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Marangoni-driven singularities via mean-curvature flow

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Abstract
In this work, it is demonstrated that the existence and topology of the recently observed interfacial singularities driven by Marangoni effects can be deduced using mean-curvature flow theory extended to account for variations of interfacial tension. This suggests that some of the physical mechanisms underlying the formation of these interfacial singularities may originate from/be modeled by the surface tension flow. The proposed approach also determines the conditions on the surface tension material behavior under which singularities may form, as well as the asymptotic behavior near singularities. Besides the application to Marangoni-driven singularities, the offered generalization of the mean-curvature flow is also of independent geometric interest.

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1. Introduction

1.1. Marangoni-driven singularities

There are numerous physical situations in which surface tension varies along the interface, which can be due to the presence of surface-active substances (soap molecules, i.e. so-called surfactants), temperature gradients along the interface or a non-uniform electric field. In these cases, surface tension gradients drive the Marangoni[1] flow, as illustrated in the case of soap molecules at the interface in figure 1.

Marangoni-driven flows exhibiting interfacial singularities were found recently [1–4], as shown in figures 2(*b*) and (*c*). In the first case (figure 2(*b*)), the chemical reaction at the water–oil interface produces a surfactant, which drives the Marangoni flow along the interface leading to a conical symmetry of the system with a singular cone tip [1, 2]. In the

[1] The effect is named after Carlo Marangoni, the Italian physicist, who re-explained the phenomenon of 'tears of wine' in 1871 after the original explanation by James Thomson (1855).

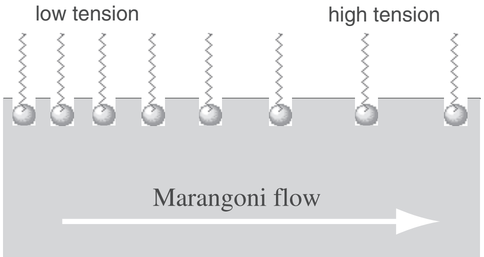

Figure 1. On the physics of surfactant-driven Marangoni flows. Surfactant molecules: the 'disks' are polar heads and the 'springs' are hydrophobic tails. Due to polarization of surfactant molecules, they tend to repel from each other in the areas of high concentration (low surface tension) thus dragging the surrounding fluid and leading to the Marangoni flow.

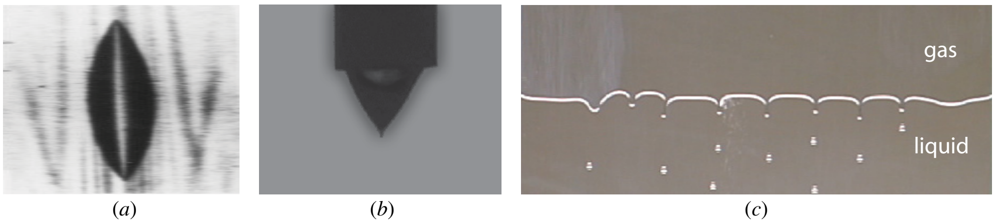

Figure 2. Physical examples of interfacial singularities. (*a*) Drop deformed in an extensional flow [7]: viscous stresses in the surrounding phase deform the drop, which forms pointed ends. (*b*) Chemical reaction-driven tip-streaming [1, 2]: acid–base reaction at the interface of the drop drives the Marangoni flow. (*c*) Surfactant-driven fingering in a Hele-Shaw cell [3, 4]: soapy water displaces air in the narrow space between two glass plates (Hele-Shaw cell) and eventually leads to fingering.

second situation (figure 2(*c*)), the surfactant-laden water phase displaces air in the narrow space between two parallel glass plates, the so-called Hele-Shaw cell, which leads to a Marangoni-driven instability [2, 4] and thus to fingering and the formation of cusps between the fingers. Another basic topological type of singularity, shown in figure 2(*a*), has not been found yet in the context of flows, where Marangoni effects would be the primary driving force for interfacial deformations. However, this type of singularity was observed under the conditions of external forcing starting with the classical experiments by Taylor [5]. Even though the primary source of drop deformation in Taylor's and other studies [6, 7] is due to externally imposed shear or extensional flow, the works of de Bruijn [6] and Eggleton *et al* [8] revealed that tip-streaming occurs due to the presence of surface-active contaminants, i.e. surface tension gradients, which makes the present analysis relevant to this situation as well.

