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Surface currents and slope selection in crystal growth

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Abstract

We face the problem of determining the slope dependent current during the epitaxial growth process of a crystal surface. This current is proportional to $\delta = p_+ - p_-$, where $p_\pm$ are the probabilities for an atom landing on a terrace to attach to the ascending (p_+) or descending (p_-) step. If the landing probability is spatially uniform, the current is proved to be proportional to the average (signed) distance traveled by an adatom before incorporation in the growing surface. The phenomenon of slope selection is determined by the vanishing of the asymmetry δ. We apply our results to the case of atoms feeling step edge barriers and downward funnelling, or step edge barriers and steering. In the general case, it is not correct to consider the slope dependent current j as a sum of separate contributions due to different mechanisms. ***To cite this article: P. Politi, C. R. Physique 7 (2006).***

Résumé

Courants de surface et sélection de la pente en croissance cristalline. La croissance épitaxiale d'une surface cristalline peut être caractérisée par un courant de surface J, dont la partie j qui dépend de la pente est étudiée. Celle-ci est proportionnelle à $\delta = p_+ - p_-$, où $p_\pm$ sont les probabilités qu'un atome déposé sur une terrasse se colle à la marche montante (p_+) ou descendante (p_-). Si la probabilité spatiale d'atterrissage est uniforme, le courant est aussi proportionnel à la distance moyenne (avec signe) parcourue par chaque atome. Le phénomène de la sélection de la pente est déterminé par la condition $\delta = 0$. Les résultats ainsi obtenus sont appliqués aux cas barrière de marche plus downward funnelling et barrière de marche plus braquage (steering). Dans le cas général, le courant j ne peut pas être considéré comme la somme de contributions séparés dues aux différents mécanismes. ***Pour citer cet article : P. Politi, C. R. Physique 7 (2006).***

Keywords: Crystal growth; Surface current; Diffusion

Mots-clés : Croissance cristalline ; Courant de surface ; Diffusion

1. Introduction

Molecular Beam Epitaxy is a well known and widespread technique to grow layers of metal and semiconductor crystals. The growth process of a high symmetry crystal surface can be described through a ballistic flux of particles

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impinging on the growing surface and the following thermal diffusion of adatoms, which finally attach to a preexistent step or nucleate new terraces [1–3].

One crucial aspect of the growth process is its possible unstable character, due to deterministic mechanisms which prevent the growing surface remaining flat. In this Note we focus on mound formation and we will discuss its description via a mesoscopic surface current J, which enters in the evolution equation $\partial_t z = Ft - \partial_x J$, where F is the flux of incoming particles and z is the local height. Experiments show that dynamics can produce coarsening, with the formation of mound facets of constant slope m^* [4].

In the theoretical seminal papers [5,6] the cause of mound formation was identified in the existence of step edge barriers, i.e., an additional barrier hindering the descent of steps. Since then, many efforts [2] have been devoted to the formalization of this idea and to the derivation of quantitative predictions for the evolution of mounds. These efforts have often combined phenomenological approaches with attempts of a rigorous derivation of J starting from the microscopic dynamics of adatoms.

The main part of the current J is the so-called slope dependent current $j(m)$, depending on the local slope m of the surface. J also contains terms depending on higher order derivatives, but the rising of the instability and the possible formation of mounds with a constant slope m^* only depend on $j(m)$. Therefore, it is natural that special attention has been devoted to its determination. Analytical approaches have followed two mainstreams: coarse-graining procedures [7–9] to pass from step-dynamics to mesoscopic dynamics (see the next section) and the evaluation of $j(m)$ through the average (signed) distance [10,11] walked by adatoms before being incorporated in the growing crystal (see Section 5).

