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# Singularities in nonlocal interface dynamics

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An effective interface dynamics is derived for a Stefan-type moving-boundary-value problem corresponding to electrostatic aggregation in two dimensions. With the help of conformal-mapping techniques the problem may be reduced to a many-body-type problem described by a system of ordinary differential equations. The method allows one to show that the Mullins-Sekerka instability in this case leads to generation of cusp singularities in the interface in a finite time.

### I. INTRODUCTION

Recently there has been a renewed interest in the study of dynamical structures, and in particular, processes of pattern formation in physical, chemical, and biological systems. Unfortunately the understanding of such common phenomena as solidification or dendritic growth, which has always been important to metallurgists, is hampered by the mathematical complexity of the problem. Indeed, the process of solidification is equivalent to the Stefan problem: a diffusion problem for the temperature (or impurity concentration) with boundary values specified on the moving interface, the local velocity of which is in turn determined by the heat (or impurity) flux.<sup>1</sup> In its general case it represents a complicated, highly nonlinear problem, which is not readily amenable even to modern numerical simulations. Thus, much of the recent work proceeds in the direction of simplifying the problem in order to gain understanding of the basic mechanisms involved and facilitate the numerical analysis. One such route is to reduce the "moving boundary value" problem to an interface dynamics problem. Such phenomenological models were proposed by Ben-Jacob, Goldenfeld, Langer, and Schon<sup>2</sup> and Brower, Kessler, Koplik, and Levine.<sup>3</sup> These models provided valuable insight into the process of dendritic growth and the importance of various physical effects, such as anisotropy and surface tension, at the expense of sacrificing some of the nonlocality of the problem. Still, the basic mechanisms at work are not fully understood.

In this paper we investigate some of the basic nonlocal mechanisms involved in diffusion-controlled growth. We shall derive the evolution equation for the interface for a Stefan problem in the quasistationary limit and neglecting surface tension. This will allow us to study the Mullins-Sekerka instability<sup>4</sup> (an instability of "bumps" on the interface) analytically beyond the linear stability analysis and show that it leads, in finite time, to the appearance of 2/3 power cusp singularities in the surface. The model that we consider is most closely related to Saffman-Taylor instability of the interface between air and water in the Hele-Shaw cell.<sup>5</sup> The latter in the absence of surface tension represents a realization of the model with different geometry.<sup>6</sup> The method used in this paper involves mapping the interface conformally onto the unit circle in the complex plane. A surprising feature of the partial differential equation (PDE) governing interface dynamics is that it may be reduced to a set of ordinary differential equations (a kind of a manybody problem) for the dynamics of the critical points of the conformal map. This facilitates analytic and numerical investigations and also points to the possibility of chaotic behavior in the evolution of the interface (the problem which requires much further study).

## II. MOVING-BOUNDARY-VALUE PROBLEM AND INTERFACE DYNAMICS

Consider a cylindrical piece of "metal" with a crosssection shape given by the contour  $\vec{y}$ . It is kept at a constant potential and is immersed in a dilute electrolyte. The cylinder grows by electrodeposition at a rate proportional to the electrostatic field. We assume that the problem is effectively two dimensional and that the growth is slow enough so that a quasistatic approximation is valid. More significant is the assumption that the electrolyte is dilute so that there is no screening of the field. The evolution of the contour is then determined by

$$\partial_t \vec{\gamma}(t) = - \vec{\nabla} \phi(\vec{\gamma}) \quad , \tag{1}$$

where the electrostatic potential satisfies

$$\Delta \phi = 0, \quad \phi(\vec{\gamma}) = 0 \quad . \tag{2}$$

Since we consider the model in two dimensions it is only natural to use complex variables and conformal mappings. The domain bounded by the contour  $\vec{y}$  is simply connected and by the Riemann theorem may be conformally mapped onto the unit disk. Let w = x + iy parametrize the "physical" plane and let  $z = f^{-1}(w)$  be a conformal map, mapping the exterior of  $\vec{y}$  onto the exterior of the unit disk in the z plane. This map provides a parametrization of the contour  $\gamma(s) = f(e^{is})$  and the evolution of the contour may be specified by the evolution of the mapping.

