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Singular response to a dopant of an evaporating crystal surface

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Moving crystal surfaces can undergo step-bunching instabilities when subjected to an electric current. We show analytically that an infinitesimal quantity of a dopant may invert the stability, whatever the sign of the current. Our study is relevant for experimental results [S. S. Kosolobov *et al.*, JETP Lett. **81**, 117 (2005)] on an evaporating Si(111) surface, which show a singular response to Au doping, whose density distribution is related to inhomogeneous Si diffusion.

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Introduction. Crystal surfaces may undergo dynamical instabilities while growing or evaporating.[1,2] An important class of instabilities is that determined by an asymmetric current of adatoms diffusing on terraces. This asymmetry may be intrinsic, e.g., determined by asymmetric attachment to steps[3] or by surface reconstruction,[4] or it may be extrinsic, e.g., due to impurities[5] or to an electric current.[6] Extrinsic instabilities may have the advantage to be tunable and therefore to be suitably switched on and off. In this Rapid Communication we consider an example of extrinsic instability displaying a phenomenon which is of double interest: an interest not only for fundamental nonequilibrium physics, because we propose a model showing a singular response to doping, but also for applications, because we suggest that recent experimental results[7] on evaporating Si(111) surfaces under an electric current, showing a singular response when exposed to a variable quantity of Au, can be understood in light of our model.

Vicinal surfaces are obtained cutting a crystal along an orientation which is close to a high-symmetry one, resulting in a morphology similar to a flight of steps. During evaporation, atoms detach from steps and diffuse on terraces until possible desorption. The application of an electric current induces an electromigration force, whose direction may depend on temperature T: this is the case for the widely studied case of Si(111), where a sequence of stable and unstable regions are found when varying T.[8] Here we are interested in analyzing how stability is affected by doping. We assume (and later we discuss) that doping induces inhomogeneity in the diffusion process, both in its symmetric part and in the drift term, the latter modifying the electromigration force. In particular, we show that a divergent drift in a region of vanishing size may induce a change of stability, even if the sign of the drift does *not* change. This result might not appear as surprising as it is, because step decoration,[9] i.e., changing attachment kinetic coefficients, may have a similar effect. However, step decoration misses two important features: first, in our model and in the Si(111) experimental system, doping induces a change of stability for both signs of the current, meaning that doping cannot be reduced to a fixed stabilizing or destabilizing effect; second, an increasing quantity of dopant may induce further stability-instability transitions. In this Rapid Communication we focus on the very first transition, occurring at a vanishing critical density of the dopant. However, we also argue as to how a larger quantity of dopant may further change the stability of the surface.

Model. The simplest model of adatom diffusion in the presence of evaporation and drift [see Fig. 1(a)] requires us to solve the following stationary diffusion equation for the adatom density $p(x)$:

$$Dp''(x) - 2a_0^{-1}\delta p'(x) - \gamma^2 p(x) = 0, \tag{1}$$

where a_0 is the lattice constant; D is the diffusion constant; $\delta/a_0 = q_{\text{eff}}ED/(k_BT)$ is the drift, with q_{eff} being the effective charge of adatoms and $q_{\text{eff}}E$ being the electromigration force; and $\gamma^2 = 1/\tau$ is the evaporation rate. As illustrated in Fig. 1(b), we adopt a simple model for inhomogeneity: each terrace is separated in two regions with different values for D and δ. Boundary conditions are expressed in terms of the surface current

$$J(x) = -Dp'(x) + 2a_0^{-1}\delta p(x), \tag{2}$$

which at steps is proportional to the supersaturation

$$J_{\text{step}} = \pm K(p_{\text{step}} - p_{\text{eq}}). \tag{3}$$

Here the plus (minus) sign applies to the ascending (descending) step. We expressly chose the same kinetic coefficient K for the ascending (K_+) and descending (K_-) step to avoid stabilizing or destabilizing effects due to their asymmetry, $K_+ \neq K_-$.[1,2]

