

Evaporative Flux Reversal of Binary Droplets: From Edge to Apex

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In this Letter, we directly measure the spatial variation of the saturation concentration along the interface of a binary droplet, showing that it is not uniform. This observation challenges the core assumption of classical evaporation theory, where a uniform interfacial saturation concentration leads to the edge-dominant evaporative flux responsible for the well-known coffee-ring effect in single-component droplets. Our interferometric measurements reveal that this assumption breaks down in multicomponent droplets, requiring a different framework to describe their evaporation behavior. By comparing the local evaporative flux with internal flow fields obtained using micro-particle image velocimetry, we show that changes in interfacial composition and flow pattern are closely coupled, leading the maximum evaporative flux to shift away from the contact line during evaporation. Recognizing that the maximum flux no longer resides at the edge is therefore essential for developing a correct and predictive understanding of evaporation in binary and more complex multicomponent droplets.

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For single-component droplets, evaporation has long been regarded as a remarkably elegant physical problem. Decades ago, the discovery that a sessile droplet evaporates with a strongly edge-enhanced evaporative flux—driving the characteristic outward coffee-ring flow and ultimately leaving a nonuniform ring-shaped deposition—highlighted the intrinsic edge-dominated nature of single-component evaporation [1,2]. The classical diffusion-limited model [3,4] not only established a clear theoretical foundation but also inspired a broad range of applications, including inkjet printing [5,6], spray cooling [7], evaporation-driven self-assembly [8,9], and various diagnostic techniques [10,11]. Because of its simplicity and predictive power, this model has become a cornerstone of modern droplet-based technologies.

However, droplets encountered in practical applications are rarely single-component. Most consist of mixtures—inks, polymer solutions, biofluids, and volatile-nonvolatile solvent blends—whose evaporation dynamics are far more intricate. When multiple components with different volatilities coexist, selective evaporation leads to strong concentration gradients inside the droplet [12–15] or even phase segregation in certain systems [16,17]. These compositional variations drive unsteady internal flows governed by solutal Marangoni stresses [18–21] and density-driven

convection [22–24], causing the evaporation to become inherently transient [25–28]. Although numerical and theoretical studies have suggested that such effects could fundamentally reshape the evaporative flux profile [18,29,30], direct experimental evidence of how the local flux is altered in a binary droplet has remained elusive.

In this Letter, using laser interferometry [31–33], we directly measure the time-dependent vapor concentration distribution along the liquid-gas interface of an evaporating binary droplet, here chosen to be an acetone-water mixture. From these measurements, we find that the saturation concentration of the volatile component is not always constant. Based on the measured concentration field, we reconstruct the local evaporative flux and identify a distinct flux-profile reversal: as evaporation proceeds, the location of the maximum flux gradually shifts from the edge toward the apex, as sketched in Fig. 1. Additionally, micro-particle image velocimetry (μ -PIV) measurements show that this

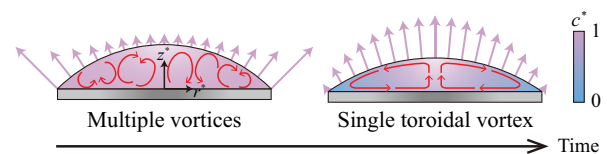


FIG. 1. Schematic illustration of the evolution of evaporative fluxes (light purple arrows) and internal flow patterns (red arrows) in an evaporating binary droplet. At early times (left), selective evaporation and the edge-dominated maximum flux generate multiple vortices. As evaporation progresses (right), the reversal of the evaporative flux leads to the formation of a single toroidal vortex. Here, c^* denotes the normalized concentration of the volatile component.

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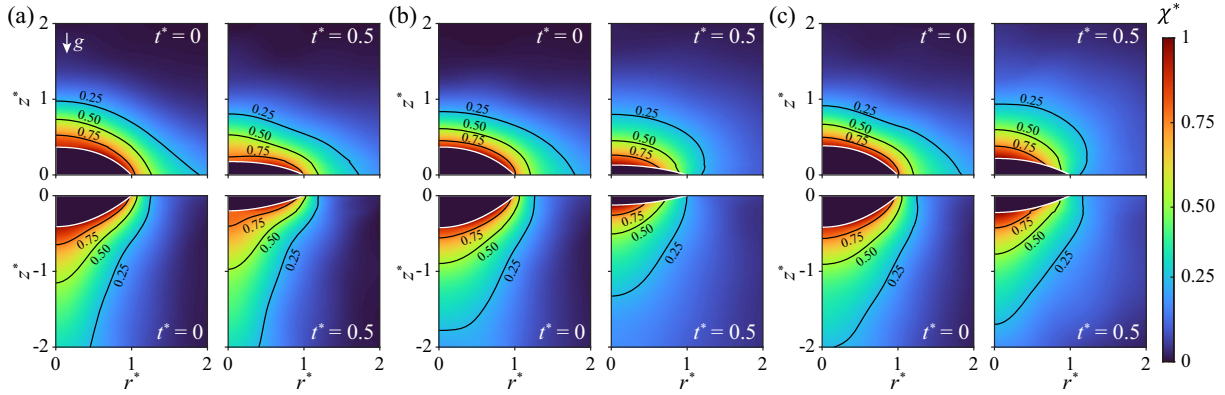


FIG. 2. Normalized mole fraction contours $\chi^* = \chi(r^*, z^*)/\chi_{\max}$ of acetone vapor around sessile (top) and pendant (bottom) droplets for initial concentrations of (a) 100%, (b) 75%, and (c) 50% acetone in a water droplet. Early ($t^* = 0$) and intermediate ($t^* = 0.5$) stages are shown, where $t^* = t/t_f$ and t_f is the total acetone evaporation time determined from the interferometry measurement. Both r and z are normalized by droplet radius $R = 2.5$ mm, i.e., $r^* = r/R$ and $z^* = z/R$. Gravity acts downward.

reversal coincides with a reorganization of the internal flow field, indicating that the evolving flux profile serves as a fundamental boundary condition governing the evaporation dynamics of binary droplets.