Let us introduce a complex potential  $V(w) = \phi(x,y)$ +  $i\psi(x,y) = V(f(z))$  and solve the problem outside the unit disk in the z plane

$$V(w) = \ln f^{-1}(w) - 1 \quad . \tag{3}$$

The complex electrostatic field at the interface is then given by

$$E = -\left(\frac{\partial V(w)}{\partial w}\right)^* = -\frac{i}{(zdf/dz)^*}$$
(4)

for  $z = \exp(is)$  and Eq. (1) may be rewritten as

$$\partial_t \gamma'(s) = -i \frac{\partial_s f(e^{is})}{|\partial_s f(e^{is})|^2} \quad . \tag{5}$$

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The prime over  $\gamma$  in Eq. (5) is there to remind us that this equation does not preserve the right parametrization of the contour. The reason is that the right-hand side (RHS) of Eq. (5) represents the normal component of the velocity, which describes only the change in the geometry of the contour. The correct dynamical equation has to include a tangential component of velocity corresponding to a continuous reparametrization of the contour necessary in order to ensure that  $\gamma(t,s) = f_t(e^{is})$  remains the boundary value of an analytic function (which specifies the parametrization of the contour as well as its geometry). The exact form of the reparametrization term is found by considering the infinitesimal mapping  $f_{\Delta t}(z)$ , induced by the evolution [Eq. (5)], and defined by  $f_{t+\Delta t}(z) = f_{\Delta t} f_t(z)$ . We shall only give the result

$$\partial_{t}\gamma(t,s) = -i\partial_{s}\gamma(t,s)\{|\partial_{s}\gamma(t,s)|^{-2} + iC[|\partial_{s}\gamma(t,s)|^{-2}]\}$$
(6)

The integral operator C is best described by its action on the Fourier transform of some real periodic function g(s):

$$g(s) = a_0 + \sum_{n=1}^{\infty} \left( a_n e^{i2\pi ns} + a_n^* e^{-i2\pi ns} \right) , \qquad (7)$$

$$iC[g(s)] = -\sum_{n=1}^{\infty} (a_n e^{i2\pi ns} - a_n^* e^{-i2\pi ns}) \quad . \tag{8}$$

The integral operator term in Eq. (6) reflects the nonlocality of the interface dynamics. This equation may easily be studied numerically; however, the analytic approach may be carried one step further.

## **III. DYNAMICS OF CONFORMAL SINGULARITIES**

Let us interpret Eq. (6) as the evolution equation for the mapping function f(z). If some contour is specified as an initial condition, one may determine the corresponding con-

$$\frac{d}{dt}\ln\partial_z f(z) = |\alpha_0|^{-2} \sum_{i=1}^N A_i + |\alpha_0|^{-2} \sum_{i=1}^N \frac{\alpha_i}{z - \alpha_i} \left( \sum_{j=1}^N A_j + 2 \sum_{j \neq i} \frac{\alpha_j (A_j + A_i)}{\alpha_i - \alpha_j} \right)$$

The LHS of Eq. (14) decomposes into the sum over simple poles,

$$\frac{d}{dt}\ln\partial_z f(z) = \frac{\dot{\alpha}_0}{\alpha_0} - \sum_{i=1}^N \frac{\dot{\alpha}_i}{z - \alpha_i} , \qquad (15)$$

and we can read off the dynamical equations for the critical points,  $\alpha$ 's, by equating the residues of the poles on both sides of Eq. (14). This yields a system of ordinary differential equations:

$$\dot{\alpha}_0 = \alpha_0 |\alpha_0|^{-2} \sum_{i=1}^N A_i$$
 (16)

$$\dot{\alpha}_i = -\alpha_i |\alpha_0|^{-2} \left( \sum_{j=1}^N A_j + 2 \sum_{j \neq i} \frac{\alpha_j (A_j + A_i)}{\alpha_i - \alpha_j} \right) \quad . \tag{17}$$

This result is quite surprising: the nonlinear and nonlocal evolution equation [Eq. (6)] conserves the number of critical points and is reduced to the problem of their dynamics!

formal map (Riemann mapping theorem). We shall consider sufficiently simple contours, such that the map is of the form

$$f(z) = a_0 z \prod_{i=1}^{N} (1 - a_i z^{-1}) \quad . \tag{9}$$

(While the method works for the polynomial mappings of arbitrary order, we have not succeeded in dealing with more general structures.) The requirement that f(z) maps the exterior of the unit disk onto the exterior of the contour conformally demands that the critical points of the map, zeroes of its derivative, all lie within the unit disk. It is convenient therefore to deal with the factorization of  $\partial_z f(z)$  directly:

$$\partial_z f(z) = \alpha_0 \prod_{i=1}^N (1 - \alpha_i z^{-1})$$
 (10)

We shall now reduce the partial differential equation [Eq. (6)] to a system of ordinary differential equations governing the dynamics of the "critical points." For the purpose of the derivation that follows it is convenient to rewrite the term on the RHS of Eq. (6) in the integral form

$$g(s) + iC[g(s)] = \lim_{\epsilon \to 0} \int_0^{2\pi} \frac{d\phi}{2\pi} \frac{1 + e^{i(\phi-s)}}{1 + \epsilon - e^{i(\phi-s)}} g(\phi) \quad .$$

$$(11)$$

Upon evaluating the integral one may obtain the following result.

