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Continuum Limit of a Step Flow Model of Epitaxial Growth

R.V. Kohn[1], T.S. Lo[1] and N.K. Yip[2]
Courant Institute of Mathematical Sciences, New York University,
251 Mercer Street, New York, NY 10012, U.S.A.[1]
Department of Mathematics, Purdue University, West Lafayette, IN 47907, U.S.A.[2]

ABSTRACT

We examine a class of step flow models of epitaxial growth obtained from a Burton-Cabrera-Frank (BCF) type approach in one space dimension. Our goal is to derive a consistent continuum model for the evolution of the film surface. Away from peaks and valleys, the surface height solves a *Hamilton-Jacobi equation* (HJE). The peaks are *free boundaries* for this HJE. Their evolution must be specified by boundary conditions reflecting the microscopic physics of nucleation. We investigate this boundary condition by numerical simulation of the step flow dynamics using a simple nucleation law. Our results reveal the presence of *special structures* in the profile near a peak; we discuss the relationship between these structures and the continuum equation. We further address the importance of evaporation for matching the local behavior near the peak to the solution of the continuum equation.

INTRODUCTION

Epitaxial growth has many important technological and industrial applications. To understand and control the properties of thin film materials, accurate and efficient modeling of the growth process is essential. Continuum modeling of epitaxial growth has received much attention in the past two decades, but our understanding remains incomplete; see e.g. [1] for a recent account. From the computational point of view, continuum models, in the form of partial differential equations (PDEs), are preferred over the microscopic ones, such as Monte-Carlo or molecular dynamics models, because of their time efficiency. Furthermore, continuum equations can model the surface morphologies at larger spatial scales with relative ease.

Continuum models of epitaxial growth very often start with the conservation equation [2,3]: $h_t = -\nabla \cdot \mathbf{J} + F$. Here $h(x,t)$ is the film surface height, $\mathbf{J}$ is the surface mass current density, and F is the source term. A constitutive equation for $\mathbf{J}$ is needed. Based on phenomenological or physical reasoning, $\mathbf{J}$ is typically taken to depend on the derivatives of the surface height function. One common example of such $\mathbf{J}$ is given by $K\nabla(\triangle h) + f(|\nabla h|^2)\nabla h$. Work has been done attempting to derive such an equation from an underlying atomic-scale stochastic model, see e.g. [4], but there is as yet no systematic procedure for doing so.

Our approach is different. It emphasizes the mesoscopic features, i.e. spatial structures whose length scale is large compared to the lattice size but small compared to the sample. They provide potential links between the microscopic and macroscopic phenomena. Working in one space dimension for simplicity, we treat the film surface as a collection of mounds, i.e. a series of peaks and valleys. On vicinal terraces far from any peak or valley, the dynamics is well-described by a step flow model. In the presence of evaporation, the associated continuum equation is a HJE: $h_t = H(|\nabla h|)$. This HJE does not apply, however, across the peaks; rather, they must be modeled as free boundaries characterized by appropriate boundary conditions. From the physical point of view, it is natural to treat the peaks separately since they are the places where nucleation is most likely to occur; the physics there is considerably different from the rest of the film.

The main goal of the present work is to explore the proper treatment of peaks. We start by (i) formulating a HJE model for epitaxial growth by examining a simple step flow dynamics; then we (ii) explore what boundary conditions should be applied at the peak to capture the consequences of nucleation. We find that the proper continuum solution is obtained by specifying the vertical velocity of the peak, V_p. Our work includes simulations of the structure of the peak region, which show the presence of two rather different local structures — "rarefaction waves" and "shock waves" — depending on the value of V_p. These local structures play a crucial role in matching the local behavior near the peak to the solution of the continuum equation.

Our viewpoint requires that there be at least a little evaporation; in the (very singular) zero-evaporation limit, one loses the ability to specify different values of V_p relative to the overall growth rate of the film.

