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Shocking facets in interface growth

V. Tsemekhman[a,*], J.S. Wettlaufer[a,b]

[a] *Applied Physics Laboratory, University of Washington, Box 355640, Seattle, WA 98105, USA*
[b] *Department of Physics, University of Washington, Box 351560, Seattle, WA 98105, USA*

Abstract

We find that a certain class of geometric models of crystal growth predicts the formation of an expanding facet with a discontinuous edge adjacent to rough orientations—a shocking facet. However, we show that although such structures are observed experimentally, aspects of their long-time evolution are not entirely captured by the same class of geometric models that predict their formation. We suggest a simplified description of interfacial evolution that reproduces the experimentally observed behavior. © 2002 Elsevier Science B.V. All rights reserved.

Crystal growth provides a physical and practical testing ground for general mathematical developments that seek to understand the relationships between interfacial convexity, non-locality, and singularity formation. Recently, the framework of so-called *geometric* models has been developed to study *interface controlled growth*, wherein the phase boundary motion is controlled solely by local surface properties and molecular kinetics. In such circumstances the geometric description is naturally applicable *ab initio* (e.g., Reference [3] reviews geometric models with the primary focus on solidification. Some papers of note in this regard include Ref. [1]). A principal motivation for such work is to understand the evolution of a partially faceted initial seed, which is a test problem with a long history in the crystal growth community (see e.g., Refs. [2–4]). Transient growth of such a seed has been observed quantitatively [5] within the context of new theoretical advances that predict an evolution termed *global kinetic faceting* whereby the rough orientations grow out of existence with *decreasing* curvature [6]. By geometric requirement a *decrease* in curvature must be accompanied by *jumps* in the surface slope—corners on the surface. Here we relate the appearance, and motion, of the corners on the surface to the expansion of facets; phenomena providing a number of tests of geometric models. We show that a certain class of geometric models predicts the formation of a "shocking facet"—an expanding facet with an edge that forms a corner with rough orientations. Moreover, it is shown that the

*Corresponding author. Tel.: +1-206-685-3734; fax: +1-206-616-3142.
E-mail address: dima@apl.washington.edu
(V. Tsemekhman).

long time evolution of a "shocking facet" cannot be described by any geometric model that predicts its formation. Finally, we demonstrate the conditions under which interface controlled growth breaks down and a complete dynamical description of long-ranged diffusion must be invoked.

It is necessary to provide the theoretical background (e.g., Refs. [4–7]) which leads to the principal issue dealt with here of shocking facets and the breakdown of interface-controlled growth. We distinguish between geometric models whereby the local velocity can depend on curvature, i.e., like motion by mean curvature [8], and those wherein the velocity does not depend on curvature [6]. For simplicity, we treat the case of a two-dimensional crystal. Hence, our goal is to understand the properties of the solutions to the evolution equation

$$\frac{\partial \vec{C}}{\partial t} = -V\vec{N},\qquad(1)$$

where $\vec{C} = \vec{C}(u,t)$ is an evolving plane curve parametrized by a variable u, $\vec{N}$ is the (inward pointing) unit normal vector, and V is the normal growth velocity. The arclength s and u are related by $s(u,t) = \int_0^u |\partial \vec{C}(u',t)/\partial u'| \mathrm{d}u'$. The growth dynamics within this class of models are such that the velocity depends only on the local orientation of the surface normal: $V = V(\theta)$, where θ is an angle between the positive x-axis and the unit tangent vector $\vec{T}$. We expect that the variety of possible functional forms of the growth function $V(\theta)$ will lead to a rich behavior of solutions to Eq. (1).

