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Kinetic model for step flow growth of [100] steps

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A kinetic model for the step flow growth of [100] steps is derived. This model extends the implicit close-to-equilibrium assumption in the classical Burton-Cabrera-Frank theory to out-of-equilibrium growth regimes common in molecular beam epitaxy. In equilibrium, the kink density coincides with Burton, Cabrera, and Frank [Philos. Trans. R. Soc. London, Ser. A **243**, 299 (1951)] but in a planar steady state solution for a periodic sequence of steps the kink density derivates and we analyze its scaling behavior with the step Peclet number. Furthermore, we observe a discontinuity of adatoms on the steps that might give a new explaination for step bunching.

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Many modern technologies depend on the ability to grow thin crystalline films in a nearly perfect manner. Precise control of the growth and thus of the properties of the crystalline films therefore requires the detailed knowledge of atomic processes at the surface. The understanding of the elementary physical phenomena and their incorporation into a continuum model, which is able to reach time and length scales of interest for device applications, is therefore of utmost importance.

Due to the discrete translational symmetry imposed by the crystal lattice, atomic steps exist at the surface of a crystal, which separate terraces that differ in height by a single lattice spacing. At sufficiently low temperatures, these steps are suitable for a mesocopic description of the surface morphology. The concept to describe the growth of a crystal by the movements of existing steps, their nucleation, or annihilation was presented by Burton, Cabrera, and Frank (BCF) [1] and serves today as a standard model for a discrete-continuum description of out-of-equilibrium growth. With the advent of powerful numerical tools for the solution of these problems [2–4], step dynamics have become an attractive alternative to atomistic modeling approaches to describe out-of-equilibrium growth. However, in the classical BCF theory, the formulation of the mass currents that leave the terraces via the steps, contains the implicit assumption that the steps are close to equilibrium. There have been recent attempts to formulate kinetic models that due not require this assumption [5]. In this Brief Report we derive a terrace-edge-kink model from kinetics, which generalizes this model by allowing for the kink Ehrlich-Schwoebel barrier [6,7], nucleation on the level of edge adatoms [8,9], the direct incorporation of adatoms into kinks and lattice sites [10], and which is not restricted to very small kink densities. The theory is formulated in terms of adatom densities on terraces, edge-adatom densities on steps, which are not considered as part of the crystal, and a density of kinks along the steps. In deriving the model we neglect the influence of second-nearest neighbors on the step structure as well as overhangs and kinks of multiple height. The model is related to the mean-field theory of habit change phenomena during growth of two-dimensional islands [11] and is valid for steps close to the [100] direction.

The adatom diffusion equation on a terrace is

$$\partial_t \rho - D_T \nabla^2 \rho = -\tau^{-1}\rho + F - M, \tag{1}$$

in which ρ is the adatom density, D_T is the adatom diffusion coefficient on a terrace, F is the deposition flux rate, M is the loss due to nucleation of adatom islands, and τ^{-1} is the desorption rate. The boundary conditions for ρ at the steps are given by the flux to the steps

$$v\rho_+ + D_T \mathbf{n} \cdot \nabla \rho_+ = -f_+, \tag{2}$$

$$v\rho_- + D_T \mathbf{n} \cdot \nabla \rho_- = f_-, \tag{3}$$

where v is the velocity of the step in the direction of the normal $\mathbf{n}$, and ρ_+ and ρ_- are the values of ρ at the step from the upper and the lower terrace, respectively. The terms $v\rho_\pm$ describe the convection of adatoms due to the motion of the step and are needed for mass conservation. The fluxes $f_\pm$ are defined per unit area along the step. We will now relate them to kinetic fluxes due to exchange processes between adatoms, step adatoms, kinks, and the crystal itself in an atomistic picture. Here the fluxes are defined per unit area in [100] direction. In order to transform them to the continuum description we relate them to the macroscopic fluxes by $f_\pm = f^\pm \cos(\theta)$ (see Fig. 1). In our notation, lower indices denote the macroscopic flux rate whereas the upper indices denote

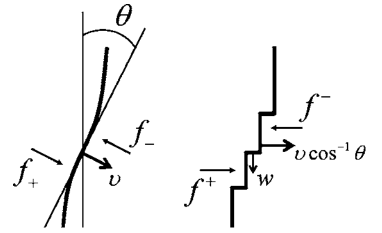

FIG. 1. Relation between the macroscopic (left) and microscopic (right) descriptions of a moving step.

