Crystal symmetry, step-edge diffusion and unstable growth

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We study the effect of crystal symmetry and step-edge diffusion on the surface current governing the evolution of a growing crystal surface. We find there are two possible contributions to anisotropic currents, which both lead to the destabilization of the flat surface: terrace current $\vec{j}_t$, which is parallel to the slope $\vec{m} = \nabla z(x,t)$, and step current $\vec{j}_s$, which has components parallel ($\vec{j}_{s\parallel}$) and perpendicular ($\vec{j}_{s\perp}$) to the slope. On a high-symmetry surface, terrace and step currents are generically singular at zero slope, and this does not allow to perform the standard linear stability analysis. As far as a one-dimensional profile is considered, $\vec{j}_{s\perp}$ is irrelevant and $\vec{j}_{s\parallel}$ suggests that mound sides align along [110] and [110] axes. On a vicinal surface, $\vec{j}_s$ destabilizes against step bunching; its effect against step meandering depends on the step orientation, in agreement with the recent findings by O. Pierre-Louis et al. [Phys. Rev. Lett. 82, 3661 (1999)].

I. INTRODUCTION

The kinetic stability of a crystal growing homoepitaxially by Molecular Beam Epitaxy is determined primarily by the possible existence of a slope-dependent mass current $\vec{j}(\vec{m})$ along the surface, i.e. by a current which does not vanish in the limiting case of a constant slope $\vec{m} = \nabla z(x,t)$, where $z(x,t)$ is the local height \footnote{In the limit of very large ES effect, $\vec{j}_s$ is no more anisotropic and therefore no more singular at $\vec{m} = 0$.}. Such a current is generally ascribed to the so-called Ehrlich-Schwoebel (ES) effect at step-edges, which hinders interlayer diffusion \footnote{Corresponding author. Present address: INFM, L.go E. Fermi 2, I-50125 Florence. E-mail: politi@fi.infn.it}.

On singular surfaces, experimental results (mainly on metal growth) show that the instability leads to mound formation and often to a coarsening process, where the typical size $L$ of the mounds increases in time (generally with a power law: $L(t) \sim t^n$) \footnote{different coarsening exponents $n$}. The template of the mound structure is already formed in the early stages of growth (the so-called 'linear regime'), and here crystal structure should determine shape and orientation of mounds. For example, cubic crystals are characterized by a four-fold and a six-fold symmetry, respectively on (100) and (111) faces: experimental analysis by Scanning Tunneling Microscopy has indeed shown square based mounds on Fe and Cu(100) \footnote{Fe and Cu(100) \cite{5–7} and triangular based ones on Rh and Pt(111) \cite{8,9}. The relevance of the in-plane symmetry} and on Rh and Pt(111) \footnote{The relevance of the in-plane symmetry}. The template of the mound structure is already formed in the early stages of growth (the so-called 'linear regime'), and here crystal structure should determine shape and orientation of mounds. For example, cubic crystals are characterized by a four-fold and a six-fold symmetry, respectively on (100) and (111) faces: experimental analysis by Scanning Tunneling Microscopy has indeed shown square based mounds on Fe and Cu(100) \footnote{The relevance of the in-plane symmetry} and triangular based ones on Rh and Pt(111) \footnote{The relevance of the in-plane symmetry}. The relevance of the in-plane symmetry for the later stages of the growth process has been definitely proven by Siegert \footnote{The relevance of the in-plane symmetry}, who has shown –through a continuum description of the surface– that unstable currents with different in-plane symmetries may give rise to different coarsening exponents $n$. For vicinal surfaces ES barriers at steps are known to stabilize against step bunching and to destabilize against step meandering \footnote{The relevance of the in-plane symmetry}.

It is therefore extremely important to determine what are the microscopic mechanisms giving rise to slope-dependent currents $\vec{j}$, what is the expression of $\vec{j}$, and how lattice symmetry enters in it. One of the main results of the present paper is the finding of two contributions to the slope-dependent current, one due to terrace diffusion ($\vec{j}_t$) and one due to step diffusion ($\vec{j}_s$). Both contributions are singular at zero slope \footnote{The relevance of the in-plane symmetry}. This is at odds with the usual phenomenological expressions of $\vec{j}_s$, used in the continuum description of surface growth \footnote{The relevance of the in-plane symmetry}, which all reduce to the simple isotropic form $\vec{j}_s = a\vec{m}$ in the small slope regime ($\vec{m} \to 0$). We will see that our expressions for $\vec{j}_s$ and $\vec{j}_s$ remain anisotropic even in this limit, and that implies a singular behaviour in $\vec{m} = 0$. Other important results concern the step current $\vec{j}_s$, which is found to destabilize layer-by-layer growth against mound formation on a high symmetry surface, and step-flow against step bunching on a vicinal surface. Step-flow is stable or unstable against step meandering, depending on the step orientation.