Experiments—For this Letter, droplets of 10 ± 0.5 μL with initial acetone concentrations of 100%, 75%, and 50% (v/v) were prepared using acetone (99.9%, Sigma-Aldrich) and deionized water (Merck Millipore Direct-Q3 UV) [see the mixture properties in Fig. S1 and Sec. I of the Supplemental Material (SM) [34]]. Each droplet was placed on a circular quartz substrate (radius 2.5 mm, thickness 1 mm), whose hydrophilic surface ensured spreading and pinning at the rim, fixing the contact radius at $R = 2.5$ mm. Experiments were performed in both sessile and pendant configurations to examine gravity effects. The initial contact angles were $44^\circ \pm 1^\circ$ (sessile) and $40^\circ \pm 1^\circ$ (pendant). All tests were conducted at $22^\circ\text{C} \pm 1^\circ\text{C}$ and $40\% \pm 5\%$ relative humidity.

Using a laser interferometer (see Appendix A for optical setup), we directly observed the vapor distribution of acetone around evaporating binary mixture droplets. The interference patterns were processed with a two-dimensional Fourier transform to extract phase shifts, which were then tomographically reconstructed into an axisymmetric refractive-index field using the Abel inverse transform. This refractive-index field was subsequently converted into the acetone mole fraction distribution through the Lorentz-Lorenz equation, and the evaporative flux was determined by applying Fick's first law of diffusion with a Stefan-flow correction (see Sec. II of SM [34] for details and Refs. [31,32]).

To quantify the internal flow patterns, we performed $\mu\text{-PIV}$ measurements by seeding fluorescent particles (2 μm , FMMG-1.3, Cospheric, USA) at a volume fraction of 0.003% (v/v) in the 75% and 50% binary droplets. The focal plane was positioned 25 μm above the quartz substrate, near the base of the droplet (see Fig. S6 of SM [34] for the PIV setup).

Comparison of acetone vapor distribution of evaporating single- and multicomponent droplets—To examine how the evaporation evolves over time, we compared the vapor distribution at the initial ($t^* = 0$) and intermediate ($t^* = 0.5$) stages of evaporation, where $t^* = t/t_f$ and t_f denote the total evaporation time of acetone. Here, t_f is defined as the moment at which acetone is fully evaporated from the droplet and no longer detectable in the interferometry measurement. The values of t_f were 75 s (sessile) and 63 s (pendant) at 100%, 186 s and 242 s at 75%, and 292 s and 305 s at 50%, respectively (see Figs. S4 and S5 of SM [34]).

For single-component droplets, the vapor distribution is primarily governed by diffusion, with the acetone vapor being slightly influenced by gravity due to the higher density of the acetone-air mixture relative to the surrounding air [Fig. 2(a)], consistent with previous observations [31,32,48]. Notably, the shape of each vapor cloud persists at both $t^* = 0$ and $t^* = 0.5$. In contrast, binary droplets exhibit markedly different behavior [Figs. 2(b) and 2(c)]. At the initial stage ($t^* = 0$), their vapor distributions resemble those of single-component droplets. However, by the intermediate stage ($t^* = 0.5$), the vapor field changes substantially, showing a pronounced reduction in vapor concentration near the contact line region in the 50% and 75% mixtures.

Generally, for sessile droplets with a small contact angle ($\theta < \pi/4$), the diffusion-limited model yields the evaporative flux as $J(r) \approx [2Dc_s(1 - \varphi)/\pi\sqrt{R^2 - r^2}]$, where D is the vapor diffusivity in air, φ is the relative humidity of a species, and c_s denotes the saturation concentration in the vapor phase, regarded as a constant [2]. The saturation concentration is defined by $c_s = P_{\text{vap}}M/R_uT$, where P_{vap} , M , R_u , and T are the vapor pressure, the molecular weight, the universal gas constant, and the ambient temperature, respectively. To satisfy the classical diffusion-limited model, c_s must remain spatially uniform along the interface.

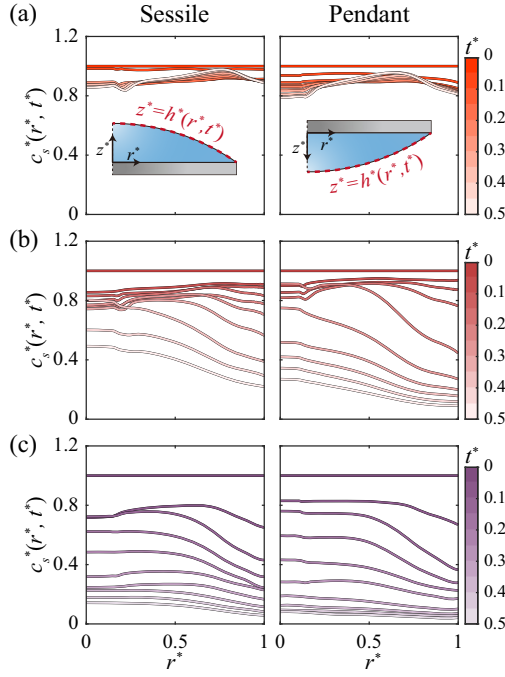


FIG. 3. Normalized saturation concentration profiles at the evolving droplet interface, $c_s^*(r^*, t^*) = c_s(r^*, t^*)/c_s(r^*, 0)$, evaluated at $z^* = h^*(r^*, t^*)$ (see the insets), as a function of the normalized radial position r^* and normalized time t^* . The left and right columns correspond to sessile and pendant droplets, respectively. Panels (a)–(c) show results for initial acetone concentrations of 100%, 75%, and 50%. Colors denote t^* , with lighter shades corresponding to later times.

To examine this condition, we extracted the interfacial mole fraction profile from the reconstructed vapor mole fraction field $\chi(r^*, z^*, t^*)$ as $\chi_s(r^*, t^*) = \chi(r^*, z^*, t^*)|_{z^*=h^*(r^*, t^*)}$, where $h^*(r^*, t^*) = h(r^*, t^*)/R$ denotes the normalized droplet height profile. Assuming local vapor-liquid equilibrium at the droplet interface, the corresponding saturation concentration was obtained as $c_s(r^*, t^*) = (P_{\text{atm}}M/R_uT)\chi_s(r^*, t^*)$ for an ideal gas mixture. For relative comparison, we normalized the profiles by their initial values as $c_s^*(r^*, t^*) = c_s(r^*, t^*)/c_s(r^*, 0)$ and examined their temporal and spatial variations (see Fig. 3).

