FACETED CRYSTALS GROWN FROM SOLUTION – A STEFAN TYPE PROBLEM WITH A SINGULAR INTERFACIAL ENERGY

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Abstract. We present a one-phase quasi-steady Stefan problem with Gibbs-Thomson and the kinetic effects when the interfacial energy is singular so that the equilibrium shape is a cylinder. We derive this model to describe crystal growth from vapor or solution. We summarize mathematical results on this model. Among other results we prove that a cylindrical shape is preserved if the initial cylindrical shape of a crystal is close to the equilibrium shape. Our formulation allows the possibility that cylindrical shape may break.

1 Introduction

The purpose of this note is to highlight what we have achieved in [10]-[14] while studying a Stefan type problem with singular interfacial energy which describes an evolution of a crystal grown from vapor or solution.

We consider a crystal growing from vapor or solution. The common feature of the growth is that concentration of atoms outside crystal is very small, compared with that in the crystal. This feature is different from the crystal solidifying from the melt where there are as many atoms outside the crystal as inside it. In the latter case the driving force of the crystal growth is the supercooling while in the former case it is a supersaturation of pressure or concentration.

We formulate our problem as a one-phase Stefan type problem like it is done by many physicists e.g. [24]. To be specific let us consider the crystal growth from solution since the problem can be mathematically formulated in the same way while physical meaning of quantities is different. Let $D(t)$ be a bounded open set in $\mathbf{R}^3$ depending on $t \geq 0$. It describes the crystal at time t. Let $C = C(x,t)$ represent the concentration of atoms at place $x \notin D(t)$ and time $t \geq 0$. Let C_e be the saturated concentration which is a constant (independent of x and t). Let $V = V(x,t)$ be the normal velocity of the crystal surface $S(t) = \partial D(t)$ at a point $x \in \partial D(t)$ in the direction of the unit outer normal $\mathbf{n} = \mathbf{n}(x,t)$. Let us present the physical setting.

(i) Atoms in a solution are transported by diffusion and this transport is much faster than the motion of the crystal surface $S(t)$. Mathematically speaking, we consider the Laplace equation

$$\Delta C = 0 \quad \text{in} \quad \mathbf{R}^3 \backslash \overline{D(t)} \tag{1.1}$$

which is derived from the heat equation with the diffusion coefficient $D_C > 0$, by dropping the term $\partial C/\partial t$.

(ii) The crystal grows by catching atoms. In this process the number of atoms is conserved. It can be written

$$v_c D_C \frac{\partial C}{\partial \mathbf{n}} = V \quad \text{on} \quad S(t), \tag{1.2}$$

where v_c represents the volume of an atom of the crystal and D_C is the diffusion coefficient of atoms in the solution. This is a Stefan-like condition. These two conditions are rather standard as in [24].

The next condition models the surface kinetics of adatoms on the crystal surface.

(iii) We assume that the normal velocity is proportional to supersaturation plus anisotropic curvature depending on anisotropy of the crystal structure. The curvature effect is stable under small scale perturbation. This effect is called the Gibbs-Thomson effect. The equation reads

$$\tilde{\beta}(\mathbf{n})V = C - C_e - \mathrm{div}_S \tilde{\xi}(\mathbf{n}) \quad \text{on} \quad S(t), \tag{1.3}$$

where div_S denotes the surface divergence [22]. The vector $\tilde{\xi}$ is the gradient field of a given interfacial energy (density) $\tilde{\gamma}(p)$ and $\tilde{\beta}$ is the kinetic coefficient depending on $\mathbf{n}$. Both reflect the anisotropic structures of crystals.

It is natural to assume that $\tilde{\beta} > 0$. For the interfacial energy $\tilde{\gamma}$ we postulate that $\tilde{\gamma}$ is positively homogeneous of degree one, i.e.

$$\tilde{\gamma}(\lambda p) = \lambda \tilde{\gamma}(p)\ ,\ p \in \mathbf{R}^3\ ,\ \lambda > 0$$

and $\tilde{\gamma}$ is convex and positive for $p \neq 0$. These assumptions are also natural. The quantity $-\mathrm{div}_S \tilde{\gamma}(\mathbf{n})$ is often called a weighted mean curvature. In fact, if $\tilde{\gamma}(p) = |p|$, then $-\mathrm{div}_S \tilde{\gamma}(\mathbf{n})$ is nothing but the (two times) mean curvature. The equation (1.3) is consistent with the derivation of interface equations by [17]; note that it can be replaced by nonlinear dependence in V i.e.

$$V = f(\mathbf{n}, C - C_e - \mathrm{div}_S \tilde{\xi}(\mathbf{n})) \tag{1.3$'$}$$

for a given f, nondecreasing in the second variable with $f(\mathbf{n}, 0) = 0$.