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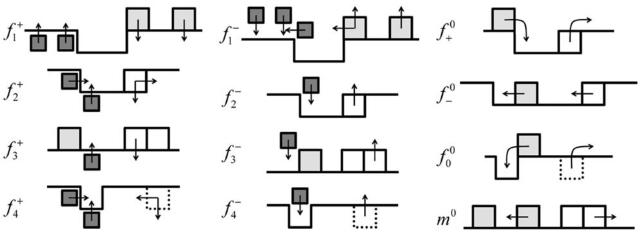

FIG. 2. Microscopic fluxes $f^{\pm}$ between the upper and lower terraces and a step, and f^0 and m^0 between steps and kink sites that explicitly change the densities of adatoms, step adatoms, and kinks. Adatoms, step adatoms, and atoms in kinks are shown in gray, light gray, and white, respectively. The dotted contours depict atoms inside the bulk. Mirrored configurations are not shown.

their microscopic origin. The fluxes are decomposed into four parts $f^{\pm}=(f_1^{\pm}+f_2^{\pm}+f_3^{\pm}+f_4^{\pm})$, see Fig. 2, in which

$$f_1^+ = (1-ak)(D_{TE}^+ a\rho_+ - D_{ET}^+\varphi)a^{-2},$$

$$f_2^+ = 2(1-ak)^2 k(D_{TK}^+\rho_+ - D_{KT}^+ a^{-2}),$$

$$f_3^+ = 2(1-ak)[D_{TK}^+ a(1-ak)^2\rho_+\varphi - D_{KT}^+ k_l k_r]a^{-1},$$

$$f_4^+ = 3[D_{TB}^+ a\rho_+ k_l k_r - D_{BT}^+(1-ak)^2 a^{-3}],$$

and

$$f_1^- = (1-a^2k^2)(D_{TE}^- a\rho_- - D_{ET}^-\varphi)a^{-2},$$

$$f_2^- = (1-ak)^2 k(D_{TK}^-\rho_- - D_{KT}^- a^{-2}),$$

$$f_3^- = 2(1-ak)[D_{TK}^- a(1-ak)^2\rho_-\varphi - D_{KT}^- k_l k_r]a^{-1},$$

$$f_4^- = D_{TB}^- a\rho_- k_l k_r - D_{BT}^-(1-ak)^2 a^{-3},$$

where a is the lattice constant, φ is the step adatom density, $k=k_l+k_r$ is the density of kinks consisting of left- and right-facing kinks, k_l and k_r, respectively, with $k_r-k_l=a^{-1}\tan(\theta)$. $D_{TX}^{\pm}$ is the diffusion coefficient for the transition from the upper $(+)$ or lower $(-)$ terrace (T) to the step (E), a kink (K), or to a crystal site (B), and $D_{XT}^{\pm}$ the corresponding inverse transition. The differences in D_{TE}^+ and D_{TE}^-, D_{TK}^+ and D_{TK}^-, D_{TB}^+ and D_{TB}^-, as well as in their inverse counterparts, model the Ehrlich-Schwoebel effect. The step adatoms diffuse along the step according to

$$\partial_t\varphi - D_E\partial_s^2\varphi = f_+^1 + f_-^1 - f_0 - m, \tag{4}$$