In the case of pure acetone droplets (100%), c_s^* remains nearly uniform along the droplet interface throughout the early to intermediate stages of evaporation ($0 \leq t^* \leq 0.5$), while its overall magnitude decreases slightly as it evaporates, as shown in Fig. 3(a). In short, the saturation concentration along the interface can be considered effectively constant during evaporation.

In contrast, for binary droplets (75% and 50% acetone in water), the saturation concentration around $t^* = 0$ remains nearly uniform for both sessile and pendant droplets. As evaporation proceeds, however, c_s^* becomes nonuniform, as shown in Figs. 3(b) and 3(c), exhibiting a pronounced decrease near the contact line region due to the preferential depletion of acetone

there. By $t^* = 0.5$, the highest local saturation concentration appears near the apex (see Movie 1), indicating that the classical diffusion-limited model is not applicable to multicomponent droplet evaporation.

Transition of evaporative flux in drying binary droplets—From the vapor distribution measurements, we obtained snapshots of the evaporative flux profiles for each case and observed a clear reversal where the maximum flux shifts from the edge to the apex (see Fig. S3 of SM [34] and Movie 2). To examine the mechanism underlying this reversal, we compared the temporal evolution of the evaporative fluxes at different radial locations.

Figure 4 shows the fluxes near the apex ($0 < r^* < 0.25$) and the contact line ($0.75 < r^* < 1$) for sessile and pendant droplets at various initial acetone concentrations. For pure acetone droplets (100%), the contact line flux remains consistently higher throughout the early evaporation stage ($0 \leq t^* \leq 0.5$), in agreement with the classical edge-dominated model. Binary droplets behave differently. Although they initially follow the single-component trend, the evaporative flux at the contact line rapidly decreases as acetone is preferentially depleted near the edge, while the apex flux remains relatively high. The normalized flux crossover time $\tau^* = \tau/t_f$, at which the apex flux surpasses the edge flux, occurs earlier for lower acetone fractions (e.g., $\tau^* = 0.34$ and 0.21 for 75%, and $\tau^* = 0.07$ for 50%). After the crossover, the apex becomes the dominant evaporation site, consistent with the vapor distribution measurements. Despite differences in droplet orientation, the progression—edge dominance, crossover, and apex dominance—is qualitatively the same.

The corresponding internal flow structures evolve through three regimes: (I) multiple vortices, (II) transition, and (III) single toroidal vortex prior to acetone depletion ($0 < t^* < 1$) (see Figs. S7 and S8 of SM [34]). The yellow-shaded region in Fig. 4 marks the transition interval between regimes (I) and (III). After the depletion of acetone ($t^* > 1$), a coffee-ring flow was observed. Importantly, the flow transition between (I) and (III) coincides with the flux crossover, indicating that the reorganization of the internal flow is directly linked to the reversal of the evaporative flux profile.

As evaporation proceeds, the overall flux decreases with the bulk concentration. Near the end of regime (I), however, the evaporative flux at the contact line J_{edge} undergoes a sudden drop once the surface tension gradient becomes insufficient to sustain Marangoni-driven chaotic flow and the decreasing droplet height enhances viscous damping [27]. The weakened flow fails to replenish acetone from the center to the near edge, further accelerating the decline of J_{edge} . Consistently, PIV measurements show that the early-stage vortices dissipate without regeneration (see Movies 3–6). This abrupt loss of edge flux initiates the evaporative flux-profile reversal, marking the transition to apex-dominated evaporation ($J_{\text{apex}} > J_{\text{edge}}$).

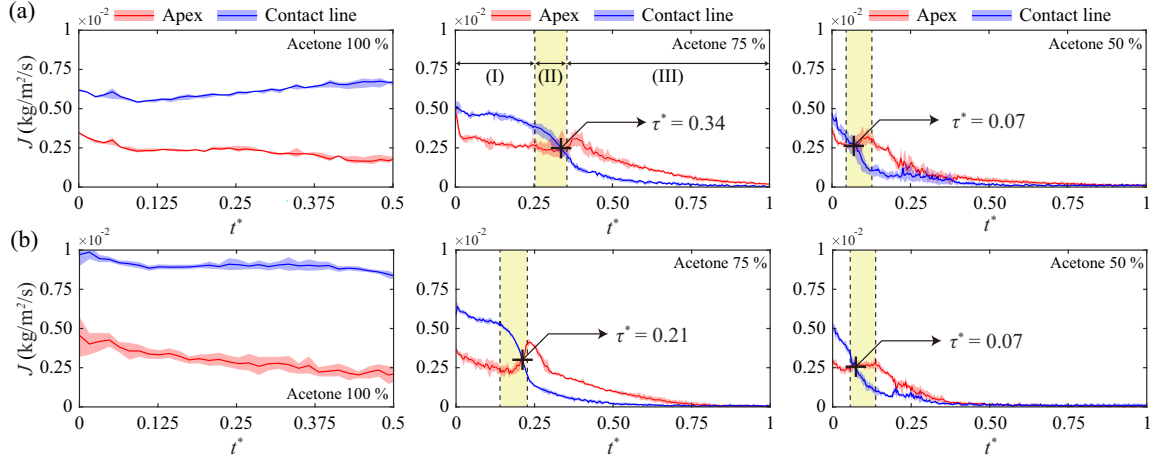


FIG. 4. Evolution of evaporative flux near the apex (red, $0 < r^* < 0.25$) and contact line (blue, $0.75 < r^* < 1$) for (a) sessile and (b) pendant droplets at initial acetone concentrations of 100%, 75%, and 50%, where $t^* = t/t_f$. Solid lines show averaged values, with shaded bands denoting min-max across three interferometric measurements. The flux crossover time τ^* is marked by +, and the yellow-shaded area indicates the flow transition period obtained from PIV experiments.

