

Master 2 Internship

Title: Coalescence at the Nanoscale: the Puzzling Role of Thermal Fluctuations

Type: Experimental and Theoretical

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Project: The coalescence of liquid droplets is an important scientific issue with numerous practical implications, such as fog nets for water harvesting, drying of smart textiles, or wing-coating engineering in aeronautics, to name a few. The underlying principles are now well established. However, fundamental and applied developments in nanofluidics raise questions about the validity of classical, continuum fluid dynamics since compositional and surface fluctuations near interfaces become increasingly important at small scales.

To address such a matter, a promising strategy is to use critical thermodynamic systems - in our case a near-critical phase-separated liquid-liquid mixture - as (i) fluctuations diverge at the critical point (so-called critical opalescence) and thus can be significantly amplified at will to reach the typical sub-millimetric length scales accessible to optical imaging methods [1], and (ii) universality in terms of Ising class is expected, so that any result is conceptually transposable to any liquid belonging to the same class. The focus of this project is thus on the coalescence of out-of-equilibrium wetting droplets in such phase-separated critical mixtures [2]. Experimentally, we plan to use the optical radiation pressure of laser waves at a near-critical liquid-liquid interface to produce jets [4] and eventually out-of-equilibrium wetting droplets [3], close enough to trigger their coalescence by the optical force of a laser beam located in between them. Due to near-criticality, the physical properties of these droplets are dictated by the temperature proximity to the critical point, and thus tunable by the chosen temperature. After a fluctuation-free calibration far enough from the critical point, we will investigate the near critical deviations from the latter, and their relations with the increasing roles of density fluctuations, gravity and evaporation when the critical point is neared by performing coalescence experiments for varying optical forces at temperatures closer and closer to the critical one.

Theoretically, asymptotic and/or numerical solutions of stochastic lubrication models [5] will be constructed for the current geometry [6], and compared quantitatively to experimentations. A collaboration with the group of Prof. Andreas Carlson at Univ. Oslo, Norway, is envisioned for the numerical aspects. Altogether, the goal is to establish for the first time on solid experimental & theoretical grounds and quantitative comparisons, the fundamental crossover between the capillary-dominated and fluctuation-dominated regimes [7] of dynamical wetting and coalescence.

We plan to further develop and extend this work within the frame of a PhD.

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[3] Saiseau, R., Pedersen, C., Benjana, A., Carlson, A., Delabre, U., Salez, T., & Delville, J.-P. Near-critical spreading of droplets. *Nature Communications*, 13, 7442 (2022).

[4] Giroto, A., Petit, J., Saiseau, R., Guérin, T., Chraïbi, H., Delabre, U., & Delville, J.-P. Conical interfaces between two immiscible fluids induced by an optical laser beam. *Physical Review Letters*, 122, 174501 (2019).

[5] Pedersen, C., Niven, J. F., Salez, T., Dalnoki-Veress, K. & Carlson, A. Asymptotic regimes in elastohydrodynamic and stochastic leveling on a viscous film. *Physical Review Fluids*, 4, 124003 (2019)

[6] Davidovitch, B., Moro, E., & Stone, H. A. Spreading of viscous fluid drops on a solid substrate assisted by thermal fluctuations. *Physical review letters*, 95, 244505 (2005).

[7] Petit, J., Rivière, D., Kellay, H., & Delville, J.-P. Break-up dynamics of fluctuating liquid threads. *Proceedings of the National Academy of Sciences*, 109, 18327 (2012).