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Thermal evolution of step stiffness on the Si(001) surface: Temperature-rescaled Ising-model approach

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We study the equilibrium fluctuation properties of a step on the 2×1-reconstructed Si(001) surface, by using the two-dimensional restricted solid-on-solid model. We propose a new approach to calculate the angle-dependent step tension $\gamma(\theta)$ (θ: mean running direction of the step, relative to the crystal axis) where the numerical renormalization-group technique and the temperature-rescaled Ising-model-interface ansatz for step fluctuations are combined. From the calculated $\gamma(\theta)$ we obtain the step stiffness $\gamma(\theta)+\partial^2\gamma(\theta)/\partial\theta^2$, which is in good agreement with experimental observation.
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Recent developments in atomic-scale observations of crystal surfaces have allowed us to determine various microscopic quantities of the surface, for example, the kink-formation energy $\epsilon^{\rm kink}$. As for the 2×1-reconstructed Si(001) surface, Swartzentruber and co-workers[1,2] measured the interkink distance distribution of a step on the surface, from which they determined $\epsilon^{\rm kink}$. Bartelt, Tromp, and Williams[3] observed step fluctuation width and equilibrium ''crystal shape'' (island shape, to be precise) on the Si(001) surface, to obtain step tension γ and step stiffness $\tilde{\gamma}$ [$\propto$1/(squared scaled fluctuation width) (Ref. 4)]; from the obtained γ and $\tilde{\gamma}$ they also estimated $\epsilon^{\rm kink}$, which is roughly equal to but is slightly different from the one in Refs. 1 and 2.

We should remark here that $\epsilon^{\rm kink}$ is not a directly measurable quantity, and for its determination, we need a theoretical formula for an observable quantity as a function of $\epsilon^{\rm kink}$ (=fitting parameter) to perform ''best fitting'' of the observed data. Hence, reliability of the formula used is crucial for that of the determined value of $\epsilon^{\rm kink}$. Use of the one-dimensional (1D) solid-on-solid (SOS) model, which strictly forbids the (in-plane) overhang configurations, may, therefore, be inadequate; for correct orientation-angle dependence of γ, which is essential in determining the island shape and step stiffness $\tilde{\gamma}=\gamma+\gamma''$ ($'$ denotes the angle derivative), we should properly take account of the non-SOS configurations.[5,6]

Considering that the non-SOS configurations become dominant on raising the temperature and that the experiment[3] is made at relatively high temperatures (as compared with Refs. 1 and 2), we may attribute the non-negligible discrepancy in the value of $\epsilon^{\rm kink}$ between Refs. 1 and 2 and Ref. 3 to the inaccuracy of the 1D-SOS-model formula of $\tilde{\gamma}$ (Ref. 7) used for data analyses.

In this paper we present a method to calculate the orientation-dependent $\gamma(\theta)$ (θ: mean running direction of a step, relative to the crystal axis). We then apply the method to a ''faithful'' model of the Si(001) surface: The *two-dimensional* (square-lattice) SOS model, with both nearest-neighbor (nn) and next-nearest-neighbor (nnn) interactions. Due to the 2×1 reconstruction, the lattice constant of the model corresponds to the two-atom unit of the actual Si(001) surface. The reconstructed structure also allows us to neglect configurations with large nn height differences, because such configurations are energetically rather unstable. Hence, we restrict each nn height difference Δh to take only $0,\pm1$. The SOS models of this type are often called restricted SOS (RSOS) models.

Note that the long-range step-step interactions that are relevant for a surface with finite step density, modifying the coefficient of the (step density)^3 term in the surface free energy, do not affect the single-step fluctuation properties. Hence, for our purpose, we can adopt a RSOS model with only short-range interactions.