The system (1.1)-(1.3) is a quasi-steady approximation of one-phase Stefan problem with Gibbs-Thomson and the kinetic effects. This system is supplemented with a prescribed concentration at the space infinity

$$\lim_{|x| \to \infty} C(x,t) = C_\infty \qquad (> C_e). \tag{1.4}$$

It is also supplemented with initial condition for a crystal shape

$$D(0) = D_0. \tag{1.5}$$

We are interested in solvability for (1.1)-(1.5) when γ may not be C^1 so that the curvature term in (1.3) is a nonlocal quantity. Such singular energy is important for material sciences and for crystal growth in low temperature; see e.g. [17], [23] and [7] Although there are several articles related to a quasi-steady approximation of a one-phase Stefan problem with Gibbs-Thomson and the kinetic effects, as mentioned in [10] none of the existing articles study such a situation with singular energy except for ours; see papers cited in [10]. The equation (1.3′) is often used (see e.g. [24]) to study the growth when $\mathbf{n}$ directs to the singularities of γ.

We consider (1.1)-(1.5) when the Frank diagram

$$\text{Frank } \tilde{\gamma} = \{p \in \mathbf{R}^3; \tilde{\gamma}(p) \leq 1\}$$

consists of two straight cones with a common base (in the $p_1 p_2$ plane), which is a disk centered at the origin of the plane. This hypothesis implies that its polar set-Wulff shape

$$W_{\tilde{\gamma}} = \bigcap_{|m|=1} \{x \in \mathbf{R}^3 \ ; \ x \cdot m \leq \tilde{\gamma}(m)\}$$

is a regular (circular) cylinder. (This set is important in the sense that it represents an equilibrium of the crystal shape.) These assumptions of course simplify the issue but such a kind of simplification is often done in the theory of crystal growth; see e.g. [25]. For $\tilde{\beta}$ we mostly consider the case that W_β is also a dilation of γ.

We now summarize what we have achieved. First, we note that the formulation of the problem itself is troublesome because the curvature term in (1.3) must be a nonlocal quantity.

(a) **Solvability of the averaged problem.** We construct a local-in-time unique solution when initial shape is a cylinder (may not be $W_{\tilde{\gamma}}$) under the assumption that $D(t)$ *stays as a cylinder* (cf. [10]).

(b) **Berg's effect.** We prove that if $D(t)$ in (a) is growing, the concentration must be monotone in a crystal surface, in the sense that at the center it must be smaller while at the edge it must be larger (cf. [11]).

(c) **Existence of self-similar solutions.** We prove that for a special choice of $\tilde{\gamma}$ (satisfying $\tilde{\gamma}\tilde{\beta} = \text{const}$) there is a self-similar solution in the sense of (a) (cf. [12]).

(d) **Stability of facet for self-similar solutions.** We examine the equation (1.3) carefully and derive a necessary and sufficient condition guaranteeing that none of the parts of the surface of the cylinder (called facets) does not bend. By Berg's effect there is a tendency that the edge grows faster than the center. If the crystal shape is near equilibrium, the (nonlocal) curvature in (1.3) prevents such a bending. But if the size is large, this cannot be achieved. If the size is smaller than equilibrium so that crystal shrinks, the facet may break unless the shape is very small. We have proved these phenomena in a rigorous framework in [13].

(e) **Stability of facet for general situation.** Without assuming that the solution is self-similar we prove that a facet does not bend near equilibrium shape. For this purpose we study the phase plane of the system of ordinary differential equations, describing the averaged problem in (a). This is the main goal of [14].

