on the other. K governs the Ising-type order, i.e., the structure of the annealed BCSOS lattice. L governs the BCSOS-type order. This leads to the phase diagram shown in Fig. 2.

In the limit $K\to-\infty$ the Ising spins are antiferromagnetically (AF) ordered. Each bond contains an Ising Bloch wall. The model reduces to the exact solvable BCSOS model on a square lattice. For $\exp(L)>2$, the surface is flat; the column heights alternate between two values. For $\exp(L)<2$, the surface is rough. The roughening transition is a KT transition.[2]

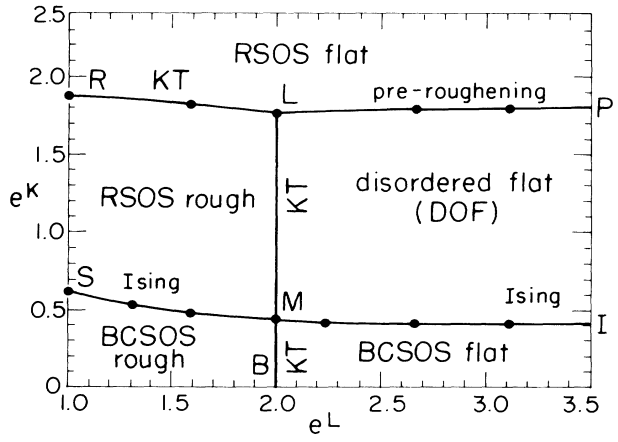

FIG. 2. Phase diagram of the RSOS model. L and K are proportional to $1/(k_BT)$.

For $K\ll 0$, the Ising spins remain AF ordered. The Bloch walls still form a square array, but with missing bonds (closed loops) at length scales smaller than the Ising correlation length. Such imperfections will not change the universality class of the roughening transition.

For $K\approx 0$, the Ising spins are disordered. The Bloch walls form a disordered array. Besides many disconnected finite clusters, one infinitely large Bloch-wall cluster remains, because long-range ferromagnetic Ising-spin order remains absent. This backbone cluster sets the roughness of the surface. The BCSOS model on the backbone undergoes a conventional roughening transition. Its rough phase at small values of L represents the conventional rough phase of the RSOS model. Its flat phase at large values of L represents the DOF phase. In the DOF phase, the surface contains a disordered array of steps, but remains flat on average because the height fluctuations are limited by the BCSOS order in the backbone.

For $K\gg 0$, the Ising spins are ferromagnetically ordered. The BCSOS lattice has fallen apart into many finite lattices. Therefore the arrows are disordered for all finite L. The infinite cluster of ferromagnetically ordered Ising spins, where sites have the same height, has taken over the role of backbone. Hence the surface is flat. This represents the conventional RSOS flat phase.

Conventional-type renormalization arguments[1] can be applied to (1) and predict that the roughening transition lines R-L-M-B belong to the KT universality class.[5]

The $K<0$ part of Fig. 2 describes the coupling be-

tween ordering of Ising-type surface degrees of freedom and surface roughening.[3,5] We expect that everywhere about S-M-I the BCSOS lattice remains a single connected cluster. Notice that a loop of ferromagnetic Ising bonds does not break the BCSOS lattice. At the transition line S-M-I these loops become infinitely large, but do not yet coalesce into objects dense enough to break up the BCSOS lattice. Hence this transition belongs to the Ising universality class. In the rough phase, along S-M, the Ising-type fluctuations induce a singularity in K_g.[3]

In the limit $L \to \infty$, each BCSOS model becomes totally ordered. The arrows in each cluster can be arranged in two ways and therefore (2) reduces to

$$Z_{\mathrm{RSOS}} = \sum_{\{\text{closed loops}\}} e^{-KN_l} 2^{N_c}, \tag{3}$$

with N_l the total step length, and N_c the number of clusters. About point I (see Fig. 2) (3) reduces to the conventional nn Ising model; 2^{N_c} contributes only a factor of 2 since the Bloch walls form one connected cluster.