The destabilizing effect of $\vec{j}_s$ on a singular surface has been observed independently by O. Pierre-Louis et al. \footnote{The relevance of the in-plane symmetry} and by Ramana Murty and Cooper \footnote{The relevance of the in-plane symmetry}. The former have also studied analytically the effect on step meandering. Here we provide a unified treatment of these diverse effects within a continuum description of the surface, we predict the new phenomenon of step bunching induced by step currents, and we analyze the different anisotropic behaviours of $\vec{j}_s$ and $\vec{j}_s$.
II. TERRACE CURRENT

In Fig. 1 we draw a piece of a vicinal surface corresponding to a constant slope $\tilde{m}$, and a piece of a step. Once adatoms have landed on the surface, they perform a diffusion process until they stick to the upper or lower step. The attachment rate $D'$ from below is considered extremely fast ($D'/D = \infty$, $D$ being the diffusion constant on the terrace); the rate $D''$ from above defines the ES length $\ell_{ES} = (D/D' - 1)$ (in units of the lattice spacing) [3,18,19]. This should be compared to the diffusion length $\ell_D$ representing the minimal distance between nucleation centers on a high-symmetry surface [20]. Under the usual conditions of crystal growth we have $\ell_D \gg 1$, while both the cases $\ell_{ES} \ll \ell_D$ (weak ES effect) and $\ell_{ES} \gg \ell_D$ (strong ES effect) may take place [3].

In one dimension (1d) we write the ES current (due to terrace diffusion) as $j_t = m f(m^2)$, and at small slopes (in the sense that $m \ll 1/\ell_D$) we have the linear behaviour $j_t = am \equiv f(0)m$, with $a = F \ell_{ES} \ell_D^2/[2(\ell_{ES} + \ell_D)]$, $F$ being the intensity of the external flux (i.e. the number of particles landing on the surface per unit time and lattice site). In two dimensions (2d), if we neglect in-plane anisotropy we can generalize and write $\tilde{j}_t = \tilde{m} f(m^2)$. Let us now discuss the microscopic origin of anisotropy and how it modifies $\tilde{j}_t$. Throughout we will consider a (100)-surface with fourfold symmetry, and take the $x$ ($y$) axis along the [100] ([010]) orientation, denoting by $\hat{x}$ and $\hat{y}$ the corresponding in-plane unit vectors. The extension to other crystal symmetries is straightforward in principle.

In the absence of surface reconstructions, terrace diffusion by itself is an isotropic process, at least in its continuum description. In contrast, the sticking of an adatom to a step depends on the microscopic environment, which depends on the step orientation. So, the ES barrier seen by an adatom approaching a step depends on the orientation of the surface and this dependence translates into an orientation-dependent ES length $\ell_{ES} = \ell_{ES}(\theta)$, where $\theta = \arctan(m_x/m_y)$ is the angle of the step relative to the $x$-axis. Assuming straight steps, the expression for a one-dimensional surface can be taken over, and for small slopes ($m \ell_D \ll 1$) we obtain

$$\tilde{j}_t = a(\theta)\tilde{m} = \frac{F \ell_{ES}(\theta) \ell_D^2}{2(\ell_{ES}(\theta) + \ell_D)} \tilde{m}.$$ (1)

The coefficient $a$ becomes independent of $\theta$ only in the regime of strong ES barriers, $\ell_{ES}(\theta) \gg \ell_D$ (in this limit, $a = F \ell_{ES}(\theta) \ell_D^2/2$). For weak barriers in-plane anisotropy is therefore present even in the 'linear' regime $m \ell_D \ll 1$ ($a = F \ell_{ES}(\theta) \ell_D^2/2$). Through the dependence of $\theta$ on $\tilde{m}$, Eq. (1) is manifestly non-analytic at $\tilde{m} = 0$.