A morphological stability (d), (e) is discussed theoretically without taking into account the curvature effect by physicists, for example by [19]. In [16] the experimental studies are performed. Our conclusion agrees with [16] in the sense that if the crystal is close to near equilibrium, the cylindrical shape is preserved. However, according to their observation cylindrical shape again seems stable if it is large but not too large. Our model does not foresee such phenomena at least for self-similar solutions. We note that one may easily replace (1.3) by (1.3$'$) in a trivial way and that (a)-(e) hold even if (1.3) is replaced by (1.3$'$) provided that $f(\mathbf{n}, 0) = f(-\mathbf{n}, 0)$.

We close this introduction by mentioning a recent review article by Adams [1] on morphological stability of snow crystals, a crystal grown from vapor. Several models are proposed there. Some are close to ours. However, the (nonlocal) curvature term in (1.3) is missing. Often the geometric model without curvature term i.e. $\beta V = C - C_e$ is studied to discuss the behavior of facets assuming that C is a constant. We note that if C is not a constant, one cannot keep a facet if there is no nonlocal curvature.

There is another review [15] but the present one provides more physical background.

2 The averaged problem

We normalized the problem (1.1)-(1.5) by scaling time to get

$$\Delta\sigma = 0 \quad \text{in} \quad \mathbf{R}^3\backslash\overline{D(t)} \tag{2.1}$$

$$\frac{\partial\sigma}{\partial\mathbf{n}} = V \quad \text{on} \quad S(t) \tag{2.2}$$

$$\beta(\mathbf{n})V = \sigma - \mathrm{div}_S\xi(\mathbf{n}) \quad \text{on} \quad S(t) \tag{2.3}$$

$$\lim_{|x|\to\infty} \sigma(x,t) = \sigma_\infty \tag{2.4}$$

$$D(0) = D_0 \tag{2.5}$$

with supersaturation $\sigma = (C - C_e)/C_e$. Here β and γ is a positive constant multiple of $\tilde{\beta}$ and $\tilde{\gamma}$, respectively, and $\sigma_\infty = (C_\infty - C_e)/C_e$.

We set

$$\gamma(p_1,p_2,p_3) = (p_1^2+p_2^2)^{1/2}\gamma_\Lambda + |p_3|\gamma_{TB}\ ,\ \gamma_\Lambda,\gamma_{TB} > 0 \tag{2.6}$$

so that the Wulff shape

$$W_\gamma = \{x \in \mathbf{R}^3\ ,\ x_1^2+x_2^2 \le \gamma_\Lambda^2\ ,\ |x_3| \le \gamma_{TB}\}.$$

For β, we at least assign the value $\beta_T = \beta_B > 0$ at $(0,0,\pm1)$ and $\beta_\Lambda(>0)$ at $(m_1,m_2,0), m_1^2+m_2^2 = 1$. Since γ is not C^1, we shall introduce a subdifferential

$$\partial\gamma(p) := \{q \in \mathbf{R}^3\ ;\ \gamma(p+h)-\gamma(p) \ge h\cdot q \text{ for all } h \in \mathbf{R}^3\}$$

instead of the usual gradient $\nabla\gamma$. The interpretation for (2.3) is

$$\beta(\mathbf{n})V = \sigma - \mathrm{div}_S\xi(x)\ ,\ \xi(x) \in \partial\gamma(\mathbf{n}(x))\ ,\ x \in S(t). \tag{2.7}$$

(The vector field ξ is often called Cahn-Hoffman vector.) Our problem is now formulated in the form of

(P) : (2.1), (2.2), (2.4), (2.5), (2.6), (2.7).

We would like to consider (P) with initial data

$$D_0 = \{(x_1, x_2, x_3) \ ; \ x_1^2 + x_2^2 < R_0^2 \ , \ |x_3| < L_0\}. \tag{2.8}$$

which is a regular cylinder. Our formulation (P) allows the possibility that cylindrical shape may bend.

