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Advacancy-induced step bunching on vicinal surfaces

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Step bunching at the surface of a sublimating crystal Joule-heated by a dc electric current is investigated in the presence of both adatoms and surface vacancies (advacancies). We show that the inclusion of advacancies in the step flow model of Burton, Cabrera, and Frank is crucial in order to reproduce the high-temperature behavior of real Si(111) vicinal surfaces. This provides a complete qualitative picture of the morphologies reported by Latyshev *et al.* [Surf. Sci. **213**, 157 (1989)], and strongly supports the hypothesis of electromigration. Agreement with experiments is obtained only assuming that the force exerted on adatoms and advacancies by the electric current is opposite to the latter.

Several observations were reported in the past[1–5] concerning step bunching on the Si(111) vicinal surface, when the sample is heated by a dc electric current. The surface morphology is seen to depend on the direction of the electric current with respect to the step staircase, as summarized in Fig. 1. According to Latyshev *et al.*,[1,2] when the current direction coincides with the direction of step motion during sublimation (step-up current), step bunches appear in the temperature range $\approx 1300-1500$ K, disappear in the range $\approx 1500-1600$ K, and again reappear above 1600 K (the melting temperature of Si is about 1685 K). Reversing the current direction (step-down current) leads to step bunching only between $\approx 1500-1600$ K, as sketched in Fig. 1.

This current-dependent behavior convinced experimentalists that *electromigration* of silicon adatoms was involved. Electromigration is a well-known phenomenon in metals, both in the bulk and at the surface. When an electric field is applied to a metal sample, impurities and interstitials migrate preferentially parallel to the field (not necessarily to its direction). At the surface, electromigration is known to take place even for metals deposited on silicon (111) (whose 7×7 reconstructed structure has metallic properties).[6,7]

Theory[8,9] predicts that interstitials (to which adatoms can be assimilated) should drift in the direction of the motion of the electric carriers, due to transfer of momentum from collisions with the carriers themselves. Thus, migration should take place preferentially *against* the field direction (and against the electric current) for conduction due to electrons.

Stoyanov[10] proposed a simple but attractive model to describe the dc current-induced step bunching. It is based on the Burton, Cabrera, and Frank[11] step flow model, with the addition of two ingredients: (i) the adatoms are subject to an electromigration (drift) force; (ii) steps are not perfect sinks for adatoms, i.e., adatoms are absorbed and emitted by steps at a finite rate. This model describes step bunching at low temperature and for step-up current, and predicts step debunching when the adatom diffusion length becomes of the order of the average terrace size. In fact, when this occurs, surface diffusion is no longer able to couple neighboring steps, which have no more tendency to bunch.

However, the model as it stands cannot account for reappearance of bunches at still higher temperature. We address this question in this paper.

Recent work[12,13] has pointed to the role of advacancies in surface dynamics near the melting point. The presence of advacancies on sublimating silicon surfaces is confirmed by the observation of advacancy islands.[1] Thus it is necessary to account for advacancies in the high-temperature regime, as already suggested by Stoyanov.[14]

In fact, assuming, as it is reasonable, that the electric current exerts on the advacancies a force opposite to that on the adatoms, advacancies can be expected to cooperate in the mechanism by which step bunching takes place, and to be essential for the "reentrant" step bunching at high T. We shall indeed show that incorporation of advacancies in a microscopic model of adatom electromigration allows for a complete and even semiquantitative description of the observed surface morphology as a function of temperature.

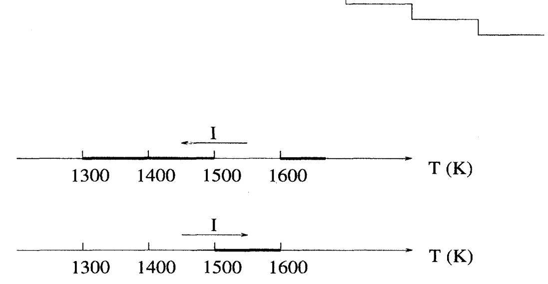

FIG. 1. Temperature range of step-bunching instability for ascending and descending current. Thick lines represent unstable regions.

