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Faceted needle crystals: an analytical approach

Mokhtar Adda-Bedia([1]) and Vincent Hakim([1,2])

([1]) Laboratoire de Physique Statistique, 24 rue Lhomond, F–75231 Paris Cedex 05, France
([2]) D.A.M.T.P., University of Cambridge, Silver Street, Cambridge CB3 9EW, U.K.

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Résumé. — Des dendrites facettées ont été observées dans quelques expériences de croissance cristalline. Il a été antérieurement proposé de décrire ces nouvelles formes de croissance limitée par la diffusion au moyen des équations classiques de la croissance dendritique en utilisant des conditions aux limites modifiées sur l'interface. Nous analysons ce nouvel ensemble d'équations en négligeant la tension de surface sur les parties rugueuses. Dans cette limite, nous trouvons qu'il n'est pas possible d'imposer à la fois un raccord tangentiel des parties rugueuses et facettées et de maintenir les interfaces rugueuses avant et arrière à la même temperature de fusion. Une solution exacte est obtenue en relâchant une de ces conditions physiques. Nous discutons les conséquences de ce résultat pour le problème complet et une solution approchée en est proposée.

Abstract. — Faceted dendrites have been observed in some crystal growth experiments. It has previously been proposed that these diffusion-limited growing shapes obey the classical equations of dendritic growth with modified boundary conditions on the interface. We analyse this new set of equations when capillary effects are neglected on the rough parts. In this limit, we find that it is not possible to require both a tangential matching of the rough and faceted parts and the same melting temperature on the front and trailing rough interface. An exact solution is obtained when one of these physical constraints is relaxed. The consequences of this result for the full problem are considered and an approximate solution is proposed.

1. Introduction.

During the last decade, much progress has been made in the understanding of interfacial pattern formation. In particular, a consistent theory now exists for the simplest steady state patterns of diffusion limited growth like the viscous fingers of diphasic flow in Hele-Shaw cells or the needle crystals of dendritic growth [1]. In the latter context, it has mostly been applied to systems with small and smooth capillary anisotropy. It seems interesting to see whether the theory is able to describe slightly more complex situations. In this paper, we consider the case of a dendrite growing below the roughening temperature. This has recently been the subject of some experimental investigations and observations of 'faceted dendrites' have been reported [2]. On the theoretical side, the classical Gibbs-Thomson condition relating the interface curvature to its undercooling has been generalized to the diffusion-limited growing faceted case [3, 4] following earlier work [5]. At present, apart from some general dimensional considerations [3], the resulting set of equations has also been applied to examine the linear stability of a moving facet [6]. Our aim is to analyse the case of a two dimensional steady-state needle crystal. In the low undercooling limit [7], we analytically obtain a faceted generalization of Ivantsov's classical solution [8] by neglecting capillary effects on the rough parts of the interface. Our rather unexpected result is that such needle crystals with symmetric facets on each flanks mathematically exist, in this limit, only if a physical constraint is relaxed. We obtain a solution by allowing either a non-tangential matching of the rough and faceted parts, or a different melting temperature of the front and trailing rough parts. The possible implications of this result for the complete physical problem (with capillary effects on the rough parts) are briefly analysed. It is suggested that our analytical solutions with large facets are good approximations to the solutions of the full equations in the limit where capillary effects are formally small (large C limit (see Eq. (6) below)).

2. Diffusion-limited faceted growth in the low supersaturation limit.

We analyse the one-sided model of solidification where diffusion in the solid phase is neglected. It is well adapted to experiments where impurity diffusion is the main growth limiting factor [2]. Since we intend to look for needle crystal solutions it is convenient to non-dimensionalize lengths so that the equation of the asymptotic parabolic behavior at infinity be simply $x \sim -y^2$. That is, we choose 2ρ as length unit where ρ is the tip radius of curvature of the asymptotic parabola. A needle moving at constant velocity V in the x-direction satisfies the following equations in its reference frame (see [1])

$$\nabla^2 u + 4\mathrm{Pe}\frac{\partial u}{\partial x} = 0 \qquad (1)$$

$$n \cdot \nabla u = -4\mathrm{Pe}\cos\theta \qquad (2)$$

$$u_{\mathrm{int}} = \Delta - \frac{d_0}{2\rho R}[w(\theta) + \frac{\mathrm{d}^2 w}{\mathrm{d}\theta^2}] \qquad (3)$$