Let us now derive an averaged problem. Suppose now that $D(t)$ is a cylinder of the form

$$D(t) = \{(x_1, x_2, x_3) \ ; \ x_1^2 + x_2^2 < R(t)^2 \ , \ |x_3| < L(t)\}, \tag{2.9}$$

with $R(t) > 0$, $L(t) > 0$ for $t \in [0, T)$. We distinguish three parts of the surface $S(t)$: top S_T, bottom S_B and the lateral part S_Λ i.e.

$$S_T = S(t) \cap \{x_3 = L(t)\} \ , \ S_B = S(t) \cap \{x_3 = -L(t)\}$$
$$S_\Lambda = S(t) \cap \{x_1^2 + x_2^2 = R(t)^2\}.$$

The normal to S_i is denoted by $\mathbf{n}_i$, $i = \Lambda, B, T$. If $D(t)$ is of the form (2.9), then the normal velocity V is spatially constant V_i on each S_i , $i = \Lambda, B, T$ and $V_T = V_B$. (The converse is also true.) Let $|S_i|$ denote the area of S_i. We average (2.7) over S_i to get

$$\beta_i V_i = \frac{1}{|S_i|} \int_{S_i} \sigma \mathrm{d}S + \kappa_i \tag{2.10}$$

with $\beta_i = \beta(\mathbf{n}_i)$. Here κ_i denotes the crystalline curvature defined by

$$\kappa_\Lambda = -2\frac{\gamma(\mathbf{n}_\Lambda)}{R(t)} \ , \ \kappa_T = -\frac{\gamma(\mathbf{n}_L)}{R(t)} - \frac{\gamma(\mathbf{n}_T)}{L(t)}$$

In fact, we first note that if $\xi(x) \in \partial\gamma(\mathbf{n}(x))$ on $S(t)$, then ξ on $S_i \cap S_j \in \partial\gamma(\mathbf{n}_i) \cap \partial\gamma(\mathbf{n}_j)$. This implies that the normal trace of ξ in S_i to its geometric boundary is uniquely determined. By this observation we are able to prove ([[13], Proposition 2.1]) that

$$\int_{S_i} \mathrm{div}_S \xi \mathrm{d}S = -\kappa_i |S_i|. \tag{2.11}$$

under suitable regularity assumption on ξ. The formula (2.10) follows immediately from (2.11)

We call the problem

$$\text{(A)} \;:\; (2.1), (2.2), (2.4), (2.5), (2.6), (2.10) \text{ with } (2.9)$$

the *averaged problem* of the original problem. Since $V_\Lambda = \mathrm{d}R/\mathrm{d}t$, $V_T = V_B = \mathrm{d}L/\mathrm{d}t$, our averaged problem is a system of ordinary differential equations for R and L. In fact, let f_i be the unique weak solution of the Neumann problem

$$-\Delta f_i = 0 \quad \text{in} \quad \mathbf{R}^3 \backslash \overline{D(t)}\ ,\ \frac{\partial f_i}{\partial \mathbf{n}} = -\delta_{ij} \quad \text{on} \quad S_j$$

with $\lim_{|x|\to\infty} f_i(x) = 0$ for $i = T, B, \Lambda$. The solution of (2.1), (2.2), (2.4) can be expressed as

$$\sigma = -\sum_{i\in I} V_i f_i + \sigma_\infty\ ,\ I = \{T, B, \Lambda\}. \tag{2.12}$$

By definition of f_i

$$\int_{S_i} \varphi \mathrm{d}S = \int_{\mathbf{R}^3\backslash\overline{D(t)}} \nabla\varphi \cdot \nabla f_i = ((\nabla\varphi, \nabla f_i))$$

for all φ with $\nabla\varphi \in L^2(\mathbf{R}^3\backslash\overline{D(t)})$. Since f_i is determined by R and L, we plug (2.12) into (2.10) to get an ODE for $R(t)$ and $L(t)$:

$$\sum_{i\in I} V_i((f_i, f_j)) - |S_j|\sigma_\infty = |S_j|\kappa_j - \beta_j V_j |S_j| \qquad j = T, \Lambda, B. \tag{2.13}$$

Actually, this ODE system is locally uniquely solvable since $(R, L) \mapsto ((f_i, f_j))$ is locally Lipschitz, [10]. We thus observe that

Theorem 1([[10], Theorem1]). *The averaged problem admits a unique local-in-time solution for arbitrary initial cylinder D_0 of the form (2.8).*