III. STEP CURRENT

Next show that crystal symmetry manifests itself also (and perhaps mainly) through step diffusion. Once adatoms have reached a step, they can diffuse along it at a rate $D_s$ and stick to a kink edge (see Fig. 1). Similarly to terrace diffusion, only if there is an asymmetry between $D'_s$ and $D''_s$, a net step current $\tilde{j}_s$ exists; the strength of the asymmetry determines an ES length along the step which will be called $\ell_s$, the subscript $k$ standing for kink. Step diffusion biased by kink barriers is similar to terrace diffusion along a one-dimensional surface [14], but some differences are worth to be stressed.

i) All the possible in-plane orientations $\theta$ of the step, with the correct symmetries, should be taken into account, because –especially for a high-symmetry orientation– all the $\theta$ are found on the same surface. This may be true also
for a vicinal surface, if steps are subject to a strong meandering. In particular, orientations corresponding to \( \theta = 0 \) and \( \theta = \pi/4 \) will be seen to behave in a qualitatively different way. ii) Adatoms arrive at the step at a rate \( F_s \) depending on the terrace size \( \ell \). For equally spaced steps \( F_s = F \ell \). However, since (in 2d) the surface current is defined as the number of atoms crossing per unit time a segment of unit length, orthogonal to the current, the actual expression for the current is obtained by multiplying the ‘single-step current’ by the number of steps per unit length, i.e. \( 1/\ell \). This factor cancels the factor \( \ell \) appearing in \( F_s \), since the current is proportional to \( F_s \) as well. iii) A step is a one-dimensional object, and therefore it has a larger roughness than a two-dimensional surface. In the expression for the unstable current, the diffusion length gives the minimal distance between steps (in 2d) or kinks (in 1d) along a high-symmetry orientation. In 2d, steps are created by nucleation and growth, and \( \ell_D \) is generally given by an expression as \( \ell_D \approx (D/F)\gamma \) with the exponent \( \gamma \) depending on the details of the nucleation process. In 1d, the corresponding expression \( \ell_D = (D_s/F_s)\gamma^s \) should be compared to the distance between thermally excited kinks, and the smaller one (called \( \ell_d \)) be chosen. In most of our discussion we will assume that \( \ell_d \) is sufficiently large so that double or multiple kinks can be neglected.

The high symmetry in-plane orientations \([100]\) and \([110]\) for a step are fairly different in the mechanisms giving rise to a step-edge current. Along a \([100]\) segment, step diffusion takes place between nearest neighbours lattice sites at a rate \( D_s \), and the analogy with a one dimensional surface is appropriate. In particular, an asymmetry in the sticking rates to a kink determines a net current along the straight segments of the step, i.e. in the \( \hat{\textbf{D}} \) direction; when \( \theta \neq 0 \) this current does not vanish and it has a component along the slope \( \hat{\textbf{m}} \), which will be seen to destabilize the flat surface.

Conversely, along a \([110]\) orientation, diffusion is a two-steps process and it is very much slower, because it requires detachment from a high coordination site. As a first approximation, it may even be reasonable to assume that no detachment at all takes place. This does not prevent a nonzero step-edge current, for the following reason. Along the \([100]\) orientation, kinks are due to nucleation, or they must be thermally activated, because a kink increases the total length of the step. Along and close to the \([110]\) orientation, the total length of the step does not depend on the specific sequence of \([100]\) and \([010]\) terraces (see Fig. 3), and therefore the step is rough even at zero temperature. The path for going from the origin \( O \) to \( P \) is equivalent to a directed random walk, where the asymmetry \( p \) between the probabilities \( p_- = (1-p)/2 \) and \( p_+ = (1+p)/2 \) to go respectively in the \( \hat{x} \) and \( \hat{y} \) directions, is nothing but the tangent of the angle \( \beta = \pi/4 - \theta \) formed by the average orientation of the step with the \([110]\) direction. Since step diffusion does take place along \([100]\) and \([010]\) segments, each step segment longer than one lattice constant contributes to the \( \hat{x} \) and \( \hat{y} \) components of the step current. This implies that \( \tilde{j}_s \) is nonzero also for \( \theta = \pi/4 \); in this case \( \tilde{j}_s \) is exactly parallel to \( \hat{\textbf{m}} \) and it has a destabilizing character. In the following we will consider separately the cases of small \( \theta \) and \( \theta \) close to \( \pi/4 \), and afterwards we will write down a general expression for \( \tilde{j}_s \), valid for any value of the angle \( \theta \).