Equation (1) is the stationary diffusion equation in a moving frame for a suitably non-dimensioned diffusion field u. The Péclet number is the ratio of the parabola tip radius to the diffusion length $\mathrm{Pe} = \rho V/2D$. Equations (2) and (3) are the boundary conditions on the interface. Equation (2) describes the conservation of the diffusing quantity at the interface and determines the normal gradient of u at the interface as a function of the angle θ between the local normal to the interface and the x-axis (see Fig. 1). Equation (3) is the quasi-equilibrium Gibbs-Thomson condition which determines u itself on the interface. The departure of u from the (non-dimensioned) undercooling (or supersaturation) Δ is due to capillary effects which are given by the capillary length d_0, the interface radius of curvature R and $w(\theta)$ which describes the surface tension dependence on direction ($w = 1$ in the isotropic case). These equations can be simplified in the small Péclet limit where the diffusion length $2D/V$ is much larger than the parabola tip radius of curvature [7]. Then, the problem can be decomposed into an asymptotic region, where capillary effects can be neglected and the shape is a simple parabola, and a tip region. In the neighbourhood of the needle tip, one can take the limit $\mathrm{Pe} \to 0$ and solve a Laplacian problem for the field $\phi = \frac{\Delta - u}{4\mathrm{Pe}}$ which has a finite nontrivial limit

$$\nabla^2 \phi = 0 \qquad (4)$$

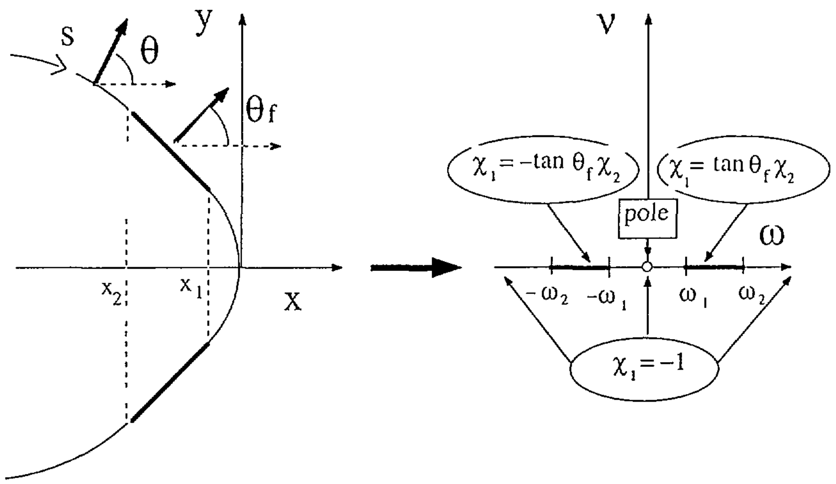

Fig. 1. — Faceted needle crystal in the physical z-plane and in the Ω-coordinate system.

$$n \cdot \nabla\phi = \cos\theta \tag{5}$$

$$\phi_{\text{int}} = \frac{1}{CR}[w(\theta) + \frac{\mathrm{d}^2 w}{\mathrm{d}\theta^2}] \tag{6}$$

This reduced problem depends on the single parameter $C = 8\rho\mathrm{Pe}/d_0$. It has a solution only for special values of C which is thus a kind of nonlinear eigenvalue [7]. The velocity V and length scale ρ are completely determined once the Ivantsov relation for the asymptotic parabola is taken into account $\Delta \asymp \sqrt{\mathrm{Pe}\ \pi}$.

