Taylor Dispersion Governs the Compositional Evolution of Marangoni-Contracted Droplets

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Marangoni contraction of sessile droplets occurs when a binary mixture of volatile liquids is placed on a high-energy surface. Although the surface is wetted completely by the mixture and its components, a quasi-stationary non-vanishing contact angle is observed. This seeming contradiction is caused by Marangoni flows that are driven by evaporative depletion of the volatile component near the edge of the droplet. If the residual component has a lower surface tension, Marangoni flows are directed toward the center of the droplet, leading to a contraction until balanced by opposing capillary flows. This mechanism is widely used for drying and cleaning in technological applications, for instance in semiconductor or printing industries, but its physical explanation remains debated in literature. Here we show that the compositional evolution of such droplets is governed by Taylor-Aris dispersion. Breaking with precedent, we demonstrate that Taylor-Aris dispersion can consistently be included in a long wave expansion for the evolution of thin films. Coupled to diffusion limited evaporation, this model quantitatively reproduces not only the apparent shape of Marangoni contracted droplets, but also their internal flows.

Introduction.—Three-phase contact line motion of liquid mixtures is abundant in natural phenomena and technological applications \cite{1-5}. Many examples are found in everyday life situations, for instance, biological fluids like blood \cite{6} and tears \cite{7}, inks for inkjet printing \cite{8, 9}, and paints for artistic techniques \cite{10, 11}. The motion of such liquids over solid surfaces gives rise to intricate phenomena, involving both macroscopic hydrodynamics and molecular interactions \cite{4, 12}. Despite growing computational capacities, this multi-scale nature requires approximate models, utilizing the principle of separation of scales, to incorporate both micro- and macrophysics simultaneously \cite{13}. Pinned contact line conditions \cite{14-18} avoid this complexity and are well studied \cite{19-23}. In contrast, moving contact lines keep to challenge both experimentalists and theoreticians. Long-wave approximations \cite{12, 23, 24} offer a greatly reduced computational complexity for shallow droplets, and can be derived systematically, for instance by a gradient dynamics formalism \cite{23, 25-27}. All existing long-wave models to date treat the liquid as homogeneus in vertical direction, ignoring, for instance, Taylor-Aris dispersion \cite{28-31}. Thus, these models have been very limited in their application to bulk liquid mixtures.

Marangoni contraction is a mechanism by which mixtures of volatile liquids seemingly dewet from substrates that are fully wetted by the individual components \cite{32-35}, see Fig. 1 (a, c). The dewetting of the mixture originates from the evaporation of the component with the highest surface tension near the contact line. The local change in composition causes a surface tension gradient and an inward Marangoni flow. Balancing with capillary flows, this leads to a quasi-stationary liquid morphology of a macroscopic drop surrounded by a microscopic wetting film. Marangoni contracted droplets have a number of fascinating properties, for instance, an exceedingly high mobility \cite{34} or vapor-mediated interactions \cite{32, 36, 37}. The underlying physical principle remains debated \cite{33, 34}, mainly because a systematic analysis of moving contact lines with evaporation and compositional gradients is lacking from literature.

Here we show that small vertical compositional gra-
diabets are in agreement with the usual assumptions in a long-wave expansion, and lead to a general expression for Taylor-Aris dispersion in thin liquid films. We show that Taylor-Aris dispersion actually dominates the compositional evolution for shallow evaporating droplets of binary mixtures. Our long-wave model allows to resolve several orders of magnitude in space with very little computational cost and does not require any coarse-grained boundary condition on moving contact lines. We demonstrate that the model is in quantitative agreement with experimental measurements of both, the internal flows, and the apparent shape of Marangoni-contracted droplets. We expect our analysis to be relevant well beyond droplet studies, as it offers general evolution equations for thin films of liquid mixtures.

