

Roughening and preroughening transitions in crystal surfaces with double-height steps

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We investigate phase transitions in a solid-on-solid model where double-height steps as well as single-height steps are allowed. Without the double-height steps, repulsive interactions between up-up or down-down step pairs give rise to a disordered flat phase. When the double-height steps are allowed, two single-height steps can merge into a double-height step (step doubling). We find that the step doubling reduces repulsive interaction strength between single-height steps and that the disordered flat phase is suppressed. As a control parameter a step doubling energy is introduced, which is assigned to each step doubling vertex. From transfer matrix type finite-size-scaling studies of interface free energies, we obtain the phase diagram in the parameter space of the step energy, the interaction energy, and the step doubling energy.

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Much attention has been paid to the phase transitions in crystal surfaces since they show rich critical phenomena. The interplay between roughening and reconstruction results in interesting phases, such as a disordered flat (DOF) phase, as well as flat and rough phases [1]. In the DOF phase the surface is filled with macroscopic amount of steps which are disordered positionally but have up-down order. Several solid-on-solid (SOS) type models have been studied, which reveals that the DOF phase is stabilized by the repulsive step-step interactions [2-4] or by specific topological properties of surfaces, e.g., Si(001) [5].

The SOS type model studies have been done in cases where the nearest-neighbor (NN) height difference Δh is restricted to be equal to or less than 1 in units of the lattice constant. However, in real crystals there also appear steps with $|\Delta h| > 1$. For example, double-height steps on W(430) become more favorable than single-height steps at high temperatures since they have lower kink energy [6]. In this paper we investigate the phase transitions in crystal surfaces in the presence of the double-height steps with $|\Delta h| = 2$, especially focusing on the stability of the DOF phase. We study a generalized version of the restricted solid-on-solid (RSOS) model on a square lattice with the Hamiltonian given in Eq. (2). We study the model under the periodic and antiperiodic boundary conditions, from which various interface free energies are defined. The interface free energy is calculated from numerical diagonalizations of the transfer matrix, and the phase diagram is obtained by analyzing their finite-size-scaling (FSS) properties.

In the RSOS model the surface is described by integer-valued heights h_r at each site $\mathbf{r}=(n,m)$ on a square lattice. (The lattice constant in the z direction is set to 1.) Only the single-height step (S step) with $|\Delta h| = 1$ is allowed. It was found that the RSOS model with NN and next-nearest-neighbor (NNN) interactions between height displays the DOF phase when the NNN coupling strength is large enough [2]. The NNN coupling accounts for the repulsive interac-

tions between parallel (up-up or down-down) step pairs. Parallel step pairs cost more energy than antiparallel (up-down or down-up) step pairs.

The double-height step (D step) is incorporated into the RSOS model by relaxing the restriction on the NN height difference to $|\Delta h| = 0, 1, 2$. We only consider quadratic NN and NNN interactions between heights since they are sufficient to describe the key feature of the phase transitions. The total Hamiltonian is written as

$$H_0 = K \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} (h_{\mathbf{r}} - h_{\mathbf{r}'})^2 + L \sum_{(\mathbf{r}, \mathbf{r}'')} (h_{\mathbf{r}} - h_{\mathbf{r}''})^2, \quad (1)$$

where $\langle \rangle$ and $()$ denote the pair of NN and NNN sites. With this Hamiltonian, a D step costs more energy than two separate S steps by an amount of $2K + 4K$ per unit length. Even though the D steps are energetically unfavorable, we will show that their effect is not negligible. We also consider a step-doubling energy E_D to study the effect of the step doubling. It is assigned to each vertex where two S steps merge into a D step (see Fig. 1). The electronic state at step edges may be different from that at a flat surface, which contributes to the step energy. When two S steps merge into a D step, the electronic state near the vertex may be changed. The change leads to an additional energy cost, which is reflected by E_D . When E_D is positive (negative), it suppresses (enhances) the step doubling. The Hamiltonian including H_0 and the step-doubling energy is then given by

$$H = H_0 + E_D N_D, \quad (2)$$

where N_D is the total number of step-doubling vertices. (For notational convenience the energy is measured in unit of $k_B T$.) The model with the Hamiltonian Eq. (2) with $E_D = 0$

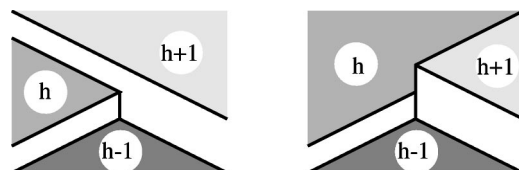


FIG. 1. Step doubling vertices.