How are these equations modified in the faceted case ? Then, $w(\theta)$ has a conic point for some specific direction θ_{f} with two different derivatives $\mathrm{d}w/\mathrm{d}\theta\ |_+$ and $\mathrm{d}w/\mathrm{d}\theta\ |_-$ and equation (6) is obviously ill defined there. In reference [3], the replacing condition [5] was very simply obtained by imagining the conic point as the limit of more and more peaked smooth anisotropy for which equation (6) is still valid. Then, averaging equation (6) in the region of rapid variation gives (recalling that $1/R = -\mathrm{d}\theta/\mathrm{d}s$)

$$\int_{\text{facet}} \mathrm{d}s\ \phi_{\text{int}} = \frac{1}{C}[\mathrm{d}w/\mathrm{d}\theta\ |_+ - \mathrm{d}w/\mathrm{d}\theta\ |_-] \tag{7}$$

where the integration is taken over the facet length. This single condition on the average field replaces the local Gibbs-Thomson condition on the facet. It can be thought of as determining the only unknown geometric characteristic of the facet i.e. its length. For simplicity, we suppose in the following that the Wulff plot contains no missing angles (i.e. no orientations for which $w + w" < 0$ [9]), as in the experimental case of reference [2]. The facets should therefore match tangentially the contiguous rough parts.

So, the determination of a faceted needle crystal is a mixed problem where the interface geometry is unknown but the field is known (Eq. (6)) on the rough parts of the interface while the inverse is true on the facets. In order to obtain some analytic understanding of this problem, we follow a strategy which has proved useful in the rough case. We first analyse a 'zero'-surface tension case so defined as to keep the main specificity of the problem. That is, we neglect the surface tension on the rough parts where we replace equation (6) simply by

$$\phi = 0 \tag{8}$$

but we still require that the shape has facets of length L normal to the directions $\pm\theta_{\rm f}$ (see Fig. 1). On the facets, the diffusive field conservation still holds (Eq. (5)) but ϕ is unknown and only obeys the constraint (7). More precisely, our solution corresponds to the case for which $w" + w = 0$ on the rough parts but for which w has a cusp. An illustrative example with $\theta_{\rm f} = \pi/4$ is given by the even function $w_1(\theta)$ such that

$$\begin{aligned} w_1(\theta) &= \alpha\cos(\theta),\ 0 \le \theta \le \frac{\pi}{4} \\ w_1(\theta) &= \alpha\cos(\pi/4-\theta),\ \frac{\pi}{4} \le \theta \le \frac{\pi}{2} \end{aligned} \tag{9}$$

where α is a real positive number related to the magnitude of the cusp. We then discuss what we expect in the large C limit where capillary effects are not neglected but are formally small on the rough parts.

3. Faceted needle-crystals.

As usual, it is useful to transform the moving free boundary problem into a problem with conditions on a known boundary by changing variables. In the classical zero-surface tension case, the interface is simply the line $\phi = 0$ in a (ϕ,ψ)-coordinate system ($\phi + i\psi$ being the complex velocity potential). This is not true here since we want to neglect surface tension on the rough parts but keep the facets and the constraint (7). Therefore, ϕ is not prescribed on the facets and should be found simultaneously with the shape of the rough parts. However, it is still true that the interface is a line of flow in a fluid mechanical analogy. That is, one has $\psi = y$ on the interface [10]. So, the interface is located on the real axis of the complex $\hat{\Phi}$-plane where $\hat{\Phi} = \hat{\phi} + i\hat{\psi} = \phi + i\psi - (x+iy)$ is the complex velocity potential in the reference frame of the moving interface. In these coordinates, the free boundary problem is transformed in the more easily tackled problem of finding $z = x + iy$ as a function of $\hat{\Phi}$.

An instructive first step is to see how the classical parabolic needle crystal is obtained in this framework. In this case, the requirements on $z(\Phi)$ are that:

i) $z(\hat{\Phi})$ should be an analytic function of $\hat{\Phi}$,

ii) on the positive real $\hat{\Phi}$ axis, $x = -\hat{\phi}$ (since $\phi = 0$ on the interface),

iii) $z(\hat{\Phi})$ should grow linearly at infinity and not faster so as to obtain an univalued complex potential,

iv) the only allowed singularities of $z(\hat{\Phi})$ are located where the velocity $\frac{{\rm d}\hat{\Phi}}{{\rm d}z}^*$ vanishes in the moving interface frame, that is at the needle tip $\hat{\Phi} = 0$. In order to obtain a smooth interface shape when going around $\hat{\Phi} = 0$ from the lower real axis to the upper real axis this should be a square root singularity (to transform the 2π turn into a normal tangent).

The solution is simply

$$z(\hat{\Phi}) = -\hat{\Phi} + ia\sqrt{\hat{\Phi}} \tag{10}$$

where a is a real constant. On the real positive $\hat{\Phi}$-axis this gives the classical parabolic shape $x = -\hat{\phi}, y = \pm a\sqrt{\hat{\phi}}$.