Exact solutions for this type of evolution equation can be expressed in terms of characteristics which represent straight rays along which the orientation of the normal is preserved [3]. It is also possible to derive a local equation for the evolution of curvature κ using methods of differential geometry to obtain

$$\frac{\partial \kappa}{\partial t} = -\tilde{V}(\theta)\kappa^2.\qquad(2)$$

Eq. (2) describes the evolution of curvature along the trajectory of the location on the boundary with fixed orientation of the normal θ, and $\tilde{V} = V + V''$ where the primes denote differentiation with

respect to θ. The derivation of this equation relies on the fact that the growth function V depends only on θ and is based on the approach of Gage and Hamilton [9] as outlined in Ref. [6]. The solution of Eq. (2) is given by

$$\kappa(\theta,t) = \frac{\kappa_0(\theta)}{1 + \tilde{V}(\theta)\kappa_0(\theta)t},\qquad(3)$$

where $\kappa_0(\theta)$ is the curvature at the location on the initial seed crystal's boundary with orientation θ of the normal. One observes that the curvature decreases with time for all orientations with $\tilde{V} > 0$. The curvature grows continuously and diverges at finite time for all orientations with $\tilde{V} < 0$.

If the characteristics intersect each other, a shock forms. A wide range of geometric models may predict the formation of shocks. The propagation of a shock results in the disappearance of surface orientations and thus, manifests itself as the development of a corner on the crystal surface. We have shown in Refs. [7,10] that the moment when a shock is initiated it coincides with the smallest curvature blow-up time corresponding to the orientation where $\tilde{V}$ is minimum. We were also able to prove that at any later time, curvature divergence is preempted by the formation of a shock: all orientations with $\tilde{V} < 0$ collide with the shock and hence always disappear prior to the blow-up of curvature for the corresponding orientation.

In the geometric evolution of crystal shapes we can observe the formation and/or expansion of facets. In particular, in Ref. [10] we suggested a simple geometric model that predicts an expansion of facets and that was able to capture major features observed experimentally in the earlier stages of surface evolution [5]. In this model the local normal velocity $V(\theta, \delta\mu)$ depends on a spatially uniform driving force, $\delta\mu$, and local orientation θ as follows:

$$V(\theta, \delta\mu) = V_{\mathrm{f}}(\delta\mu)\xi(\theta) + V_{\mathrm{r}}(\theta, \delta\mu).\qquad(4)$$

Here $V_{\mathrm{f}}(\delta\mu)$ is the normal growth rate at facet orientations

$$V_{\mathrm{f}}(\delta\mu) = c_{\mathrm{f}} g(\delta\mu)\exp\left(-\frac{\pi\sigma^2}{kT\delta\mu}\right),\qquad(5)$$

whereas for non-singular orientations we express the linear response to the growth drive as

$$V_r(\theta, \delta\mu) = c_r\mu(a|\sin 2\theta| - \sin^2 2\theta). \qquad (6)$$

Additionally, $\xi(\theta)$ is a smooth function that is maximum at facet orientations and decreases to zero at the orientations with the highest step density thus determining the nature of the transition between facet like and rough growth. In the expressions above c_f and c_r are mobility coefficients, $g(\delta\mu)$ is a smooth pre-exponential function of the growth drive, σ is the free energy of a critical nucleus on the facet (e.g., Refs. [11,12]), T is the temperature, and $\xi(\theta)$ is a smooth function periodic in $\pi/4$, with the properties: $\xi(0) = 1$ and $\xi(\pi/4) = 0$. In contrast to the form of $V(\theta, \delta\mu)$ suggested earlier [6], Eq. (4) incorporates a cusp at singular orientations, and hence as a result the facets expand. According to this model, if $2 < a < \frac{7}{3}$, the areas where the density of steps is maximum, grow fastest; nevertheless, curvature around $\theta = \pi/4$ *decreases*, while it *increases* at vicinal orientations. A shock is initiated in finite time at a location very close to the facet, and almost immediately collides with a facet. Thus, certain classes of geometric growth models, built around the velocity function that has a cusp at facet orientations, display a behavior in which a facet expands and collides with the shock in finite time. This leads to the formation of a "shocking facet"—an expanding facet separated from the rough orientations by a corner.

The evolution of a "shocking facet", as observed in the experiments [5], may result in a sharp decrease in the curvature within a certain distance from a corner and in the appearance of a "dynamical facet" at the roughest orientation. On the scale observed in optical microscopy, such "facets" are flat regions of the boundary characterized by a constant step density. Depending on experimental conditions, this may sometimes be followed by development of concavities that replace the dynamical facets [13].