**FIG. 2.** Step profile, when its average orientation is close to \([110]\).

For the moment, we will suppose that the slope \( m = |\hat{\textbf{m}}| \) of the surface is larger than \( 1/\ell_D \), i.e. we are in the ‘vicinal’ regime. The step current \( \tilde{j}_s \) can always be written as the sum of \( \tilde{j}_s^0 \) and \( \tilde{j}_s^\perp \), where \( \tilde{j}_s^0 = (\tilde{j}_s \cdot \hat{\textbf{m}})\hat{\textbf{m}}/m^2 \) and \( \tilde{j}_s^\perp = (\tilde{j}_s \cdot \hat{\textbf{m}})\hat{\textbf{m}}/m^2 \). The vector \( \hat{\textbf{m}} \) is orthogonal to \( \hat{\textbf{m}} \).

If we are close to the \([100]\) orientation, the current \( \tilde{j}_s \) is simply \( \tilde{j}_s^\perp = j_{1d}(m_s)\hat{x} \), where \( j_{1d}(m_s) \) is the usual unstable current for a one-dimensional surface whose slope is \( m_s = \tan \theta = m_x/m_y \). For small \( \theta \), \( j_{1d} = a_s m_s \), with \( a_s = (F_s/\ell)k_\perp^2/2(\ell_k + \ell_d) \). By decomposing \( \tilde{j}_s^\perp \) along \( \hat{\textbf{m}} \) and \( \hat{\textbf{m}} \), we obtain

\[
\tilde{j}_{s0}^\perp = \frac{j_{1d}(m_s)}{m^2}[m_x\hat{\textbf{m}} - m_y\hat{\textbf{m}}].
\]
The uphill component is \((\vec{j}_{[100]} \cdot \vec{m}/m) \approx a_s m_s^2/m^2 > 0\), for small \(m_s\). The positive value of this component explains why step-edge current is enough to destabilize a flat, high symmetry surface. More details are given in Sec. III.

If we are close to the [110] orientation, as explained above, the current originates from entropic fluctuations around the straight step, which create segments along [100] and [010] directions. Each segment of length \(s\) contributes a local current proportional to \((s - 1)\). Therefore, the components \(j_x\) and \(j_y\) of the step current along the \(\hat{x}\) and \(\hat{y}\) axes are simply proportional to the probabilities (per step site) \(p^x_\perp\) and \(p^y_\perp\) to have a couple of consecutive step sites in the horizontal and vertical direction, respectively:

\[
\begin{align*}
\vec{j}_{[110]} &= \frac{F}{\ell^2} (p^x_\perp \hat{x} - p^y_\perp \hat{y}) = F \left( \frac{1 - p}{2} \right)^2 \hat{x} - F \left( \frac{1 + p}{2} \right)^2 \hat{y} .
\end{align*}
\]  

By using the relations \(p = \tan(\theta - \pi/4)\) and \(m_s = \tan \theta\), after some easy algebra we obtain

\[
\begin{align*}
\vec{j}_{[110]} &= \frac{F}{\sqrt{1 + m_s^2}} \left[ \frac{m_s}{1 + |m_s|} \vec{m} + \frac{1 - |m_s|^2}{(1 + |m_s|)^2} \vec{m}_\perp \right] .
\end{align*}
\]

The expressions (2) and (3) are valid close to the [100] and [110] orientations: they can be generalized to any value of \(m_s\), i.e. to any angle \(\theta\), by writing

\[
\vec{j}_s = A(\theta) \vec{m}/m + B(\theta) \vec{m}_\perp/m .
\]

Both \(A\) and \(B\) are periodic in \(\theta\), with period \(\pi/2\) (see Fig. 3). Their behaviours close to \(\theta = 0\) and \(\theta = \pi/4\) are derivable from \(\vec{j}_{[100]}\) (Eq. (3)) and \(\vec{j}_{[110]}\) (Eq. (4)). More precisely, \(A(\theta)\) is always nonnegative, it has a minimum for \(\theta = 0\) and a maximum for \(\theta = \pi/4\). The function \(B(\theta)\) vanishes at \(\theta = 0, \pi/4\), it has a positive slope in \(\theta = 0\) and a negative slope in \(\theta = \pi/4\). These properties are all that we need in the following, and they do not depend on the specific model assumed to derive Eqs. (2,4), because they are mainly due to symmetry considerations. For example, if multiple kinks are allowed when the step is close to the [100] orientation, \(A(\theta)\) is no more zero at \(\theta = 0\), but \(A(0) > 0\) and \(\theta = 0\) is still a minimum. Finally note that in contrast to the terrace current \(j_t\) (3), the step contribution (4) is independent of the surface slope \(m\), i.e. the step distance, in the vicinal regime (\(m\ell_D \gg 1\)).