in which D_E is the step adatom diffusion coefficient on a step, s is the arc length along the step, $f_{\pm}^1=f_1^{\pm}\cos(\theta)$ is the flux from the incoming adatoms, $f_0=(f_+^0+f_-^0+f_0^0)\cos(\theta)$ is the loss of step adatoms due to their incorporation into kinks or a crystal site, and $m=(m^0+f_3^++f_3^-)\cos(\theta)$ is the loss due to nucleation of kink pairs from edge adatoms (see Fig. 2). The quantities are

$$f_+^0 = (1-ak)^2 k(D_{EK}^+ a\varphi - D_{KE}^+)a^{-2},$$

$$f_-^0 = (1-ak)^3 k(D_{EK}^- a\varphi - D_{KE}^-)a^{-2},$$

$$f_0^0 = 2(1-ak)[D_{EB}k_l k_r\varphi - D_{BE}(1-ak)^2 a^{-3}],$$

$$m^0 = 4(1-ak)^2[D_{EK}^-(1-ak)^2\varphi^2 - D_{KE}^- k_l k_r]a^{-1},$$

where $D_{EX}^{\pm}$ is the diffusion coefficient for the transition from the upper $(+)$ or lower $(-)$ step (E) to a kink (K), or to a crystal site (B), and $D_{XE}^{\pm}$ the corresponding inverse transition. The differences in D_{EK}^+ and D_{EK}^-, as well as D_{KE}^+ and D_{KE}^-, model the kink Ehrlich-Schwoebel effect. We assume only small differences in these coefficients, which satisfies the mean-field approach in the treatment of nucleation [12]. For simplicity we assume the same diffusion coefficients for left- and right-facing kinks. Due to the attachment of adatoms and step adatoms into kinks, the kinks move along the step. The velocities for left- and right-facing kinks are $-w$ and w, respectively. The resulting convective flux of kinks with respect to s is $w(k_r-k_l)$. By taking account of nucleation and breakup the kink density evolves according to [5]

$$\partial_t k + \partial_s[w(k_r-k_l)] = 2(g-h), \tag{5}$$

where $w=(a/k)(f_+^0+f_-^0+f_2^++f_2^-)$ is the kink velocity, $g=\frac{1}{2}m^0+f_3^++f_3^-$ the gain due to nucleation of kink pairs, and $h=f_4^++f_4^-+f_0^0$ the loss due to annihilation of kinks.

By neglecting deposition flux, desorption and nucleation in (1) we can derive an equation for the normal velocity v of the step from conservation of mass

$$v = awk\cos(\theta)\frac{1+(2g+h)a/wk}{1+\varphi\kappa a^2}. \tag{6}$$

This expression allows for a simple physical interpretation. The main term $awk\cos(\theta)$ corresponds to the growth of crystal by incorporation of atoms into kinks. The $(2g+h)a/wk$ term in the numerator accounts for the additional contribution of one-dimensional nucleation of step edge dimers and the filling of step vacancies to the growth, and the $\varphi\kappa a^2$ term in the denominator comes from the change of the arc length of a moving curved step, with κ being the curvature. If we employ the assumption that densities of kinks are small, i.e., $ak \ll 1$, the following simplifications of the model are possible: (a) The factor $1-ak$ can be approximated by unity. (b) The components $f_2^{\pm}$, $f_3^{\pm}$, and $f_4^{\pm}$ of the adatom fluxes can be neglected if compared with $f_1^{\pm}$. (c) The step adatom flux f_0^0 can be neglected if compared with $f_{\pm}^0$. The same holds for the nucleation term m^0. (d) If further nucleation events are neglected in the definition of the normal velocity and the step is assumed to be approximately straight, the growth is approximated by incorporation of adatoms into kinks and the step velocity becomes

$$v = awk\cos(\theta), \tag{7}$$

which corresponds to the velocity law derived in [13]. The resulting approximations give equations very similar to those obtained in [5]. There is, however, one minor discrepancy in the equation for edge adatom diffusion. In [5] the step velocity is adopted in the form of Eq. (7) and used to define the flux f_0 of atoms to kinks from macroscopic mass conservation arguments. In our model, we define the flux f_0 microscopically and use macroscopic arguments to derive the expression for the step velocity v. While both approaches seem equally justifiable, we choose the latter one, because by doing so, we maintain the same consistency principle throughout derivation of governing equations in our model, where all fluxes come from discrete microscopic considerations.