The general solution of Eq. (1) for the piece of terrace $x \in [\alpha\ell,\ell]$ is given by

$$p(x) = Ae^{\lambda_1 x} + Be^{\lambda_2 x}, \tag{4}$$

with

$$\lambda_{1,2} = \frac{\delta \pm \sqrt{\delta^2 + a_0^2 D\gamma^2}}{Da_0}, \tag{5}$$

and analogously for $x \in [0,\alpha\ell]$, with coefficients A',B' in Eq. (4). The unknowns A,B,A',B' are determined from two conditions (3) at steps $x = 0,\ell$ and from continuity of current (2) and density $p(x)$ at $x = \alpha\ell$. Before going further, let us recall what is the relevant function determining the stability of the evaporating surface. Each step moves with a velocity proportional to the sum of the upper and lower step current, J_{step}, as given by Eq. (3). Perturbing a perfect train of steps, we find[10] the stability being governed by the function

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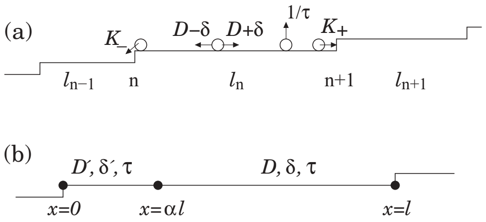

FIG. 1. Profiles of (a) a one-dimensional surface and (b) a terrace, relevant to the inhomogeneous model. All symbols are explained in the text.

$\phi(\ell) = p(\ell) - p(0)$: the system is stable (unstable), if $\partial_\ell \phi(\ell)$ is positive (negative).

We now assume a diverging drift in the region close to the descending step ($\delta' \to \infty$), whose size vanishes in such a way that the product

$$\frac{\delta' \alpha \ell}{a_0 D} = c \tag{6}$$

is constant. This model, as discussed at length in the experiment section, is motivated by a possible strong impact on drift of a dopant highly localized near steps. As shown in detail in the Supplemental Material,[11] we do not really need a "singular" model with a diverging δ' in order to get reversal stability, but the experimental system does show a singular behavior.

We also considered the case of a diverging D, but it resulted in having no effect when restricted to a region of vanishing size. For this reason, the parameter D' does not appear in the following equations. For ease of notation, we define the scaled quantities $\tilde{\delta} = \delta/(a_0 D)$, $\tilde{\gamma} = \gamma/\sqrt{D}$, and $\tilde{K} = K/D$, all of dimension $[L^{-1}]$.

Solving the two coupled diffusion problems in the above limit, we have found that the resulting effect is equivalent to introducing a new boundary condition at the descending step ($x = 0$),

$$J_{\text{step}} = -K(p_{\text{step}} e^{-2c} - p_{\text{eq}}). \tag{7}$$

The new condition arises because the density profile in proximity to the descending step becomes infinitely steep in the limit $\alpha\ell \to 0$, changing from some value p_{step} at $x = 0$ to a value $p_{\text{step}} e^{2c}$ at $x = \alpha\ell \to 0^+$. Therefore, $p_{\text{step}} e^{2c}$ becomes our new postulated value $p(x = 0)$, after taking the limit $\alpha\ell \to 0$. Because of this redefinition of J_{step}, the function $\phi(\ell)$, which governs the stability of surface dynamics, has to be redefined as $\phi(\ell) = p(\ell) - p(0)e^{-2c}$.

By solving Eq. (1) with the usual boundary condition (3) at $x = \ell$ and new boundary condition (7) at $x = 0$, and assuming

$$\tilde{\delta}\ell \ll \tilde{\gamma}\ell \ll 1, \tag{8}$$

we obtain

$$\partial_\ell \phi(\ell\,) = \frac{2\tilde{K}^2 e^{-2c} p_{\text{eq}} (4\tilde{\delta} - \tilde{K}(1 - e^{-2c}))}{(\tilde{K} + \tilde{K}e^{-2c} + 2\tilde{\delta}\tilde{K}\ell + \tilde{K}^2 e^{-2c}\ell)^2}. \tag{9}$$

Therefore, the instability condition $\partial_\ell \phi(\ell) < 0$ reads

$$4\tilde{\delta} < \tilde{K}(1 - e^{-2c}), \tag{10}$$

which reduces to the usual condition $\tilde{\delta} < 0$, when the boundary layer is absent ($c = 0$). It is worth noting that Eq. (10) does not depend on the desorption rate γ. In fact, it is possible to study a model where evaporation is neglected: in this case, simpler calculations allow us to keep δ' large, but finite. This calculation, which gives the same result and therefore proves the robustness of our model, can be found in the Supplemental Material.[11] In the experimental system[7] we consider here, desorption is not negligible, see Eq. (8), but we have shown it does not affect the stability reversal process.