1.2. Geometric flows

In this work, it is proposed to study certain aspects of Marangoni-driven singularities—topological types and asymptotics—with the help of geometric flows. Geometric flows of surfaces and curves have proven to be useful not only as mathematical ideas exploited, for example, in differential geometry and in the proof of Poincaré's conjecture, but also as a practical tool for solving various physical problems such as flame propagation, droplet dynamics, crystal growth, image processing [9], motion of grain boundaries [10], soap froth

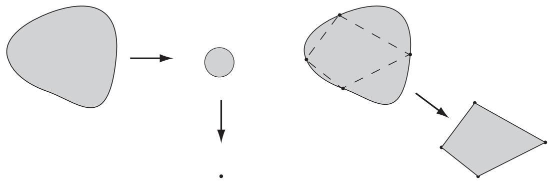

Figure 3. Mean-curvature flow with and without nonuniform surface tension. (*a*) Mean-curvature flow. (*b*) The simplest case of (5) when $\sigma(s) = 0$ at $s = s_1, s_2, \ldots, s_k$.

[11] and general relativity [12]. They also found application in the development of numerical methods such as widely used level-set techniques [13], where level sets evolve by the mean curvature [14, 15]. Such a variety of applications is due to the basic fact that in many physical problems interfaces propagate with a speed proportional to the local curvature κ. While the mean-curvature flow can be used in level-set methods, which were recently extended to interfacial flows with surfactants [16], there has been no work on understanding Marangoni-driven singularities at fluid interfaces from the viewpoint of the mean-curvature flow.

1.2.1. Basic theory. The most well-studied geometric flow is the mean-curvature flow[2] in the plane $\mathbb{R}^2$:

$$\frac{\partial \boldsymbol{x}}{\partial t} = -\kappa\, \boldsymbol{n}, \tag{1}$$

where $\boldsymbol{x}(s,t) \equiv (x, y) : S^1 \times [0, T) \to \mathbb{R}^2$ is a point on the curve with $s \in S^1$ being the natural parametrization (arclength) of the curve and $t \in [0, T)$ the time. Equation (1) describes the evolution of a curve on which every point moves in the direction opposite to the outward unit normal $\boldsymbol{n}$ with a speed equal to the mean curvature κ. As suggested in [17], a visual illustration of this flow could be the evolution of an elastic band in honey. As one can expect, this is a curve shortening flow and has smoothing properties due to the parabolic character of (1), i.e. similar to that of the heat equation [18] since equation (1) can be rewritten as

$$\frac{\partial \boldsymbol{x}}{\partial t} = \frac{\partial^2 \boldsymbol{x}}{\partial s^2}, \tag{2}$$

where the standard fact from the geometry of curves that $\kappa\, \boldsymbol{n} = -\partial \boldsymbol{t}/\partial s$ with $\boldsymbol{t} = \partial \boldsymbol{x}/\partial s$ being the unit tangent vector has been taken into account. It is known that for the convex initial data $\boldsymbol{x}(s, 0)$, the curve enclosing a compact set contracts smoothly to a single point in a finite time T and becomes circular at the end of the contraction [18], see figure 3(*a*); this result was generalized for a convex hypersurface in $\mathbb{R}^n$, $n \geqslant 3$, in [19]. However, if the initial data for (2) are non-convex, then singularities can occur [20] in the dynamics of (2). In this work we demonstrate that singularities may also form for convex initial data if (2) is modified to account for the complex behavior of interfaces with interfacial tension varying along the interface.