The ODE system (2.13) for $(R(t), L(t))$ admits a unique equilibrium point

$$z_0 = \frac{2}{\sigma_\infty}(\gamma_\Lambda, \gamma_{TB}).$$

Fortunately, this ODE system can be expressed as

$$\frac{dz}{dt} = Az + F(z)$$

with $z = (R, L) - z_0$. Here A is 2×2 matrix having negative determinant and $F(0) = 0$ with

$$\lim_{\sigma \to 0} \sup_{|x|,|y| \le \sigma} |F(x) - F(y)| \ \ /|x - y| = 0.$$

Then by a standard theory of ODEs, [18], we observe that there are one dimensional stable and unstable manifold through z_0 as discussed in [14]. This will help to bound $|V_\Lambda / V_T|$ near the equilibrium point, which is useful to conclude that a facet does not bend near equilibrium even for original problem.

3 Relation to the original problem

The original problem (P) and the averaged one may be different since there may not exist $\xi \in \partial\gamma(\mathbf{n}(x))$ satisfying (2.7). In fact, as noted in [13] the solution of (A) is a solution of (P) if and only if for each i there exists ξ satisfying

$$\sigma - \mathrm{div}_S \xi(x) = \ \text{const} \quad \text{on} \quad S_i, \tag{3.1}$$

$$\xi(x) \in \partial\gamma(\mathbf{n}_i) \quad \text{on} \quad S_i(\backslash S_j)(i \neq j), \tag{3.2}$$

$$\xi(x) \in \partial\gamma(\mathbf{n}_i) \cap \partial\gamma(\mathbf{n}_j) \quad \text{on} \quad S_i \cap S_j (i \neq j). \tag{3.3}$$

If such ξ does not exist, the crystal shape $D(t)$ of (P) cannot stay as a cylinder. Either lateral or top may become bent. If such ξ exists it is a minimizer of an obstacle type variational problem: minimize $\int_{S_i} |\mathrm{div}_S \xi - \sigma|^2 \mathrm{d}S$ with a constraint $\xi \in \partial\gamma(\mathbf{n}(x))$ (cf. [13]). We say S_i is *stable* if S_i admits ξ satisfying (3.1)-(3.3). Using this variational characterization, we are able to obtain a criterion for the stability of a facet. We just give it for top. We suppress the time variable since the minimization problem does not depend on time explicitly.

Theorem 2([[13], Theorem 4.6]). *Assume that $\sigma = \sigma(\sqrt{x_1^2 + x_2^2}, x_3)$ satisfies $\nabla\sigma \in L^2(\mathbf{R}^3 \backslash \overline{D(t)})$ and that σ is even in x_3. Facet S_T is stable if and only if*

$$-\gamma(\mathbf{n}_\Lambda) \le \frac{r}{R}\gamma(\mathbf{n}_\Lambda) + \frac{r}{2}(\overline{\sigma_r} - \overline{\sigma_R}) \le \gamma(\mathbf{n}_\Lambda) \quad \text{for all} \quad r \in [0, R]. \tag{3.4}$$

Here $\overline{\sigma_r}$ is the average of σ over $S_T(r) = S_T \cap \{x_1^2 + x_2^2 \le r^2\}$, i.e.,

$$\overline{\sigma_r} = \frac{1}{|S_T(r)|} \int_{S_T(r)} \sigma \ \mathrm{d}S.$$

This is a general criterion for existence of ξ satisfying (3.1)-(3.3) for general given σ which may not satisfy (2.1), (2.2), (2.4).

If σ fulfills (2.1), (2.2) and (2.4), we observe Berg's effect which goes back to [5] and was proved by [21] when the crystal shape is a regular polygon. It asserts that if the crystal shape is growing, then supersaturation near a corner or an edge is larger than that in the center. This is also extended by [20] for a cylinder. Here we need a stronger version.

Theorem 3 [[11], Theorem 1]. *Let $D = D(t)$ be a cylinder of the form (2.9). Let σ be a unique solution to*

$$\Delta\sigma = 0 \quad \text{in} \quad \mathbf{R}^3 \backslash \overline{D}$$

$$\frac{\partial\sigma}{\partial\mathbf{n}} = V_i \quad \text{on} \quad S_i \quad i = \Lambda, T, B,$$

where $\sigma = \sigma(r, x_3)$, $\sigma(r_1, -x_3) = \sigma(r, x_3)$ and V_i , $i = \Lambda, T, B$, are constants. Moreover, $V_T = V_B$. Then we have
(a) If $V_T > 0$ then $\partial\sigma/\partial x_3 > 0$ for $x_3 > 0$ on S_Λ.
(b) If $V_\Lambda > 0$ then $\partial\sigma/\partial r > 0$ on $S_T \bigcup S_B$.

