# Why Monatomic Steps on Si(001) Are Always Rough

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High temperature scanning tunneling microscopy (STM) measurements are performed to study the behavior of monatomic step edges on Si(001) at elevated temperatures. The freeze out temperature $T_f$, at which the roughness of these step edges is frozen out, is estimated to be well above the intrinsic roughening temperature $T_r$ of the edges. This means that these step edges are always rough at room temperature. The 1D random walk behavior of the step edges, as determined from room temperature STM images, suggests that the step-step interaction is very weak compared to the kink formation energies.

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A considerable amount of research has recently been devoted to monatomic step edges on Si(001) surfaces. This interest is due to the important role of these steps in several surface phenomena, such as preferential reactivity and growth of epitaxial overlayers. One interesting aspect of step edges is that they can roughen above a certain temperature. On an atomic scale the roughening of a step edge is defined as the proliferation of kinks by thermal generation at a roughening temperature $T_r$ at which the free energy for creation of a kink becomes zero.

The general idea of a roughening transition of crystal *surfaces* was first suggested by Burton, Cabrera, and Frank [1] (BCF) in a classical paper. This roughening transition has important consequences for crystal growth theory. Crystal growth on a relatively flat surface well below $T_r$ is a difficult process, requiring the formation of a critical nucleus cluster. If a surface is at its roughening temperature, then according to the BCF theory [1] the critical fluctuations produce clusters of arbitrary size and hence the nucleation barrier to crystal growth disappears. In the BCF theory the crystal surface is mapped on a 2D Ising spin system. Using this concept Burton and coworkers predicted that there would be large fluctuations in the surface structure at the Ising model's critical temperature. However, they made one important simplification in their description: They ignored the existence of additional layers. As a result this transition is not similar to that of the 2D Ising model but rather turns out to be of the Kosterlitz-Thouless type [2,3]. This means it is a phase transition of infinite order; the singularity in the surface free energy per unit area at $T_r$ is extremely weak.

For a vicinal surface [4,5], a surface with a regular array of steps and terraces, the situation is in principle only slightly different. It is a well-known result that an isolated step is not stable and roughens at any nonzero temperature. A repulsive step-step interaction is necessary to explain the relative stability of vicinal surfaces. The roughening of a step edge is expected to occur at a significantly lower temperature as compared to the roughening of the surface itself. On an (001) surface, for example, the creation of an adatom-vacancy pair needs four broken bonds whereas the creation of a pair of kinks along a step edge only needs one broken bond.

Recently we developed an UHV (ultrahigh vacuum) high temperature STM (scanning tunneling microscope) [6,7] for the observation of surfaces at elevated temperatures. The general design of this instrument and its modifications are described elsewhere [6]. All STM images were taken with a sample bias of −1.5 or −2 V and a tunneling current of 0.5 nA. The $5\times20$ mm$^2$ Si(001) substrates were cut from commercially available wafers (Wacker, FZ, B-doped 8-12 $\Omega$cm) and ultrasonically rinsed in ethanol before loading into the vacuum system. The misorientation of the surface with respect to (001), as determined by x-ray diffraction, is about 0.5° in the [110] direction and 0.1° in the $[\bar{1}10]$ direction. Inside the UHV chamber, which has a base pressure below $1\times10^{-10}$ torr, the samples were heated resistively. Temperatures were measured with an infrared pyrometer (calibrated against a NiCr-NiAl thermocouple) with an absolute accuracy of about 25 K. After outgassing sample and holder for several hours the sample was thermally cleaned at 1500 K during several seconds. This procedure results in a $(2\times1)$ reconstructed atomically clean Si(001) surface [6-8] with an average terrace length of about 150 Å.

The Si(001) surface reconstructs by forming surface dimers that are arranged in parallel rows. The dimers can be oriented along two possible directions, depending on the plane where the crystal is cut. Hence the surface has two degenerate phases, which are related by a 90° rotation; their surface periodicity is either $2\times1$ or $1\times2$. A slight misorientation with respect to the [001] direction in the [110] (or $[\bar{1}10]$) direction results in a vicinal surface with two types of monatomic steps. Step edges parallel to the dimer rows of the upper terrace are straight (*A*-type steps), whereas the others are ragged (*B*-type steps), i.e., they exhibit a high density of thermally excited kinks.