By $h_{i,j}$ ($=0,\pm1,\pm2,\ldots$) we denote the height variable of the RSOS model at the lattice point (i,j). The RSOS Hamiltonian H, under the restriction on nn height differences, is given by

$$H=\epsilon_x\sum_{i,j}|h_{i+1,j}-h_{i,j}|+\epsilon_y\sum_{i,j}|h_{i,j+1}-h_{i,j}|+\epsilon_2\sum_{i,j}(|h_{i+1,j+1}-h_{i,j}|+|h_{i-1,j+1}-h_{i,j}|), \quad (1)$$

where ϵ_x (respectively, ϵ_y) is the nn broken-bond energy of the x direction (respectively, the y direction), and ϵ_2 accounts for the corner energy of the step.[1,2]

To obtain $\gamma(\theta)$ of the RSOS model, we take the following steps: (1) For special directions corresponding to the crystal axes ($\theta=0,\pi/2$), we obtain γ by analyzing the vicinal surface using the numerical renormalization-group method.[8–10] (2) For general θ, we make the ''temperature-rescaled Ising-model ansatz'' for anisotropic step tension.[11] The RSOS calculation made in step 1 is used to determine the rescaled temperature.

In the first step, we apply a ''field'' $\boldsymbol{\eta}=(\eta_x,\eta_y)$ conjugate to the surface gradient, by adding the terms

$$-\eta_x\sum_{i,j}(h_{i+1,j}-h_{i,j})-\eta_y\sum_{i,j}(h_{i,j+1}-h_{i,j}) \tag{2}$$

to the RSOS Hamiltonian (1).[12] Then the thermal averages $p_x=\langle h_{i+1,j}-h_{i,j}\rangle$, $p_y=\langle h_{i,j+1}-h_{i,j}\rangle$ [independent of (i,j), assuming uniformity] give the surface gradient vector $\boldsymbol{p}=(p_x,p_y)$. Below T_{R} (roughening temperature) we expect the square-root ''critical behavior''[13] in the $\boldsymbol{p}$-$\boldsymbol{\eta}$ curve. For instance, along the x direction [i.e., $\boldsymbol{\eta}=(\eta_x,0)$],

$$p_x=\begin{cases}=0 & \eta_x\leqslant\eta_x^c\\ \sim\text{const}\times\sqrt{\eta_x-\eta_x^c} & \eta_x>\eta_x^c \text{ and } \eta_x\approx\eta_x^c\end{cases} \tag{3}$$

This square-root behavior is characteristic of the Gruber-Mullins-Pokrovsky-Talapov universality class[14] to which the vicinal surface belongs. This behavior occurs along a general direction of $\boldsymbol{\eta}$ near the corresponding critical field $\boldsymbol{\eta}^c=(\eta_x^c,\eta_y^c)$. An important point is that, for $\boldsymbol{\eta}$ parallel to the crystal axis, the amplitude $|\boldsymbol{\eta}^c|$ itself is the step tension γ for the step running perpendicular to that direction. To obtain an accurate $\boldsymbol{p}$-$\boldsymbol{\eta}$ curve, we employ the numerical renormalization-group method[8–10,15] for the transfer-matrix diagonalization. In the actual calculation we map[16] the RSOS model to a three-state vertex model and applied the product-wave-function renormalization-group (PWFRG) method,[10,15] which is a variant of White's density-matrix renormalization group (DMRG).[8] The PWFRG method is suited for our problem where the vicinal surface—a massless system—is considered.[15] Although the method is originally developed for Ising-type models,[10] we can easily generalize the method to handle the vertex models [the actual implementation parallels the quantum-version of the PWFRG Refs. (15 and 17)]. To test the accuracy of the method we have applied it to the body-centered cubic SOS model (BCSOS model) (Ref. 18) which is exactly solvable; we have found that the PWFRG gives a close-to-exact $\boldsymbol{p}$-$\boldsymbol{\eta}$ curve, even with a small number (~ 12) of ''retained bases'' (in the DMRG/PWFRG terminology).