As has been shown previously [7], geometric models, wherein the normal growth velocity depends only on the local orientation of the normal to the interface, preserve the convexity of the crystal shape. Additionally, they predict a gradual evolution of curvature at any surface orientation ($\kappa \sim t^{-1}$ for sufficiently large time t), only possibly interrupted by the sudden disappearance of the corresponding orientation altogether due to the formation of a shock. Any rapid drop of curvature at a given orientation while the latter exists cannot be a result of geometric evolution.

Thus, geometric models, while working well when applied to the problem of surface growth before the formation of a shocking facet, cannot describe the entire evolution of the shocking facets observed in Ref. [5].

The incompleteness of geometric models of crystal growth is not surprising because they are essentially kinematic in nature. They cannot be applied when the field variables are modified by the interface motion itself. As we show below, the very appearance of a shocking facet results in the onset of a long-range thermal diffusion in the melt between the facet and rough orientations. The supercooling at a given orientation changes with time: it increases as the location with this orientation approaches the facet. Therefore, the problem of shape evolution becomes intrinsically non-local and needs to be described using a more complete approach.

Firstly, let us review certain subtle features that can be discerned from the experiments of Maruyama [5]: (i) Before the shock reaches the facet ($t = t_0 \approx 4$ s), the velocity of a step at the edge of the facet, equal to the rate of facet expansion, is $v_{edge} \approx 12\,\mu m/s$; the velocity of the step at the roughest orientation $\theta = \pi/4$, is $v_{\pi/4} \approx 1\,\mu m/s$. (ii) After the formation of a "shocking facet", at $t > t_0$, $v_{edge} \approx 2-3\,\mu m/s$, while $v_{\pi/4}$ does not change, being equal to $1\,\mu m/s$; (iii) At $t > t_c \approx 17$s, when the arclength between the edge of the facet and the location on the surface with $\theta = \pi/4$ is of the order of $100\,\mu m$, the curvature around the roughest orientations drops sharply; v_{edge} does not change, while at the same time $v_{\pi/4}$ increases rapidly to become equal to v_{edge} when the dynamic facet forms at $t \approx 20$ s. After that, the entire rough region represents a plane with the orientation $\theta = \pi/4$, and all steps along this dynamic facet move with the same velocity. One of our goals is to qualitatively explain such distinct features of surface growth.

Below the roughening transition the surface grows due to the motion of steps. A step velocity is determined by the temperature field within the melt at the interface. Molecules attach almost exclusively to steps: attachment kinetics at terraces and facets are determined by nucleation processes which are exponentially slow at small growth drives. Under typical conditions, the rate of molecular attachment at the steps is much higher than the rate of removal of heat generated in the process of crystallization at the steps. Steps therefore behave mainly as heat sources while heat removal becomes a limiting factor in growth processes. The component of the temperature gradient normal to the interface is small in the vicinity of the surface areas with higher density of steps due to large effective density of heat sources there. Correspondingly, steps move slowly at rough orientations. On the other hand, step velocities at vicinal orientations are high due to the low density of heat sources that lead to a relatively large supercooling of the melt near the interface, taking its largest value at facet orientations. Normal growth velocities are nevertheless maximum at rough orientations: steps there need not move far to produce a noticeable motion of the interface in the normal direction. In the vicinity of the facet, on the other hand, even a significant displacement of a step results in a very little shift of the interface in the normal direction.

Before the formation of a corner, the step density at any given orientation changes very slowly. The thermal conditions at the interface change on a timescale much longer than the experimental one. In particular, on the experimental timescale, a total displacement of the interface in the normal direction is very small compared to a typical lengthscale on which the temperature changes [10], hence the effect of such a motion on the thermal field and normal growth velocities is negligible, and the temperature field and hence the normal growth velocity remain steady. These are the velocities that enter Eq. (1). In the following, we use the term "quasisteady state" to describe a thermal field before the formation of a shocking facet. When a corner forms between two rough regions with different but high step density, small changes in the local

growth velocity might only be expected in the narrow vicinity of a corner. Long-range effects are negligible, and the growth velocity itself can still be treated in the quasisteady context.