![FIG. 3. Plots of \(A(\theta)\) and \(B(\theta)\), which are defined in Eq. (5).](image)

Before going on, let us generalize the expression of \(\vec{j}_s\) to any value of the surface slope \(\vec{m}\). In the limit \(m \ll \ell_D\), both \(\vec{j}_x\) and \(\vec{j}_y\) go to zero, because contributions coming from steps and terraces of opposite sign compensate. In a region of small slope, \(\vec{j}_{s1} \bigg|_{\text{small slope}} = \frac{N_+ - N_-}{N} \vec{j}_{s1} \bigg|_{\text{large slope}}\), where \(N_{\pm}\) is the number of positive and negative steps (for \(j_x\)) or terraces (for \(j_t\)) and \(N = N_+ + N_-\). Since \((N_+ - N_-)/N = m\ell_D\), in the small slope regime: \(\vec{j}_{s1} = A(\theta) \ell_D \vec{m}\) and \(\vec{j}_{s\perp} = B(\theta) \ell_D \vec{m}_\perp\). For a generic slope, we can write

\[
\begin{align*}
\vec{j}_{s1} &= A(\theta) g(m^2) \vec{m} \quad & \vec{j}_{s\perp} &= B(\theta) g(m^2) \vec{m}_\perp
\end{align*}
\]

where \(g(m^2) \to \ell_D\) for \(m\ell_D \ll 1\) and \(g(m^2) \to 1/m\) for \(m\ell_D \gg 1\). The simplest function interpolating between the two limiting values is \(g(m^2) = \ell_D/\sqrt{1 + m^2\ell_D^2}\), but its actual form does not need to be specified.

\[\text{Strictly speaking, a slope-dependence is maintained through the } \ell\text{-dependence in } \ell_{Dz}, \text{ but when } \ell \text{ is small, } \ell_{Dz} \text{ should be replaced by the distance between thermally activated kinks (see the main text).}\]
IV. STABILITY OF VICINAL SURFACES

Let us now perform a linear stability analysis of a growing vicinal surface of average slope \( \bar{m}_0 \). The local height is \( z(\vec{x}, t) = \bar{m}_0 \cdot \vec{x} + \epsilon(\vec{x}, t) \) and the local slope is \( \bar{m} = \bar{m}_0 + \nabla \epsilon \). The evolution, as determined by the step current, is given by the equation \( \partial_t z = -\nabla \cdot \vec{j}_s \). By using the general properties given above, for \( A \) and \( B \), and working in the special cases \( \theta_0 = 0 \) and \( \theta_0 = \pi/4 \), we obtain:

\[
\partial_t z = -\nabla \cdot \vec{j}_s = -g(m_0^2) [A(\theta_0) + B'(\theta_0)] \partial^2_{\perp} \epsilon - A(\theta_0)(\partial/\partial m_0)[m_0 g(m_0^2)] \partial_{\parallel}^2 \epsilon
\]

(7)

where \( \partial_{\perp} \) and \( \partial_{\parallel} \) are directional derivatives perpendicular and parallel to \( \bar{m} \) (i.e. parallel and perpendicular to the step). Thus the coefficient of \( \partial_{\parallel}^2 \) gives informations on step meandering, and that of \( \partial_{\perp}^2 \) on step bunching.

Since \( A(\theta) \geq 0 \) and \( (\partial/\partial m_0)[m_0 g(m_0^2)] > 0 \), the current \( \vec{j}_s \) (more precisely \( \vec{j}_s^\parallel \)) has always a destabilizing character against step bunching; if multikinks are not allowed along the \([100]\) orientation, along the \( \hat{x} \) in the two cases. For \( \theta_0 = 0 \) and \( \theta_0 = \pi/4 \), we obtain:

\[
\partial_t z = -\nabla \cdot \vec{j}_s = -g(m_0^2) |A(\theta_0) + B'(\theta_0)| \partial^2_{\perp} \epsilon \]

(7)

where \( \partial_{\perp} \) and \( \partial_{\parallel} \) are directional derivatives perpendicular and parallel to \( \bar{m} \) (i.e. parallel and perpendicular to the step). Thus the coefficient of \( \partial_{\parallel}^2 \) gives informations on step meandering, and that of \( \partial_{\perp}^2 \) on step bunching.

