Continuum limits of step flow models

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Abstract. We present continuum equations derived from microscopic steps-and-terraces growth models. These can be used in the modeling of epitaxial thin film growth. Assuming certain attachment and detachment mechanisms at the steps, we have obtained a Hamilton-Jacobi equation for the surface height which is coupled with a diffusion equation for the edge adatom density. These equations are supplemented with boundary conditions to describe the evolution of peaks and valleys on the surface.

1 Introduction

The purpose of this paper is to present continuum limits of step flow models used in the study of crystal growth. We are mainly interested in *epitaxial growth* so that the atomic structure plays an important role. A simple example of interest to us is *molecular beam epitaxy* (MBE) [Tsa] in which atoms (single or in molecules) are delivered under *ultra-high vacuum* conditions onto the surface where they diffuse until they meet a step and are incorporated into the crystal lattice. Other more complicated growth methods such as *chemical vapor deposition* (CVD) can also be described by the step flow models studied in this paper.

The foundation for the study of crystal growth was laid out in the paper of Burton, Cabrera and Frank [BCF] which also establishes the starting point of the present work. Recognizing that the vapor-solid interface consists of three different kinds of geometric objects – *terraces*, *steps(edges)* and *kinks* of dimensions *two, one* and *zero* respectively – BCF postulates that atoms in the vapor phase land on the terraces and then diffuse until they meet a step along which they continue to diffuse until they are incorporated into the kink sites along the steps. (Figure 1.) The diffusing atoms on the interfaces are called *adatoms.* Thus the surface grows by the movements of steps which are induced by the movements of kinks. Of course, in order to have *effective vertical growth*, new steps must be generated. This can be caused by nucleation or the presence of screw-dislocations. A modern account of the theory of crystal growth can be found in [PV].

At the finest scale, crystal growth should be described by the hopping of adatoms between different lattice sites on the interface. The BCF theory operates at the next scale where the terraces are treated as a continuum. Our principal objective is to develop a theory that operates at the next larger scale at which the interface is described by a continuous height function which can have peaks and valleys. More generally, we can move to even larger scale over which many peaks and valleys are homogenized. Presumably, this is the scale where macroscopic mass transport of material on the surface (such as surface diffusion) takes place.

There is an abundance of continuum equations in the literature describing the evolution of the height function in epitaxial growth. Typical examples include the KPZ, EW and Villain equations. These are described in [PV]. Our work is different in two aspects. Firstly, we derive these equations more or less rigorously from step flow models. Secondly, the equations we obtain form a *coupled system* of equations for the *height function* and the *adatom density.* Having adatom density in the equations is important if one wants to treat non-trivial surface chemistry involving the reactions between different chemical species.

In this paper, we concentrate on the $1+1$ dimensions. Extensions to $2+1$ dimensions can be found in [EY].

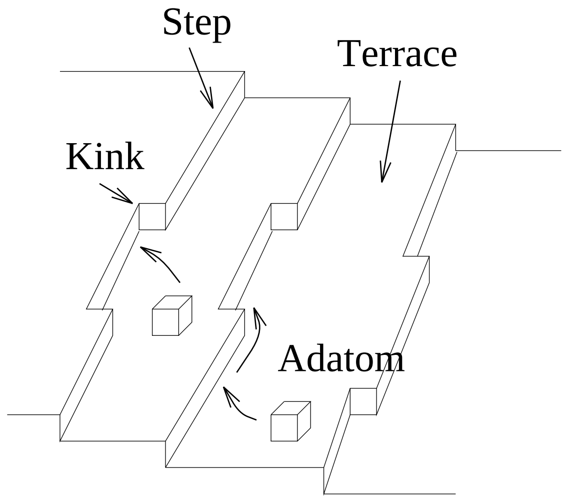

Figure 1

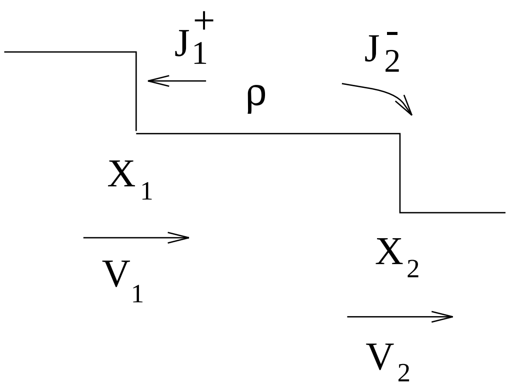

Figure 2

2 BCF Theory and Its Generalization

In $1+1$ dimensions, the surface consists of terraces and steps only. We first consider *vicinal terraces* which are bounded by one up-step (at x_1) and one-down step (at x_2) (Figure 2.)