Experiments.—We measured the apparent shape and the internal flows of Marangoni-contracted droplets inside an atmospheric control chamber (size \( \sim 10\,\text{cm} \times 10\,\text{cm} \times 10\,\text{cm} \)), at room temperature. The droplets were composed of mixtures of water ("Milli-Q", resistivity 18 MΩ cm) and a carbon diol (Sigma Aldrich, \( \geq 98\% \)). Piranha cleaned microscopy glass coverslips (170 µm thick) were used as substrates (see supplement for details [35]). The humidity was set by continuously injecting a well-defined mixture of dry and water-saturated nitrogen behind gas-permeable membranes at the side-walls of the chamber. Droplets (initial volume 0.5 to 1 µl) were deposited with a clean gas-tight syringe (Hamilton 1700). High resolution micro particle image velocimetry (µPIV, Fig. 1(b)) was performed with an inverted fluorescence microscope (Nikon Eclipse Ti2E) and a high-aperture water-immersion objective (20x, NA 0.95) to allow for diffraction limited imaging even in the bulk of the droplets. Polystyrene microspheres (Thermo Fisher Scientific F8809, diameter 200 nm) were used as flow tracers, with a mass fraction of 7.8 \times 10^{-5} of the particle stock solution in the binary mixture. Images of the particles were captured with a high-speed camera (Phantom VEO 4k 990L) at 600 to 1000 FPS, quickly switching between two planes by automating the closed-loop piezo focus system of the microscope. Simultaneous side-view imaging of the drop was performed with a telecentric lens (Fig. 1(b)).

Fig. 1(c) shows the evolution of the apparent contact angle \( \theta_{\text{app}} \) of spreading drops of pure water (diol mass fraction \( \phi = 0 \)), pure 1,2-Propanediol (\( \phi = 1 \)) and of their mixtures. Pure liquids spread into complete wetting. In contrast, the binary mixtures reach a stationary non-equilibrium apparent contact angle \( \theta_{\text{app}} > 0 \) shortly after deposition. The drops stay in this contracted state for several minutes. This wetting behavior has been described previously [22][31], showing that \( \theta_{\text{app}} \) depends on \( \phi \) and the ambient relative humidity RH.

Fig. 2(a) shows the velocity field inside the droplet, as determined by µPIV. The arrows indicate the velocities that have been measured in different \( z \)-planes. The insets show dense velocity fields for two exemplary \( z \)-planes. Close to the free surface, the flow is directed inward, precisely balanced by an outward flow close to the substrate, leading to a quasi-stationary shape. The surface tension gradient can be derived from the shear stress at the free surface, using the viscosity \( \eta \) from literature [38][41].

Fig. 3 shows the experimentally derived surface tension gradient as a function of the distance \( d \) to the contact line, for various diols, compositions, and relative humidities. All curves follow a power law \( \partial_r \gamma \sim d^{-\gamma/2} \). Despite significant differences in composition, surface activity, and evaporation rates, the curves nearly collapse in physical units. To understand this seemingly universal behavior of Marangoni-contracted droplets, we derive in the following a long-wave expansion for liquid films of mixtures, subject to diffusion-limited evaporation.

Lubrication theory.—We consider the general case of a thin liquid film on a flat solid surface. The liquid is composed of a volatile solvent and a non-volatile solute (local mass-fraction \( \phi \)). Surface tension \( \gamma(\phi) \) shall depend instantaneously on the bulk composition (for generalizations see, e.g., [23][26]). The free surface is described by \( h(\vec{r}) \), where \( \vec{r} \) is the location in the substrate plane (see Fig. 1(a)). Incompressible Stokes flow without body forces is governed by

\[
\eta \nabla^2 \vec{u} = \nabla p, \\
\nabla \cdot \vec{u} = 0
\]

where \( \vec{u} \) is the fluid velocity, \( \eta \) is the dynamic viscosity...
of the fluid, and $p$ is the fluid pressure. The evolution of the solute field $\phi$ is given by

$$\partial_t \phi = \nabla \cdot \left( D \nabla \phi - \bar{u} \phi \right),$$

with $t$ as time and $D$ as diffusion coefficient of the solute. For simplicity we limit the following analysis to isothermal, isochoric, and isoviscous cases. We consider a no-slip and no-flux boundary condition at $z = 0$, kinematic and stress boundary conditions at $z = h$, and a Stefan-type boundary condition that links composition and evaporation (see supplemental material for details [38]).