At equilibrium, $\rho_{\pm}=\rho$. The principle of detailed balance requires that all fluxes $f_i^{\pm}$, $i=1,2,3,4$ as well as $f_{\pm}^0$, f_0^0, and m^0 must vanish. From these assumptions equilibrium values for ρ and φ can be derived. Assuming that the step edge is parallel to the lowest index crystallographic direction, i.e., [100], implies $k_l=k_r=k/2$, resulting in an expression for the equilibrium density of kinks

$$k=a^{-1}(1+\sqrt{D_{EK}^{\pm}/D_{KE}^{\pm}}/2)^{-1}, \tag{8}$$

which coincides with BCF's expression for the case when only kinks of height ±1 are considered.

Assuming the crystal surface consists of a periodic sequence of steps, separated by distance L and moving at velocity v along [010] direction, we can construct a planar steady state solution. Under a quasisteady state assumption, by neglecting nucleation of two-dimensional islands on terraces as well as desorption, and applying the requirements for detailed balance, the solution of the diffusion equation for adatoms on the terrace

$$-D_T\nabla^2\rho=F \tag{9}$$

is sought in the form

$$\rho(x,t)=-\frac{F}{2D_T}[(x-vt)^2-L^2]+C_1(x-vt)+C_2$$

with unknown coefficients $C_1=(\rho_+-\rho_-)/2L$ and $C_2=(\rho_++\rho_-)/2$, where $\rho_{\pm}=\rho(vt\pm L)$. Boundary conditions become

$$D_TC_1-FL=-f_+,\quad D_TC_1+FL=f_-. \tag{10}$$

The diffusion equation for edge adatoms reduces to

$$f_1^++f_1^--f_0-m=0, \tag{11}$$

and the equation for kinks becomes

$$g-h=0. \tag{12}$$

Solution of the system gives the planar steady state values of k, φ, ρ_+, and ρ_-. The system is solved numerically for an atomistic solid-on-solid model, with different values of F between 0 and 1, L between 10 and 10 000, D_E between 10^4 and 10^7, and D_T taken to be 10^{12}. We define the step Peclet number as $P_{step}=2FL/D_E$ and plot the kink denity as a function of P_{step} and analyse the scaling behaviour $k\sim P_{step}^{\gamma}$ in Fig. 3. Out of equilibrium, the kink density deviates from the value in (8) and is lower than predicted in [5], resulting from the $(1-ak)$ correction, which accounts for the effect, that if the kink density increases, less and less area remains to create edge adatoms. The bold lines show the full system. A scaling exponent asymptotically approaching 1/3 from below with decreasing P_{step} is observed. The value 1/3 reflects the two-dimensional nature of the system. Terrace-kink, terrace-edge, and edge-kink interactions are never decoupled and the system does not break up into 1+1 dimensionality. Under the assumption that densities of kinks are small, i.e., $ak\ll 1$ and by neglecting processes related to the detachment from kinks and bulk [14] also predict a scaling exponent of 1/3 for the model in [5]. The lower lines in Fig. 3 are the solution of a restricted system: processes of attachment to and detachment from kinks and edge vacancies are neglected; the only way to create kinks is one-dimensional nucleation by edge diffusion of edge adatoms, the only way of kink annihilation is attachment of edge adatoms into edge vacancies. In this case direct terrace-kink interaction is neglected, which allows decoupling of terrace-edge and edge-kink interactions. The scaling exponent in this case is 1/4, which is consistent with the one-dimensional nucleation theory [15,16] and is also observed in [17] in the limit of rare detachment events. This case corresponds to diffusion mass transport mechanism. The upper line in Fig. 3 corresponds to neglected one-dimensional nucleation by edge diffusion of edge adatoms. In this case, the only way to create kinks is one-dimensional nucleation by attachment of an adatom next to an edge adatom. This case corresponds to the evaporation-condensation mass transport mechanism and gives an asymptotic scaling exponent of 1/2. For both special cases the solution of the exact system does not converge to either one within a wide range of P_{step}.