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The model of Burton, Cabrera, and Frank extended so as to incorporate creation and recombination of adatom-advacancy pairs[12] and electromigration[10] reads

$$D\frac{\partial^2 c}{\partial z^2} - \frac{c}{\tau} - \frac{DF}{k_BT}\frac{\partial c}{\partial z} - Kc\sigma + Kc_0\sigma_0 = 0, \tag{1}$$

$$\Lambda\frac{\partial^2 \sigma}{\partial z^2} + \frac{DF}{k_BT}\frac{\partial \sigma}{\partial z} - Kc\sigma + Kc_0\sigma_0 = 0 \ , \tag{2}$$

where c and σ designate the adatom and advacancy densities, respectively, c_0 and σ_0 their equilibrium values, D and Λ are the adatom and advacancy diffusion coefficients, τ is the average residence time of an adatom before desorption, F the electromigration force, k_BT the thermal excitation energy, and K is a kinetic reaction coefficient. Here, for the sake of simplicity, we assume that adatoms and advacancies are subject to the same electromigration force (in absolute value).

The above equations are to be supplemented with kinetic equations describing attachment at the steps. In an Onsager picture these equations take the form[15]

$$-\frac{\partial c}{\partial z} + \frac{c}{\xi} = \mp\frac{c-c_0}{d}, \tag{3}$$

$$-\frac{\partial \sigma}{\partial z} - \frac{\sigma}{\xi} = \mp\frac{\sigma-\sigma_0}{d_v} \ , \tag{4}$$

where $\xi = k_BT/F$, $d = D/k$, and $d_v = \Lambda/k_v$, each having the dimension of a length. The quantities k and k_v are the adatoms and advacancies attachment-detachment kinetic coefficients at the steps. We adopt the simplifying assumption—which does not affect the main conclusion—that the step kinetics are symmetric, that is, there is no Schwoebel effect (see below). The + and − signs in Eqs. (3) and (4) hold for the lower and the upper side of a step, respectively. Finally to complete the description, the step normal velocity v_n is related to the mass currents $J_\pm = [D(\partial c/\partial z - Fc/k_BT) - \Lambda(\partial\sigma/\partial z + F\sigma/k_BT)]_{z=0^\pm}$ (J_+ and J_- are the currents from the lower and upper terraces, respectively) by

$$v_n = \Omega(J_+ - J_-) \ . \tag{5}$$

Ω is the atomic area of the solid. The set of Eqs. (1)–(5) completely describes step dynamics for straight steps.

This problem is nonlinear, for two reasons. (i) An obvious nonlinearity stems from the adatom-advacancy reaction term. (ii) There is a hidden nonlinearity which is peculiar to any moving-boundary problem (a Stefan-like problem). This nonlinearity can be accounted for rather easily, while the one originating from Eq. (1) precludes a full analytical treatment. However, before resorting to a brute-force numerical treatment, we can gain some insight by assuming weakly out-of-equilibrium conditions. That is, we assume that the actual values σ and c are close to their equilibrium values, σ_0 and c_0. This is reasonable, because the evaporation rate is small even at quite a high temperature. Quantitatively, this can be expressed by saying that the adatom diffusion length before desorption, $x_s = \sqrt{D\tau}$, is at least as large as the terrace size even at high T.[16]

Expanding Eq. (1) to first order in $c_1 = c - c_0$ and $\sigma_1 = \sigma - \sigma_0$, we obtain

$$\frac{\partial^2 c_1}{\partial z^2} - \xi^{-1}\frac{\partial c_1}{\partial z} - (x_s^{-2} + \ell_1^{-2})c_1 - \ell_2^{-2}\sigma_1 - x_s^2 c_0 = 0, \tag{6}$$

$$\frac{\partial^2 \sigma_1}{\partial z^2} + \xi^{-1}\frac{\partial \sigma_1}{\partial z} - \ell_3^{-2}\sigma_1 - \ell_4^{-2}c_1 = 0 \ , \tag{7}$$

where $\ell_1 = \sqrt{D/K\sigma_0}$, $\ell_2 = \sqrt{D/Kc_0}$, $\ell_3 = \sqrt{\Lambda/Kc_0}$, and $\ell_4 = \sqrt{\Lambda/K\sigma_0}$. Note that the relation $\ell_1\ell_3 = \ell_2\ell_4$ holds.