In the second step, we make an ansatz that the anisotropy of the step tension is the same as that of the interface tension of an Ising model with suitable temperature rescaling. In the present case, we consider the interface of the nnn Ising model (two-level approximation of the RSOS model), to obtain an analytic expression of the Ising-model interface tension $\gamma_I(\theta,\beta)$. We then assume that, with suitable ''rescaled inverse temperature'' $\tilde{\beta}_I$, the relation

$$\tilde{\beta}_I\gamma_I(\theta,\tilde{\beta}_I)=\beta\gamma(\theta,\beta). \tag{4}$$

holds. The remarkable fact is that, in the case of the BCSOS model, this relation holds *exactly* with $\gamma_I(\theta,\beta_I)$ being the interface tension of the square-lattice nn Ising model.[11] Noting that the BCSOS model and the Ising model belong to totally different universality classes as for the critical behavior (i.e., anomalous temperature dependence of physical quantities), we can expect the ''universality of angle dependence'' of $\gamma(\theta,\beta)$, which lies behind Eq. (4) among a large class of models belonging to different universality classes. Some other exactly calculated examples[19] are also in support of this. Further, for a general (well-behaved) surface model, relation (4) actually holds at two different limits: $T\to 0$ where non-Ising excitations in the surface model are energetically suppresed, and $T\to T_R-0$ where both γ_I and γ are essentially isotropic. These facts strongly support the validity of relation (4), as a good approximation at least. In the actual calculation, we use Eq. (4) at a particular value of $\theta=\theta_0$ (e.g., $\theta_0=0$ or $\pi/2$) for which the right-hand side of Eq. (4) has been obtained in the step 1, to determine $\tilde{\beta}_I$. We should note that the validity of Eq. (4) can also be tested at this stage, by checking the θ_0 independence of $\tilde{\beta}_I$ thus determined. Full θ dependence of $\gamma(\theta)$ can then be derived by using the Ising-model result.

Since the 2D nnn Ising model is not exactly solvable, calculation of $\gamma_I(\theta,\beta)$ itself is nontrivial. For this purpose, just as we have done in our recent paper[20] where the Ising-model treatment of Si(001) is made, we adopt the ''imaginary path weight'' (IPW) random-walk method, which is based on the well-known Feynman-Vdovichenko random-walk solution of the two-dimensional Ising models.[21]

Let us give here a brief description of the IPW method (for details, see Refs. 5, 11, 20, and 22). We consider the low-temperature phase of an Ising model (either solvable or nonsolvable) with an interface, and regard each microscopic configuration of the interface as a path of a random walker. We then specify the connectivity matrix $A=[A(\boldsymbol{r},\mu|\boldsymbol{r}',\nu)]$ characterizing the random-walk problem, whose nonzero elements are given in terms of the elementary path-weights $\{\exp(-\beta\Delta E)\}$ (ΔE: bond-breaking energy). In the multiple index $(\boldsymbol{r},\mu)$, $\boldsymbol{r}$ represents the lattice points (or, the unit-cell position, in general), and μ the ''inner degrees of freedom'' (intra-unit-cell position, direction of the elementary walk, etc.). The essential point in the IPW method is to assign the imaginary ''corner weight'' $\exp(i\phi/2)$ to each turn of the walk with angle ϕ; with this simple recipe, contributions from unwanted path configurations are completely (for solvable cases) or nearly completely (for nonsolvable cases) removed. The interface partition function is then given by the lattice Green's function $G=(I-A)^{-1}$. For systems with translational invariance where $A(\boldsymbol{r},\mu|\boldsymbol{r}',\nu)$ can be written as $A(\boldsymbol{r}-\boldsymbol{r}')_{\mu\nu}$, we can introduce the spatially Fourier-transformed connectivity matrix $\hat{A}(\boldsymbol{k})=[\hat{A}(\boldsymbol{k})_{\mu\nu}]$. The long-distance limit of G determining γ, is essentially given by the integral $\int \mathrm{d}^2k\,\exp(i\boldsymbol{k}\boldsymbol{r})/D(\boldsymbol{k})$ where $D(\boldsymbol{k})=D(k_x,k_y)=\det[I-\hat{A}(\boldsymbol{k})]$. We can easily evaluate the integral at the *pure-imaginary saddle point* $\boldsymbol{k}^*=i\boldsymbol{\omega}=(i\omega_x,i\omega_y)$.