As mentioned earlier, atoms from vapor phase land on the terraces and diffuse. This is described by the following diffusion equation:

$$\rho_t = D\rho_{xx} - \frac{\rho}{\tau} + F \tag{1}$$

where ρ is the terrace adatom density; D is the diffusion constant; τ is the mean survival time before a terrace adatom evaporates; F is the deposition flux. The boundary conditions for (1) are given in terms of the fluxes:

$$J_1^+ = D\rho_x|_1^+ + v_1\rho_1^+; \quad J_2^- = -(D\rho_x|_2^- + v_2\rho_2^-); \tag{2}$$

where v_1 and v_2 are the velocities of the steps at x_1 and x_2.

In addition, we need to give constitutive relations for the fluxes J and the velocities v. These will depend on the mechanisms of attachment and detachment at the steps. We will consider the following two models.

2.1 T-Model: Attachment Only

In this model, the terrace adatoms near a step has certain probability of attaching to the step. After attachment, they will stay in the lattice forever. Thus we impose the following condition:

$$J_1^+ = \alpha_+\rho_1^+, \quad J_2^- = \alpha_-\rho_2^- \tag{3}$$

where α_+ and α_- are the *attachment rates* to the upper- and lower-steps. The step velocity is given by the conservation of adatoms:

$$\frac{1}{a^2}v = J^+ + J^- = \alpha_+\rho^+ + \alpha_-\rho^-. \tag{4}$$

where a is the dimension of an adatom.

This model ((1), (2), (3) and (4)) is called the **T-Model** as only terrace adatoms are considered.

2.2 TE-Model: Attachment and Detachment

In this model, the terrace adatoms can *attach to* and *detach from* a step. To capture this mechanism, we introduce the notion of *edge adatoms* which reside on and diffuse *along* a step. Its density is denoted by φ which is a function at each step. Now the fluxes are postulated to be

$$J_1^+ = \alpha_+\rho_1^+ - \beta_+\varphi_1, \quad J_2^+ = \alpha_-\rho_2^- - \beta_-\varphi_2 \tag{5}$$

where β_+ and β_- are the rates at which edge adatoms *detach* and go back to the lower- and upper-terraces. The velocity of each step in this model is given by

$$v = a^2k(\gamma\varphi - \delta) \tag{6}$$

where k is the average kink density along a step; γ and δ are the attachment and detachment rates of edge adatoms to and from the kink sites. To complete the model, the rate of change of the edge adatom density is given by the following conservation law:

$$\frac{d\varphi}{dt} = J^+ + J^- - \frac{v}{a^2} \tag{7}$$

We call this model ((1), (2), (5), (6) and (7)) the **TE-Model** as it considers both the terrace and edge adatoms.

3 Continuum Limits of Vicinal Surfaces

We now present the continuum limit for a vicinal surface which consists of a sequence of vicinal steps so that the surface is *monotonically decreasing* or *increasing*. Let the step locations be at $\{x_j(t)\}_{j=-\infty}^{\infty}$. We want to derive the evolution equation of a *continuous function* $h(x,t)$ such that $h(x_j(t),t) - h(x_{j+1}(t),t) \approx a$. Here a is treated as the step height. The limiting process we consider is the following:

$$a \longrightarrow 0, \quad l_j(t) \,(\text{terrace width}) \, = x_{j+1}(t) - x_j(t) = \frac{a}{|h_x|} = O(a). \tag{8}$$

3.1 T-Model Without Evaporation

Under the assumption of no evaporation, and in the regime where the step velocity is much smaller than the hopping rate of the terrace adatoms, (1) and (2) can be simplified to the following:

$$0 = D\rho_{xx} + F, \quad J^+ = D\rho_x^+, \quad J^- = -D\rho_x^-. \tag{9}$$

Now the set of equations (9), (3) and (4) can be easily solved on each individual terrace. (The details are recorded in [EY].) By performing the limiting process (8), we obtain the following simple evolution equation for $h(x,t)$:

$$\frac{\partial h}{\partial t} = Fa^3. \tag{10}$$

Remark. Equation (10) states that the height increases uniformly across the surface. This is simply the consequence of the fact the terrace adatoms are eventually incorporated onto either the upper or lower step *irreversibly*. In particular, this model does not permit an adatom to diffuse across a step.