The situation changes dramatically with the formation of a shocking facet; a cold region of the melt above the facet appears in close proximity to the warm surface layer of the melt near rough orientations. The effect leads to the breakdown of the quasisteady state and the onset of thermal diffusion in the melt towards the facet. As a result, the temperature of the melt near the edge of the facet grows, while the melt temperature in the vicinity of rough orientations gradually decreases. Hence, the velocity of the tangential motion of the facet becomes lower, and the normal growth velocity at rough orientations increases.

As a shocking facet continues to move, further absorbing rough orientations, larger and larger areas of the surface are affected by its motion. In order to quantitatively describe the evolution of an interface containing a shocking facet, one has to solve a complete diffusion problem with a moving boundary. Our goal here is to illustrate the basic effects, and we do so using a simplified description based on the following assumptions. We start from the quasisteady state wherein temperature field is considered known at any location. The discrete distribution of steps is replaced by their continuous distribution, and we assume that a facet spreads with a constant velocity $v \equiv v_{\text{edge}}$. The propagation of a shocking facet is accompanied by the loss of rough orientations, which can be viewed as an elimination of steps and the replacement of an area of finite step density with a facet. Since each step effectively plays the role of a line heat source, the elimination of steps implies the disappearance of heat sources. The area of the surfaces that is absorbed by the shock during the initial stage of interfacial evolution consists mostly of the vicinal orientations and is therefore, to a first approximation, not very different from the flat surface. We may then visualize the process of elimination of vicinal orientations as a replacement of a flat area covered with the heat sources (steps) by a flat area with no heat sources (facets). Disappearance of the heat sources along the vicinal surface can then treated by considering an

expanding facet as a surface covered with the heat sinks whose surface density is identically the same as that of the heat sources that are being absorbed. The motion of such a surface leads to the annihilation of heat sources. We approximate the density of heat sources along the vicinal surface and hence, that of the heat sinks along the spreading facet as being constant. We also assume that this density does not change with time.

The last assumption requires explanation. Volume heat sources typically enter inhomogeneous terms of the diffusion equation. Surface sources are usually treated by imposing Stefan boundary condition on the jump of the heat flux at the surface: this jump is proportional to the normal growth velocity. In our simplified description we treat the surface sources as inhomogeneous terms. Such a treatment is clearly justified on a timescale during which the thermal field in the melt, and hence the normal velocity of the surface, is not modified by the motion of the interface itself; that is, when the geometric models are valid. Once the field begins to change, the normal velocities change as well, and hence the source terms in the diffusion equation are modified. Our goal below is to find when such modifications *begin* at different locations on the interface, marking the *end* of the applicability of geometric models.

As a shocking facet spreads, the area where no crystallization processes take place and hence, no heat is generated, expands. This enhances the thermal flux from the surviving rough orientations, decreases the surface melt temperature there, and leads to increasing normal growth velocities at rough orientations. The temperature field around an expanding surface with a constant density of heat sinks can be computed using Green's functions for the two-dimensional diffusion problem. The only phenomenon that affects the temperature at the interface is the motion of a shocking facet. According to the discussion above, we are focusing on timescales such that this effect is still small. Therefore, we compute the effect of this motion on the melt temperature at the interface as a perturbation to the thermal field around the surface before the formation of the shocking facet. Denoting $u = (T - T_\infty)/T_\infty$, where T_∞ is the melt temperature far away from the interface, we write

for the field at a point (x, z) at a time t (see Fig. 1)

$$u(x, z, t) = u_{\mathrm{st}}(x, z) - \int_0^t \mathrm{d}t' \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \mathrm{d}x' \mathrm{d}z'$$
$$\times G(x - x', z - z', t - t')Q(x', z', t'). \tag{7}$$

Here u_{st} is a thermal distribution before the formation of a shocking facet at $x = 0$,

$$G(x, z, t) = \frac{1}{4\pi\kappa t}\exp\left(-\frac{x^2 + z^2}{4\kappa t}\right) \tag{8}$$

is the Green's function, κ is the thermal diffusivity of the melt, and

$$Q(x, z, t) = q\theta(vt - x)\theta(x)\delta(z), \tag{9}$$

where $\theta(x)$ is Heaviside's step-function. The constant q is proportional to the amount of heat generated from a unit area of the surface in unit time, has the units of velocity and is related to the normal growth velocity V_n via $q = V_n S$, where $S = L/cT_\infty$, is a Stefan number, L is the latent heat of fusion, and c is the specific heat. Note that q is proportional to the assumed surface density of heat sinks.