The above-described IPW method leads to the following set of ''constitutive equations'' to determine $\gamma(\theta)$:[5,11]

$$D(\mathrm{i}\boldsymbol{\omega})=0, \tag{5}$$

$$\frac{\partial D(\mathrm{i}\boldsymbol{\omega})}{\partial\omega_y}\Big/\frac{\partial D(\mathrm{i}\boldsymbol{\omega})}{\partial\omega_x}=\tan\theta, \tag{6}$$

$$\beta\gamma_I(\theta,\beta)=\omega_x\cos\theta+\omega_y\sin\theta. \tag{7}$$

The equilibrium crystal shape drawn in the x-y plane is just the *imaginary zeros* of D:[5,11,22]

$$D(i\beta\lambda y,i\beta\lambda x)=0, \tag{8}$$

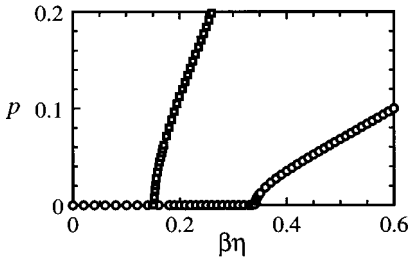

FIG. 1. Surface gradient p vs external field η ($\times\beta$) of the RSOS model calculated by PWFRG, along the x direction (open circles) and the y direction (open squares) at $k_BT/\epsilon_x=0.5$. The number of retained bases in the PWFRG is 12.

where λ is the Lagrange multiplier associated with the volume-fixing constraint in the Wulff construction. It should be noted that the IPW method not only gives us exact $\gamma_I(\theta,\beta)$ for solvable cases (any planar lattice without bond crossings[11,22]) but also serves as a fairly accurate approximation scheme.[5]

The Ising-model approximant of the present RSOS model has the coupling constants J_x, J_y (nn couplings) and J_2 (nnn coupling) which are related to the RSOS broken-bond energies as $\epsilon_x=2J_x$, $\epsilon_y=2J_y$, and $\epsilon_2=2J_2$. All the relevant Ising-model expressions can be found in Ref. 20. In particular, the function $D(\boldsymbol{k})$ has the form

$$D(k_x,k_y)=M-a\cos k_x-b\cos k_y-c_1\cos(k_x+k_y)-c_2\cos(k_x-k_y), \tag{9}$$

where M, a, b, c_1, and c_2 are temperature-dependent quantities that are explicitly given in Ref. 20. The ansatz (4) leads to renormalization of these quantities.

Due to the 2×1 reconstruction, there are two types of steps, the S_A step (running along the x direction) and the S_B step (running along the y direction), whose behaviors are rather different from each other. In fact, there is a large difference in the kink formation energies $\epsilon_A^{\rm kink}$ and $\epsilon_B^{\rm kink}$, with the ratio being $\epsilon_A^{\rm kink}/\epsilon_B^{\rm kink}=90/28$.[1] We adopt this ratio, and use the refined estimates[2] of $\epsilon_B^{\rm kink}$ and the kink-corner creation energy c, deduced from the data at 350 °C: $\epsilon_B^{\rm kink}=40$ meV/(two atoms) and $c=60$ meV. These values determine the RSOS parameters to be $\epsilon_x=188$ meV/(two atoms), $\epsilon_y=100$ meV/(two atoms) and $\epsilon_2=-30$ meV/(two atoms) [note that the two-atom length corresponds to 7.68 Å (Ref. 1)].

In Fig. 1, we show the p-$\beta\eta$ curves calculated by the PWFRG along the x direction and the y direction at temperature $k_BT/\epsilon_x=0.5$. The square-root behavior (3) is clearly seen. Reading off the values of the critical fields η_x^c and η_y^c from these curves drawn at each temperature T, we obtain $\gamma(0)$ (S_B step) and $\gamma(\pi/2)$ (S_A step) as a function of T, which is shown in Fig. 2(a). Then, using Eq. (4) we obtain the rescaled inverse temperature $\tilde{\beta}_I$ as shown in Fig. 2(b). At first sight, in using Eq. (4), $\tilde{\beta}_I$ depends on θ. Hence, to verify the validity of ''temperature-rescaled Ising-model ansatz,'' we should check θ independence of the rescaled temperature.