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and with the restriction $|\Delta h|=0,1$ will be referred to as the RSOS3 model, and the model with the Hamiltonian Eq. (2) and with $|\Delta h|=0,1,2$ will be referred to as the RSOS5 model.

In a continuum description phase transitions in crystal surfaces are described by the sine-Gordon model

$$H = \int d^2\mathbf{r} \left[\frac{1}{2} K_G (\nabla \phi)^2 - \sum_{q=1}^{\infty} u_q \cos(2q\pi\phi) \right], \quad (3)$$

where $\phi(\mathbf{r}) \in (-\infty, \infty)$ is a real-valued local average height field, K_G the stiffness constant, and u_q the fugacity of q charge [7]. In the renormalization group sense u_1 is irrelevant at high temperatures where the model renormalizes to the Gaussian model with a renormalized stiffness constant $K_G < \pi/2$ describing the rough phase. As temperature decreases, u_1 becomes relevant at a roughening transition temperature. There appear two kinds of low temperature phases depending on the sign of u_1 : For positive u_1 the Hamiltonian favors an integer average height and hence the surface is flat. For a negative u_1 it favors a half-integer average height. Since the microscopic height is integer-valued, the surface can take the half-integer average height by forming steps with up-down order, i.e., the surface is in the DOF phase. As temperature decreases further, the sign of u_1 changes and the surface falls into the flat phase. At the roughening transition between the rough phase and the flat or DOF phase, the renormalized stiffness constant takes the universal value of $\pi/2$. The flat and DOF phases are separated by the preroughening transition characterized by $u_1=0$ [7].

The phase boundaries can be obtained using FSS properties of the interface free energies. Consider the model on a finite $N \times M$ square lattice rotated by 45° under the various boundary conditions (BC's): The periodic BC, $h(n+N, m) = h(n, m) + a$ with integer a , and the antiperiodic BC, $h(n+N, m) = -h(n, m) + a \pmod{2}$ with $a=0$ and 1. They will be referred to as (\pm, a) BC's [the upper (lower) sign for the (anti)periodic BC's]. The free energy is obtained from the largest eigenvalue of the transfer matrix. Detailed description of the transfer matrix setup can be found in Refs. [2,5]. The boundary conditions except for the $(+, 0)$ BC induce a frustration in the surface. The interface free energy η_κ is defined as the excess free energy per unit length under the κ BC with $\kappa = (\pm, a)$ from that under the $(+, 0)$ BC:

$$\eta_\kappa = -\frac{1}{M} \ln \frac{Z_\kappa}{Z_{(+,0)}} \quad (4)$$

with Z_κ the partition function satisfying the κ BC.

The interface free energies have characteristic FSS properties in each phase. In the rough phase they show the universal $1/N$ scaling in the semi-infinite limit $M \rightarrow \infty$ as

$$\eta_{(+,a)} = \frac{\zeta}{2} \frac{K_G a^2}{N} + o\left(\frac{1}{N}\right) \quad (5)$$

$$\eta_{(-,a)} = \frac{\pi\zeta}{4N} + o\left(\frac{1}{N}\right), \quad (6)$$

where $K_G \leq \pi/2$ is the renormalized stiffness constant of the Gaussian model and ζ is the aspect ratio of the lattice con-