Since we work in (1+1) dimension, our film surface consists of a sequence of terraces and steps (Figure 1). The BCF model [5] determines the surface evolution by computing the fluxes of surface adatoms to the steps. In the quasistatic approximation one obtains a step velocity law of the form

$$V_n = a[f(l_+) + g(l_-)] \ , \tag{1}$$

where V_n is the velocity of the nth step; a is the lattice spacing; l_+ (l_-) is the width of the terrace ahead of (behind) the nth step; and f and g are the fluxes of adatoms to the nth step from the terrace ahead and behind, respectively. The continuum limit in this paper is taken in the sense of letting $a \to 0$ and $a/l_\pm \to |h_x|$, see e.g. [6]. In this limit, we have $V_n \approx h_t/|h_x|$ and the principal-order continuum equation for (1) is a *first order* HJE. (If higher-order terms in a are kept, one gets a higher order PDE; we shall return to this point in the last section.)

This article is organized as follows. We first introduce the use of a HJE for the modeling of step flow and nucleation. Then the importance of evaporation is discussed. Next some simulation results and interpretations are presented. Finally we relate our work to other models.

THE HAMILTON-JACOBI EQUATION AND NUCLEATION

In this section, we explain the use of a HJE to model step flow and nucleation at the continuum level. The step velocity law (1) is our starting point. The term $g(l_-)$ can be neglected if, as we shall assume throughout, the Schwebel barrier is infinite. To capture the main phenomena — attachment and evaporation of surface adatoms — with minimal complexity, we use the following simple form for V_n:

$$V_n = af(l_n) = \frac{aFl_el_n}{l_e + l_n} \tag{2}$$

where $l_e = \sqrt{D\tau_e}$ is the diffusion length and l_n is the width of the nth terrace. (D and τ_e denote the diffusion constant and evaporation time for the surface adatoms.) Note that

$V_n \approx aFl_e$ for $l_n \gg l_e$ and $V_n \approx aFl_n$ for $l_n \ll l_e$. (See [1] for alternative formulas — with similar qualitative behavior — derived directly from the BCF framework.) The continuum limit of (2) is the following firstorder HJE:

$$h_t = \frac{a^2 F l_e |h_x|}{a + l_e |h_x|} = H(|h_x|)\ . \tag{3}$$

This equation describes the evolution of vicinal steps relatively well. However, at the peak of the surface profile, i.e. the top terrace, we encounter another important physical phenomenon — nucleation. It is through this process that the film surface gains new height. The modeling of this phenomenon requires the prescription of the peak vertical velocity V_p. (In the present work, we assume that nucleation only occurs on the top terrace.) The overall shape of the profile near the peak depends on the magnitude of V_p (Figure 2).

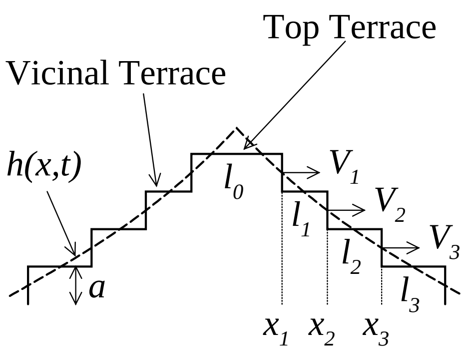

Figure 1.

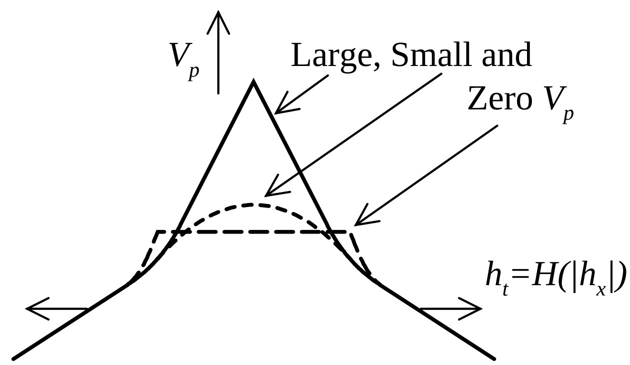

Figure 2.

If the conventional viscosity theory of HJEs [7] is used to solve (3), there is a unique solution which in fact corresponds to $V_p = 0$, i.e the height *does not* grow. To model the vertical growth of the peak, it is thus natural to consider the following version of (3):

$$h_t = H(|h_x|) + V_p \mathbf{1}_{\text{peak}}(x) \tag{4}$$

where $\mathbf{1}_{\text{peak}}$ is a function defined to be equal to 1 at the position of the peak and 0 otherwise. The use of such a singular term can also be found in [8]. In effect, the peak growth is being treated separately from the vicinal growth. Sometimes a regularized version of the above equation is considered. It would be interesting to develop a theory of viscosity-type solutions for (4), and to explore its approximation by various regularizations and numerical solution schemes.