Our conclusion regarding the \([100]\) steps agrees with the analysis of Pierre-Louis et al. [14], while in the case of \([110]\) steps they find stability (instability) for large (small) values of the kink ES length \( \ell_k \). Our expression (7) is valid if adatoms are not allowed to turn around corners and this effectively sets \( \ell_k = \infty \). Its generalization to any \( \ell_k \) indeed shows that \( A(\pi/4) \) becomes a minimum (and the quantity \( [A(\pi/4) + B'(\pi/4)] \) gives, using Eq. (4), a negative result \((-F/\sqrt{2})\). Therefore, steps along the \([110]\) orientations are stabilized against step meandering.

Concerning step meandering, we must distinguish between \( \theta_0 = 0 \) and \( \theta_0 = \pi/4 \), because \( B'(\theta_0) \) has opposite signs in the two cases. For \( \theta_0 = 0 \) the derivative is positive and therefore steps along the \([100]\) orientation are destabilized. On the contrary, the evaluation of \( [A(\pi/4) + B'(\pi/4)] \) gives, using Eq. (4), a negative result \((-F/\sqrt{2})\). Therefore, steps along the \([110]\) orientations are stabilized against step meandering.

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Ramana Murty and Cooper [15] have performed Monte Carlo simulations of a vicinal surface, with steps along the \([100]\) axis. Step meandering is indeed observed, even if the terrace current \( \vec{j}_t \) is absent. Conversely, no step bunching seems to occur, suggesting that their simulations correspond to \( A(0) = 0 \).

V. STABILITY OF SINGULAR SURFACES

The analysis of a high-symmetry surface is complicated by the non-analytic behavior of \( \vec{j}_s \) and \( \vec{j}_t \) in \( \bar{m} = 0 \). In the small slope regime \( (m \ll 1/\ell_D) \), \( \vec{j}_s^\parallel \) and \( \vec{j}_s^\perp \) become (see Eq. (1))

\[
\vec{j}_s^\parallel = \ell_D A(\theta) \bar{m} \quad \vec{j}_s^\perp = \ell_D B(\theta) \bar{m}_\perp \, .
\]

(8)

It should be stressed that the singularity is physically justified, as we now try to argue. Close to an extremum of the profile, \( z(x, y) = z_0 + (c_1/2)x^2 + (c_2/2)y^2 + c_3 xy, \bar{m} = (c_1 x + c_3 y, c_2 y + c_3 x) \) and \( m_n = (c_1 r + c_3)/|c_2 + c_3 r| \), where \( r = x/y \). The prefactors \( A \) and \( B \) are therefore manifestly non-analytic functions at \( x = y = 0 \). The reason is that close to an extremum, steps are closed lines and as the top (or the bottom) of the profile is approached, the step orientation is no more defined. The angular dependence of \( A \) and \( B \) also implies that the evolution equation does not become linear in the small-slope regime, and hence arbitrary profiles cannot be treated as superpositions of harmonic ones.

The problem of non-analyticity does not appear when we consider a one-dimensional profile, i.e. a profile varying only in one direction (for example, \( z = z(x, t) \)), because the prefactors \( A \) and \( B \) are now constants\(^3\). This implies that the divergence of the current is easily evaluated:

\[
\partial_t z = -\nabla \cdot (\vec{j}_s^\parallel + \vec{j}_t) = -\nabla \cdot (\ell_D A(\theta) \bar{m} + \ell_D B(\theta) \bar{m}_\perp + a(\theta) \bar{m}) = -[\ell_D A(\theta_0) + a(\theta_0)] \nabla^2 z \equiv -\nu(\theta_0) \nabla^2 z \, .
\]

(9)

The component of the step current parallel to the step \( (\vec{j}_s^\parallel) \) does not contribute, because \( \nabla \cdot \bar{m}_\perp \equiv 0 \), while the component parallel to the slope \( (\vec{j}_s^\perp) \) destabilizes the flat surface, analogously to \( \vec{j}_t \).