Let us now consider the less elementary faceted case. The above stated conditions i), iii) and iv) are unchanged. Condition ii) is still true on the rough parts (this is our zero surface tension condition) but has to be modified on the facets. There ϕ is unknown but the profile is known : it is a straight line segment perpendicular to the $\pm\theta_{\rm f}$ directions (the conic singularities of the Wulff plot. As the complex velocity $(\frac{{\rm d}\hat{\Phi}}{{\rm d}z}^*)$is parallel to the profile, this gives a relation between the real (χ_1) and imaginary (χ_2) parts of the analytic function $\chi \equiv \frac{dz}{d\hat{\Phi}}$ on the facets

$$\chi_1 = \pm \tan\theta_f \ \chi_2 \tag{11}$$

The plus and minus signs refer to the lower and upper facets respectively (see Fig. 1). Since the conditions (11) are simply expressed in terms of the derivative of the velocity potential, we first determine χ rather than directly $z(\hat{\Phi})$. We also find it convenient to avoid the square root singularity and the allied cut plane by using instead of $\hat{\Phi}$ the complex variable $\Omega = \omega + i\nu$ with $\hat{\Phi} = \Omega^2$. The requirements on $\chi(\Omega)$ are easily obtained from the above conditions on $\hat{\Phi}$:

i) χ is analytic in the upper half Ω-plane,

ii) $\chi_1 = -1$ on the real Ω-axis outside the segments $[-\omega_2, -\omega_1]$ and $[\omega_1, \omega_2]$ corresponding to the facets and equation(11) should be satisfied on the facets,

iii) χ should tend to a constant as $\Omega \to \infty$ in the upper half Ω-plane,

iv) χ has a simple pole singularity at the origin $\Omega = 0$.

Finding χ is the classical problem of determining an analytical function given a linear relation between its real and imaginary parts on the real line. It can be solved following standard techniques (see e.g. [11]) . We simply give the solution in our case

$$\chi = -1 + \frac{i}{Y(\Omega)} \left\{ a\Omega + \frac{b}{\Omega} - 2\cos\theta_f \ \Omega \int_{\omega_1}^{\omega_2} \frac{d\omega'}{\pi} \frac{Y_r(\omega')}{\omega'^2 - \Omega^2} \right\} \tag{12}$$

with

$$Y(\Omega) = (\Omega^2 - \omega_1^2)^{1-\theta_f/\pi}(\Omega^2 - \omega_2^2)^{\theta_f/\pi}, Y_r(\omega) = (\omega^2 - \omega_1^2)^{1-\theta_f/\pi}(\omega_2^2 - \omega^2)^{\theta_f/\pi} \tag{13}$$

The determination of $Y(\Omega)$ is chosen such that is real positive for Ω real and larger than ω_2. The real constants a and b are still to be determined. Equation (12) is a parametric description of the faceted needle since on the real Ω-axis it gives (separating real and imaginary parts)

$$\begin{aligned} \frac{dx}{d\hat{\phi}} &= \frac{dx}{2\omega d\omega} = -1 \\ \frac{dy}{d\hat{\phi}} &= \frac{dy}{2\omega d\omega} = \frac{1}{Y(\omega)} \left\{ a\omega + \frac{b}{\omega} - 2\cos\theta_f \ \omega \int_{\omega_1}^{\omega_2} \frac{d\omega'}{\pi} \frac{Y_r(\omega')}{\omega'^2 - \omega^2} \right\} \end{aligned} \tag{14}$$

for ω outside the facet intervals $[-\omega_2, -\omega_1]$, $[\omega_1, \omega_2]$. On the facets, one has

$$\begin{aligned} \pm\frac{1}{\sin\theta_f}\frac{dx}{d\omega} &= \frac{1}{\cos\theta_f}\frac{dy}{d\omega} = l(\omega) \\ &\equiv 2\omega \left(\mp \sin\theta_f + \frac{1}{Y_r(\omega)} \left\{ a\omega + \frac{b}{\omega} - 2\cos\theta_f \ \omega \ P \int_{\omega_1}^{\omega_2} \frac{d\omega'}{\pi} \frac{Y_r(\omega')}{\omega'^2 - \omega^2} \right\} \right) \end{aligned} \tag{15}$$