stants in the horizontal and vertical directions [2,8]. In the flat phase $\eta_{(+,a)}$ and $\eta_{(-,1)}$ are finite because at least one step is induced under the $(+, a)$ and $(-, 1)$ BC's, while $\eta_{(-,0)}$ is exponentially small in N since the $(-, 0)$ BC may not induce any steps [2]. In the DOF phase the $(-, 1)$ BC does not induce any frustration in the step up-down order, but the $(+, a)$ and $(-, 0)$ BC's do. So $\eta_{(-,1)}$ is exponentially small in N , and $\eta_{(+,a)}$ and $\eta_{(-,0)}$ are finite [2]. From these FSS properties the roughening points can be estimated from

$$\eta_{(+,1)} = \frac{\pi\zeta}{4N}, \quad (7)$$

where the universal value of $K_G = \pi/2$ at the roughening transition used in Eq. (5). The preroughening points between the flat and the DOF phase can be estimated from the crossing behaviors of $N\eta_{(-,0)}$ or $N\eta_{(-,1)}$, which converges to zero in one phase and diverges to infinity in the other phase as N grows.

The estimation of transition points using the interface free energies suffers from slow convergence due to corrections to the scaling. They may smooth out the crossing behaviors of $N\eta_{(-,0)}$ and $N\eta_{(-,1)}$ at the preroughening transitions for small N . But one can safely cancel out leading corrections to scaling by taking the ratio or the difference of them, which can be seen as follows. Consider the lattice version of the continuum model in Eq. (3). It is obvious, using the transformation $\phi \rightarrow \phi - 1/2$, that the model under the $(-, 0)$ BC is the same as that under the $(-, 1)$ BC with u_q replaced by $-u_q$ for odd q . It yields the relation

$$\eta_{(-,0)}(u_1, u_2, u_3, \dots) = \eta_{(-,1)}(-u_1, u_2, -u_3, \dots). \quad (8)$$

So if one neglects all higher order contributions from $u_{q \geq 3}$, the location of $u_1=0$ is found from the condition $\eta_{(-,0)} - \eta_{(-,1)} = 0$ or $R=1$ with

$$R \equiv \frac{\eta_{(-,0)}}{\eta_{(-,1)}}. \quad (9)$$

It is not influenced by correction-to-scalings from u_2 . Therefore the relation $R=1$ can be used to get the $u_1=0$ point more accurately. One can easily see that $R > 1$ for negative u_1 and $R < 1$ for positive u_1 . It approaches 1 in the rough phase and at the preroughening transition points, diverges in the DOF phase, and vanishes in the flat phase as $N \rightarrow \infty$.

In the RSOS3 model the exact point with $u_1=0$ is known along the line $L=0$ [7]. It is called the self-dual point and is located at $K = K_{SD} = \ln[\frac{1}{2}(\sqrt{5}+1)]$. From numerical studies of the RSOS3 model transfer matrix, we could obtain the exact value of K_{SD} with error less than 10^{-12} by solving $R=1$ even with small system size $N=4$, which indicates that R is a useful quantity to determine the preroughening transition points accurately. It will be used in the analysis of the RSOS5 model.

We first consider the RSOS5 model in a special case of $E_D=0$ and compare its phase diagram with that of the RSOS3 model to have insight into the role of the D step. At low temperatures the D step is unfavorable due to larger free energy cost than the S step. So the nature of the low tem-

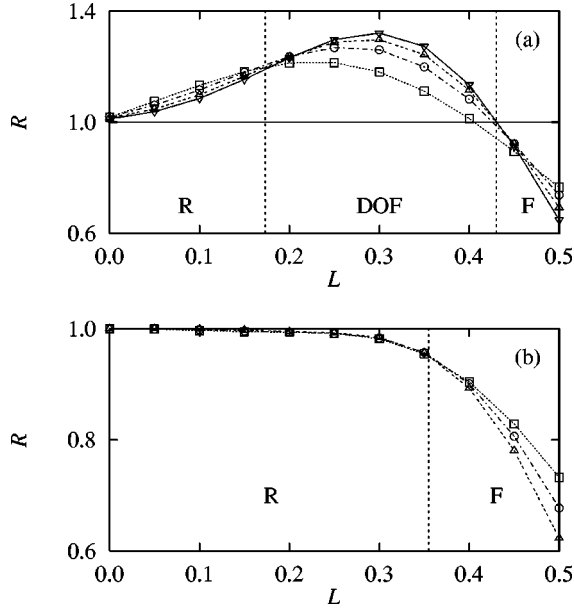


FIG. 2. The ratio R defined in the text for the RSOS3 model (a) and the RSOS5 model (b) along the line $L=5K$. The rough, DOF, and flat phases are denoted by R, DOF, and F, respectively. (\square for $N=4$, \circ for $N=6$, \triangle for $N=8$, and ∇ for $N=10$.)