To derive evolution equations in terms of vertically averaged quantities, we take the limit of long waves, where the characteristic horizontal scale, $r_0$, shall be much larger than the characteristic vertical scale, $h_0$: $\epsilon_h = h_0 / r_0 \ll 1$. We define the vertically averaged velocity $\bar{u}$, the total hydrodynamic flux $\Phi$, the vertically averaged composition $\bar{\phi}$, and the effective solute height $\Psi$ through

$$\Phi = \int_0^h dz \, \bar{u} = \bar{u} h, \quad \Psi = \int_0^h dz \, \phi, = \bar{\phi} h,$$

and the deviations from the average as

$$\bar{u} = \bar{u} + \delta \bar{u}, \quad \phi = \bar{\phi} + \delta \phi.$$

We scale all horizontal coordinates as $\bar{r} = r_0 \bar{r}'$, and all vertical coordinates as $z = \epsilon_h r_0 z'$. Velocities are scaled analogously with $u_0$. Time is scaled as $t = t_0 t'$. Pressure and surface tension are scaled as $p = p_0 p'$ and $\gamma = \gamma_0 \gamma'$, respectively, where $\gamma_0 = \gamma(\phi = 0)$, the surface tension of the pure solvent. The diffusivity is treated similarly, $D = D_0 D'$ with $D_0 = D(\phi = 0)$. We scale $j = j_0 j'$ for the evaporation rate, where $j$ and $j_0$ are determined according to [12] for the diffusion limited regime.

Whether the averaged quantities alone are sufficient to describe the evolution of the film, depends on the magnitude of $\delta \phi$. Thus we scale $\delta \phi = \epsilon_0 \delta \phi'$, requiring $\delta \phi' = O(1)$, seeking an expression for $\epsilon_0$ below. In the following we will omit the primes for readability and work exclusively with scaled quantities.

The derivation of the evolution equation for the film height follows the standard procedure described in the canonical reviews [12, 24]. One obtains

$$\partial_t h = -\nabla \cdot \Phi - E \cdot j,$$

with $E = j_0 \eta / (\epsilon_h^4 \gamma_0)$, from integrating [2] along $z$. Note that $\Phi$ does not involve any approximation. The long wave expansion is used only in the expressions for the evaporation rate $j$ (see [12]) and the horizontal hydrodynamic flux [43]

$$\Phi = \frac{h^3}{3} \nabla p + \frac{1}{\epsilon_h^2} \frac{h^2}{2} \nabla \gamma + O(\epsilon_h^2),$$

Here we have defined $u_0 = \epsilon_h^2 \gamma_0 / \eta$, $p_0 = \epsilon_0^2 \gamma_0 / r_0$, and $t_0 = r_0 \eta (\epsilon_h^3 \gamma_0)$. The pressure $p = \gamma h^2 - \Pi(h)$ contains capillary and surface (disjoining) forces.

The two terms in $\Phi$ are capillary- and Marangoni flux $\Phi_C$ and $\Phi_M$, associated with Poiseuille- and Couette-type velocity profiles, respectively [38]. These will shear any horizontal compositional gradient, such that a vertical gradient arises naturally. Combined with molecular diffusion from equation 3 this leads to Taylor-Aris dispersion [28, 29]. In addition, the Stefan boundary condition for evaporation at the free surface requires a vertical compositional gradient [44, 45]. It is commonly accepted that vertical compositional gradients are beyond the limit of the lubrication expansion [26, 27, 43, 46–50]. We challenge this paradigm, identifying three regimes, depending on aspect ratio and Péclet number: i. A regime of faint vertical compositional gradients where previous long-wave models hold [43]; ii. An intermediate regime of small but not negligible vertical gradients for which we derive a previously unknown evolution equation for $\Phi$ including Taylor-Aris dispersion; iii. A regime of large vertical gradients where the full advection-diffusion problem has to be solved [43].