Consider a meandering step edge trapped between two step edges separated by a distance $2L$, $L$ being the average spacing between two adjacent step edges. Repulsive interactions between the step edges will tend to limit the amount of meandering by making closely spaced steps unfavorable. Restricting step fluctuations, however, decreases the entropy term and thus increases the free energy. Alerhand and co-workers [9,10] have determined an expression for the energy per unit step length due to step-step interactions on the Si(001) surface

$$E = -C \ln\left[\frac{L}{\pi a}\right]. \tag{1}$$

$C$ is a constant [(58 meV)/$2a$] [11] and $2a = 7.7$ Å, the width of a dimer row. $C$ is determined by the surface-stress anisotropy due to the $2\times1$ reconstruction, the elastic constants of the medium (silicon), and the geometry of the domains (the $2\times1$ reconstruction rotates by 90° at each monatomic step edge). The potential felt by a wandering step is [12], for a small deviation $x$ measured from the middle between two step edges, given by $\varepsilon_s x^2$ [where $\varepsilon_s \sim (C/L^2)$, using Eq. (1)].

Step edges of the Si(001) surface are stable at temperatures up to 525 K on a time scale of at least several minutes [6]. Around 725 K, however, the step edge meandering on Si(001) changes slowly on the same time scale as shown in Fig. 1 of Ref. [7]. A value of about 2 eV can be derived for the kink detachment barrier from these images [7]. This value of 2 eV is in good agreement with data published by Elswijk *et al.* [13]. Since the kink detachment barrier of a monatomic step edge is relatively high (2 eV) and since in our preparation method the samples are radiation quenched from 1500 K to about 500 K in a few seconds, the step structure is frozen in at a temperature higher than 600–700 K.

Another interesting feature of kinks in monatomic step edges on vicinal Si(001) [7] is that they tend to offset the step edge in a direction perpendicular to the edge by $2a$ (7.7 Å, the width of a dimer row) or multiples of $2a$. We therefore represent a monatomic step edge by a 1D array of integers specifying the number of dimer pairs in each column perpendicular to the step edge and parallel to the surface plane, or equivalently by the length of the column ($h_i$) relative to the flat $T=0$ reference step edge. The blocks in the columns interact with their neighbors with an energy $\varepsilon_{L,\parallel}$ (solid-on-solid model). The anisotropic interaction energies $\varepsilon_{L,\parallel}$ determined from the rough $B$-type step edge and straight $A$-type step edge are 0.24 and 0.38 eV, respectively [8].

If we now reintroduce the repulsive energy $\varepsilon_s$ again (per unit length between two step edges) and if we make no difference between an $A$- and $B$-type step edge one obtains, after taking the sum over all step edges, the following Hamiltonian (called $H_{\mathrm{VGL}}$, after Villain, Grempel, and Lapujoulade [4])

$$H_{\mathrm{VGL}} = \sum_{m,r} \frac{\varepsilon}{2}[h_r(m) - h_{r-1}(m)]^2 + \varepsilon_s [h_r(m) - h_r(m-1)]^2, \tag{2}$$

where $r$ is the coordinate along a step, $m$ the step index, and $h_r(m)$ the displacement of the $m$th step at the coordinate $r$. Both $h$ and $r$ are measured in units of $2a$ (=7.7 Å). This Hamiltonian is known to have a roughening transition. The transition is of infinite order and is in the Kosterlitz-Thouless universality class [4]. Important to note is that only kinks with length $\pm1$ are allowed in this model. This condition is almost satisfied for the $A$-type step edges; for the $B$-type step edges, however, this condition is definitely not satisfied.

The roughening temperature of a sequence of $A$- or $B$-type step edges can be obtained from the following equation [4]:

$$\frac{\varepsilon_s}{kT_r} e^{\varepsilon/2kT_r} = \frac{1}{2}\pi^2. \tag{3}$$

Taking $\varepsilon = 0.24$ eV and $\varepsilon_s = 1.5\times10^{-4}$ eV [$L=20$, for 0.5° off Si(001)] gives a roughening temperature of 200–300 K for a $B$-type step edge using Eq. (3). The roughening temperature of an $A$-type step edge ($\varepsilon = 0.38$ eV) is also below 300 K. This temperature is significantly lower than the freeze out temperature of the monatomic step edges on Si(001) which is estimated to be in the range 600–700 K. Changing the miscut angle, and hence $\varepsilon_s$, from, say, 0.1° to 2.5° or $C$ ($\varepsilon_s \sim C/L^2$) by 1 order of magnitude does not alter our conclusion because $T_r$ stays below 400 K for both type of step edges. Hence we conclude that $T_r$ is below $T_f$, so a rough state of the step edges will normally be observed in room temperature STM images of vicinal Si(001) surfaces after annealing.