perature phase in the RSOS5 model is not different from that in the RSOS3 model, i.e., the flat phase. At high temperatures, the surface is in the rough phase in the RSOS3 model. Since the rough phase is critical and there is no characteristic length scale, there will be no difference between S and D steps. So the RSOS5 model will also have the rough phase as a high temperature phase. There is significant difference in intermediate temperature range, where the repulsive step interactions stabilize the DOF phase in the RSOS3 model. Without the D steps the parallel steps have less meandering entropy than antiparallel ones. It is energetically unfavorable for parallel steps to approach each other closer than the interaction range while antiparallel steps can approach each other at will [2]. However, if one allows the D step, two parallel S steps can approach each other and form a D step without the interaction energy cost. Provided that the energy cost of the D step is not too high, the presence of the D step reduces repulsive interaction strength effectively and enhances the meandering entropy of parallel steps. Then it will suppress the DOF phase.

To see such effects quantitatively, we calculate the ratio R for the RSOS3 model and the RSOS5 model with $E_D=0$ along a line $L=5K$ (see Fig. 2). The strip width for the transfer matrix is $N=4, 6, 8$, and 10 for the RSOS3 model and $N=4, 6$, and 8 for the RSOS5 model. The RSOS3 model displays the roughening and the preroughening transitions along the line $L=5K$, which is manifest in Fig. 2(a). There are three regions where N dependence of R is distinct with each other. The surface is in the rough phase with negative u_1 in the small L (high temperature) region, where R approaches 1 from above. And the surface is in the DOF (flat) phase for the intermediate (large) L region, where R grows (vanishes). The roughening and preroughening transition points are estimated from Eq. (7) and $R=1$ with R in Eq. (9), respectively, which are represented by broken vertical lines.

The situation changes qualitatively in the RSOS5 model.

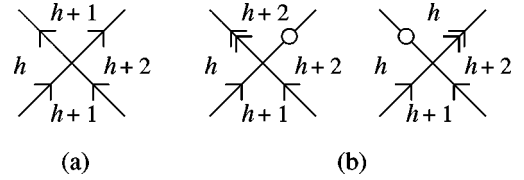


FIG. 3. Comparison of configurations of two parallel steps which cost the interaction energy (a) and from a D step (b). S (D) steps are denoted by single (double) arrows.

As can be seen in Fig. 2(b), R is always less than 1, and there are only two regions with distinct N dependence of R . In the small L region R approach 1 from below, and in the large L region R vanishes as N increases. They correspond to the rough phase with positive u_1 and the flat phase, respectively. The roughening transition point is estimated from Eq. (7) and represented by the broken vertical line. It shows that the DOF phase is suppressed in the presence of the D step. We have also checked that R is always less than 1 ($u_1 > 0$) and the DOF phase does not appear at any values of K and L in the RSOS5 model with $E_D=0$.

We can argue the reason why the DOF phase disappear in the presence of the D step as follows. Consider two parallel S steps merging at a vertex. If the D step is not allowed, the possible vertex configuration is as shown in Fig. 3(a) and the energy cost for such configuration is $2K+4L$. On the other hand, if the D steps is allowed, the step doubling may occur in two ways as shown in Fig. 3(b) with the energy cost $3K+5L$. Though the step doubling costs more energy ($K+L$), entropic contribution of the step doubling ($-\ln 2$) may lower the free energy of parallel steps below than the value without the step doubling. Our numerical results above show that the step doubling suppresses the DOF phase entirely in the $E_D=0$ case. In our model a D step costs more energy than two separate S steps. The two energy scales may be comparable to each other in a more realistic model, where the suppression effect will be stronger.