Apex-dominant evaporation and single toroidal vortical flow—Upon the reversal of the evaporative flux profile ($J_{\text{apex}} > J_{\text{edge}}$) [see (III) in Figs. 4(a) and 4(b)], we observed a single toroidal vortex (see Figs. S7 and S8 of SM [34] and Movies 3–6). The direction of internal flow reversed depending on droplet orientation: sessile droplets exhibited inward flow near the substrate, whereas pendant droplets showed outward flow, consistent with previous observations [22,23].

Previous interpretations of density-driven flow in binary droplets have invoked edge-dominant evaporation, inherited from the flux singularity of single-component droplets, whereby the contact line is assumed to sustain the strongest flux and thus the primary site of volatile depletion [21,22,26]. Our measurements demonstrate, however, that this maximum evaporative flux at the edge is rapidly suppressed by preferential acetone depletion near the contact line, leading to an evaporation profile in which the maximum flux shifts toward the apex.

The internal flow direction is therefore governed not by an instantaneous depletion due to edge singularity but by an internal composition gradient already established within the droplet, with an acetone-rich apex and a water-rich edge, independent of droplet orientation. In sessile droplets, this configuration is gravitationally stable and consistent with experimental observations [13–15], whereas in pendant droplets its persistence remains uncertain.

To elucidate the development of nonuniform internal composition quantitatively, we introduce the evaporative Péclet number as

$$\text{Pe}_{\text{evap}} = \frac{\tau_{\text{evap}}}{\tau_{\text{flow}}} = \frac{\rho_l w_{\text{bulk}} H U_{\text{avg}}}{J_{\text{avg}} R}, \quad (1)$$

which characterizes the competition between evaporation-driven depletion and flow-induced replenishment. Here, the

evaporation timescale is given by $\tau_{\text{evap}} = \rho_l w_{\text{bulk}} H / J_{\text{avg}}$, where ρ_l is the mixture density, w_{bulk} is the bulk acetone mass fraction, and $J_{\text{avg}} = 1/S \int_S J(r, t) dS$ is the spatially averaged evaporative flux where S and H are the surface area and maximum droplet height, respectively. The flow timescale is defined as $\tau_{\text{flow}} = R / U_{\text{avg}}$, where $U_{\text{avg}} = 1/A \int_A |\vec{V}| dA$ is the area-averaged flow velocity (see Fig. S9 of SM [34]) over the in-plane cross-sectional area A , and $\vec{V} = (u, v)$ denotes the in-plane velocity vectors.

Figure 5 represents the evolution of Pe_{evap} , calculated based on experimental measurements, including U_{avg} and J_{avg} . During the early stage with edge-dominant evaporation ($J_{\text{apex}} < J_{\text{edge}}$), we find $\text{Pe}_{\text{evap}} \approx \mathcal{O}(10^2) \gg 1$, indicating that Marangoni-flow-driven replenishment far outpaces evaporation, so that any local depletion of the volatile component is rapidly homogenized and the droplet maintains an approximately uniform composition.

By contrast, after transition to the apex-dominant evaporation, Pe_{evap} falls below unity, indicating that the rate of acetone depletion at the contact line already exceeds bulk replenishment by internal flow. Consequently, density-driven convection has a negligible effect on the internal composition distribution relative to evaporation, leaving acetone depletion at the edge uncompensated and establishing a persistent nonuniform concentration gradient along the interface until complete selective evaporation of acetone.

We further provided a scaling analysis of Pe_{evap} in Appendix B and found an order-of-magnitude reduction depending on the dominant flow-generation mechanism: a Marangoni-driven convection [see Eq. (B1)], which provides strong replenishment [$\text{Pe}_{\text{evap}} > \mathcal{O}(10^2)$], and a buoyancy-driven regime [see Eq. (B2)], in which the flow is much weaker [$\text{Pe}_{\text{evap}} \lesssim \mathcal{O}(10^1)$]. This transition explains

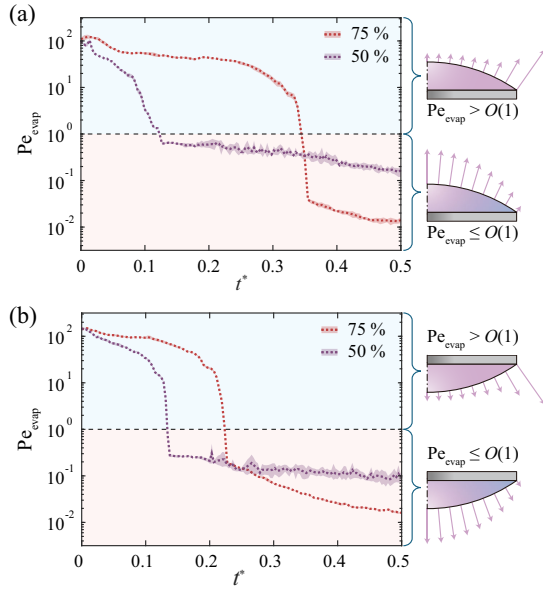


FIG. 5. Evolution of evaporative Péclet number $Pe_{\text{evap}} = \rho_l w_{\text{bulk}} H U_{\text{avg}} / (J_{\text{avg}} R)$ for sessile (a) and pendant (b) droplets. Dotted lines show averages; shaded bands span min-max across three interferometric measurements.

the loss of compositional homogenization and the emergence of persistent interfacial concentration gradients, which ultimately underlie the flux reversal.

Conclusion—In classical descriptions of droplet evaporation, the diffusion-limited model predicts a flux singularity at the contact line, making the edge the dominant site of mass loss. In contrast, by directly measuring the evaporative flux using laser interferometry, we show that the flux profile of binary droplets evolves over time, with the location of the maximum flux shifting from the edge to the apex—a flux reversal that occurs irrespective of droplet orientation. Moreover, this phenomenon is observed in several binary systems consisting of volatile-nonvolatile pairs (see Appendix C). For the representative acetone-water case, the evolution of the flux profile is strongly correlated with the development of internal flow, as elucidated by the evaporative Péclet number, which captures the balance between evaporation-driven depletion and flow-induced replenishment.