A recent paper by Li and Evans [12] has renewed the interest on the slope dependent current. Authors claimed that standard phenomenological continuum theories are inappropriate to describe mound slope selection. Afterwards, some of their claims have been corrected [13], but their work has shown that $j(m)$ should be evaluated with great care. For these reasons, in the following we reconsider the problem, giving a formulation as general as possible for the slope dependent current and for the evaluation of the selected slope m^*, determined by the condition $j(m^*) = 0$. We prove that the current $j(m)$ is proportional to the asymmetry $\delta(L) = p_+(L) - p_-(L)$ between the probabilities for an atom deposited on a terrace of size L to be incorporated into the upper (p_+) and lower (p_-) steps. This proof does not refer to any specific microscopic mechanism. An important by-product is that, in general, $j(m)$ can not be considered as a sum of contributions due to separate mechanisms. Finally, we also discuss the alternative formulation for the current in terms of the averaged (signed) distance walked by adatoms before incorporation.

2. The current

In the case of conserved growth (no desorption, no overhangs), a useful concept to study the dynamics of the surface is the mesoscopic current J, entering via the evolution equation $\partial_t z = Ft - \partial_x J$ for the local height z. It is worth stressing that the dynamics of the surface, and therefore the current J, are determined by the dynamics of steps: adatoms enter only through their attachment (and detachment) rate to steps.

A piece of surface of slope m looks differently, according to the value of the slope. For large m (Fig. 1(a)) we have a sequence of all uphill or downhill terraces of size $L \simeq 1/|m|$. For small m (Fig. 1(b)) we have a mix of different types of terraces of size $L \simeq \ell_D$, where ℓ_D is the nucleation length [2]: in this case, the average slope is determined by

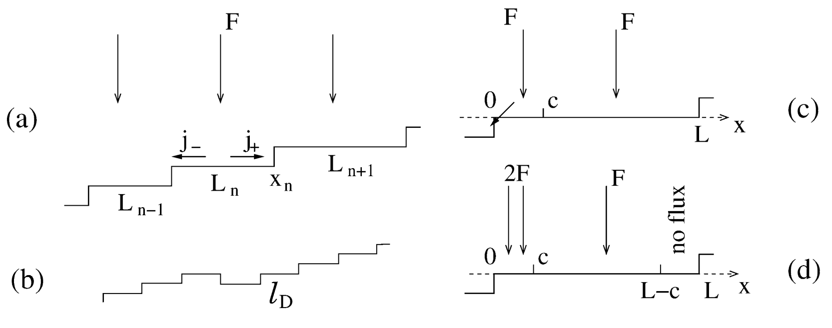

Fig. 1. (a,b) Schematic of a growing one-dimensional surface at (a) large and (b) small slope (Fig. 1(b) has been shrunk). (c, d) Coordinates for the models studied in Section 3: (c) Downward Funnelling and Ehrlich–Schwoebel effect; (d) steering and Ehrlich–Schwoebel effect.

uncompensated uphill and downhill terraces. The two pieces of surface look different because nucleation on terraces prevent them being larger than ℓ_D.

The analytical determination of the current J starts from the large slope case (see Fig. 1(a)). In a one-dimensional picture, the flux FL of atoms landing on the terrace splits in two currents $j_\pm(L) = FLp_\pm(L)$, where $p_\pm(L)$ are the probabilities that an atom attaches to the ascending (p_+) or descending (p_-) step. The velocity of the nth step is simply equal to $[j_-(L_{n+1}) + j_+(L_n)]$. The sum $(p_+ + p_-) = 1$, while $\delta = (p_+ - p_-)$ defines the asymmetry δ.