Inserting [5] into [3] and integrating over the film height gives

$$h \partial_t \bar{\phi} = Pe^{-1} \nabla \cdot \left( D h \nabla \bar{\phi} + \epsilon_0 \int_0^h dz \, D \nabla \delta \phi \right)$$

$$- \Phi \cdot \nabla \bar{\phi} - \epsilon_0 \nabla \cdot \int_0^h dz \, \delta \phi \bar{u}$$

$$+ E \left( \bar{\phi} + \epsilon_0 \delta \phi(h) \right) j,$$
Pe = \frac{u_0 r_0}{D_{0}} is the Péclet number for the horizontal scales \( r_0 \) and \( u_0 \). \( \overline{D} \) is the vertically averaged diffusion coefficient. Care has to be taken when changing the order of integration and differentiation or applying the boundary conditions, see \([38]\). In contrast to Eq. 6 terms with non-averaged quantities remain. These terms scale as \( \sim \epsilon_\phi \), while the next-order terms in \( \mathcal{O} \) with \( \mathcal{O} \) scale as \( \sim \epsilon_h^2 \). Thus, the magnitude of \( \epsilon_\phi \) relative to \( \epsilon_h \) determines which terms in \( \mathcal{O} \) should be kept.

Limit i. \( \epsilon_\phi \lesssim \epsilon_h^2 \). We may ignore all terms \( \sim \epsilon_\phi \) and recover the previously known evolution equation \([43]\):

\[
h \frac{\partial \bar{\phi}}{\partial t} = \text{Pe}^{-1} \nabla \cdot \overline{D} h \nabla \bar{\phi} - \vec{\phi} \cdot \nabla \bar{\phi} + \vec{\epsilon} \bar{\phi} \nabla j + \mathcal{O}(\epsilon_\phi).\] (9)

Limit iii. \( \epsilon_\phi \sim 1 \). No simplified evolution equation for vertically averaged quantities can be derived, and the full problem has to be solved \([43]\).

Limit ii. \( \epsilon_\phi \sim \epsilon_h \). In this case, terms up to \( \sim \epsilon_\phi \) have to be retained. We consider a convection dominated problem i.e., \( \text{Pe} \gg 1 \) and \( \mathcal{E} \ll 1 \). Far below the boiling point, and with typical \( D_0 \sim 10^{-9} \text{m}^2/\text{s} \) and \( u_0 \sim 10^{-3} \text{m/s} \), this holds for most sessile nano- to microliter droplets with small contact angles. Eq. 8 simplifies further \([38]\):

\[
h \frac{\partial \bar{\phi}}{\partial t} = \text{Pe}^{-1} \nabla \cdot \overline{D} h \nabla \bar{\phi} - \vec{\phi} \cdot \nabla \bar{\phi} - \epsilon_\phi \nabla \cdot \int_0^h dz \Delta \bar{\phi} \bar{u} + \mathcal{E} \bar{\phi} \nabla j + \mathcal{O}(\epsilon_\phi^2).\] (10)

The remaining term with \( \Delta \bar{\phi} \) scales as \( \epsilon_\phi \). Thus a governing equation for \( \Delta \bar{\phi} \) can be truncated to \( \mathcal{O}(1) \). Inserting \( \mathcal{O} \) into \( \mathcal{O} \) multiplying with \( h \), and subtracting \( \mathcal{O} \) gives the leading order governing equation for \( \Delta \bar{\phi} \) \([38]\):

\[
\overline{D} \frac{\partial^2 \Delta \bar{\phi}}{\partial z^2} = \mathcal{E} \bar{\phi} \nabla j/h + \Delta \bar{u} \cdot \nabla \bar{\phi} + \mathcal{O}(\epsilon_\phi),\] (11)

where we have identified the natural scale \( \epsilon_\phi \) as \( \epsilon_\phi^2 \text{Pe} = \text{Pe}_z \), the Péclet number for the characteristic vertical scales \( \epsilon_h r_0 \) and \( \epsilon_h u_0 \). Eq. 11 describes a stationary advection-diffusion problem in the comoving frame of the mean flow and can be integrated. The resulting expressions for \( \Delta \bar{\phi} \) and the integral in \( \mathcal{O} \) can be found in the supplement \([38]\). In cases of axial or translational symmetry, Eq. 10 reduces to \( \mathcal{O} \) with \( \overline{D} \) replaced by \( \overline{D}_{\text{eff}} \) to account for Taylor-Aris dispersion:

\[
\overline{D}_{\text{eff}} = \overline{D} + \frac{e^2 \text{Pe}^2 \gamma}{105} \left( \frac{2 \epsilon_\phi^2 \text{Pe}_c}{20} + \frac{\epsilon_\phi \text{Pe}_M}{30} \right).\] (12)

The requirements for limit ii became apparent now: The problem must be convection dominated in horizontal direction (\( \text{Pe} \gg 1 \)), but diffusion dominated in vertical direction (\( \text{Pe}_z \ll 1 \)), as was already noted by Taylor and Aris in their treatments of pipe flow \([28, 29]\).

