On the structure of Marangoni-driven singularities

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Introduction History of the problem and motivation



• The fact that chemically-induced Marangoni effects can transfer chemical into mechanical energy directly has been known for a long time, e.g. in the context of camphor scrapings (Mensbrugghe 1869, Rayleigh 1890) or ameba-like motions at the oil-water interface (Magome and Yoshikawa, 1996).

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It is this latter circumstance that is of central interest to this work, namely, the formation of interfacial singularities due to Marangoni effects.

Motivation Paradigm Key problem

Introduction Paradigm problem: Marangoni-driven tip-streaming^a

Rediscovered in the course of pendant drop measurements with acid/alkaline reaction at the oil/water interface.



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- The latter drives Marangoni flow and sweeps surfactant towards the tip of the conical drop. The resulting ultra low interfacial tension in the tip area allows the interface to tear up and to create a thin thread through which the phase 1 is ejected into phase 2.

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It is remarkable that this physical system demonstrates a substantial separation of scales: the pendant drop is of 0.5 mm diameter, while the thread is about 5 μ m thick, i.e. the tip area appears as a singularity.

Introduction Key problem



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 - the key problem is a resolution of the following paradox: (I) on one hand, in order to get a conical drop with a pointed end of an infinite curvature one needs the interfacial tension to diverge, $\sigma \to \infty$ as $r \to 0$, which follows from the self-similar solution construction;

(II) on the other hand, in order to get tip-streaming, one needs the interfacial tension $\sigma \rightarrow 0$, since $p \sim \sigma/r$ and, for the fixed finite pressure p and small size emitted droplets, σ should be small too.

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Thus, the problem is to reconcile (I) and (II).

Problem setup Self-similarity Singularity structure

Marangoni-driven tip-streaming Problem setup



Figure: Cone set-up in spherical system of coordinate; θ^* is the cone semi-angle.

Marangoni-driven tip-streaming Problem setup



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Since the tangential boundary condition (Marangoni stresses) drives the phenomena, the appropriate nondimensional notations (without introduction of new variables) read

$$r
ightarrow l_{c} r, \ \mathbf{v}
ightarrow rac{\sigma_{0}}{\mu} \ \mathbf{v},$$
 $p
ightarrow rac{\sigma_{0}}{l_{c}} \ p, \ \sigma
ightarrow \sigma_{0} \sigma,$

where $l_c = \sqrt{\sigma_0/\rho g}$ is the capillary length, μ the dynamic viscosity of each medium, and σ_0 the interfacial tension in the clean interface case.

Marangoni-driven tip-streaming Steady tip streaming self-similarity: $\Psi = r^n \varphi(\cos \theta)$



In the Stokes regime,

$$Mo_1 \ll Mo_2 \ll 1, Mo = g\mu^4 / \rho \sigma_0^3,$$

only the case n = 1 gives a physically realizable self-similar solution

$$\Psi_i = r\varphi_i(\cos\theta), \ \sigma = \frac{\sigma_{min}}{r}, \ p = \frac{\pi(\cos\theta)}{r^2},$$

where

$$\begin{split} \varphi_1 &= -\frac{\sigma_{\min}\delta}{2} \frac{(1-x)(x-\xi)}{\sqrt{1-\xi^2}} \frac{1+\xi}{1+\delta-\xi(1-\delta)},\\ \varphi_2 &= -\frac{\sigma_{\min}}{2} \frac{(1+x)(x-\xi)}{\sqrt{1-\xi^2}} \frac{1-\xi}{1+\delta-\xi(1-\delta)}. \end{split}$$

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Marangoni-driven tip-streaming Steady tip streaming self-similarity: meaning and existence



Meaning: The case n = 1 conforms with the experimental observation that the flow is driven from the base of the drop towards its tip. It is also the lowest order solution in the following senses: (a) $\sigma \sim r^{-1}$ is the slowest decay of interfacial tension as $r \to \infty$, which allows for the flow towards the drop tip; (b) $\psi \sim r$ is the least singular at r = 0. The alongthe-axis singularity is observed for n > 1.

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- **Existence** is evidenced experimentally as well as surfactant transport equation supports the self-similar solution feasibility.

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Marangoni-driven tip-streaming Thread solution in tip-streaming: slender jet approximation





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Interfacial boundary conditions (with $\epsilon = h_{\infty}/l_c \ll 1$):

$$-p + \sigma \left[\frac{\epsilon}{h\sqrt{1 + \epsilon^2 h_z^2}} - \epsilon^3 h_{zz} \right] = -2\epsilon^2 \left[u_\rho - h_z w_\rho \right] + O(\epsilon^4),$$
$$w_\rho = -\epsilon\sigma_z + O(\epsilon^2),$$
$$u - wh_z = 0.$$

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Singularity structure Thread solution in tip-streaming

The leading order equation is

$$-\epsilon \frac{h^4}{2} \left[\frac{\sigma_z}{h} - \frac{\sigma h_z}{h^2} \right] - \sigma_{\min} \tan \theta^* \left(h^2 - \hat{h}_\infty^2 \right) = 0,$$

$$z \to -\infty : \quad h_z \to 0,$$

$$z \to +\infty : \quad h_z \to 2\sigma_{\min} \tan \theta^* / \epsilon.$$

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The scaling for the thread radius:

$$h_\infty \sim \mathit{l_c} an heta^* \, \sigma_{\min}$$

Since the interfacial tensions in the clean and surfactant interface cases differ by two orders of magnitude, this equation gives the right estimate for the experimentally observed difference between the drop size $\sim 0.5\,\mathrm{mm}$ and the thread diameter $\sim 5\mu\mathrm{m}.$

Conclusions

 In this work a systematic study of Marangoni-driven singularities is presented, which involved finding a self-similar solution in the neighborhood of a singularity as well as the resolution of this singularity via the construction of a thread solution with singular perturbation analysis.

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- The scaling law for the thread thickness is determined,

$$h_{\infty} \sim l_c \tan \theta^* \sigma_{\min},$$

which demonstrates the dependence on the lowest value of the interfacial tension, σ_{\min} , the cone semi-angle θ^* .

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 While the analysis is done in the context of surfactant Marangonidriven singularities, the results are general and independent of the nature of the Marangoni stresses. However, one has yet to discover experimentally interfacial singularities driven by temperature and electric field gradients.

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