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\(^3\)The angle \( \theta \) may indeed take the values \( \theta_0 \) and \( (\theta_0 + \pi) \), corresponding to \( \tan \theta = \pm |m_s| \), but because of the \( \pi/2 \) periodicity, \( A(\theta_0) = A(\theta_0 + \pi) \). This is no more true for the \((111)\) surface of a cubic crystal.
The instability gives rise to pyramid-shape mounds, whose orientations $\theta_i$ should be the most unstable ones, i.e. correspond to the maxima of $\nu(\theta)$. In this respect, the step current favours the orientations forming $45^\circ$ with the crystallographic axes, while the anisotropy induced by the terrace current depends on the microscopic details of the sticking processes. Since the presence of more kinks along the step should favour the descent of the adatom, it is reasonable to think that $\ell_{ES}(\theta)$ is maximum in $\theta = 0, \pi/2$ [22], and therefore the two contributions to $\nu(\theta)$ should compete. Close to $\theta = 0$, we have

$$\nu(\theta) = a_s m_s \frac{m_x}{m} + a(\theta) = a_s \theta^2 + a(\theta) .$$  \hspace{1cm} (10)

By using expression (2) for $a(\theta)$ and the expression of $a_s$ given above Eq. (5), we obtain

$$\nu'(0) = F \left[ \frac{\ell_{ES}^3 \nu''(0)}{2(\ell_{ES} + \ell_D)^2} + \frac{\ell_k \ell_d^2}{\ell_k + \ell_d} \right] .$$  \hspace{1cm} (11)

Mounds will align along the crystallographic axes if $\nu(\theta)$ has a maximum in $\theta = 0$, i.e. if $\nu''(0) < 0$. It is apparent that for a sufficiently large ES effect at steps this condition is not fulfilled, because the anisotropic character of $\vec{j}_i$ disappears. On the other hand, $\ell_{ES}$ should also not be too small, otherwise $\vec{j}_i$ itself would be negligible. Therefore $\nu(\theta)$ will have maxima in $\theta = 0, \pi/2$ only if several conditions are simultaneously satisfied: i) $\ell_{ES}(0)$ must be negative; ii) $\ell_{ES}/\ell_D$ should be neither too large nor too small; iii) $\ell_k/\ell_d$ should be small.

Refs. [13] and [22] recently reported simulations on high-symmetry surfaces, taking into account step-edge diffusion. In both cases, if kink barriers are present the mound sides align along [110] and equivalent axes. Since in [13] there are no ES barriers at steps and in [22] the barriers are infinite and therefore isotropic, mound orientation is determined only by $\vec{j}_s$, in agreement with our picture.

VI. DISCUSSION

Some aspects of the subject considered in the present paper have not been sufficiently clarified and they deserve further analysis. First of all, the non-analytic behaviour of the surface current remains problematic, because it implies that the continuum evolution equation $\partial_z z + \nabla \cdot \vec{j} = 0$ is not defined at $\vec{m} = 0$. As we have argued above, this non-analyticity is an inescapable consequence of the persistence of crystal anisotropy in the ‘linear’ regime of the instability; if it could be removed, e.g. through a more careful treatment of the interpolation between vicinal and singular surfaces, this would also imply that mounds are initially isotropic and develop their anisotropic shapes only in the nonlinear regime. It is however also conceivable that, as in the case of equilibrium surface relaxation below the roughening temperature [23], the appearance of a singularity in the continuum evolution equation carries a real physical message: That the surface is not well described by a smooth function $z(\vec{r}, t)$ near its maxima and minima.

A second important aspect concerns vicinal surfaces. We have seen that for [110] steps $\vec{j}_s$ has a stabilizing character with respect to step meandering and a destabilizing character with respect to step bunching. It would be interesting to evaluate quantitatively these effects and to compare them with the opposing effects of the terrace current. This comparison has been done for step meandering [14], and it seems that the effect of $\vec{j}_s$ may dominate $\vec{j}_i$. At any rate the predicted step bunching instability should be clearly visible in simulations of models which have no ES barriers but only asymmetric sticking at kinks [13]. Finally, in this work we have not addressed the effects of crystal anisotropy on the smoothing terms in the continuum evolution equation, which are crucial in determining the actual length scale of the instability [8][21]. Under far from equilibrium conditions, the dominant smoothing mechanism is believed to be due to random nucleation [18], which is manifestly isotropic; the anisotropy of the equilibrium step free energies will however be felt if detachment from steps becomes significant.

\[\text{In simulations of solid-on-solid models the step edge barriers are often implemented such as to reduce the probability for adatoms to approach steps, rather than to descend from them [2][24]. In this case the barrier at a [110] step may in fact (slightly) exceed that of the close packed [100] step [2].}\]
VII. CONCLUSIONS

In this paper we have studied the different contributions to the surface current on a (100)-surface, which depend only on the slope \( \vec{m} \). Such contributions come from biased surface diffusion, both on terraces (\( \vec{j}_t \)) and along steps (\( \vec{j}_s \)), where the bias mechanism is an Ehrlich-Schwoebel barrier at steps and kinks, respectively.