Integrating over z' and replacing the θ-functions with the limits of integration over x', we obtain for $\tilde{u}(x, z, t) = u(x, z, t) - u_{\mathrm{st}}(x, z)$

$$\tilde{u}(x, z, t) = -\frac{q}{4\kappa}\int_0^t \mathrm{d}t' \int_0^{vt} \mathrm{d}x' \frac{\mathrm{e}^{-((x-x')^2+z^2)/4\kappa(t-t')}}{t - t'}$$
$$= -\frac{q}{4\kappa}\int_0^t \mathrm{d}t' \frac{\mathrm{e}^{-z^2/4\kappa(t-t')}}{t - t'} I_1, \tag{10}$$
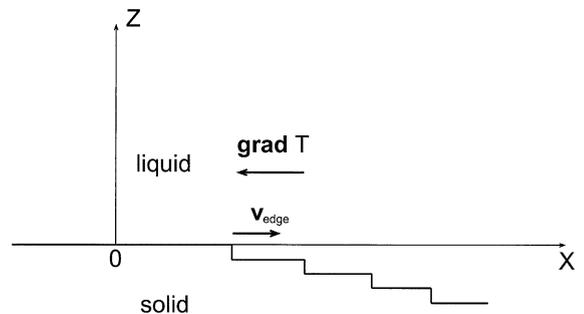

Fig. 1. A shocking facet forms at $x = 0$ at time $t = 0$, and moves with a constant velocity v_{edge}. Its motion leads to a large thermal flux away from rough orientations facilitating the removal of heat generated there in the process of crystallization, and thus increasing local normal growth velocities.

where

$$I_1 = \sqrt{4\kappa(t-t')} \int_{-a}^{-a+\varepsilon} dy\, e^{-y^2}, \tag{11}$$

where $a = x/\sqrt{4\kappa(t-t')}$ and $\varepsilon = vt/\sqrt{4\kappa(t-t')}$. Under experimental conditions, after the formation of a shocking facet $v < 5\,\mu m/s$. Since $\kappa \sim 10^{-7}\,m^2/s$, for any $t < 100\,s$, that is on the entire experimental timescale, we will have $vt \ll \sqrt{4\kappa t}$. Assuming then that small values of $t - t'$ do not contribute to the integral over t', so that $\varepsilon \ll 1$, we obtain

$$I_1 = \sqrt{4\kappa(t-t')} \int_{-a}^{-a+\varepsilon} dy\, e^{-y^2}$$
$$\approx \varepsilon \sqrt{4\kappa(t-t')}\, e^{-a^2}, \tag{12}$$

from which it is found that

$$\tilde{u}(x,z,t) \approx -\frac{qvt}{4\kappa} \int_{(x^2+z^2)/4\kappa t}^{\infty} \frac{d\eta}{\eta} e^{-\eta}. \tag{13}$$

It is seen that large values of η do not contribute to the above integral, thereby justifying our earlier assumption that $(t - t')/t \sim 1$. Two limits are of interest now. If $(x^2 + z^2)/4\kappa t \ll 1$, evaluation of Eq. (13) leads to the following expression:

$$\tilde{u}(x,z,t) = -\frac{q}{4\kappa} vt \left(-\gamma + \log \frac{4\kappa t}{x^2+z^2} \right), \tag{14}$$

where γ is the Euler constant. At short times, $t \ll (x^2 + z^2)/4\kappa$, $\tilde{u}$ is exponentially small. Since $\kappa \sim 10^{-7}\,m^2/s$, and the typical size of the rough part of the sample in Ref. [5] is approximately $0.1\,mm$, Eq. (14) is already applicable to the entire rough part of the interface at $t \sim 1\,s$.