At $K \approx 0$, the density of ferromagnetic Ising bonds becomes sufficient to break up the Ising-Bloch-wall cluster. One infinite cluster remains, besides many finite ones, as long as the ferromagnetic Ising order parameter vanishes. The preroughening transition L-P, where this backbone cluster disintegrates, does not belong to the Ising universality class. The step interactions establish a preference for up-down-up-down step order inside clusters of intertwined terraces, but disconnected terraces, outside the interaction range, lack an up-down bias. This reveals itself by the factors Z_{BCSOS} in (2) and 2^{N_c} in (3), which favor big clusters to break up into many small ones and significantly influences the critical fluctuations. Equation (3) is similar in appearance to the graph expansions of the O(n) and q-state Potts model,[6] but differs from both. Variation of L [see (2)] resembles variation of n and q in these models. This suggests that L-P might be a line of continuously varying critical exponents.

We studied the finite-size-scaling properties of surface tensions $\eta^{\pm}(a)$ in semi-infinite strips of widths N up to 10, by calculating the largest eigenvalue of the transfer matrix of (1) for periodic step boundary conditions $h(N,t) = h(0,t) - a$ with $a = 1,2$ (which impose a steps in the surface), and antiperiodic step boundary conditions $h(N,t) = -h(0,t) + a$ with $a = 0,1$ [inversion of $h(\mathbf{r})$ with respect to integer or half-integer values]. The $\eta^{\pm}(a)$ are the differences in free energy (per unit length) between these and periodic boundary conditions $h(N,t) = h(0,t)$.

We decompose the η's as follows: $\eta^{+}(1) = \eta_f^F \pm \eta_f^A + \eta_s^{+}(1)$; $\eta^{+}(2) = \eta_f^F(2) + \eta_s^{+}(2)$; $\eta^{-}(0) = \eta_s^{-}(1)$; $\eta^{-}(1) = \eta_f^F \pm \eta_f^A + \eta_s^{-}(0)$, with $+$ $(-)$ for even (odd) values of N. η_f^F is associated with the ferromagnetic Ising-spin order, and η_f^A with the AF Ising-spin order. η_f^F is the free energy of one Ising Bloch wall, and is finite in the RSOS flat phase and zero everywhere else. The argument a in $\eta_f^F(a)$ for $a > 1$ indicates that the boundary condition $(+,a)$ induces a distinct Ising Bloch walls in the surface. η_f^A is the free energy of a string of ferromagnetic Ising spins and is finite in the BCSOS flat and rough phases and zero everywhere else. The $\eta_s^{\pm}(a)$ are associated with the BCSOS sector. In the RSOS and BCSOS rough phases, they scale as[3,7] $S_s^{+}(a) = N\eta_s^{+}(a) = K_g a^2/2$ and[5] $S_s^{-}(a) = N\eta_s^{-}(a) = \pi/4$. In the BCSOS and DOF flat phases, they are finite, except for $\eta_s^{-}(0)$ which vanishes. In the RSOS flat phase, where all BCSOS lattices are finite, all $\eta_s^{\pm}(a)$ vanish.

We found the following symmetry in the RSOS model: $\eta^{+}(1) = \eta^{-}(1)$ for all values of N along the line $\exp(L) = 2$.[5] Since inside the rough phase $S^{-}(1) = \pi/4$ and $S^{+}(1) = K_g/2$, with $K_g < \pi/2$, this implies that the roughening line L-M-B is located at $\exp(L) \leq 2$, and that the preroughening transition must exist. Moreover, our numerical results for $S^{+}(2)$ (equal to π at roughening) and for $S^{+}(1) - S^{-}(1)$ (equal to zero at roughening) strongly indicate that L-M-B coincides with $\exp L = 2$.