EVAPORATION AND VERTICAL PEAK VELOCITY

From the previous PDE viewpoint, V_p is a free variable for the HJE (4). We claim however that this freedom of choosing V_p can be realized physically by varying the value of F. The argument rests on identifying the *nucleation length* l_c — the typical length of the top terrace when a new layer nucleates. An argument similar to that of [9] gives

$$l_c^2\left(\frac{l_c}{2} + l_e\right) = \frac{aD}{Fl_e} \quad \text{and} \quad V_p = aF^2 l_c^2 \tau_e\ . \tag{5}$$

Let us briefly summarize the derivation of (5). We assume that a nucleation event occurs whenever two adatoms occupy the top terrace simultaneously. Then the nucleation rate $1/\tau_n$ is the product of the adatom arrival rate and the top terrace occupation probability. Assuming an infinite Schwoebel barrier and using the quasistatic approximation, the adatom density on the top terrace is $\rho = F\tau_e$. Thus when the top terrace has length l the nucleation rate is

$$\frac{1}{\tau_n(l)} = (Fl) \cdot (F\tau_e l) \ . \tag{6}$$

Now the nucleation length l_c is determined by

$$af(l_c/2) = \frac{l_c/2}{\tau_n(l_c)} \tag{7}$$

since the terraces just below the peak have length approximately $l_c/2$. This gives the first part of (5). The second part follows from the obvious relation $V_p = a/\tau_n(l_c)$.

The relation (5) simplifies in the extremes $\frac{aD}{Fl_e^4} \ll 1$ ($l_c \ll l_e$) and $\frac{aD}{Fl_e^4} \gg 1$ ($l_c \gg l_e$), as follows:

No evaporation ($l_e \to \infty$) or **large deposition flux** ($F \to \infty$) : $l_c = \left(\frac{aD}{Fl_e^2}\right)^{\frac{1}{2}}$ and $V_p = a^2 F$ (8)

Strong evaporation ($l_e \to 0$) or **small deposition flux** ($F \to 0$) : $l_c = \left(\frac{2aD}{Fl_e}\right)^{\frac{1}{3}}$ and $V_p = 2^{\frac{2}{3}} a^{\frac{5}{3}} F^{\frac{4}{3}} \tau_e^{\frac{2}{3}} D^{\frac{1}{3}}$ (9)

We now draw two crucial conclusion. (i) The peak velocity V_p can have various scalings with respect to F. On the other hand, the rate of height growth given by (3) is always proportional to F. This clearly shows that V_p is a free variable from the point of view of the HJE. (ii) The presence of evaporation ($l_e < \infty$) is crucial for the freedom to prescribe V_p. Without evaporation, $V_p = a^2 F$ is constant and equal to the overall film growth rate.

Our discussion accounts for nucleation on the top terraces but not on vicinal terraces. This is of course an idealization. It is however reasonable in the large-Schwoebel-barrier limit, since vicinal terraces are drained of adatoms by attachment at steps, but the top terrace loses adatoms only by nucleation and evaporation.

There has recently been work on the modeling of nucleation events in terms of stochastic atomic-scale processes such as diffusion and collision of adatoms [10]. Such work could potentially provide improved peak models for coupling to our continuum approach.

LOCAL PATTERNS AT THE PEAK

To exemplify and validate our viewpoint, we perform numerical simulations of the step flow model defined by (2) using a simple but reasonable nucleation rule: *a new terrace is introduced at the center of the top terrace when its width reaches a prescribed value* l_c. (In all our simulations, the initial height profile is assumed to be symmetric about the peak.) By varying l_c, we can monitor the value of V_p. Our results are summarized in Figures 3 and 4, and they support the qualitative picture of Figure 2.