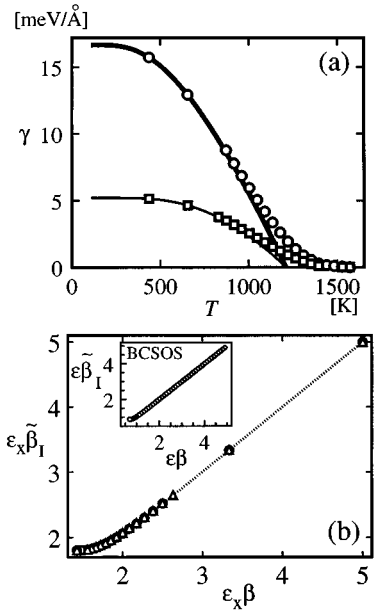

FIG. 2. (a) Temperature dependence of $\gamma(\theta)$ ($\theta=0,\pi/2$) of the RSOS model calculated by the PWFRG: Open circles correspond to $\gamma(0)=\gamma_B$ [S_B step of Si(001)], and open squares to $\gamma(\pi/2)=\gamma_A$ [S_A step of Si(001)]. Solid lines are the Ising-model results without the temperature rescaling (Ref. 20). The ratio γ_A/γ_B calculated at 676 K ($k_BT/\epsilon_x=0.310$) is 0.364, and the one at 873 K ($k_BT/\epsilon_x=0.400$) is 0.398, which agrees with the observed ratios of step free energies (Refs. 23 and 24); for instance, in the recent observation of the equilibrium island shape by Ichimiya, Tanaka, and Hayashi (Ref. 24), the ratio is reported to be $\simeq5/13=0.38$ at 400–600 °C. (b) The rescaled inverse temperature of the Ising model vs the inverse temperature of the RSOS model. Open circles correspond to S_B step ($\theta=0$), and open triangles to S_A step ($\theta=\pi/2$). The inset shows the rescaling curve for BCSOS model (Ref. 11) with the ϵ being the basic excitation energy of the model.

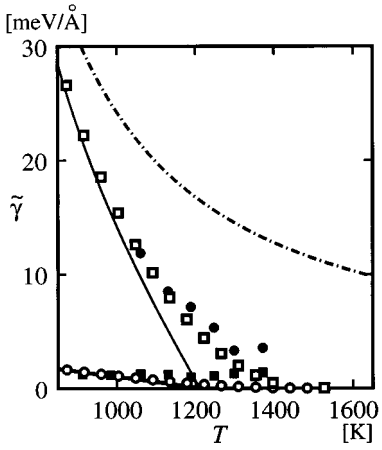

FIG. 3. Temperature dependence of the step stiffness $\tilde{\gamma}$ calculated by the present approach. Open squares and open circles are the step stiffness of S_A step and S_B step, respectively. The filled squares and the filled circles are the experimental data taken from Bartelt, Tromp, and Williams (Ref. 3). The bold and thin solid curves are the ones calculated by the Ising model without the temperature rescaling (Ref. 20). The dot dashed curve is the 1D SOS approximation (Ref. 7) for S_A step. 1D SOS curve for S_B step is omitted for convenience.

We see in Fig. 2(b) that the two rescaling curves, one for $\theta = 0$ and the other for $\theta = \pi/2$, are almost the same, which supports the validity of the ansatz.

The step stiffness $\tilde{\gamma}(\theta)$ of the RSOS model calculated as the Ising-model stiffness at the rescaled temperature, is shown in Fig. 3, where comparison with experiment[3] is also made. One can see a satisfactory agreement between the calculated curve and the experimental one.

To summarize, in this paper we have proposed an efficient method to calculate orientation-dependent step free energy on a surface. In the method, we combine the analytical calculation for the Ising-model interface and the numerical-renormalization-group calculations made for the 2D SOS model, where the latter are utilized to determine the temperature rescaling of the Ising-model approximant of the SOS model. The method is applied to the Si(001) surface, which gives a consistent explanation for experiments; by the use of the kink formation energies determined by Swartzentruber and co-workers,[1,2] the calculated step stiffness is in good agreement with the data of Bartelt, Tromp, and Williams,[3] dissolving the existing discrepancy. Since the present approach based on the temperature-rescaled Ising-model ansatz have a wide range of applicability, it will be helpful in discussing fluctuation properties of steps on various crystal surfaces.

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