The normal growth velocity of the interface between the crystal and its melt is determined by the normal to the surface component of the gradient of temperature computed at the interface. Considering for simplicity a rough part of the interface of circular shape with radius R, we calculate the temperature gradient at the interface using Eq. (14), and find

$$(\vec{\nabla}\tilde{u}(t))_n = \frac{q}{2\kappa} \frac{vt}{R}. \tag{15}$$

We observe that in the simplified picture we are exploring here, the change of the temperature gradient and hence, the change of the local normal growth velocity are independent of the location along the interface. In general, deviations from our approximation may only lead to a weak dependence of $(\vec{\nabla}\tilde{u})_n$ on the interface coordinate, which implies that, with the exception of a very narrow region near the edge of the facet, the effect of the expanding shocking facet on the normal growth velocity at any given time varies weakly with location along the rough part of the interface. We have already employed the fact that, in the quasisteady state, the normal growth velocities observed in Ref. [5] and predicted by Eq. (6), vary only slightly within rough orientations. Since both the initial local interface velocities and their change due to the formation of a shocking facet vary weakly along the rough orientations, and the change grows almost linearly with time, we expect that significant deviations of the interface normal growth velocity from its values in the quasisteady state will begin within a short period of time on the entire interface.

From Eq. (15) we can compute the timescale over which the geometric models are valid: these are the times when $(\vec{\nabla}\tilde{u})_n \ll (\vec{\nabla}u_{st})_n$. Since $V_n = \kappa(\vec{\nabla}T)_n/ST_\infty$ and $(\vec{\nabla}\tilde{u})_n = (\vec{\nabla}T)_n/T_\infty$, we find $(\vec{\nabla}u_{st})_n = SV_n/\kappa = q/\kappa$. Then, $(\vec{\nabla}\tilde{u})_n/(\vec{\nabla}u_{st})_n = vt/2R$. In the experiments of Maruyama [5], the velocity of the edge of the facet $v \approx 3\,\mu m/s$ and $R \sim 100\,\mu m$. Hence, the above inequality starts to fail at $t \sim 10\,s$, and our model predicts that this is the time when the deviations from geometric evolution begin to appear.

Such a behavior is in fact observed in Ref. [5] where the deviations from the geometric evolution for the entire interface are initiated only shortly before $t = 20\,s$, when a dynamic facet is formed. At $t > 15\,s$, changes in the growth velocities begin and velocities of the areas closer to the edge of the shocking facet increase faster than those of the roughest part of the interface leading to the flattening of the surface. Prior to $t = 15\,s$, the motion of the entire interface can be described within the geometric model with a high degree of accuracy.

One may notice that Eq. (15) gives only linear time deviations in $(\vec{\nabla}\tilde{u})_n$, and in the experiments of Maruyama [5], the linear time regime is very short. This is not surprising: our simple model assumes that the heat flux from the surface does not change

with time, and is therefore applicable only when the deviations from the quasisteady state are small. However, as the thermal field near the surface changes, the growth velocity increases, and so does the amount of heat emitted from a unit area of the interface in unit time. Therefore, q in Eq. (9) grows as well, and non-linear effects start to be noticeable. A detailed description of such a non-linear evolution can only be obtained by solving a complete transient diffusion problem, but the essence of the non-local influence of facet evolution is captured herein.

Clearly, the early stages of the evolution of a crystal surface below the roughening transition, when major changes on the surface take place without any significant normal growth, are well described by the geometric models. A certain class of these models wherein the normal growth velocity has a cusp at facet orientations, predicts the expansion of the facet, the development of a corner, and the possible formation of a shocking facet. We have shown that the same geometric model is applicable even for some time after the formation of a shocking facet. Deviations from the geometric evolution due to a thermal diffusion flux away from the rough orientations towards the facet are predicted to appear almost simultaneously along the entire rough part of the surface. Such deviations lead to the formation of dynamic facets observed in the experiments.

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