The expressions of \( \vec{j}_{s,t} \) are relevant in two respects: they determine the linear stability of the flat surface and – in the case of unstable growth – they also determine the shape and the orientation of the emerging structure.

The terrace current is parallel to the slope, while the step current has components parallel and perpendicular to the slope, because step diffusion takes place along the [100]- and [010]-segments that constitute the step.

A first important result is that the anisotropic character of \( \vec{j}_s \) and \( \vec{j}_t \) persists in the small slope regime \( \vec{m} \to 0 \): this means that they are both non-analytic at \( \vec{m} = 0 \) and consequently this does not allow a complete description of the evolution of a high-symmetry surface.

Concerning the stability of a singular surface, \( \vec{j}_s \) is found to be destabilizing because it has a positive component in the direction of \( \vec{m} \). The most unstable orientations form angles of 45° with the crystallographic axes, while \( \vec{j}_t \) is usually (but not always, see footnote 4) thought to select the \( \hat{x}, \hat{y} \) axes. If a competition exists \( \vec{j}_s \) should prevail (see discussion below Eq. (11)).

The stability of a vicinal surface is more complex. The terrace current \( \vec{j}_t \) is known to stabilize against step bunching and to destabilize towards step meandering, whatever is the orientation of the surface. Surprisingly, the step current is instead found to generally favor step bunching (along the crystallographic axes \( \vec{j}_s \) has no effect if multiple kinks are not present). Finally the effect of \( \vec{j}_s \) on step meandering strongly depends on the surface orientation and the strength of the ES effect at kinks: [100] steps are destabilized, while [110] steps may be stabilized if the kink ES-barrier is not too weak.

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APPENDIX A: LIST OF SYMBOLS

| Symbol | Description |
|--------|-------------|
| \( z(\vec{x}, t) \) | local height of the surface at point \( \vec{x} \) and time \( t \) |
| \( \vec{m} \) | local slope |
| \( \vec{m}_\perp = (-m_y, m_x) \) | vector orthogonal to the slope |
| \( \theta = \arctan \left( \frac{m_x}{m_y} \right) \) | angle between the average orientation of a step and the \( \hat{x} \) axis |
| \( m = |\vec{m}| = |\vec{m}_\perp| \) | modulus of the local slope |
| \( \ell = 1/m \) | terrace size |
| \( \vec{m}_0 \) | average slope of a vicinal surface |
| \( \vec{j}_t \) | terrace current |
| \( \vec{j}_s \) | step current |
| \( \vec{j}_s^\parallel \) | step current parallel to the slope \( \vec{m} \) |
| \( \vec{j}_s^\perp \) | step current perpendicular to the slope \( \vec{m} \) |
| \( F \) | intensity of the external flux of particles |
| \( F_s = F\ell \) | rate of particles arriving to the step |
| \( D \) | diffusion constant on the terrace |
| \( D_s \) | diffusion constant along a step |
| \( D' \) | attachment rate of an adatom on a terrace to the ascending step |
| \( D'' \) | attachment rate of an adatom on a terrace to the descending step |
| \( D'_s \) | attachment rate of an adatom on a step to the ascending kink |
| \( D''_s \) | attachment rate of an adatom on a step to the descending kink |
| \( \ell_{ES} \) | ES length on a terrace |
| \( \ell_k \) | ES length along a step |
| \( \ell_D \) | diffusion length on a terrace |
| \( \ell_{D_s} \) | diffusion length along a step |
| \( \ell_d \) | the minimum between \( \ell_{D_s} \) and the distance between thermally activated kinks |
In the directed random walk picture of a step close to the [110] orientation, probabilities that the step goes in the $\hat{x}$ ($p_-$) and $\hat{y}$ ($p_+$) directions

$$p = p_+ - p_-$$

asymmetry in the probabilities $p_\pm$