The roughness of a step edge is most simply characterized by the deviation-deviation correlation function, $G(r) = \langle (h_o - h_r)^2 \rangle$, where $r$ is a distance measured parallel to the step edge and $h_r$ is the deviation of the step edge in the perpendicular direction (in the surface plane). When the step edge is smooth and below the roughening temperature $T_r$, the asymptotic limit of $G(r)$ is a constant [4,5]. Above $T_r$ and if step-step interactions are included, $G(r)$ is predicted [4,5] to diverge logarithmically. At the roughening temperature the prefactor of the logarithmic term has the universal value $2/\pi^2$.

$G(r)$ was determined by counting the distribution of $h_i$ for several $A$- and $B$-type step edges, each with a length of at least 150 dimer rows (see Fig. 1). Plotting of $G(r)$ vs $r$ for low $r$ values (see Fig. 2) results in straight lines. These results are in disagreement with a generally accepted Kosterlitz-Thouless type of roughening transition which predicts a $\ln(r)$ dependence of $G(r)$, although we must be careful with our conclusion because the logarithmic dependence of $G(r)$ on $r$ does not hold for very low $r$ values [3]. On the other hand, recent Monte Carlo simulations of the roughening of Au(110) performed by Mazzeo *et al.* [14] indicate that already for small system sizes

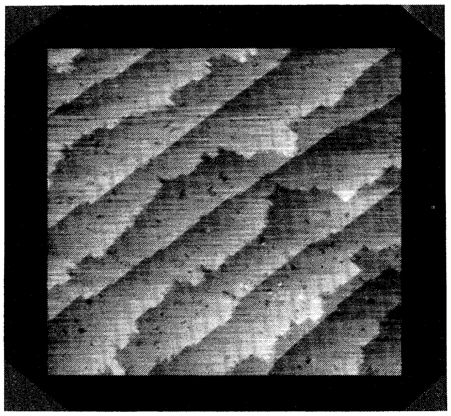

FIG. 1. Room temperature STM image of a vicinal Si(001) surface (0.5° off). Sample voltage is −2 V, the tunneling current is 0.5 nA, and the scan area is about 100×100 nm$^2$.

(12–32 units) and hence values of $r$ within the range of Fig. 2 a $\ln(r)$ dependence of $G(r)$ is observed above $T_r$. So, in conclusion we cannot completely rule out a Kosterlitz-Thouless type of roughening transition of the monatomic step edges on vicinal Si(001). This is interesting because by far the most investigated roughening transitions are of the Kosterlitz-Thouless type and so it is of interest to find examples exhibiting roughening transitions with an apparently different nature. For several examples of non-Kosterlitz-Thouless roughening transitions we refer to the review paper of van Beijeren and Nolden [15].

If step-step interactions are absent or very weak the step edge can be regarded as isolated and the mean square displacement $\langle (h_o - h_r)^2 \rangle$ diverges linearly with $r$. This result follows from the random distribution of positive and negative kinks in a step under equilibrium. The behavior of $G(r)$ for the step edges of Si(001) depicted in Fig. 2 is a strong indication that $\varepsilon_s \ll \varepsilon_{L,\parallel}$ [16]. We can write, if the formation energy of a kink with length $k$ is given by $E_k$,

$$\langle (h_o - h_r)^2 \rangle = 2Dr\,, \tag{4a}$$

with

$$2D = \langle k^2 \rangle = \sum k^2 p_k = \frac{\sum k^2 \exp(-E_k/kT)}{\sum \exp(-E_k/kT)}\,, \tag{4b}$$