1.2.2. Restriction to surface tension phenomena. Application of equation (1) to fluid drops [9] under the constraint that the enclosed volume is constant is called the *surface tension flow* and is valid only if the surface (or interfacial) tension $\sigma(s)$ is constant; in fact, there should

[2] Unless otherwise stated, all equations throughout this work are given in a non-dimensional form.

be a factor $\sigma(s)$ on the right-hand side of (1), since it is surface tension which drives the interface. Namely, the interface of the mean curvature κ and the free energy per unit area σ is urged toward its center of curvature due to the pressure difference across the interface given by $\Delta p = \sigma\,\kappa$.

This allows one to model the evolution of a soap bubble or a drop which, when perturbed, eventually assumes a spherical shape in 3D and a disk shape in 2D, as in the case of oil drops on the surface of water [21]. Of course, this curvature flow is a simplification of the physical reality, as bulk forces, e.g. due to inertia or viscous resistance, are neglected. However, in many cases the mean-curvature flow (1) provides a reasonable approximation of the interface evolution, identification of the equilibrium states and, besides its practical importance, has many fascinating geometrical properties [17], which stimulate the ongoing research. There is also a class of problems when the model (1) is accurate up to a constant factor in front of the curvature κ due to viscous resistance. Namely, when the fluid motion obeys Darcy's law [22], the fluid velocity $\boldsymbol{v}$ is proportional to the pressure gradient ∇p, i.e. in a dimensional form $\boldsymbol{v} = -(K/\mu)\nabla p$, where K is the medium permeability and μ is the fluid viscosity. Clearly, ∇p can be due to surface tension, and the factor K/μ slows down the interfacial motion. In this case, the motion occurs in the direction of the pressure gradient, which agrees with the above physical justification for the surface tension flow. In particular, Darcy's law applies to the case of fingering in a Hele-Shaw cell [23] in figure 2(*c*), which was originally designed as a model of porous medium. Notably, the fluid motion is potential in this case, as due to the mass conservation condition, $\nabla\cdot\boldsymbol{v} = 0$, one has $\nabla^2 p = 0$. In the other two cases—tip-streaming in figure 2(*b*) and drop deformation in figure 2(*a*)—the motion takes place on small scales and thus viscous effects dominate, which leads to the Stokes flow regime. The latter sometimes admits a potential flow approximation [24] and thus also justifies the application of the mean-curvature flow (1) in certain cases.

2. Extension to Marangoni phenomena

2.1. Generalization of mean-curvature flow

As pointed out in section 1.2.2, the physical driving force for the mean-curvature flow is the surface tension $\sigma(s)$, which should be incorporated as a factor on the right-hand side of (1). However, in the standard case of the mean-curvature flow, surface tension is constant and thus scaled out. In order to generalize (1) to the case of Marangoni-driven interfacial flows, note that a general interfacial force acting on the interface at some point $\boldsymbol{x}_s$ is given by [25, 26]

$$\boldsymbol{f}_s(\boldsymbol{x}_s) = -\boldsymbol{n}\sigma(\nabla_s\cdot\boldsymbol{n}) + \nabla_s\sigma, \tag{3}$$

where $\boldsymbol{n}$ and $\boldsymbol{t}$ are unit normal and tangent vectors, respectively, cf figure 4, $\nabla_s = \boldsymbol{t}\,\partial_s$ is the interfacial gradient and $\nabla_s\cdot\boldsymbol{n} = \kappa$ is the interfacial curvature. Clearly, the motion induced by the force (3) has two components, normal and tangential, which are due to the capillary pressure difference across the interface, $-\boldsymbol{n}\,\sigma\,(\nabla_s\cdot\boldsymbol{n})$, and the surface tension gradient, $\nabla_s\,\sigma$, respectively.