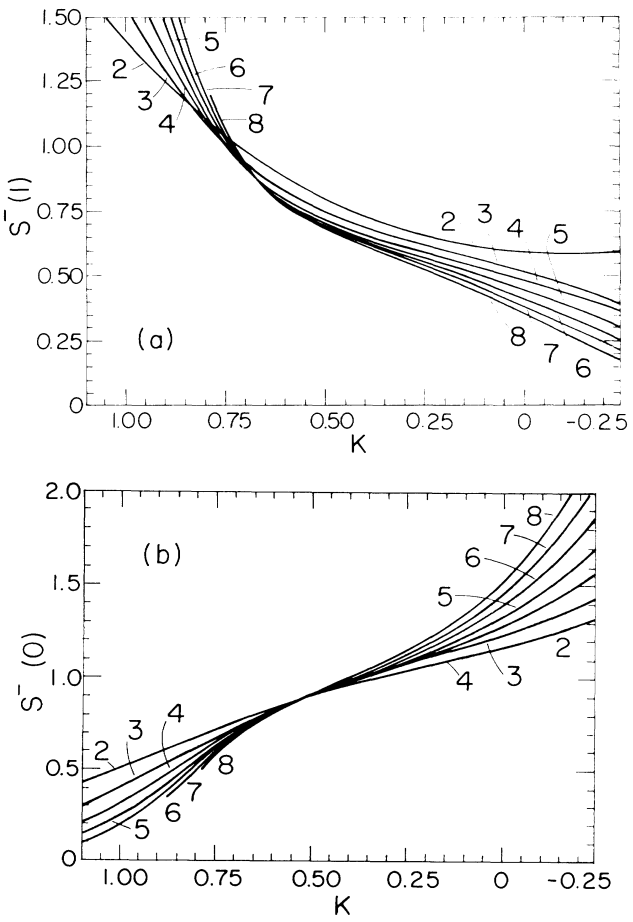

FIG. 3. Scaling behavior of (a) $S^{-}(1)$ and (b) $S^{-}(0)$ across the preroughening transition at $L \to \infty$.

The line R-L is the threshold where $\eta^{\pm}(1)$ and $\eta^{+}(2)$ vanish with universal amplitudes $S^{+}(a)=a^{2}\pi/4$ and $S^{-}(a)=\pi/4$. We used this property to locate this line numerically. Similarly, S-M-I is the threshold where η_{I}^{A} vanishes. At point I, we find numerically a universal amplitude $S_{I}^{A}/2\pi=0.1245\pm0.001$, consistent with an Ising transition.[7]

Figure 3 confirms that the DOF phase has the type of long-range order described above. In the RSOS flat phase, every $\eta_{s}^{\pm}(a)$ must vanish and η_{I}^{F} must be finite. In the DOF phase η_{I}^{F} must vanish, and $\eta_{s}^{\pm}(2)$ must be finite. Figure 3 shows their behavior across the preroughening transition at $L\rightarrow\infty$ (point P in Fig. 2). As predicted, $S^{-}(1)=S_{I}^{f}+S_{s}^{-}(0)$, Fig. 3(a), diverges in the RSOS flat phase and vanishes in the DOF phase, while $S^{-}(0)=S_{s}^{-}(1)$, Fig. 3(b), vanishes in the RSOS flat phase and diverges in the DOF phase.

At the multicritical point L, on approach from the rough side, the universal amplitudes must take the limit values: $S^{+}(a)=a^{2}\pi/4$ and $S^{-}(a)=\pi/4$. Along L-P we find numerically a monotonic variation from these values at L towards the following values at P: $S^{-}(1)=0.799\pm0.001$, $S^{-}(0)=0.770\pm0.002$, $S^{+}(1)=0.923\pm0.003$, and $S^{+}(2)=3.69\pm0.01$. At point P we find numerically a thermal critical exponent $y_{t}=0.303\pm0.001$ $(a=2-2/y_{t}=4.60\pm0.02)$. So the preroughening transition is much weaker than Ising. In a system with an intermediate DOF phase, the behavior of the specific heat will be almost identical to that in the conventional KT theory, with only a second weak singularity near its maximum. It might be useful to reanalyze previous numerical and experimental specific heat data.

The numerical convergence is slow about L. This is not surprising since the DOF phase is stabilized by an intricate entropy effect. We cannot rule out that the η's scale not only at L-P, but also inside a part of the DOF phase close to L. (The infinite Ising backbone cluster might change into a set of fractal-shaped clusters as is the case along L-P.)

The RSOS model is related to the one-dimensional spin-1 quantum chain. We can show[5] that the DOF-type order is related to the so-called Haldane gap,[8] and that the preroughening transition is analogous to one of the transitions[9] in that model.

In Fig. 2, the DOF phase appears when the ratio between the step repulsion L and the step energy K becomes larger than $L/K=1.23\pm0.04$. In the unrestricted SOS model with Gaussian nn and nnn interactions J and M, the DOF phase should appear at $M/J\approx0.75$ (since $K\approx J+2M$ and $L\approx4M$). This estimate ignores the attraction between up-down steps (of order $2M$) which favors the AF step order. We expect that the DOF phase is realized in experimental systems.

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