where $p_k$ is the probability of a kink with length $k$. The summation $\sum$ runs over all possible $k$ values. The value $\langle k^2 \rangle$, which is the local mean square length of a kink or the diffusivity (2D) of a kink [12], can immediately be extracted from an analysis of the distribution of kinks and kink lengths as demonstrated in two different papers [8] (this results in a value of about 1.5 for a $B$-type and 0.1–0.2 for an $A$-type step edge). The values $G(1) \approx 1.7$ for the rough $B$-type and $G(1) \approx 0.1$ for an $A$-type step edge as obtained from Fig. 2 correspond reasonably well with the values as derived from Ref. [8]. The 1D random walk does not hold above a certain $r$ value which depends of course on the terrace length and the type of step edge as well as the diffusivity of the kinks. This limitation can simply be understood because although step-step interactions may be very weak on Si(001), step edges never cross and hence we must regard the step edges at least as hard walls. This restricts the diffusivity of the kinks when step edges approach each other and limits the deviation-deviation correlation function to a maximum value of $(2/3 - 4/\pi^2)L^2$. The last result can easily be obtained using the probability function at low $T$: $P(x,L) = (1/L) \times \cos^2(\pi x/2L)$ [17]. $P(x,L)$ is peaked around $x=0$ due to the large contribution of the configurational entropy when the step edge is midway between the walls [17]. The highest value of the experimentally determined deviation-deviation correlation function is very close to the maximum value $(2/3 - 4/\pi^2)L^2$ indicating that step-step interactions are very weak. Important to note is that a weak step-step interaction corresponds to a low $T_r$ [see Eq. (3)], so in our case $T_r < T_f$ certainly holds.

Another feature which may be important to note is that in every $B$-type step edge we analyzed, we observed a decrease in the correlation function for $r$ values in the range 40–60 dimer positions. This may be an indication for the existence of step waviness [18], although the period of the waviness is clearly too small to compare it immediately with the observations of Tromp and Reuter [18]. Finally, we want to note that we have not addressed the influence of defects, which may act as pinning centers for kinks [7].

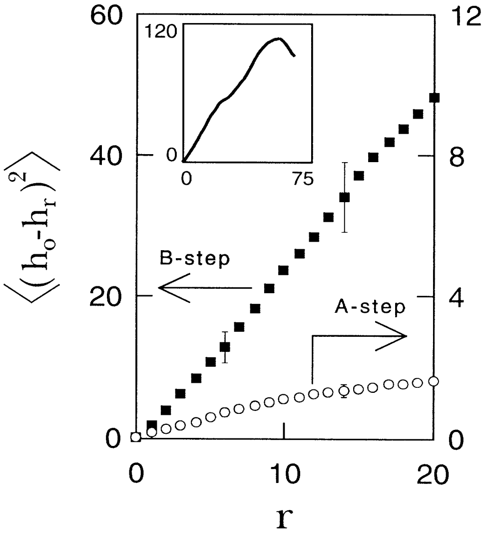

FIG. 2. Deviation-deviation correlation function $G(r) = \langle (h_o - h_r)^2 \rangle$ of an $A$- and a $B$-type monatomic step edge vs $r$. Typical error bars are indicated. In the inset $G(r)$ is given for a $B$-type step edge up to $r=70$.

Summarizing, STM measurements of vicinal Si(001) at 725 K clearly demonstrate the changes in the meandering of the monatomic step edges. The roughening temperature of the monatomic step edges (determined by dimer-dimer and step-step interactions) on this surface is, however, predicted to occur at significantly lower temperatures. So, the freeze out temperature of the monatomic step edges on Si(001) lies well above the roughening temperature and hence these step edges will always be rough at room temperature. The deviation-deviation correlation function of an *A*- or *B*-type step edge is proportional to $r$ for $r$ values up to about 20 dimer rows. This result is in agreement with a 1D random walk which suggests that step-step interaction is relatively small as compared to the kink formation energies. We found no indication for a Kosterlitz-Thouless type of roughening transition which predicts a $\ln(r)$ behavior above $T_r$.

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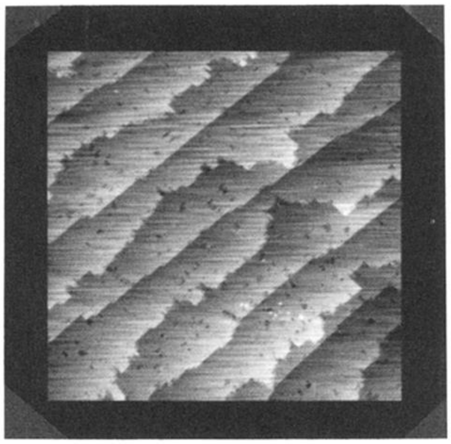

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