3.2 TE-Model Without Evaporation

Still assuming no evaporation and the step velocity much smaller than the hopping rate of the terrace adatoms, the system of equations (9), (5), (6) and (7) can also be solved quite easily. The only difference is that now ρ is expressed in terms of φ. Performing the same limiting process (8), we obtain the following continuum equations

$$\begin{aligned}
\Phi_t \pm (\Phi v)_x &= aF + a\partial_x\left(A(h_x)\partial_x\frac{\Phi}{|h_x|}\right) \pm Q\partial_x(A(h_x)\Phi) - \frac{v}{a^2}|h_x| && (11)\\
v &= a^2k(\gamma\frac{\Phi}{|h_x|} - \delta) && (12)\\
h_t &= v|h_x| && (13)
\end{aligned}$$

where

$$\begin{aligned}
\Phi &= \varphi|h_x| = \text{Edge adatom density per unit length in } x; & (14)\\
A(h_x) &= \frac{(\alpha_+\beta_- + \alpha_-\beta_+)aD}{2((\alpha_+ + \alpha_-)D|h_x| + \alpha_+\alpha_- a)}; & (15)\\
Q &= \frac{\alpha_+\beta_- - \alpha_-\beta_+}{\alpha_+\beta_- + \alpha_-\beta_+}; & (16)\\
\pm &= -\mathrm{sgn}(h_x). & (17)
\end{aligned}$$

Remark. Equations (11) – (13) constitute the coupled system we announced at the beginning. They show that the edge adatoms are able to diffuse *across the terraces*. This diffusion (described by the second order term in (11)) is much more global than just the terrace diffusion considered in the original BCF theory. The term $A(h_x)$ represents the *mobility factor*. Its form shows that the presence of steps slow down diffusion. (Note that $A(h_x) \to 0$ as $|h_x| \to 0$.) The Q term is an additional drift caused by the *Schwoebel effect* which is an additional step-edge barrier to be overcome by the adatoms in order to diffuse across the steps.

4 Boundary Conditions at Valleys and Peaks

So far we have restricted our attention to vicinal surfaces. However, on the actual growing interface, valleys (local minima) and peaks (local maxima) are generally present. At these points, $|h_x|$ equals zero or is undefined so that the continuum equations (11) and (13) become singular. In addition, the peaks can be the nucleation sites where new steps are continuously generated so that the surface height can grow up in the vertical direction. Our approach is to supplement the equations by appropriate boundary conditions at the singular points. The system then becomes a free boundary value problem.

Let $X(t)$ be the location of a valley ($h_x(X(t)^+) > 0$ and $h_x(X(t)^-) < 0$) or a peak ($h_x(X(t)^+) < 0$ and $h_x(X(t)^-) > 0$). For convenience, we rewrite (11) in the form of

$$\frac{\partial \Phi}{\partial t} = aF + \partial_x(\mathcal{J}) - \frac{1}{a^2}h_t \tag{18}$$

The boundary conditions we derive are described as follows.

Conservation Law. This is the Rankine-Hugoniot condition in conservation law:

$$[\Phi(X(t))]\dot{X}(t) = -[\mathcal{J}(X(t))] \tag{19}$$

where $[f(X(t))] = f(X^+(t)) - f(X^-(t))$.

Geometric Condition. This is the consequence of the continuity of $h(x,t)$ at $X(t)$ and is obtained by taking the time derivative of the relationship $h(X^+(t),t) = h(X^-(t),t)$:

$$\dot{X}(t)\Big|_{\text{valley}} = -\frac{|h_x v(X^+)| - |h_x v(X^-)|}{|h_x(X^+)| + |h_x(X^-)|}, \quad \dot{X}(t)\Big|_{\text{peak}} = \frac{|h_x v(X^+)| - |h_x v(X^-)|}{|h_x(X^+)| + |h_x(X^-)|} \tag{20}$$

We need at least one more condition in order to close the system. In the case of valley, we impose the condition of *continuity of* φ:

$$[\varphi(X(t))] \ (= \left[\frac{\Phi}{|h_x|}\right]) \ = 0 \tag{21}$$

which is physically reasonable by the fact that the edge adatom can freely diffuse from one side of the valley to the other without any obstruction. Now conditions (19), (20) and (21) completely determines the evolution of the coupled system at the valley.

However, we do not expect the continuity of φ to be valid at the peak due to the possibility of nucleation. In principle, we need *two more* boundary conditions – one for $X(t)$ and one for the vertical growth velocity of h. It seems that we need to look in more detail the nucleation mechanisms. This is currently work in progress.

On the other hand, if we assume that there is no nucleation and the peaks (valleys) are moving down (up) i.e. there is mass transport from peaks to valleys, then we can still impose the continuity of φ at the peak. In this case, we have all the boundary conditions needed.

As a summary, we have obtained continuum equations for epitaxial growth at a mesoscopic length scale – one that smooths out the steps and kinks but sees the individual valleys and peaks. It would also be interesting to derive a macroscopic model, on a length scale that smooths out the valleys and peaks. We suppose this can be done by suitable averaging or homogenization of our mesoscopic model, but this remains a task for future work.

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