If Eq. (10) is satisfied, the instability time τ_{ins} is set by the relation[12]

$$\tau_{\text{ins}}^{-1} = a_0 K |\partial_\ell \phi(\ell)|. \tag{11}$$

Analogously, one may consider a situation with diverging drift in the region close to the ascending step, $\delta' \ell(1 - \alpha)/(a_0 D) = c_1$ as $\alpha \to 1$. This inhomogeneity results in a new effective boundary condition at the ascending step,

$$J_{\text{step}} = K(p_{\text{step}} e^{2c_1} - p_{\text{eq}}) \tag{12}$$

and in the redefinition $\phi(\ell) = p(\ell)e^{2c_1} - p(0)$, so that the instability condition $\partial_\ell \phi(\ell) < 0$ becomes

$$4\tilde{\delta} < \tilde{K}(e^{2c_1} - 1). \tag{13}$$

Expressions (10) and (13) are a central result of our paper. In the following we discuss their application to a surprising experimental effect, observed on the evaporating surface of Si(111) in the presence of a tiny coverage on Au atoms.

Experiment.[7] Step-bunching instability of a Si(111) vicinal surface heated by a dc electric current to high temperatures is a well-established phenomenon.[13,14] In the so-called first temperature regime ($830 \leqslant T \leqslant 950\,^\circ$C), the regular vicinal surface is stable (unstable) for an uphill (downhill) current. The effect is usually understood[8] by assuming that neutral silicon adatoms acquire an effective positive charge which gives rise to an electromigration force δ. In the first temperature regime, approximations (8) are valid,[15] the density profile is linear, $p(x) \simeq \delta x$, and we have stability for $\delta > 0$.

A few years ago, it was observed by Kosolobov, Latyshev, and collaborators (KL) that a submonolayer deposition of gold on a vicinal Si(111) surface drastically affects its stability, which changes *four* times as a function of the increasing Au coverage.[7] In particular, the addition of a very small quantity of Au atoms to a clean Si surface (0.0016 ML in Ref. 16) resulted in a reversal of stability (uphill dc setup becomes unstable and vice versa). Such critical density, not enough even to decorate vicinal steps with a single line of Au atoms, hints at a phase transition at zero critical density, $\rho_{\text{Au}}^{\text{cr}} \to 0$ as $\ell \to \infty$. Moreover, the instability develops much faster than is usual at this temperature for a clean Si surface.[17]

We propose to relate this phenomenon to our model of Fig. 1(b), with a diverging drift. To do so, we note that Au adatoms have a much larger effective charge than Si adatoms,[18,19] so that their dynamics in the electric field is faster. As a consequence, Si adatoms move in an established environment of Au adatoms, whose steady profile may be strongly inhomogeneous near the border. Relating Au inhomogeneity to Si diffusion inhomogeneity causes a stability reversal, as demonstrated below.

The key observation is that the negative effective charge of Au atoms and their strong affinity[20] to the step region make them concentrate near the descending step boundary $x = 0$. The effective equilibrium profile of Au adatoms, $n(x)$, is governed by an equation of type (1), i.e.,

$$D_{\mathrm{Au}} n''(x) + 2a_0^{-1}|\delta_{\mathrm{Au}}| n'(x) - \frac{n}{\tau_{\mathrm{Au}}} = 0. \tag{14}$$

Due to the strong affinity assumption, the boundary condition is $n(0) = n^+_{\mathrm{Au}}$, where n^+_{Au} is the equilibrium density of Au adatoms close to the descending step. Within the experimental instability time scale, Au coverage remains approximately constant. Therefore, we can solve Eq. (14) assuming a negligible Au evaporation and get $n(x) = n^+_{\mathrm{Au}} e^{-\kappa x}$, where $\kappa = 2a_0^{-1}|\delta_{\mathrm{Au}}|/D_{\mathrm{Au}}$.