Our findings demonstrate that the contact line singularity is not universal in binary droplets: instead, the spatiotemporally evolving flux profile itself constitutes the fundamental boundary condition governing evaporation dynamics. We believe that our direct measurements of the flux profile in binary droplets provide a new foundation for understanding multicomponent droplet evaporation and for guiding its control in practical applications ranging from inkjet printing to uniform coating.

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Data availability—The data that support the findings of this article are publicly available [49].

- [1] R. D. Deegan, O. Bakajin, T. F. Dupont, G. Huber, S. R. Nagel, and T. A. Witten, Capillary flow as the cause of ring stains from dried liquid drops, *Nature (London)* **389**, 827 (1997).
- [2] R. D. Deegan, O. Bakajin, T. F. Dupont, G. Huber, S. R. Nagel, and T. A. Witten, Contact line deposits in an evaporating drop, *Phys. Rev. E* **62**, 756 (2000).
- [3] H. Hu and R. G. Larson, Evaporation of a sessile droplet on a substrate, *J. Phys. Chem. B* **106**, 1334 (2002).
- [4] H. Hu and R. G. Larson, Analysis of the microfluid flow in an evaporating sessile droplet, *Langmuir* **21**, 3963 (2005).
- [5] D. Lohse, Fundamental fluid dynamics challenges in inkjet printing, *Annu. Rev. Fluid Mech.* **54**, 349 (2022).
- [6] P. Calvert, Inkjet printing for materials and devices, *Chem. Mater.* **13**, 3299 (2001).
- [7] J. Kim, Spray cooling heat transfer: The state of the art, *Int. J. Heat Fluid Flow* **28**, 753 (2007).
- [8] W. Li, C. Zhang, and Y. Wang, Evaporative self-assembly in colloidal droplets: Emergence of ordered structures from complex fluids, *Adv. Colloid Interface Sci.* **333**, 103286 (2024).
- [9] J. Pyeon, S. M. Park, J. Kim, J.-H. Kim, Y.-J. Yoon, D. K. Yoon, and H. Kim, Plasmonic metasurfaces of cellulose nanocrystal matrices with quadrants of aligned gold nanorods for photothermal anti-icing, *Nat. Commun.* **14**, 8096 (2023).
- [10] R. Chen, L. Zhang, D. Zang, and W. Shen, Blood drop patterns: Formation and applications, *Adv. Colloid Interface Sci.* **231**, 1 (2016).
- [11] M. J. Hertaeg, R. F. Tabor, A. F. Routh, and G. Garnier, Pattern formation in drying blood drops, *Phil. Trans. R. Soc. A* **379**, 20200391 (2021).
- [12] X. Cai, N. Xie, Z. Qiu, J. Yang, M. He, K. S. Wong, B. Z. Tang, and H. Qiu, Aggregation-induced emission luminescence-based direct visualization of concentration gradient inside an evaporating binary sessile droplet, *ACS Appl. Mater. Interfaces* **9**, 29157 (2017).
- [13] H. Kim and H. A. Stone, Direct measurement of selective evaporation of binary mixture droplets by dissolving materials, *J. Fluid Mech.* **850**, 769 (2018).
- [14] A. K. Bell, J. Kind, M. Hartmann, B. Kresse, M. V. Höfler, B. B. Straub, G. K. Auernhammer, M. Vogel, C. M. Thiele, and R. W. Stark, Concentration gradients in evaporating binary droplets probed by spatially resolved Raman and NMR spectroscopy, *Proc. Natl. Acad. Sci. U.S.A.* **119**, e2111989119 (2022).
- [15] A. Erb, J. Kind, T. L. Zankel, R. W. Stark, and C. M. Thiele, Visualization and quantification of local concentration gradients in evaporating water/glycerol droplets with micrometer resolution, *Proc. Natl. Acad. Sci. U.S.A.* **122**, e2423660122 (2025).