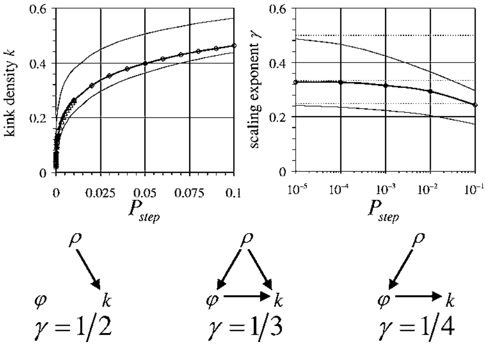

FIG. 3. Kink density k and scaling behavior $k\sim P_{step}^{\gamma}$ as a function of the step Peclet number. From top to down without edge diffusion approching 1/2, full model approaching 1/3, and without adatom-kink interaction approching 1/4.

Besides the kink density, we analyze the boundary value of the adatom density on a step. In Fig. 4 we plot the difference in the adatom densities on the lower and the upper terrace. The adatoms density on the lower terrace is always higher than that on the upper terrace. This comes from the fact that once we classify steps into kinks and kink-free straight segments, we see that the number of ways for attach-

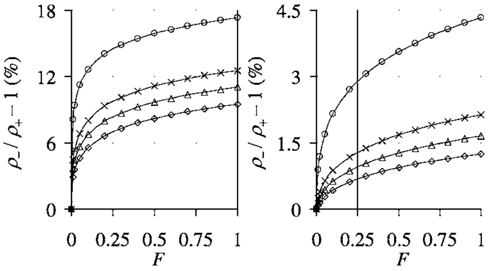

FIG. 4. Discontinuity in adatom density at steps as a function of deposition flux. (left) $D_E=10^4$, (right) $D_E=10^7$.

ment of an adatom onto a straight segment, into a kink site and into a step vacancy is different for upper and lower terrace. This asymmetry is intensified by the differences in detachment rates from kink sites and edge adatoms to adatoms. The larger the kink density gets, the more pronounced this effect will be. The observed difference in the adatom densities is inverse to the Ehrlich-Schwoebel effect and might be a natural way to explain step bunching. The effective inverse Ehrlich-Schwoebel barrier also has stabilizing effect against step meandering [18]. If we, in addition, allow for differences in the diffusion coefficients $D^+_{TX} < D^-_{TX}$ and their inverse counterparts $D^+_{XT} < D^-_{XT}$, with $X = E, K, B$ we also have the opposite effect of a meandering instability [19] but a stabilizing effect against step bunching. Therefore the coexistence of a bunching and meandering instability, as experimentally observed in [20] might be possible. To further analyze the interplay of the different instabilities as well as the influence of the kink Ehrlich-Schwoebel effect we need to solve the full time-dependent model.

We derived a continuum model for epitaxial growth from kinetics, allowing for the kink Ehrlich-Schwoebel barrier, nucleation on the level of edge adatoms, and the direct incorporation of adatoms into kinks and lattice sites. This model enables numerical studies of different mass transport mechanisms on a continuum scale. The model is valid for equilibrium as well as out-of-equilibrium growth regimes. The obtained steady state solution demonstrates scaling behavior and an asymmetry in the adatom density. We believe the model to be an important tool to bridge atomistic and continuum scales in epitaxial growth. All quantities entering the model are related to atomistic processes, and are accessible via *ab initio* calculations. The derived model can therefore be seen as an alternative to material specific kinetic Monte Carlo simulations with the potential to reach larger length and time scales.

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