If we now impose that the space integral of $n(x)$ equals the total number of deposited Au atoms,

$$\rho_{\mathrm{Au}} \ell = \int_0^\ell n(x)dx = \frac{n^+_{\mathrm{Au}}}{\kappa}, \tag{15}$$

we get the relation $\kappa = n^+_{\mathrm{Au}}/\rho_{\mathrm{Au}}\ell$, which diverges for $\rho_{\mathrm{Au}} \to 0$. Finally, the equilibrium density profile writes

$$n(x) = n^+_{\mathrm{Au}} \exp\left(-\frac{n^+_{\mathrm{Au}} x}{\rho_{\mathrm{Au}} \ell}\right). \tag{16}$$

Analogously, for downhill current and small density of Au adatoms we obtain a distribution function peaked near the upper step boundary, $x = \ell$,

$$n(x) = n^-_{\mathrm{Au}} \exp\left(\frac{n^-_{\mathrm{Au}}(x - l)}{\rho_{\mathrm{Au}} \ell}\right). \tag{17}$$

As anticipated, we relate the gradient in the density of Au atoms to an additional drift (effective electromigration force) of Si adatoms as[21]

$$\delta' = \delta - c^* \frac{\partial n(x)}{\partial x}. \tag{18}$$

This renormalizes the drift in the region where the gradient is sufficiently large (see Fig. 2), getting

$$\delta(x) = \delta' = c^* n^+_{\mathrm{Au}} \kappa, \quad \text{for} \quad x \in [0, \kappa^{-1}]. \tag{19}$$

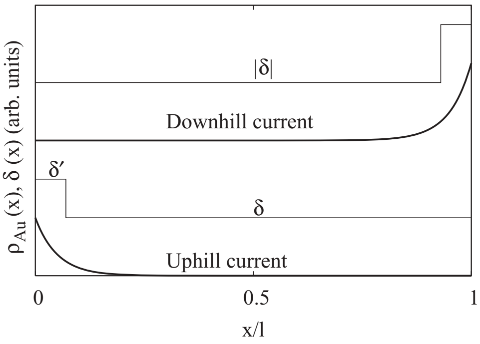

FIG. 2. Schematic profiles of the density of dopant adatoms (thick lines) and drifting forces $\delta(x)$ (thin lines) at small dopant concentration, $\rho_{\mathrm{Au}}\ell \ll 1$. For uphill (downhill) current $\delta > 0$ ($\delta < 0$), the dopants are concentrated near the left (right) boundary. The $\delta < 0$ curves are shifted for better vision.

If we compare Eq. (19) with Eq. (6) and identify the size ($\alpha\ell$) of the vanishing region of divergent drift with κ^{-1}, we find the relation $c = c^*(n^+_{\mathrm{Au}}/a_0 D)$. In conclusion, we have related the problem of Si diffusion in the presence of a tiny quantity of Au doping to the model of inhomogeneous diffusion, discussed in the previous part of this Rapid Communication.

We stress that our results do not change qualitatively if the parameter c has a slow dependence on other quantities such as the local gradient or average density of Au adatoms: the decisive role is played by the sign of c. Note also that the limit of a diverging drift and gradient is a simplifying assumption chosen for clarity of presentation. Simulation of a discrete model with strictly *finite* drifts and gradients yields qualitatively the same results as the continuous model (to be discussed elsewhere).

Discussion. The parameters given in Refs. 22 anbd 23 lead to dimensionless estimates $\tilde{\delta}\ell \approx 10^{-6}$, $\tilde{\gamma}\ell \approx 10^{-4}$, and $\tilde{K}\ell \approx 10^{-2}$, where $\ell \approx 100$ nm is a typical size of a terrace. This means that assumptions (8) are satisfied and Eqs. (9)–(13) do apply to the KL experiments.

For a tiny quantity of gold, Eqs. (10) and (11) should be evaluated. Since $\tilde{K}/\tilde{\delta} \approx 10^4$, Eq. (10) is surely satisfied for finite c (more precisely, for $c > 2 \times 10^{-4}$). The microscopic origin of the strong Au-induced increasing of the effective drift $\delta(x)$ for silicon adatoms may be a hard-core exclusion interaction between Au and Si adatoms: the presence of Au adatoms near the step excludes the presence there of Si adatoms and thus generates an effective enhanced drift of Si adatoms close to the step,[24] proportional to the local density gradient, as in Eq. (18).

Another microscopic origin might be the recharging effect,[16] according to which a neutral Au adatom subtracts negative charge from the substrate, while the substrate, to compensate, transfers an equal charge of the opposite sign to the Si adatoms. Since $q^{\mathrm{eff}}_{\mathrm{Si}} \approx 0.004e$[18] and $q^{\mathrm{eff}}_{\mathrm{Au}} \approx -e$,[19] we expect that recharging effect may lead to a strong renormalization of δ. Note that the drift enhancement due to recharging is proportional to the local density of the Au adatom itself, and not to its gradient as in the case of the hard-core exclusion effect alone. With some amendments to our line of argument [Eqs. (15)–(19)], the instability reversal can be obtained. We note that both recharging and hard-core exclusion effects contribute with the same sign to the renormalization (18), leading to the above-discussed kinetic instability reversal. We also stress that the effective charge of Si adatoms remains positive across all the transitions.