- [16] H. Sadafi, R. Rabani, S. Dehaeck, H. Machrafi, B. Haut, P. Dauby, and P. Colinet, Evaporation induced demixing in binary sessile drops, *Colloids Surf. A* **602**, 125052 (2020).
- [17] Y. Li, P. Lv, C. Diddens, H. Tan, H. Wijshoff, M. Versluis, and D. Lohse, Evaporation-triggered segregation of sessile binary droplets, *Phys. Rev. Lett.* **120**, 224501 (2018).
- [18] C. Diddens, H. Tan, P. Lv, M. Versluis, J. Kuerten, X. Zhang, and D. Lohse, Evaporating pure, binary and ternary droplets: Thermal effects and axial symmetry breaking, *J. Fluid Mech.* **823**, 470 (2017).
- [19] H. Kim, Multiple Marangoni flows in a binary mixture sessile droplet, *Phys. Fluids* **34**, 122102 (2022).
- [20] C. Li, N. Zhang, X. Wang, and X. Ye, Marangoni instability of an evaporating binary mixture droplet, *Phys. Fluids* **35**, 108745 (2023).
- [21] B. Bozorgmehr and B. T. Murray, Numerical simulation of evaporation of ethanol–water mixture droplets on isothermal and heated substrates, *ACS Omega* **6**, 12577 (2021).
- [22] A. Edwards, P. Atkinson, C. Cheung, H. Liang, D. Fairhurst, and F. Ouali, Density-driven flows in evaporating binary liquid droplets, *Phys. Rev. Lett.* **121**, 184501 (2018).
- [23] Y. Li, C. Diddens, P. Lv, H. Wijshoff, M. Versluis, and D. Lohse, Gravitational effect in evaporating binary microdroplets, *Phys. Rev. Lett.* **122**, 114501 (2019).
- [24] C. Diddens, Y. Li, and D. Lohse, Competing Marangoni and Rayleigh convection in evaporating binary droplets, *J. Fluid Mech.* **914**, A23 (2021).
- [25] J. R. Christy, Y. Hamamoto, and K. Sefiane, Flow transition within an evaporating binary mixture sessile drop, *Phys. Rev. Lett.* **106**, 205701 (2011).
- [26] X. Zhong and F. Duan, Flow regime and deposition pattern of evaporating binary mixture droplet suspended with particles, *Eur. Phys. J. E* **39**, 18 (2016).
- [27] R. Bennacer and K. Sefiane, Vortices, dissipation and flow transition in volatile binary drops, *J. Fluid Mech.* **749**, 649 (2014).
- [28] X. Ma, K. Sefiane, R. Bennacer, X. Lapert, and F. Bakir, Solutal and gravitational effects during binary mixture droplets evaporation, *Microgravity Sci. Technol.* **36**, 17 (2024).
- [29] P. Sáenz, A. Wray, Z. Che, O. Matar, P. Valluri, J. Kim, and K. Sefiane, Dynamics and universal scaling law in geometrically-controlled sessile drop evaporation, *Nat. Commun.* **8**, 14783 (2017).
- [30] S. Karpitschka, F. Liebig, and H. Riegler, Marangoni contraction of evaporating sessile droplets of binary mixtures, *Langmuir* **33**, 4682 (2017).
- [31] S. Dehaeck, A. Rednikov, and P. Colinet, Vapor-based interferometric measurement of local evaporation rate and interfacial temperature of evaporating droplets, *Langmuir* **30**, 2002 (2014).
- [32] M. Kuk, J. Pyeon, and H. Kim, Vapor distribution changes evaporative flux profiles of a sessile droplet, *J. Colloid Interface Sci.* **652**, 646 (2023).
- [33] D. Shukla and P. K. Panigrahi, Digital holographic interferometry investigation of liquid hydrocarbon vapor cloud above a circular well, *Appl. Opt.* **59**, 5851 (2020).
- [34] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/m739-56q7> for sample selection, laser-interferometry processing, evaporative flux reconstruction, droplet geometry analysis, PIV measurements, and additional information, which includes Refs. [35–47].
- [35] L. R. Pendrill, Refractometry and gas density, *Metrologia* **41**, S40 (2004).
- [36] W. Acree and J. S. Chickos, Phase transition enthalpy measurements of organic and organometallic compounds and ionic liquids. Sublimation, vaporization, and fusion enthalpies from 1880 to 2015. Part 2. C11–C192, *J. Phys. Chem. Ref. Data* **46**, 013104 (2017).
- [37] G. Lugg, Diffusion coefficients of some organic and other vapors in air, *Anal. Chem.* **40**, 1072 (1968).
- [38] R. D. Johnson III, NIST Computational Chemistry Comparison and Benchmark Database, NIST Standard Reference Database Number 101, Release 22 (2022).
- [39] S. Song and C. Peng, Viscosities of binary and ternary mixtures of water, alcohol, acetone, and hexane, *J. Dispersion Sci. Technol.* **29**, 1367 (2008).
- [40] A. Estrada-Baltazar, A. De León-Rodríguez, K. R. Hall, M. Ramos-Estrada, and G. A. Iglesias-Silva, Experimental densities and excess volumes for binary mixtures containing propionic acid, acetone, and water from 283.15 K to 323.15 K at atmospheric pressure, *J. Chem. Eng. Data* **48**, 1425 (2003).
- [41] T. Grossmann and J. Winkelmann, Ternary diffusion coefficients of glycerol + acetone + water by Taylor dispersion measurements at 298.15 K, *J. Chem. Eng. Data* **50**, 1396 (2005).
- [42] K. S. Howard and R. McAllister, Surface tension of acetone–water solutions up to their normal boiling points, *AIChE J.* **3**, 325 (1957).
- [43] M. Takeda and K. Mutoh, Fourier transform profilometry for the automatic measurement of 3-D object shapes, *Appl. Opt.* **22**, 3977 (1983).
- [44] J.-F. Lin and X. Su, Two-dimensional Fourier transform profilometry for the automatic measurement of three-dimensional object shapes, *Opt. Eng.* **34**, 3297 (1995).
- [45] M. Kalal and K. Nugent, Abel inversion using fast Fourier transforms, *Appl. Opt.* **27**, 1956 (1988).
- [46] M. El Fagrich and H. Chehouani, A simple Abel inversion method of interferometric data for temperature measurement in axisymmetric medium, *Opt. Lasers Eng.* **50**, 336 (2012).
- [47] W. Thielicke and E. Stamhuis, PIVlab—towards user-friendly, affordable and accurate digital particle image velocimetry in MATLAB, *J. Open Res. Software* **2**, e30 (2014).
- [48] P. Kelly-Zion, C. J. Pursell, N. Hasbamer, B. Cardozo, K. Gaughan, and K. Nickels, Vapor distribution above an evaporating sessile drop, *Int. J. Heat Mass Transfer* **65**, 165 (2013).
- [49] M. Kuk and H. Kim, Supporting data for article “Evaporative flux reversal of binary droplets: from edge to apex”, Version 1.0, [Dataset], Zenodo (2026), [10.5281/zenodo.20923741](https://doi.org/10.5281/zenodo.20923741).
- [50] J. Ryu, H. S. Ko, and H. Kim, Vapor absorption and Marangoni flows in evaporating drops, *Langmuir* **38**, 2185 (2022).
- [51] S. Shin, I. Jacobi, and H. A. Stone, Bénard–Marangoni instability driven by moisture absorption, *Europhys. Lett.* **113**, 24002 (2016).
- [52] B. I. Shraiman, Diffusive transport in a Rayleigh–Bénard convection cell, *Phys. Rev. A* **36**, 261 (1987).