Following the method introduced in [7], the displacement of step n during the deposition of one monolayer can be approximated as

$$x_n(t + 1/F) - x_n(t) = -\frac{1}{F}\big[j_-(L'') + j_+(L')\big] \tag{1}$$

where $L' = \frac{1}{2}(L_n + L_{n-1})$ and $L'' = \frac{1}{2}(L_n + L_{n+1})$. It is useful to sum the quantity L_n to both sides, so as to have

$$x_n(t + 1/F) - x_n(t) + L_n = -\frac{1}{2}\big[L'' + L' - 2L_n + L'\delta(L') - L''\delta(L'')\big] \tag{2}$$

The quantity on the left can be approximated as $(-1/mF)\partial_t z + (1/m) = (1/mF)\partial_x J$, where $m = 1/L$ is the slope. The quantity on the right can be worked out using the relations [7][1]

$$L'' + L' - 2L_n \approx \frac{1}{2}\big[L^2\partial_{xx}L + L(\partial_x L)^2\big] \tag{3}$$

$$L'\delta(L') - L''\delta(L'') \approx (L' - L'')\partial_L\big[L\delta(L)\big] \approx L\partial_x L\partial_L\big[L\delta(L)\big] \tag{4}$$

Finally, we get

$$\frac{1}{mF}\partial_x J = \frac{1}{m}\partial_x\left[-\frac{L\partial_x L}{4} + \frac{L\delta(L)}{2}\right] \tag{5}$$

so that the total current J comes out to be

$$J = \frac{1}{2}FL\delta(L) - \frac{1}{4}FL\partial_x L \tag{6}$$

If we use the slope m, the second term takes the form $\frac{1}{4}F(\partial_x m)/m^3$, which was already found in [7,8]. In the following we will focus on the slope dependent part:

$$j = \frac{1}{2}FL\delta(L) \tag{7}$$

We stress that Eq. (7) is the most general form, not depending on any assumption on the microscopic processes occurring at the surface. It is valid for large slope, $m > 1/\ell_D$ (Fig. 1(a)). In the opposite limit, $m < 1/\ell_D$ (Fig. 1(b)), $j(m)$ is linear [7] in m, as also expected from symmetry reasons for a slope dependent current: $j(m) = j(1/\ell_D)\ell_D m$, where $j(1/\ell_D) = \frac{1}{2}F\ell_D\delta(\ell_D)$ is evaluated according to (7).

The condition of instability of the flat surface reads $j'(0) > 0$, i.e., $j(1/\ell_D) > 0$, as trivially shown by a linear stability analysis.[2] Finally, if mounds develop facets with constant slope m^*, the current must vanish on it: $j(m^*) = 0$.

3. Downward funnelling, step edge barriers and steering

Let us now consider the following model (see Fig. 1(c)). Atoms deposited within a distance c from the descending step are incorporated in it (downward funnelling, DF), while atoms deposited in the remaining $(L - c)$ portion of the terrace diffuse freely, feeling an additional (Ehrlich–Schwoebel, ES) barrier at the descending step. Atoms deposited in the c region give a trivial contribution Fc to j_-. As for the others, we must solve the diffusion equation $F + D\rho''(x) = 0$ for $c < x < L$ and $\rho''(x) = 0$ for $0 < x < c$, with boundary conditions $\rho'(0) = \rho(0)/\ell_{ES}$, $\rho(L) = 0$,

[1] The two expressions on the left are expanded at the lowest orders.

[2] The equation $\partial_t z = -j(1/\ell_D)\ell_D\partial_{xx}z$ is solved by $z(x,t) = \exp(iqx + \omega t)$ with $\omega(q) = j(1/\ell_D)\ell_D q^2$.

and $\rho(x)$, $\rho'(x)$ continuous in $x = c$. The quantity $\ell_{ES} \geqslant 0$ is the well known Ehrlich–Schwoebel length and measures the additional barrier felt by an adatom in the sticking process to the descending step. It is straightforward to get

$$\rho'(0) = \frac{F}{2D}\frac{(L-c)^2}{L+\ell_{ES}}, \qquad \rho'(L) = -\frac{F}{2D}\frac{(L-c)(L+c+2\ell_{ES})}{L+\ell_{ES}} \tag{8}$$