Therefore, (1) should be generalized to the following motion of a curve:

$$\frac{\partial\boldsymbol{x}}{\partial t} = \partial_s\sigma(s)\boldsymbol{t} - \sigma(s)\,\kappa\,\boldsymbol{n}. \tag{4}$$

With a reparametrization of the interface $s \to \widetilde{s}(s,t)$, the tangential motion component (4) can be removed by choosing $\partial_t\widetilde{s} = \partial_s\sigma(s)$. Because of that and because we are interested in

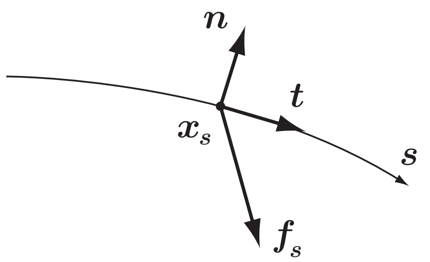

Figure 4. On a general interfacial force $\boldsymbol{f}_s$ acting on the interface at a point $\boldsymbol{x}_s$, $\boldsymbol{n}$ and $\boldsymbol{t}$ are unit normal and tangent vectors, respectively.

interfacial singularities, which can be formed only by the normal motion, we will restrict our attention to the reparametrized version of (4) dropping tildes:

$$\frac{\partial \boldsymbol{x}}{\partial t} = -\sigma(s)\,\kappa\,\boldsymbol{n} \equiv \sigma(s)\frac{\partial^2 \boldsymbol{x}}{\partial s^2}. \tag{5}$$

As shown below, it is possible to gain some insight into the origin and topological variety of Marangoni-driven singularities based on this natural modification of the mean-curvature flow equation (1), which now accounts for Marangoni effects.

It must be noted that there may often be more than one geometric flow which could be used to treat a particular problem [27], which is the case in many applications of the mean-curvature flow mentioned in the introduction. Despite this non-uniqueness of a choice of a geometric flow, it provides a useful evolutionary prospective to problems which admit equilibrium solutions [27], as all the problems under consideration here.

2.2. *Existence of singularities*

As observed in experiments [1, 2, 4], surface (interfacial) tension drops to ultra-low values[3] in the vicinity of the cusp (cf figure 2(*c*)) or pointed tip of a drop (cf figure 2(*b*)), which in turn allows for the existence of these singularities. Indeed, only ultra-low surface energy σ can sustain the high curvature κ of the interface, when the pressure is kept fixed, $p \simeq \sigma\,\kappa$. In the simplest case when the surface tension vanishes at discrete points at the interface, i.e. $\sigma(s) = 0$ at $s = s_1, s_2, \ldots, s_k$, these points are stationary, and due to the curve shortening flow of (5) one can expect the initially smooth curve to shrink to a *k*-polygon as in figure 3(*b*). To prove this expectation, let us consider the simplest case when the surface tension $\sigma(s)$ vanishes at s_i, but is a non-zero constant on each interval (s_{i-1}, s_i). Generalization of the theorem given below to the case when the surface tension function $\sigma(s) \geqslant 0$ is a convex function on (s_{i-1}, s_i) vanishing at the ends of each interval (and thus $\sigma(s)$ is continuous for all *s*) is straightforward.

Theorem 1. *(Existence) Let* $\boldsymbol{x}(0,\cdot) : S^1 \to \mathbb{R}^2$ *be a smooth simple closed curve, which is embedded in* $\mathbb{R}^2$ *and parametrized by* $s \in S^1$. *Let also the weight function* $\sigma(s) \geqslant 0$ *have* $k \geqslant 3$ *isolated zeros at* s_i, $i = 1, 2, \ldots, k$, *not located on one line, and be a non-zero constant on each interval* (s_{i-1}, s_i). *Then* $\boldsymbol{x} : S^1 \times [0,\infty) \to \mathbb{R}^2$ *exists satisfying (5) and is smooth for all t. This solution converges to the limiting shape* $\boldsymbol{x}(\infty, s)$, *which is a k-polygon with vertices* $\boldsymbol{x}(0, s_i)$, $i = 1, 2, \ldots, k$, *in the* C^∞ *norm.*

[3] ~ 1 mN m^{-1}, which is two orders of magnitude lower than in the clean interface case.