**Numerical simulations.**—We implemented the evolution equations \([6, 9]\) with the flux \([7]\) and the effective diffusivity \([12]\) in an axisymmetric finite volumes scheme with convergent numerical mobilities after \([51, 52]\). and

**FIG. 4:** Simulated fluxes and effective diffusivity as a function of \( r (\phi = 0.20 \text{ I, 2-PD, RH = 13 %, } \tau = 11.5 \text{s}) \). (a) Capillary (blue) and Marangoni (red) fluxes balance each other, resulting in a total flux \( \Phi \sim 0 \) (black). (b) Normalized effective diffusivity. The individual flux components cause strong dispersion in the drop.

We used accurate material properties \( \gamma(\phi) \), \( \eta(\phi) \), found in literature \([39, 41, 53]\), and assumed \( D \sim \eta(\phi)^{-1} \) in accordance with the Stokes-Einstein relation. Simulations were initiated with a droplet of \( \sim 0.7 \mu \text{L volume and } \sim 30^{\circ} \text{ apparent contact angle, on top of a precursor in equilibrium with the vapor field of the droplet. See \([38]\) for details.}

Fig. 1 (c) compares the apparent contact angle observed in simulations with (dashed) and without (dash-dotted) Taylor-Aris dispersion, finding a quantitative agreement respectively with experimental data. Fig. 2 (b) shows stream lines and composition for a contracted droplet. The observed difference in composition between center and edge is merely 0.2 %. The most striking feature of the simulations is a quantitative reproduction of the experimentally observed surface tension contrast (Fig. 3) for the case with Taylor-Aris dispersion.

Origin and importance of Taylor-Aris dispersion are highlighted in Fig. 4. Panel (a) shows the strong but compensating capillary and Marangoni fluxes in the droplet. The effective diffusivity scales quadratically with the individual flux components, thus leading to strong effective diffusivities (panel (b)) in regions where the fluxes are large. Taylor-Aris dispersion amplifies diffusion by more than a factor 30, making it the governing phenomenon for the distribution of solutes in Marangoni-contracted droplets.

Typical droplet scales are \( r_0 \sim 1.5 \text{mm and } h_0 \sim 0.15 \text{mm} \), for which we obtain \( \text{Pe} \sim 8 \times 10^4 \) and \( \text{Pe}_z \sim 8 \times 10^2 \). Although this should be beyond the regime of Taylor-Aris dispersion, we observe quantitative agreement between simulated and measured velocities and surface tensions (Figs. 2, 3). This is not a coincidence but caused by very small dimensionless \( \bar{u} \) and \( \bar{\phi} \) when applying the natural scales. Measured velocities are \( \sim 2 \text{ mm/s} \) while we chose \( u_0 = \epsilon_h^2 \gamma_0/\eta \sim 47 \text{ mm/s} \). In addi-
tion, compositional differences in horizontal direction are \( \ll \mathcal{O}(1) \), set by an intricate coupling of evaporative enrichment and dispersion. The appropriate \( \text{Pe}_z \), for which \( \delta \phi = \mathcal{O}(1) \), is thus much smaller (~1/10^4) than the one calculated for natural scales.

**Conclusion.** — We present experimental data of internal flow fields and the apparent shape of Marangoni contracted drops of diol-water mixtures of various liquid concentrations and ambient humidities. The measurements allowed us to derive the surface tension gradient, which follows a power law \( \sim t^{-3/2} \). Through a long wave expansion for thin liquid films, we derived a general evolution equation for the vertically averaged composition with Taylor-Aris dispersion. The applicability of this equation depends on the Péclet numbers for characteristic horizontal respectively vertical scales, requiring \( \text{Pe} \gg 1 \gg \text{Pe}_z \), which is the case for many microliter-scale droplet applications. For Marangoni contracted drops, Taylor-Aris dispersion dominates the compositional evolution. We show that the model is in qualitative agreement with the experimental findings. Our theoretical analysis provides a general tool for describing thin films of liquid mixtures.

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