The contributions of atoms deposited in the $(L-c)$ region to the currents $j_\pm$ are $D|\rho'(L)|$ and $D\rho'(0)$, respectively. Therefore

$$j_- = Fc + \frac{F}{2}\frac{(L-c)^2}{L+\ell_{ES}} \equiv FLp_-(L), \qquad j_+ = \frac{F}{2}\frac{(L-c)(L+c+2\ell_{ES})}{L+\ell_{ES}} \equiv FLp_+(L) \tag{9}$$

Their sum gives the total flux of particles arriving on the terrace: $j_+ + j_- = FL$, while their (semi-)difference gives the current $j = \frac{1}{2}FL\delta(L) = \frac{1}{2}(j_+ - j_-)$, i.e.,

$$j = \frac{F}{2}\frac{\ell_{ES}(L-2c) - c^2}{L+\ell_{ES}} = \frac{F}{2}\frac{\ell_{ES}(1-2mc) - mc^2}{1+m\ell_{ES}} \tag{10}$$

If $c = 0$, we find the well known result $j = \frac{1}{2}FL\ell_{ES}/(L+\ell_{ES}) = \frac{1}{2}F\ell_{ES}/(1+m\ell_{ES})$.

Eq. (10) shows that it is not generally possible to write j as a sum of two separate contributions, $j = j_{ES} + j_{DF}$, due to the Ehrlich–Schwoebel (ES) effect and to the downward funnelling (DF), respectively. In fact, it is instructive to consider the two limits $\ell_{ES} = 0$ and $\ell_{ES} = \infty$. For $\ell_{ES} = \infty$, $j_- = Fc$ and $j_+ = F(L-c)$, i.e., j_- is entirely due to downward funnelling and j_+ to the ES effect. It seems natural to write $j = j_{DF} + j_{ES}$, with $j_{DF} = -\frac{1}{2}Fc$ and $j_{ES} = \frac{1}{2}F(L-c)$. For $\ell_{ES} = 0$ we are induced to assume $j = j_{DF} = -Fc^2/2L$. Therefore, if we want to write $j = j_{DF} + j_{ES}$, the expression for j_{DF} depends on ℓ_{ES}: it is constant (not depending on m) for large barriers and it is linear in m for weak barriers. The conclusion is that $j(L)$ should be handled as a whole.

Let us now consider a situation where the flux of particles on the terrace is not uniform, see Fig. 1(d). Because of steering effects [14], we assume that all atoms destined to land within distance c from the ascending step are steered and finally land in the c region close to the descending step (of the upper terrace). So, this region undergoes an effective flux $2F$. Working out calculations[3] similar to those made here above, we get the result

$$j = \frac{F}{2}\frac{\ell_{ES}L - 2c(L-c)}{L+\ell_{ES}} = \frac{F}{2}\frac{2c^2m + \ell_{ES} - 2c}{1+m\ell_{ES}} \tag{11}$$

This current is always positive if $\ell_{ES} > 2c$, or it changes sign in $m^* = (1-\ell_{ES}/2c)/c$, if $\ell_{ES} < 2c$ (see Fig. 2(b), dashed line). However, in the latter case $j'(m^*) > 0$, which implies instability of the slope m^*.[4] So, in this model there is no slope selection: either the surface is always unstable or it is metastable [15].