where P denotes principal part and the upper (resp. lower) sign holds on the $[\omega_1, \omega_2]$ (resp. $[-\omega_2, -\omega_1]$) interval. Since $Y(\omega)$ vanishes at the facet extremities, equation (14) shows that for general choices of a and b, the rough parts join the facets with vertical tangencies. A tangential matching can only be obtained if the expression between parentheses in equation (14) vanishes both at $\omega = \pm\omega_1$ and $\omega = \pm\omega_2$. This gives two equations which determine a and b

$$\begin{aligned} a(\omega_1,\omega_2) &= 2\cos\theta_{\mathrm{f}} \int_{\omega_1}^{\omega_2} \frac{\mathrm{d}\omega'}{\pi} \frac{\omega'^2 Y_r(\omega')}{(\omega'^2-\omega_2^2)(\omega'^2-\omega_1^2)} \\ b(\omega_1,\omega_2) &= -2\cos\theta_{\mathrm{f}}\ \omega_1^2\omega_2^2 \int_{\omega_1}^{\omega_2} \frac{\mathrm{d}\omega'}{\pi} \frac{Y_r(\omega')}{(\omega'^2-\omega_2^2)(\omega'^2-\omega_1^2)} \end{aligned} \tag{16}$$

With such a choice a local analysis shows that indeed the facet and the rough parts match tangentially. The whole solution depends on the parameters ω_1 and ω_2 which can be advantageously traded for the more physical facet length and aymptotic parabola tip radius of curvature. Following our previous convention, we choose twice the tip radius of curvature of the asymptotic parabola as length unit (so that the shape behavior at infinity is simply $x \sim -y^2$) and explore the variety of shapes by varying the facet length. This is easily done by noting that rescaling ω_1 and ω_2 by r simply dilates the shape by r^2. We take advantage of it by choosing $[1,\lambda]$, $1 < \lambda < \infty$ as a basic family of intervals. The one parameter family with fixed asymptotics and varying facet lengths is obtained by rescaling the obtained shapes, that is by using the parameters $\omega_1(\lambda) = 1/\mid 2a(1,\lambda)\mid$ and $\omega_2(\lambda) = \lambda/\mid 2a(1,\lambda)\mid$ in the previous formulas ($a(1,\lambda)$ is defined by Eq. (16)). Some of these shapes are drawn in figure 2. The facet length $L(\lambda)$ is obtained by integrating equation (15) between $\omega_1(\lambda)$ and $\omega_2(\lambda)$

$$L(\lambda) = -\int_{\omega_1(\lambda)}^{\omega_2(\lambda)} l(\omega)\mathrm{d}\omega \tag{17}$$

It varies between 0 and a finite maximum length which depends on θ_{f} [12].

At this stage, it may appear that we have obtained a genuine solution for a faceted needle crystal. There is, however, a somewhat hidden remaining condition to satisfy. In the course of solving our problem, we have traded the condition $\phi = \mathrm{cst}$ for the new condition $\frac{\partial\phi}{\partial s} = 0$. In the usual case, this is a harmless change but here there are several disconnected rough parts. With the new condition although ϕ is constant on each rough part, it is not possible to ensure that the value of the constant is the same on different rough parts. Indeed, we obtain for the undercooling difference between the trailing and front rough parts

$$\Delta\phi = \phi_{\mathrm{trail}} - \phi_{\mathrm{front}} = x_2 + \hat{\phi}_2 - (x_1 + \hat{\phi}_1) = \omega_2^2 - \omega_1^2 - L\sin\theta_{\mathrm{f}} \tag{18}$$

This does not vanish as can be seen in figure 3 where $\Delta\phi$ is plotted as a function of the facet length. The only way to obtain the same undercooling on both rough parts would be to relax the physical requirement (mechanical equilibrium) of tangential matching between the rough and faceted parts [13]. This can of course be achieved by a different choice of the constants a and b in equations (14, 15).