To understand the above pictures at the continuum level, we observe the appearance of *inner* and *outer slopes* (m_1 and m_2) which are the slopes of the height profile near and

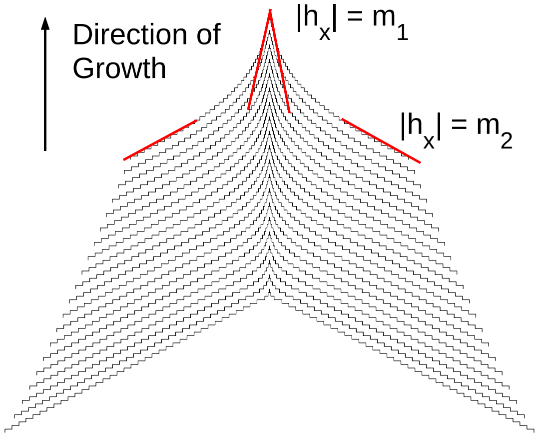

Figure 3. Small l_c, large V_p, rarefaction wave

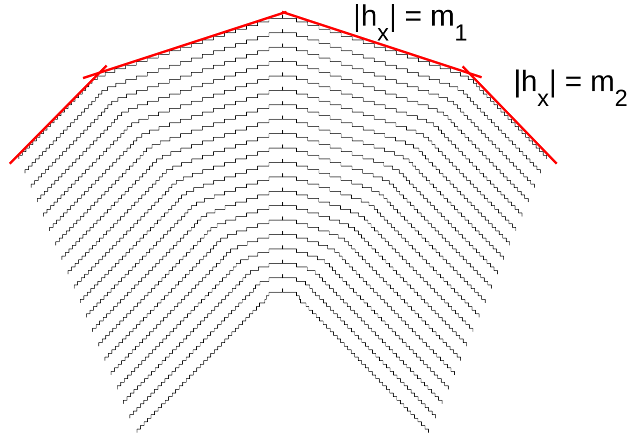

Figure 4. Large l_c, small V_p, shock wave

away from the peak. Intuitively, m_1 is determined by the nucleation phenomena and m_2 is the far-field condition. The correct solution of (4) is the one that connects these two slopes together. We call the patterns in Figures 3 and 4 "rarefaction waves" and "shock waves"; the former occurs when $m_1 > m_2$ and the latter when $m_1 < m_2$.[1] The explicit form of the rarefaction wave for the HJE (3) is given by:

$$|h_x(x,t)| = \begin{cases} m_1 & \left|\frac{x}{t}\right| \le c_1 \\ \frac{a}{l_e}\left(\sqrt{aFl_e}\left|\frac{x}{t}\right|^{-\frac{1}{2}} - 1\right) & c_1 \le \left|\frac{x}{t}\right| \le c_2 \\ m_2 & \left|\frac{x}{t}\right| \ge c_2 \end{cases}$$

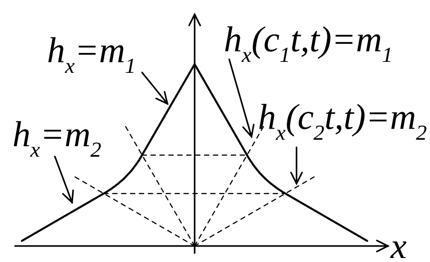

Figure 5.

where c_1 and c_2 are constants such that the $|h_x|$ is defined continuously (See Figure 5).

At the discrete level, according to (2), the step evolution is given by:

$$\dot{X}_n(t) = af(X_{n+1} - X_n) \quad \text{or} \quad \dot{l}_0 = 2af(l_1)\ , \quad \dot{l}_n = a[f(l_{n+1}) - f(l_n)]\ , \quad n = 1, 2, \ldots \tag{10}$$

where X_n and $l_n = X_{n+1} - X_n$ denote the location of the nth step and the width of the nth terrace (Figure 1). This system has a very simple *traveling wave* type solution with *constant slope*: $\dot{l}_0(t) = 2af(\frac{l_c}{2})$; $\quad l_n = \frac{l_c}{2}$; $\quad n = 1, 2, \ldots$ Only l_0 changes in time. The overall step pattern *repeats* itself after a time period of $T = \frac{l_c}{2a} f(\frac{l_c}{2})^{-1}$. This leads to

$$V_p = \frac{2a^2}{l_c} f\left(\frac{l_c}{2}\right) \ . \tag{11}$$

In this example, m_1 is explicitly given by $2al_c^{-1}$. These clearly show that V_p and m_1 are related to the nucleation phenomena.

For the case of finite but large Schwoebel barrier, we believe that a similar description still holds because the step evolution (1) can be considered as a small perturbation of (2).

[1] This terminology comes from the theory of hyperbolic conservation laws in PDEs. In fact, in terms of the new variable $u = h_x$, (3) can be written as $u_t = [H(|u|)]_x$.