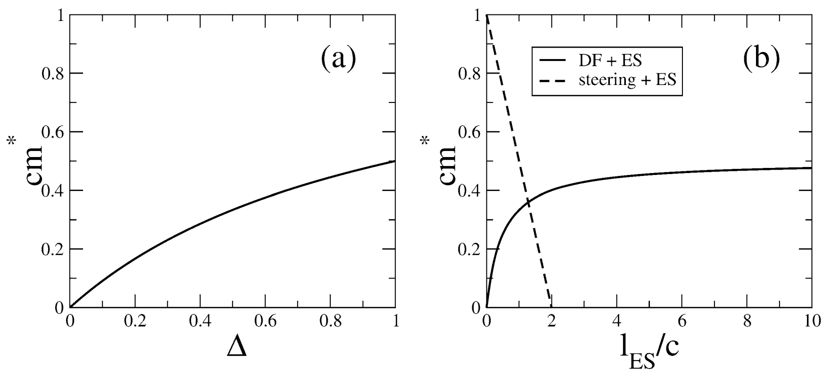

Fig. 2. (a) The selected slope cm^* as a function of Δ, Eqs. (12), (13). (b) Full line: the selected slope cm^* as a function of ℓ_{ES}/c (DF + ES model, Eq. (14)). Dashed line: the slope m^* such that $j(m^*) = 0$ for the steering + ES model (Section 3). There is no slope selection in this case, because $j'(m^*) > 0$.

[3] The use of a continuum diffusion equation at atomic scales may be questionable. However, if a very precise numerical output is not required, its use for evaluating the asymmetry δ is appropriate.

[4] If $J(m^*) = 0$, the slope m^* is stable (unstable) if $J'(m^*)$ is negative (positive).

4. Slope selection

First, let us apply the condition $j(m) = 0$ to a model studied by Li and Evans [12]:

$$j_+ = F(L-c)P_+, \qquad j_- = Fc + F(L-c)P_- \tag{12}$$

If $P_\pm$ are the probabilities for atoms deposited in the $(L-c)$ region to attach to the step, this model is similar to what we studied in the previous section. For constant $P_\pm$, the condition $j_+ = j_-$, i.e., $\delta = 0$, gives the following expression for the selected slope m^*,

$$m^* = \frac{\Delta}{c(1+\Delta)} \tag{13}$$

where $\Delta = P_+ - P_-$. Note that Δ is not the total asymmetry δ, which vanishes for $m = m^*$, but the asymmetry for atoms deposited in the $(L-c)$ portion of the terrace only. In Fig. 2(a) we report $cm^*(\Delta)$. Eq. (13) perfectly matches the numerical results given in [12].

Let us now turn back to Eq. (10). In this case, we get

$$m^* = \frac{\ell_{\mathrm{ES}}}{c\,(c + 2\ell_{\mathrm{ES}})} \tag{14}$$

In Fig. 2(b) (full line) we plot cm^* as a function of ℓ_{ES}/c. For small and large ℓ_{ES}/c, $m^* = \ell_{\mathrm{ES}}/c^2$ and $m^* = 1/(2c)$, respectively. Therefore, for large ES barrier the selected slope corresponds to a terrace size equal to $2c$, as expected by a trivial compensation of DF and ES effects.

The use of (10), as well as of (12), to find m^* is limited by the constraint $m^* > 1/\ell_D$. For Eq. (10) this means $\ell_{\mathrm{ES}} > c^2/(\ell_D - 2c) \approx c^2/\ell_D$; in the opposite limit, $\ell_{\mathrm{ES}} < c^2/\ell_D$, $j(1/\ell_D) < 0$ and no selected slope exists: this happens because the DF effect is so strong to induce a stabilizing (downhill) current at all slopes.

5. Another expression for the current

Eq. (7), $j(L) = \frac{1}{2}FL\delta(L)$, is the most general expression for the slope dependent current on a region of (large) slope $m = 1/L$. The quantity $\delta(L) = (p_+ - p_-)$ measures the asymmetry between the sticking probabilities to the upper and lower steps. The flux F is usually assumed to be spatially uniform, apart from fluctuations. However, as anticipated in the previous Section, because of atom-substrate interactions, steering effects may occur and atoms are no longer deposited uniformly on the terrace. In spite of this, the expression $j(L) = \frac{1}{2}FL\delta(L)$ still continues to be correct. In the Introduction we mentioned a possible different evaluation of $j(m)$, through the average signed distance d walked by an adatom, from the deposition to the incorporation site. In the following we are proving the equivalence of the two expressions, if the flux is uniform. If it is not uniform, the average distance d is not an appropriate quantity to determine j.