End Matter

Appendix A: Laser interferometer setup—The experimental setup utilizes a Mach-Zehnder interferometer as illustrated in Fig. 6(a). A He-Ne laser ($\lambda = 632.8$ nm, 10 mW, 1135P, JDSU, USA) serves as the light source, with its intensity modulated by a neutral density filter (NDF). To eliminate speckle noise and provide a sufficient field of view for the $R = 2.5$ mm droplet, a spatial filter (SF) expands the beam diameter to approximately 30 mm. This collimated beam is then partitioned by the first beam splitter (BS1) into a reference beam and a sample beam. The reference and sample beams travel different optical paths: path 1 for the reference beam and path 2 passes through the test section (TS) for the sample beam, respectively, and are merged at the second beam splitter (BS2). The resulting interference pattern is captured by a camera (D5500, Nikon, Japan) through an imaging system comprising an achromatic lens (L1) and a camera lens (L2, AF-P DX NIKKOR 70–300 mm, Nikon, Japan). By adjusting the tilt of the mirror (M3), the spatial frequency of the fringes is set within the range of $15 \text{ mm}^{-1} < |\mathbf{f}| < 18 \text{ mm}^{-1}$.

Figure 6(b) presents a raw interferometric image with an evaporating droplet placed in the test section. The evaporated vapor creates a refractive-index field, deforming the interference fringes. These distortions are most significant in

regions of high vapor accumulation, particularly near the droplet interface. The methodology for extracting physical quantities from raw images is provided in Sec. II of SM [34].

Appendix B: Scaling analysis of Pe_{evap} —While the evolution of Pe_{evap} in Fig. 5 is determined directly from experimental measurements using Eq. (1), we additionally perform a scaling analysis to evaluate whether the observed significant reduction in Pe_{evap} can be theoretically captured by the evaporation and flow timescales derived from thermophysical properties of the acetone-water system (see Table S1 and Fig. S1 of SM [34]). In this analysis, we focus on two quasisteady flow regimes where the flows are driven by well-defined mechanisms: (I) multiple vortices driven by surface tension gradients [25,27] and (III) a single toroidal vortex driven by density gradients [22,23].

To define Pe_{evap} based on scaling arguments, we first estimate the evaporation timescale τ_{evap} . Based on the mass loss rate of the small contact angle droplet, $\dot{m} = 4RDc_s$ [3], the evaporation timescale is expressed as $\tau_{\text{evap}} \sim (\rho_l V w_{\text{bulk}} / 4RDc_s)$, where $V \approx (\pi/2)R^2H$ denotes the droplet volume for $R \gg H$.

We then estimate the characteristic flow velocity U , and the flow timescale $\tau_{\text{flow}} = R/U$, for each regime. For (I) multiple vortices, we consider the stress balance between solutal Marangoni and viscous stresses at the free interface [19,50,51]: $(\Delta\gamma/R) \sim (\mu U/H)$, where μ is the dynamic viscosity. The surface tension difference $\Delta\gamma \sim |\partial\gamma/\partial w|\Delta w$ is induced by interfacial depletion of the volatile component, with $\Delta w = w_{\text{bulk}} - w_{\text{interface}}$. By adopting the Fick's second law of diffusion at the liquid-gas interface, the change in mass fraction during the flow timescale is identified as $\Delta w = (2J/\rho_l)(\tau_{\text{flow}}/\pi D_{\text{eff}})^{1/2}$ [51], where D_{eff} is the effective diffusion coefficient. Given the vigorous convection observed in our PIV measurements (see Movies 3–6), we utilize $D_{\text{eff}} \approx D_m \text{Pe}^{1/2} = (RUD_m)^{1/2}$ [51,52] in the limit of high Péclet number ($\text{Pe} = RU/D_m \gg 1$), where D_m is the mutual diffusion coefficient in the binary mixture. By scaling the evaporative mass flux as $J \sim (Dc_s/R)$, the surface tension difference becomes $\Delta\gamma \sim (2/\sqrt{\pi}) \cdot [(Dc_s|\partial\gamma/\partial w|)/(\rho_l R^{3/4}U^{3/4}D_m^{1/4})]$. Incorporating this into the stress balance and solving for U yields $U \sim (2/\sqrt{\pi})^{4/7} [(HDc_s|\partial\gamma/\partial w|)/\rho_l \mu D_m^{1/4} R^{7/4}]^{4/7}$. Consequently, the flow timescale is characterized as $\tau_{\text{flow}} \sim R^2 [\rho_l \mu D_m^{1/4} / HDc_s |\partial\gamma/\partial w|]^{4/7}$. Finally, the evaporative Péclet number scales as

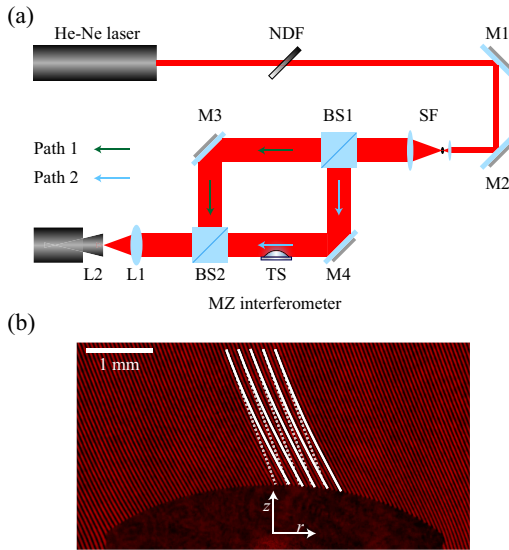


FIG. 6. (a) Schematic of the optical setup, consisting of a spatial filter and a Mach-Zehnder interferometer. The detailed optical system is described in Appendix A. (b) Typical raw interferometric image obtained with (a). The deviation of white solid lines from dashed lines highlights a gradual deformation of the interference fringe by the refractive-index field.