Proof. Since the weight function $\sigma(s)$ vanishes at s_i, $i = 1, 2, \ldots, k$, those points are stationary, and thus one needs to consider the curve evolution only on the interval (s_{i-1}, s_i). Because by a change of coordinates (rotation and translation in $\mathbb{R}^2$), the vector problem (5) on (s_{i-1}, s_i) can be transformed to a scalar problem; this is equivalent to the consideration of the following heat conduction problem:

$$\begin{aligned} &\frac{\partial x}{\partial t} = \sigma(s)\frac{\partial^2 x}{\partial s^2}, \qquad && 0 < t,\ 0 < s < l, \\ &x(0, s) = x_0(s), && x(t, 0) = x(t, l) = 0, \end{aligned} \tag{6}$$

where $l = s_i - s_{i-1}$. The solution of (6) may be found by separation of variables:

$$x(t, s) = \sum_{n=1}^{\infty} \mathrm{e}^{-\lambda_n t} E_n(x_0)(s), \tag{7}$$

where E_n is the projection on the nth eigenfunction, $E_n(f)(s) = \phi_n(s)(\phi_n, f)$ and the eigenvalues and eigenfunctions of the heat operator $A = \sigma(s)\partial_s^2$ are

$$\lambda_n = (\sigma\pi^2/l^2)n^2, \qquad \phi_n(s) = (2/l)^{1/2}\sin(\pi n s/l). \tag{8}$$

As one can see from solution (7), it decays monotonically to the zero solution, which corresponds to a flat facet of the k-polygon. Since the initial condition $x_0(s)$ satisfies the same boundary conditions as the solution does, then based on the standard property of the heat operator [28], the convergence takes place in the C^∞ norm due to the smoothing property of the semigroup e^{At} generated by the heat operator A. □

Note that there is no requirement that the initial shape $\boldsymbol{x}(0, s)$ should be convex, but singularities form for convex data too. Also, the C^0 smoothness of the initial curve $\boldsymbol{x}(0, \cdot) : S^1 \to \mathbb{R}^2$ can be relaxed to $L^2(S^1)$, because the smoothing property of the semigroup (7) generated by the heat operator [29] still allows the existence of a smooth solution of the heat equation [30]. Finally, it is important to point out that at the singularity points the curvature diverges and the normal vector is undetermined even if the initial conditions $\boldsymbol{x}(s, 0)$ are smooth and convex.

2.3. Asymptotics near the singularity

Experimental observations of Marangoni-driven singularities [1, 2, 4] demonstrate that the behavior of the interface in the neighborhood of these singularities is symmetric (self-similar), cf figure 2. Moreover, in support of this, a self-similar solution was found in the case of tip-streaming [2]. This is consistent with the fact that surface tension plays the smoothing role due to its tendency to minimize the surface area and with the general theoretical understanding that if there are no physically relevant characteristic length scales in the problem, then the solution behaves self-similarly [31, 32]. There are many physical examples of such situations, e.g. the Jeffrey–Hamel flow in a converging channel and flow in the neighborhood of a stagnation point [33]. In our case when one 'zooms into' the neighborhood of the singular point, all the outer macroscopic scales, e.g. the drop size, become irrelevant and thus one can expect a self-similar behavior near the singularity.

In order to get self-similar behavior, though, it was found in the case of tip-streaming [2] that the surface tension should have a simple power-law form. In particular, given the fact that surface tension vanishes either at some particular spatial point s_0 or at time t_0, this implies that the asymptotic behavior of surface tension has the power-law form $\sigma(s) = |s-s_0|^m$ or $\sigma(s,t) = |t-t_0|^m$, respectively[4].