A piece of evidence that c is not small and also that our theory does apply to KL experiments is the expectation that Au doping leads to a much stronger instability than simple reversing of the sign of the current. In fact, since $\tilde{K}(1 - e^{-2c}) \gg \tilde{\delta}$, according to Eqs. (9) and (11) we have that $\tau^{\mathrm{doping}}_{\mathrm{ins}} \ll \tau^{\mathrm{reversal}}_{\mathrm{ins}}$, as it is actually seen in experiments.[17]

In the Introduction we stressed that the same doping induces a change of stability for both signs of the current. Therefore, let us now consider a downhill current, which means $\delta < 0$ and that the clean Si surface is bunching-unstable, see Eq. (13) with $c_1 = 0$. Au adatoms are now driven towards the ascending step and the boundary layer (inhomogeneity) forms at the ascending step, with a profile given by Eq. (17)

FIG. 3. Stability diagram of a Si(111) surface, with an uphill current ($\delta > 0$), when doped with a variable quantity of Au. The clean surface ($\rho_{\rm Au} = 0$) and the fully covered Si surface ($\rho_{\rm Au} = 1$) are stable. A tiny Au deposition on the clean surface or a tiny removal of it from the fully covered surface induces a stability reversal and the surface becomes unstable (thick horizontal lines). In the intermediate region, the surface is expected to recover stability (thin horizontal line), because the Au profile is more flat. If the sign of the current is reversed, stability regions are also reversed (thick line ⇔ thin line).

(see also Fig. 2). Assuming an additional drift of Si adatoms, $\delta' = -c^* \frac{\partial n(x)}{\partial x} = c^* n^-_{\rm Au}\kappa$, in the region of a strong Au gradient, $(\ell - x) < 1/\kappa$, we recover our model with diverging drift δ' at the ascending step. The same considerations we had for positive δ and Eq. (10) are now applicable to negative δ and Eq. (13), providing perfectly symmetrical conclusions for a negative electromigration force: a tiny amount of Au doping allows us to stabilize the surface, and the stability is much stronger than the stability gained by simply reversing the field.

Let us now pass to reasoning further features of the KL experiments,[7] which go beyond our simple model. These experiments are done for a variable quantity of Au, from a clean Si surface ($\rho_{\rm Au} = 0$) to an almost full covering ($\rho_{\rm Au} = 1$)[24] and the authors find a total of four stability transitions while increasing $\rho_{\rm Au}$ (see Fig. 3). It is reasonable to assume that enhanced drift is suppressed with increasing Au coverage, which becomes more homogeneous. Since a model with a constant $\delta(x)$ is equivalent to a clean Si surface, we expect that increasing $\rho_{\rm Au}$ leads to a new reversal of stability, in agreement with experiments. However, why does a further increase of $\rho_{\rm Au}$ produce a further change of stability? This change of stability is better understood if large $\rho_{\rm Au}$ coverages are described in terms of "Au holes," i.e., empty places which can potentially host Au adatoms: $\rho_{\rm Au}^{\rm holes} = 1 - \rho_{\rm Au}$. A gradient of Au adatoms corresponds to a gradient of Au holes, which appears to have a strong *positive* effective charge. This change of sign compensates for the negative sign relating the two spatial derivatives, $\rho_{\rm Au}^{\prime\rm holes} = -\rho'_{\rm Au}$. Thus, removing a tiny quantity of Au from a fully covered Si surface reverses its stability. In Fig. 3 we graphically summarize the stability diagram of Au-doped Si(111).

We conclude with one prediction of our model: if dopant adatoms have a positive effective charge, such as Cu or Ag,[18] the stability reversal effect is not expected. Indeed, positively charged foreign adatoms under uphill current ($\delta > 0$) will be driven to an ascending step and eventually form a profile (17), for which Eq. (13) will apply. Proceeding along the lines of Eqs. (18) and (19), we conclude that Eq. (13) cannot be satisfied, since $\delta > 0$ and $c_1 < 0$, so an instability reversal will not happen. Indeed, for a Cu-doped surface, the reversal of bunching stability was not observed.[16]

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