The general solution of Eqs. (6) and (7) takes the form $c_1 = -c_0 + e^{\alpha z}$ and $\sigma_1 = \sigma_0 + e^{\alpha z}$, where α obeys the fourth order algebraic equation

$$\left(\alpha^2 - \frac{\alpha}{\xi} - (x_s^{-2} + \ell_1^{-2})\right)\left[\alpha^2 + \left(\frac{\alpha}{\xi} - \ell_3^{-2}\right)\right] - (\ell_2\ell_4)^{-2} = 0. \tag{8}$$

The four solutions can be written analytically or solved numerically at convenience. Let α_i ($i = 1, \ldots, 4$) label the four solutions (which are, for the large set of parameters explored so far, all real). Therefore the solution for c_1 and σ_1 takes the form

$$c_1 = -c_0 + \sum_{i=1}^{4} A_i e^{\alpha_i z}, \quad \sigma_1 = \sigma_0 + \sum_{i=1}^{4} A_i R_i e^{\alpha_i z} \ , \tag{9}$$

where the A_i's are integration constants, and $R_i = \ell_2^2[\alpha_i^2 - \alpha_i/\xi - (x_s^{-2} + \ell_1^{-2})]$.

The four integration constants A_i are determined straightforwardly from the four conditions at the step Eqs. (3) and (4). Since this involves writing huge formulas, we have not felt it worthwhile to list them in this brief exposition. Once c_1 and σ_1 are known, we can determine the growth velocity [Eq. (5)] for an arbitrary configuration of the train (i.e., for an arbitrary phase shift between two consecutive steps). This results in a set of nonlinear evolution equations for the instantaneous position of the steps (or equivalently the terrace widths; see Ref. 17). This set of equations has a steady-state solution where the steps are equidistant and move at a constant speed. We investigate next the stability of this solution. This can be accomplished by considering small departures away from the steady-state solution, i.e., small fluctuations of the terrace widths. The result can be cast into a relation linking the wave vector of each fluctuation to its growth rate in time. A positive growth rate signals an instability.

Explicitly, the step position is written as $\zeta_n = n\ell_0 + \zeta_0 e^{\omega t + inq}$, where ω is the growth rate, q the wave vector, and ℓ_0 the step separation in the steady-state regime. The rate ω can be cast in the form $\omega = \partial\psi/\partial\ell_0$, where $\psi(\ell_0)$ is given by

$$\psi = \sum_{i=1}^{4} (e^{\alpha_i\ell_0} - 1)\left(\frac{D}{d} - R_i\frac{\Lambda}{d_v}\right)A_i(\ell_0). \tag{10}$$

Here we have written ℓ_0 in the argument of A_i to remind that they explicitly depend on ℓ_0. Expression (10) is written

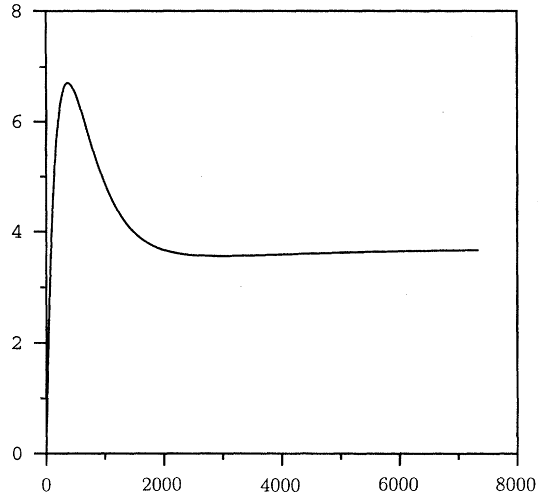

FIG. 2. The function $\psi(\ell_0)$ as a function of ℓ_0 (ℓ_0 is measured in atomic spacing unit). We have taken $x_s = e^{\beta 1.2(\mathrm{eV})}$; $d = e^{-\beta 0.5(\mathrm{eV})}$; $c_0 = e^{-\beta 0.5(\mathrm{eV})}$; $\sigma_0 = e^{-\beta 0.5(\mathrm{eV})}$; $l_1 = l_2 = l_3 = l_4 = 1.5 x_s$. The vertical unit is arbitrary.

for $q = \pi/\ell_0$ (out-of-phase fluctuations of neighboring steps), since this one is the most unstable mode (see Ref. 17).

Clearly, a positive slope of the function $\psi(\ell_0)$ corresponds to a step pairing instability. Note that this linear stability analysis only gives the instability threshold, but says nothing on the long-term surface morphology, which is dictated by the nonlinearities. To obtain this, one must numerically solve the full nonlinear problem, or perform simulations.