From the above arguments, one finds that the step doubling plays an important role in phase transitions. So we introduce a new term $E_D N_D$ in Eq. (2) with the step-doubling energy E_D and study the phase diagram in the parameter space (K, L, E_D). When $E_D < 0.0$ (> 0.0), the step doubling is favored (suppressed). One can easily expect that the DOF phase does not appear for negative E_D .

For positive E_D the step doubling is suppressed and the effect of the step interaction becomes important. So we expect there appears the DOF phase in the positive E_D side of the parameter space. In Fig. 4 we show the ratio R for $e^{-E_D}=0.2$ and along the line $L=5K$. Though the convergence is not good, compared with Fig. 2(a), one can identify three regions as the rough, DOF, and flat phases from the N dependence of R . The roughening point between the rough phase and the DOF phase is estimated using Eq. (7), and the preroughening point using $R=1$ for $N=8$. They are denoted by broken vertical lines.

We obtain the phase diagram in the whole parameter space using the conditions $\eta_{(+,1)} = \pi\zeta/4N$ for the roughening transition boundary and $R=1$ for the preroughening transition boundary. It is obtained for strip width $N=4, 6$, and 8 . Since the maximum N we can handle is small, the convergence of the phase boundary is poor especially as one ap-

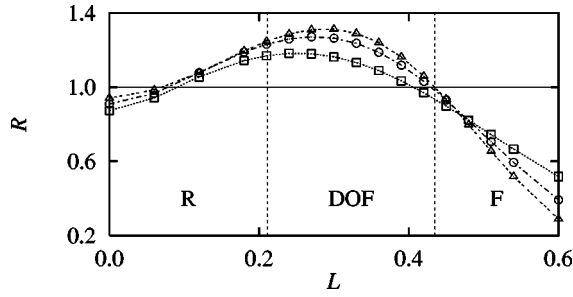


FIG. 4. The ratio R for the RSOS5 model with the step doubling energy $e^{-E_D}=0.2$ along the line $L=5K$. (\square for $N=4$, \circ for $N=6$, and \triangle for $N=8$.)

proaches $e^{-E_D}=0$. But there is no qualitative change in shape. So we only present the phase diagram obtained from $N=8$ in Fig. 5. The region under the surface represented by broken lines corresponds to the rough phase. The DOF phase is bounded by the surfaces of broken lines and solid lines. The region above the surfaces corresponds to the flat phase. One should notice that there is a critical value of E_D , approximately 0.071, smaller than which the DOF phase does not appear.

In summary, we have studied the phase transitions in the RSOS5 model with the Hamiltonian in Eq. (2) with D steps as well as S steps. We have found that the D step, which has not been considered in previous works, plays an important role in phase transitions in crystal surfaces. The presence of

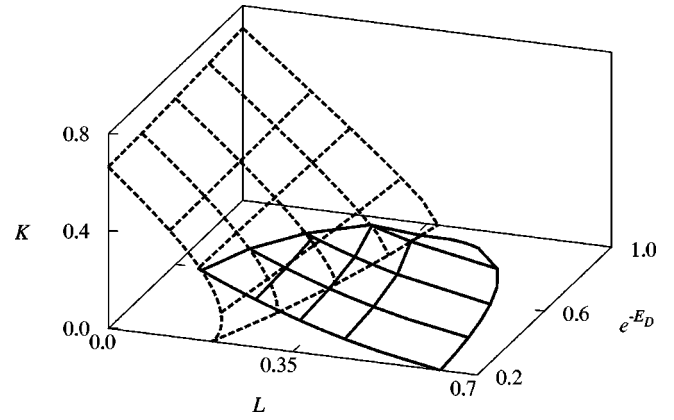


FIG. 5. The phase diagram of the RSOS5 model in the (K, L, e^{-E_D}) parameter space. The roughening transition lines are denoted by broken lines and the preroughening transition lines by solid lines.

the D step reduces the strength of the repulsive interaction between parallel steps through the step doubling, and hence suppresses the DOF phase. We also found that the step-doubling energy is an important quantity which characterizes a surface upon the roughening.

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