The convention is to take d positive if the atom attaches to the ascending step (see Fig. 1):

$$d = \frac{1}{L}\int_0^L \mathrm{d}x\,\big[(L-x)\tilde{p}_+(x) - x\tilde{p}_-(x)\big] = \int_0^L \mathrm{d}x\,\tilde{p}_+(x) - \frac{1}{L}\int_0^L \mathrm{d}x\big(\tilde{p}_+(x) + \tilde{p}_-(x)\big)x \tag{15}$$

with $\tilde{p}_\pm(x)$ being the probabilities that an adatom deposited in x attaches in $x = 0$ ($\tilde{p}_-(x)$) and in $x = L$ ($\tilde{p}_+(x)$). Since $p_\pm = (1/L)\int_0^L \mathrm{d}x\,\tilde{p}_\pm(x)$ and $(\tilde{p}_+(x) + \tilde{p}_-(x)) = 1$, we get $d = Lp_+ - \frac{1}{2}L = \frac{1}{2}L\delta(L)$, so that

$$j(L) = \frac{1}{2}FL\delta(L) = Fd \tag{16}$$

In a Kinetic Monte Carlo simulation, we can easily implement the above expression and write

$$j_{\mathrm{KMC}} = F\frac{N_r - N_l}{N_a} \tag{17}$$

where $N_{r,l}$ are the total hops in the uphill and downhill direction, and N_a is the total number of deposited atoms. Formula (17) was firstly introduced in [10] and has the merit to be valid for a surface of any slope. At author's knowledge, a rigorous derivation and a comparison with the mesoscopic current were missing.

It is worth stressing that Eqs. (16), (17) are no more applicable if the landing probability is not spatially uniform. In this case, $j(L)$ is no more proportional to d, as shown by a trivial example: no downward funnelling ($c = 0$) and infinite ES barrier, $\ell_{\text{ES}} = \infty$. The resulting current $j = \frac{1}{2}FL$ can be written as $j = Fd$ if the flux is uniform: in that case, $d = \frac{1}{2}L$. Different spatial distributions of the landing atoms modify d and therefore the expression $j = Fd$, but do not modify the correct expression $j = \frac{1}{2}FL\delta$, because the step dynamics would be unchanged.

6. Final remarks

In this Note we mainly focused on the slope dependent current and the slope selection mechanism. We have shown that a correct derivation of this current is possible and its general expression has been found. The condition $j(m^*) = 0$ determines the selected slope. More generally, the shape $z_s(x)$ (or $m_s(x) = z_s'(x)$) of stationary solutions depends on the vanishing of the full current J in all the points, $J(m_s(x)) \equiv 0$. It is worth noting that the conditions $j(m^*) = 0$ and $J(m_s(x)) \equiv 0$, when applied to a discrete or discrete-continuous model, are valid only averaging J on time scales not smaller than $1/F$ [12].

With regard to additional terms in the current, our derivation in Section 2 shows that a symmetry breaking term, having the form $J = \partial_x A(m^2)$, appears naturally when we coarsen step dynamics. This term is the only term surviving in a plain vicinal surface without any additional microscopic mechanism (no step edge barriers, no nucleation, no thermal detachment, no downward funnelling): so, in this sense, it should be considered the most fundamental one. Finally, J should also contain at least a Mullins-like term, $J \sim \partial_{xx} m$, which may be due to several mechanisms [16].

However, even if the rigour in the derivation of the mesoscopic current has improved in the course of time, the evolution equation $\partial_t z = F - \nabla \cdot \boldsymbol{j}$ is not much more than a phenomenological equation, specially in two dimensions where additional problems linked to step edge diffusion exist. The dynamics of a truly vicinal surface, which can be studied with much more rigour [17], shows that the full nonlinear equations governing the growth process of a real system are far more complicated.

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