If only adatoms are present, we find Stoyanov's result:[10] $\psi(\ell_0)$ behaves as ℓ_0 at small ℓ_0/x_s, goes through a maximum for $\ell_0 \sim x_s$ before it decays exponentially at larger ℓ_0. In other words we have step pairing at small ℓ_0/x_s, while for $\ell_0 > x_s$ stability is restored. Since x_s decreases with temperature, Stoyanov's model accounts for the appearance of step bunching at low temperature, and its disappearance at higher temperature (when $\ell_0 > x_s$). This model does not, however, account for the reappearance of the instability at still higher temperature, as reported in several experiments.[1–5]

The main outcome of the present study lies in the finding that incorporation of adavacancies, which are most likely to be active at high temperature, leads to *the reappearance of the instability* at high temperature *for step-down force*, or step-up current according to the electromigration hypothesis. This is a new result, which contrasts with previous attempts at the problem.[14] Figure 2 shows the behavior of $\psi(\ell_0)$ as a function of ℓ_0 at increasing temperature. The function ψ' (derivative of ψ with respect to ℓ_0) is proportional to the instability growth rate ω: when it is positive ω is positive and this signals an unstable situation. An instability is signaled by a positive slope. The presence of adavacancies results in a reappearance of a positive slope at large ℓ_0 (or small x_s—because only the ratio ℓ_0/x_s matters—which means at higher temperature). A convenient way to look at the figure is to fix a value of ℓ_0, and following the variation of the sign of $\psi'(\ell_0)$ at different temperatures. The result is presented in Fig. 3. We see that at a miscut of 8′ ($\ell_0 \approx 430$ in atomic distances; the full line in Fig. 3) the surface is unstable up to 1200 K, then it becomes stable until $T \approx 1700$ K, where it becomes unstable again. The region of stability is larger than in experiments (where it spans approximately 100 K), but the disagreement is not dramatic. In any case, it is clear that the model has many weak points: it is only one dimensional, since we only look at the behavior of the terrace size; it does not account for fluctuations, which are likely to enforce the instability and to decrease consequently the width of the region of stability. This is why we have not tried very hard to find the best possible—nor even a better—set of parameters for drawing Fig. 2. Note that the result displayed in Fig. 3 shows also that at a temperature below about 650 K the growth rate goes rapidly to zero (actually the behavior is exponential). This explains why step bunching occurs only above a certain temperature, in agreement with the general behavior in Fig. 1. Finally, when the current direction is reversed, we obtain a complementary picture, i.e., stable intervals in the previous situation are unstable and vice versa.

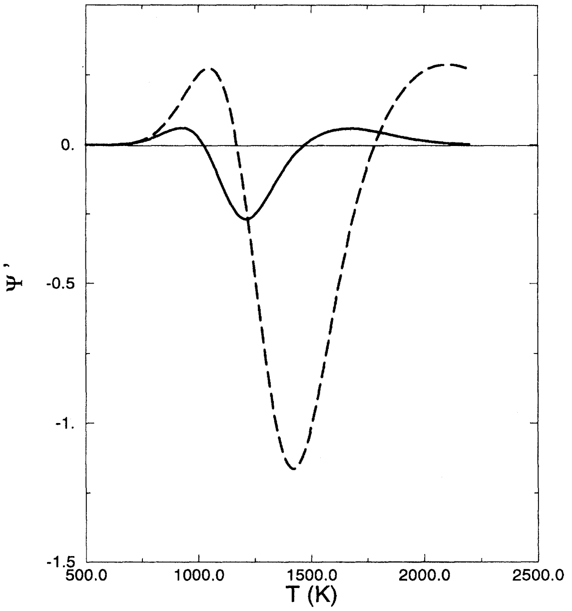

FIG. 3. Growth rate as a function of temperature for a fixed miscut (or a fixed interstep distance). Here we show two different interstep distances $\ell_0 = 1000$ (dashed line), and $\ell_0 = 430$ (full line). The sign of ψ' is the same as that of the growth rate of perturbations (an instability is signaled by a positive sign). Other parameters, the same as in Fig. 2. The vertical unit is arbitrary.

Our results obviously support, and substantiate, the hypothesis that adatom and advacancy electromigration is the key ingredient for understanding step bunching of Si(111). A coherent view is obtained by assuming that a force acts on silicon atoms, which is opposite to the electric current. This is in agreement with the semiclassical picture of the so-called "wind-force effect": the conduction electrons transfer by collision their momentum to the surface atom, which feels then a force directed along the electron drift—clearly opposite to the current.

The next step of the analysis is to deal with the subsequent development of the instability. This task requires a nonlinear treatment. This is crucial in order to settle important questions, such as the study of wavelength selection of the bunches (when this notion has a meaning), and/or static and dynamical statistical features of the surface morphology. We hope to report along these lines in the future.

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