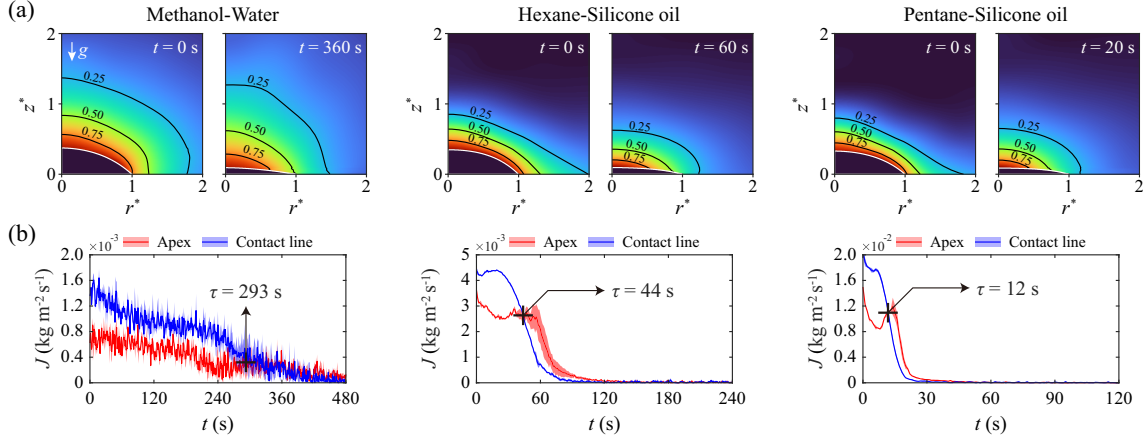


FIG. 7. (a) Normalized mole fraction contours $\chi^* = \chi(r^*, z^*)/\chi_{\max}$ of volatile component around the evaporating binary droplets for various binary systems, with an initial volume fraction of 75% for the volatile component (methanol, hexane, and pentane). Early and intermediate stages are shown. (b) Evolution of evaporative flux near the apex (red, $0 < r^* < 0.25$) and contact line (blue, $0.75 < r^* < 1$). Solid lines indicate averaged values, with shaded bands denoting min-max values. The flux crossover time τ is marked by +.

$$\text{Pe}_{\text{evap}}^{(\text{I})} \sim \frac{\pi}{8} w_{\text{bulk},0} \left(\frac{H}{R} \right) \left[\frac{H \rho_l^{3/4} |\partial \gamma / \partial w|}{\mu D_m^{1/4} (Dc_s)^{3/4}} \right]^{4/7}. \quad (\text{B1})$$

The theoretical flow velocities at the initial ($t^* = 0$), Marangoni-dominated regime, were approximately 15 and 13 mm/s for 75% and 50% initial acetone concentrations, showing good agreement with the experimentally measured maximum flow velocity of $\mathcal{O}(10 \text{ mm/s})$ [see Fig. S9(b) of SM [34]]. The corresponding $\text{Pe}_{\text{evap}}^{(\text{I})}$ values from Eq. (B1) were 757 and 491 for the 75% and 50% cases, respectively.

As the flow structure transitions into (III) single toroidal vortex, we consider the equilibrium between the gravitational and viscous forces [23], $g \Delta \rho_l \sim \mu \nabla^2 U$, where g is the gravitational acceleration. The density difference can be approximated using the solutal expansion coefficient β , $\Delta \rho_l \approx \rho_l \beta \Delta w$. Unlike regime (I), where Δw represents the local interfacial depletion that drives solutal Marangoni flow, the driving force in regime (III) is the density contrast between the bulk interior and the interface. Consequently, Δw is derived from the overall mass change during flow timescale as $\Delta w = \Delta m / m_0 \sim (R Dc_s \tau_{\text{flow}} / \rho_l V)$. Substituting this into the density relation yields $\Delta \rho_l \sim \beta (2 Dc_s \tau_{\text{flow}} / \pi R H)$. The flow velocity is then determined as $U \sim (2 Dc_s H \beta g / \pi \mu)^{1/2}$, leading to the following flow timescale: $\tau_{\text{flow}} \sim (\pi \mu R^2 / 2 Dc_s H \beta g)^{1/2}$. Ultimately, the evaporative Péclet number is derived as

$$\text{Pe}_{\text{evap}}^{(\text{III})} \sim \left(\frac{\pi}{32} \right)^{1/2} w_{\text{bulk}} \left(\frac{\rho_l^2 H^3 \beta g}{\mu Dc_s} \right)^{1/2}. \quad (\text{B2})$$

We calculated the flow velocities in regime (III) from the acetone concentrations around $t^* \approx \tau^*$, inferred from the geometric evolution of the droplet (see Fig. S4 of SM [34]),

obtaining values of 1.1 and 1.4 mm/s for the 75% and 50% cases, respectively. While these predicted velocities substantially overestimate the experimental measurements—which remain on the order of $\mathcal{O}(0.01\text{--}0.1 \text{ mm/s})$ —such discrepancies are consistent with scaling-based estimates of density-driven flows in previous literature [23]. This discrepancy is further expected here because regime (III) corresponds to a later stage of evaporation in which the mixture properties are more difficult to determine accurately. Nevertheless, the corresponding $\text{Pe}_{\text{evap}}^{(\text{III})}$ values obtained from Eq. (B2) were 16 and 32, respectively, indicating a significant reduction in Pe_{evap} from regime (I) to regime (III) by approximately a factor of $10^1\text{--}10^2$, which qualitatively agrees with the experimental trend shown in Fig. 5.

*Appendix C: Flux reversal in different binary systems—*To assess whether the observed flux reversal extends across different binary mixtures (see Table S1 of SM [34]), we carried out interferometric measurements under identical conditions for methanol-water, hexane-silicone oil (10 cSt), and pentane-silicone oil (10 cSt), each with an initial concentration of 75% of the more volatile component. The resulting vapor distributions and representative evaporative fluxes at the apex and contact line are shown in Fig. 7.

All systems exhibit a qualitatively similar flux reversal: evaporation is initially edge-dominant and later becomes apex-dominant [Fig. 7(b)]. The flux crossover time, τ , however, varies systematically among systems, reflecting changes in viscosity, surface tension gradient, and volatility. For systems with comparable viscosity and surface tension gradient, lower volatility leads to a longer τ , as seen in methanol-water (293 s) compared with acetone-water (63 s). By contrast, at comparable volatility, τ decreases as

viscosity increases, and the surface-tension gradient decreases, as seen in hexane-silicone oil (44 s); it decreases further in pentane-silicone oil (12 s), where higher volatility reinforces this trend.

These results indicate that the observed flux reversal is not limited to a specific binary mixture but can occur across multiple systems, where evaporation-driven depletion and flow-induced replenishment coexist.