$$|\partial_{z}f(z)|^{-2} + iC[|\partial_{z}f(z)|^{-2}] = \sum_{i=1}^{N} \frac{A_{i}}{|\alpha_{0}|^{2}} \frac{z + \alpha_{i}}{z - \alpha_{i}}$$
(12)

for  $z = e^{is}$  with

$$A_i^{-1} = (1 - |\alpha_i|^2) \prod_{j \neq i} (\alpha_i - \alpha_j) (\alpha_i^{-1} - \alpha_j^*) \quad . \tag{13}$$

After a straightforward if somewhat lengthy computation one may rewrite Eq. (6):

$$\ln \partial_{z} f(z) = |\alpha_0|^{-2} \sum_{i=1}^{N} A_i + |\alpha_0|^{-2} \sum_{i=1}^{N} \frac{\alpha_i}{z - \alpha_i} \left( \sum_{j=1}^{N} A_j + 2 \sum_{j \neq i} \frac{\alpha_j (A_j + A_i)}{\alpha_i - \alpha_j} \right)$$
(14)

### IV. MULLINS-SEKERKA INSTABILITY AND CUSPS IN THE INTERFACE

It is trivial to see that a circular contour with its radius growing as  $\sqrt{t}$  is an admissible solution; however, it is linearly unstable with respect to the formation of "bumps" (the Mullins-Sekerka instability). We shall now use the dynamical equations derived above to extend the study of the interface evolution beyond the linear stability analysis. Let us look at an *n*-fold perturbation of the circle:

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$$\gamma(s) = re^{is} \left[ 1 + \frac{q}{n-1} e^{-ins} \right] \quad . \tag{18}$$

The derivative of the conformal map is then

$$\partial_z f(z) = r(1 - qz^{-n}) \tag{19}$$

and the critical points are  $\alpha_0 = r$  and  $\alpha_k = \eta \exp(i2\pi k/n)$ 

with  $\eta^n = q$ . Using Eqs. (13), (16), and (17) we get

$$\frac{d}{dt} \ln \alpha_0 = \frac{|\alpha_0|^{-2}}{1 - |\eta|^{2n}}$$
(20)

and

$$\frac{d}{dt}\ln|\eta| = (1 - 2n^{-1})\frac{|\alpha_0|^{-2}}{1 - |\eta|^{2n}} \quad (21)$$

It follows that  $q = cr^{n-2}$  and, for  $q \ll 1$ ,  $r(t) \approx \sqrt{t}$ . Thus the amplitude of the *n*-fold mode grows as  $q(t) \approx ct^{n-2/2}$ . (Note that n = 2 is a special case and shows that ellipses evolve by simple dilation, just like circles.) However, care must be taken whenever the value of |q| approaches 1 and the critical points of the map hit the unit circle. This actually happens in *finite time* and corresponds to the appearance of singularities in the contour. Letting  $q \rightarrow 1$  in Eq. (18) one finds that the singularities have the form of 2/3 power cusps. (It is interesting to note that the cusps appear with only few "modes" in our "analytic" parametrization, while they correspond to an infinity of "angular harmonics.") The finite time cusp formation may also be seen in a direct numerical simulation of Eq. (10): the results for a sixfold perturbation are shown in Fig. 1.

## V. CONCLUSIONS

The appearance of finite time cusp singularities in the interface is a plausible result of the nonlocal dynamics. However, while the model may provide a reasonable description of the early stages of evolution, in a real physical system singularities will not appear because of the surface tension. (Furthermore, in our "electrodeposition" model the evolution of cusps will be opposed by screening in the regions of high electrostatic field.)

We have used the mapping method to derive the interface dynamics partial differential equation for a more realistic boundary condition in Eq. (2), which includes the effects of surface tension  $[\phi(\vec{\gamma}) = \sigma\kappa]$ , where  $\kappa$  is the local curvature of the interface]. The numerical simulations suggest that sharp bumps in that case undergo tip splitting rather then evolve into cusps. We shall report these results elsewhere.<sup>6</sup>

The conformal mapping method appears to provide a convenient tool for analytic and numerical studies. The "singularity" dynamics derived above is akin to the "pole"

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FIG. 1. The evolution of a sixfold perturbation of a circle and formation of cusps in the interface. (Result of a numerical simulation of the interface dynamics equation.)

dynamics known in the context of the Korteweg-de Vries equation and some other conservative systems. The formulation of the problem in terms of the ordinary differential equations that it provides may allow one to study the "chaotic" aspects of the interface evolution.

Note added in proof. After this paper was accepted for publication, Nigel Goldenfeld brought to the attention of the authors an article by G. H. Meyer, in *Numerical Treatment of Free Boundary Value Problems*, edited by J. Albrecht (Birkhauser, Basel, 1982). For a model of Hele-Shaw flow with suction, G. H. Meyer using the conformal mapping method, had obtained a family of analytic solutions leading to cusp singularities.

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