4. Discussion.

We have analysed the growth of a faceted needle crystal when capillary effects on the rough parts are neglected. Within this approximation, the set of equations (4-7) is overconstrained if one requires that rough parts match the facets tangentially. A solution has been obtained but only by choosing different constant values of the diffusing field on different growing rough parts. This conclusion has been reached for a needle crystal in the context of a one-sided model and for a diffusion length much larger than the tip length scale but we do not expect these simplifications to be critical features. A more difficult but physically relevant question

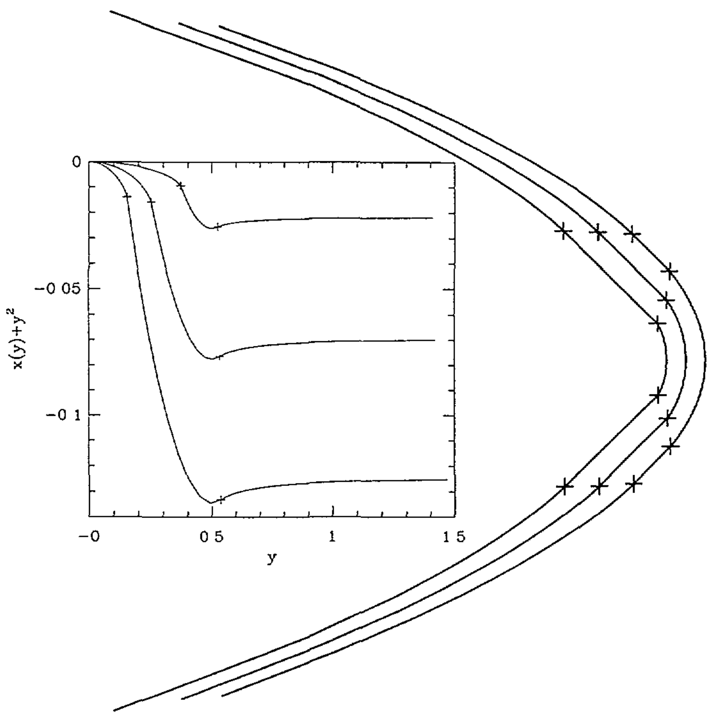

Fig. 2. — Analytical needle crystals for a faceting direction $\theta_f = 45°$ and facet lengths $L = 0.22$, 0.40 and 0.54. The facet extremities are shown by plus signs. The corresponding undercooling difference between the front and trailing rough parts have been chosen equal to 0.014, 0.046 and 0.086 respectively so that tangential matching of the rough and faceted parts are obtained. Insert: analytical correction to the Ivantsov parabola $x = -y^2$.

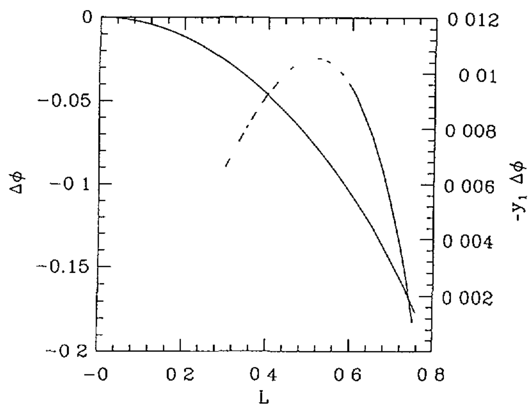

Fig. 3. — Variation of the undercooling between the trailing and front rough parts needed for tangential matching of the rough and faceted part when the facet length varies from 0 ($\theta_f = 45°$). up to the maximum facet length (full bold line). The product $-y_1 \Delta\phi$ which determines C as a function of facet length (Eq. (19)) is also plotted. A part of the curve is dashed as a reminder that equation (19) is obtained in the limit of large facets.

is to determine the importance of capillary effects on the rough parts (for definiteness, $w(\theta) = 1 + w_1(\theta)$ (Eq. (9)) can be taken) The simplest mathematical possibility is that our conclusion is still valid in this case. It would then remain to find some physical mechanism (i.e. growth kinetics) left out of the simple model (6 , 7) which could explain the experimental observations [2]. However, some other possibilities appear conceivable to us.