MODIFIED EQUATIONS AND A SINGULAR LIMIT

Our model so far involves only first order PDEs. This is in contrast with the higher order, nonlinear PDEs typically found in the literature [2,3]. In this section, we comment on the relationship between our model and associated higher-order PDEs.

As mentioned in the introduction, a second-order PDE arises naturally by keeping terms of second order in the small parameter a. For our model (2), the resulting nonlinear diffusion equation is:

$$l_n = \frac{a}{|h_x|} + \frac{a^2 h_{xx}}{2|h_x|^3}\,, \qquad h_t = af\left(\frac{a}{|h_x|}\right)|h_x| + \frac{a^3}{2}f'\left(\frac{a}{|h_x|}\right)\frac{h_{xx}}{|h_x|^2}\,. \tag{12}$$

Nonlinear diffusions of this type — usually modified near $h_x = 0$ and regularized by higher-order terms — have been used to model mounding produced by molecular beam epitaxy [3]. However equation (12) is singular when $h_x = 0$; moreover its derivation is based on vicinal step-trains, and *does not* apply at peaks or valleys. We therefore take the view that the equation should not be applied across the peaks and valleys; rather, these should be treated as free boundaries. Perhaps the singular diffusion terms could be important in giving a more precise description of the solution near the peaks and valleys.

We conjecture that inclusion of the diffusion term will be crucial for understanding the no-evaporation limit $l_e \to \infty$. This limit is singular, in the sense that our HJE (3) degenerates to $h_t = a^2F$ and it loses the ability to support different values of V_p (see (8)). The associated limit of (12) is $h_t = a^2F + a^3F\frac{h_{xx}}{2|h_x|^2}$. This equation was proposed as a continuum model of no-evaporation, infinite-Schwoebel-barrier growth in [11]; it captures the formation of sharp peaks seen in Monte-Carlo models of this growth regime. We wonder whether the solutions obtained by our viewpoint converge to those considered in [11], in a suitable limit involving $l_e \to \infty$ and $l_c \to 0$.

The singular character of the no-evaporation limit has also been recognized in [12]. That paper discusses a different model, but there too even a small amount of evaporation has a profound effect on the growth morphology.

CONCLUSION

Despite much study, we still do not have a complete understanding of the link between atomic-scale and continuum models of film growth. From both the mathematical and physical points of view, it is important to identify the correspondence between models on different scales. We have shown, for a particular class of step-flow and nucleation models in (1+1) dimension, that the appropriate continuum model is a Hamilton-Jacobi equation with specified peak velocity. We have also argued that the no-evaporation limit is singular, interesting, and remains to be properly understood. Work is in progress in other directions as well, including: appropriate modeling of valleys; convergence of the step flow solutions to those of the continuum model; alternative nucleation models with stochastic effects; and large-scale behavior such as coarsening.

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REFERENCES

1. A. Pimpinelli and J. Villain, *Physics of Crystal Growth*. (Cambridge University Press, 1998)
2. J. Villain, J. de Physique I **1**, 19 (1991); O. Pierre-Louis, C. Misbah, Y. Saito, J. Krug and P. Politi, Phys. Rev. Lett. **80**, 4221 (1998).
3. P. Politi and J. Villain, Phys. Rev. B **54**, 5114 (1996).
4. D.D. Vvedensky, A. Zangwill, C.N. Luse and M.R. Wilby, Phys. Rev. E **48**, 852 (1993).
5. W.K. Burton, N. Cabrera and F. Frank, Phil. Trans. Roy. Soc. **243**, 299 (1951).
6. W. E and N.K. Yip, J. Stat. Phys. **104**, 221 (2001).
7. L.C. Evans, *Partial Differential Equation*, Graduate Studies in Mathematics, Vol. 19. (American Mathematical Society, 1998)
8. T. Schulze and R.V. Kohn, Physica D **132**, 520 (1999).
9. I. Elkinani and J. Villain, J. de Physique I **4**, 949 (1994).
10. J. Krug, P. Politi and T. Michely, Phys. Rev. B **61**, 14037 (2000); J. Krug, Eur. Phys. J. B **18**, 713 (2000).
11. J. Krug, J. Stat. Phys. **87**, 505 (1997).
12. P. Šmilauer, M. Rost and J. Krug, Phys. Rev. E **59**, R6263 (1999).