The following result imposes certain restrictions on possible functional forms of the surface tension necessary for the occurrence of singularities.

Theorem 2. *(Asymptotics) If surface tension vanishes at the spatial point s_0 in the power-law fashion $\sigma(s) = |s-s_0|^m$ or at time t_0 as $\sigma(s,t) = |t-t_0|^m$, then a singularity in the curvature develops only if $m > 1$.*

Proof. Since the curvature is simply $\kappa = |\partial^2\boldsymbol{x}/\partial s^2| = \left[x_{ss}^2 + y_{ss}^2\right]^{1/2}$, it is sufficient to consider the scalar equation

$$x_t = \sigma(s)x_{ss}, \tag{9}$$

in order to determine the asymptotic behavior near an interfacial singularity. In what follows, we take into account the fact that the governing equation (9) is simple and has a number of symmetries, among which the dilation–contraction symmetries are most relevant for our purposes.

First, consider the case when surface tension vanishes at the spatial point s_0 as $\sigma(s) = |s-s_0|^m$. Using the affine transformations $t \to \alpha t$, $s \to \beta s$, it is easy to show that (9) is invariant if $\alpha = \beta^{2-m}$, which implies that the solution has a self-similar form $x(t,s) = x(\eta)$, $\eta = s\,t^{\frac{1}{m-2}}$. This representation of the solution reduces the partial differential equation (9) to the ordinary differential equation

$$x'' + \frac{\eta^{1-m}}{2-m}x' = 0, \tag{10}$$

the integration of which yields the solution of (5) in the self-similar form

$$\boldsymbol{x}(\eta) \sim \int \mathrm{e}^{-\frac{\eta^{2-m}}{(2-m)^2}}\,\mathrm{d}\eta, \qquad \eta = \frac{|s-s_0|}{t^{\frac{1}{2-m}}}. \tag{11}$$

Therefore, singularity in the curvature $|\boldsymbol{x}_{ss}| \sim |s-s_0|^{1-m}t^{-\frac{3}{2-m}}\,\mathrm{e}^{-\frac{\eta^{2-m}}{(2-m)^2}}$ develops only if $m > 1$.

Second, if the surface tension vanishes at time t_0 in the power-law fashion $\sigma(s,t) = |t-t_0|^m$, then the analysis similar to the above demonstrates that the solution behaves self-similarly as

$$\boldsymbol{x}(\eta) \sim \int \mathrm{e}^{-\frac{m-1}{2}\frac{\eta^2}{2}}\,\mathrm{d}\eta, \qquad \eta = \frac{s}{|t-t_0|^{\frac{m-1}{2}}}, \tag{12}$$

and thus $|\boldsymbol{x}_{ss}| \sim s\,|t-t_0|^{-\frac{3}{2}(m-1)}\,\mathrm{e}^{-\frac{m-1}{4}\eta^2}$, which also leads to the conclusion that a singularity in the curvature develops only if $m > 1$. □

In physical reality, singularities may also form dynamically under the simultaneous limits $\sigma \to 0$ and $\kappa \to \infty$ as $t \to t_0$ at some interfacial point s_0, which allows for the singular points to travel up to the singularity formation time t_0—the time at which the product $\sigma(s)\kappa(s)$ vanishes. The fact that singular points can travel along the interface, in particular, can be seen from the traveling wave solution $y(s-ct)$ of $y_t = \sigma(s,t)y_{ss}$. If $\sigma(s,t) = s - ct$, which allows for the point of vanishing surface tension to travel, then $y \sim (s-ct)^{1-c}$. Therefore, the singularity in curvature develops if $c > -1$, which then travels with the speed c along the interface.