If one imagine perturbing the zero-surface tension cusped shapes (with $\Delta\phi = 0$) then capillary effects play a dominant role in the rough/faceted matching region where curvature is large and, *a priori*, the possibility exists that a tangential matching is automatically enforced. We have however not found any convincing reason why this should be true and we believe that in this case as well tangential matching should be required as an additional independent condition on the interface shape. This leads us to consider as starting point the zero-surface tension solutions with tangential matching. One should understand how small capillary effects on the rough parts can compensate the finite undercooling difference between the front and trailing rough parts. It is of course possible that a solution only exists when C (Eq. (6)) is of order one. A more attractive possibility is that facets are large in the large C limit and almost reach the needle tip. In this limit, it seems that one can construct approximate solutions of the full problem which are quite close to our analytic solutions with large facet lengths. The idea is simply to replace the small front rough part of our analytic solution by an arc of circle. Its radius of curvature R is geometrically determined by tangential matching to the facets to be $R = y_1(L)/\sin(\theta_f)$ where $y_1(L)$ is the (positive) ordinate of the facet front extremity (see Fig. 1). The unphysical undercooling difference of the 'zero'-surface tension solution can then be viewed as coming from capillary effects and determines C as a function of facet length by equation (6) (see Fig. 3):

$$C(L) = -\frac{\sin(\theta_f)}{y_1(L)\Delta\phi(L)} \tag{19}$$

The value of the diffusing field on this new interface is the same as for our analytic solution and the shape has been distorted in a small neighbourhood of the tip only. So, the field around the modified solution is practically unchanged and equations (4-6) appear to be well satisfied [14]. Finally, equation (7) relates the facet length to the cusp in $w(\theta)$ (which should be large for consistency). It should be noted that C is large and capillary effects are formally small because they have been measured on an aymptotic parabola characteristic length scale. They remain finite on the scale of the actual tip which tends to zero as C tends to infinity in a boundary-layer like manner.

Direct numerical studies [15] of equations (4-7) will hopefully be able to confirm the validity of this large-C approximate solution and show that no subtlety has escaped our notice. It is also interesting to remark that (19) does not involve the Wulff plot shape, so this relation would presumably be more easy to test experimentally than the corresponding prediction in the weakly anisotropic case [1] (were it not only valid in two dimensions).

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[10] This is easily obtained as follows (see e.g. Saffman P.G. and Taylor G.I., *Proc. R. Soc.* **A 245** (1958) 312). Let the interface be parametrized by its curvilinear abscissa s. On one hand, $\frac{\partial y}{\partial s} = -\cos\theta$ from the definition of θ (see Fig. 1). On the other hand, $\frac{\partial \phi}{\partial n} = \cos(\theta) = -\frac{\partial \psi}{\partial s}$ from equation (5) and the fact that $\phi + i\psi$ is an analytical function. Comparing the two equations, one readily obtains $\psi = y$ on the interface with suitable choices of origin.

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[12] The large λ behavior of $a(1,\lambda)$ is found to be $a(1,\lambda) \sim -\cos(\theta_f)B(3/2-\theta_f/\pi, \theta_f/\pi)\lambda/\pi$. The maximum facet length is obtained by taking $\omega_1 = 0, \omega_2 = \frac{\pi}{2\cos\theta_f B(3/2-\theta_f/\pi,\theta_f/\pi)}$. For $\theta_f = \pi/4$, this gives $L_{\max} \approx 0.757$.

[13] If our analysis is strictly seen as a solution for the case $w(\theta) = w_1(\theta)$ (Eq. (9)), then tangential matching is not really required. In this precise case, the rough and faceted part are in equilibrium for an arbitrary matching angle as emphasized to us by C. Caroli. However, we consider it as a first step toward obtaining a solution in the large C limit. In this case, we believe it is important to implement the tangential matching requirement already at this first-approximation stage, as explained in the discussion section.

[14] This is also true around the modified tip for a different reason. For our analytic solutions with large facets, the field gradient is almost constant in the tip region so the conservation equation (5) is automatically satisfied for any interface shape. The shape is determined by satisfying equation (6). This is similar to what happens for a growing interface filling a channel (Dombre T. and Hakim V., *Phys. Rev.* **A 36** (1987) 2811). Contrary to this latter case, a simple arc of circle is obtained here because the large undercooling $\Delta\phi$ dominates the small field variation along the shape. Our modified solution as described is not completely satisfactory only in a small region near the facet rear end. There the rough part of our analytic solution matches tangentially the facet but has a large curvature which is expected to be smoothed out by capillary effects.

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