This can be proved by the maximum principle for $\partial\sigma/\partial r$ and $\partial\sigma/\partial x_3$ formally. But we should worry about the regularity of these quantities. The paper [11] overcame such difficulties.

From this version of Berg's effect it is clear for example that

$$\overline{\sigma_r} - \overline{\sigma_R} < 0 \quad \text{for all} \quad r \in [0, R] \quad \text{if} \quad V_\Lambda > 0.$$

Then the second inequality of (3.4) is automatically fulfilled. So the inequality (3.4) is reduced to

$$\frac{r}{2}(\overline{\sigma_R} - \overline{\sigma_r}) \leq \gamma(\mathbf{n}_\Lambda)(1 + \frac{r}{R}).$$

Our next question is a relation between stability of facets, size of crystals and prescribed supersaturation σ_∞. Since there are too many parameters, we first consider a self-similar solution which only exists for particular aspect ratio of Wulff shape cylinder.

Theorem 4 ([[12], Theorem 4.8]). *There exists a choice of γ and β satisfying*

$$\beta \cdot \gamma = \text{ const}$$

for which $S(t) = a(t)W_\gamma$, $a(0) = 1$ is a solution of the averaged problem (A).

Using Theorem 2 with help of Berg's effect, we are able to state our stability result depending on the size of crystal and σ_∞. We just confine ourselves just to S_T.

Theorem 5 ([[13], Theorem 4.8]). *Let γ and β be as in Theorem 4. Suppose that $\sigma^\infty > 2$ (so that crystal grows). The facet S_T is stable if and only if*

$$\frac{a(t)(\sigma^\infty a(t) - 2)C_T}{\beta_T + a(t)C_T} \leq \overline{d_T}$$

where C_T and $\overline{d_T}$ are positive constants depending only on W_γ. (A similar result holds for S_Λ).

This results says that near the equilibrium shape ($a(t) \approx 1$) a facet is stable so in this case the solution of (A) is a solution of (P). It turns out that this is in fact true for the general evolution, as proved in [14].

Theorem 6 ([[14], Theorem 4.7]). *Assume that γ is of the form (2.6) and $\beta_T = \beta_B > 0$, $\beta_\Lambda > 0$. Let (R_0, L_0) be close to the equilibrium z_0 in (2.14). Then the solution of the averaged problem (A) with initial data (2.8) solves (P) for a short time. In particular, facets S_T, S_B, S_Λ are stable.*

As alluded at the end of section 2, it is important to get a bound for velocity ratio $|V_\Lambda/V_T|$. If the evolution is self-similar, this is easy. In general, one is able to control this quantity near equilibrium.

We do not go into any technical details. Moreover, we did not intend to exhaust all main results so that this review is accessible for a large class of audience.

We conclude this paper by pointing out several open problems.

(i) Uniqueness: We do not know whether the solution of (P) is unique even if there is a solution of (A) solving (P). This seems to be an issue of clarifying the class of solutions. In related problems [6], [9], [2], [4] the correct choice of ξ is the minimizer of $\int_S |\mathrm{div}\xi - \sigma|^2 \,\mathrm{d}S$ under $\xi \in \partial\gamma(\mathbf{n}(x))$. If this is proved, the velocity must be constant provided that facets are stable. So all solutions of (P) would agree with a solution of (A), so that the solution is unique.

(ii) After bending : If the facet stability condition is violated, construction of a solution of (P) is a nontrivial business. Even if σ is given this is not trivial as pointed out by [8].

(iii) It is interesting to know which facet S_T or S_Λ becomes unstable first for the self-similar solution. This is an interesting question in physics [16]. Our criterion in Theorem 5 will answer the question provided that the values C_T and $\overline{d_T}$ as well as C_Λ and $\overline{d_\Lambda}$ are clarified. However, to know such constants we should calculate f_i's.

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