[4] This is clearly a generalization of the Taylor series assumption (which would give $m = 1$), since σ may be tending to zero at a different rate, and may also be seen as a power-law approximation of the actual behavior of the surface tension.

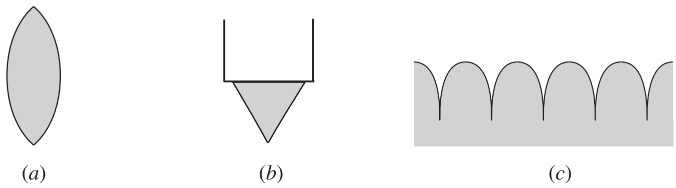

Figure 5. Three topological types of planar Marangoni-driven interfacial singularities (all the shapes are considered as planar): (*a*) a free drop, (*b*) a pendant drop, (*c*) infinite interface.

2.4. *Connection to experimental observations*

Given the fact that interfacial singularities may form for convex initial data in the case of surface tension variation with isolated zeros, one can expect, based on theoretical topological considerations, three types of (stationary) singularities in the planar case, namely volume preserving singularities—(a) a free drop, as in figure 5(*a*), and (b) a pendant drop, as in figure 5(*b*)—and infinite extent interface singularities, as in figure 5(*c*). In all these cases, the number of singular points and spacing between them along the interface may vary. In 3D the possible singular topologies are richer, but the key ones are analogous to the singularities shown in figure 5.

As discussed in section 2.1, the two anticipated types of Marangoni-driven singularities were found in experiments recently [1–3] and are illustrated in figures 2(*b*) and (*c*) (compare to figures 5(*b*) and (*c*), respectively). The first type illustrated in figure 5(*a*) has not been found yet in experiments on flows in which Marangoni effects are the leading cause of interfacial deformations, though it should be feasible from a topological viewpoint. In fact, as mentioned in section 1.1, this type of singularity was observed experimentally (cf figure 2(*a*)) when viscous stresses in the surrounding phase deform the drop leading to the formation of pointed ends [5–7] facilitated by the presence of surface-active contaminants.

3. Discussion and future work

In this communication we established a connection between the origin of Marangoni-driven interfacial singularities and the mean-curvature flow (5) extended to the surface tension varying case. Further studies are needed in order to extend theorem 1 to the case of the constant volume constraint, and to understand topological structures of 3D singularities as well as stability and time evolution of these singularities. The latter effects are especially important for understanding the transformation of chemical energy into self-agitated mechanical motion, as observed in the chemical reaction driven tip-streaming seen in figure 2(*b*), and in the classical problem of spontaneous dancing of camphor scrapings studied by Rayleigh [21]. The analysis offered here is independent of the particular material behavior, i.e. the dependence of the interfacial tension $\sigma(\gamma(t,s))$ on the surfactant concentration $\gamma(t,s)$; thus, it would be interesting to couple the dynamics of the geometric flow (5) with an unsteady transport of the surfactant at the interface.

While the offered analysis, based on the classical sharp interface theory, adequately captures the presence of Marangoni-driven singularities and asymptotic behavior near them on a macroscopic level, this approach breaks down when the interfacial curvature diverges [34–36]. Therefore, the fine microscopic structure of singularities needs to be studied separately by introducing a more refined physical model on smaller scales, in particular by taking into account the fact that even though the interfacial tension takes ultra-low values, it is not exactly zero.

In conclusion, it should be mentioned that the weighted mean-curvature flow has been used in the crystal theory, in which case the surface energy $\sigma(\boldsymbol{n})$ depends on the local orientation of the interface, i.e. the normal vector $\boldsymbol{n}$ [5, 37, 38], as opposed to the fluid interfaces (5), where the surface energy $\sigma(s)$ depends only on a position at the interface [39]. Also, sometimes the dependence $\sigma(\boldsymbol{n}, \kappa)$ on the curvature κ is introduced in the crystal theory to smooth out the crystal corners [37, 38].

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