Rubber-like elasticity in laser-driven free surface flow of a Newtonian fluid

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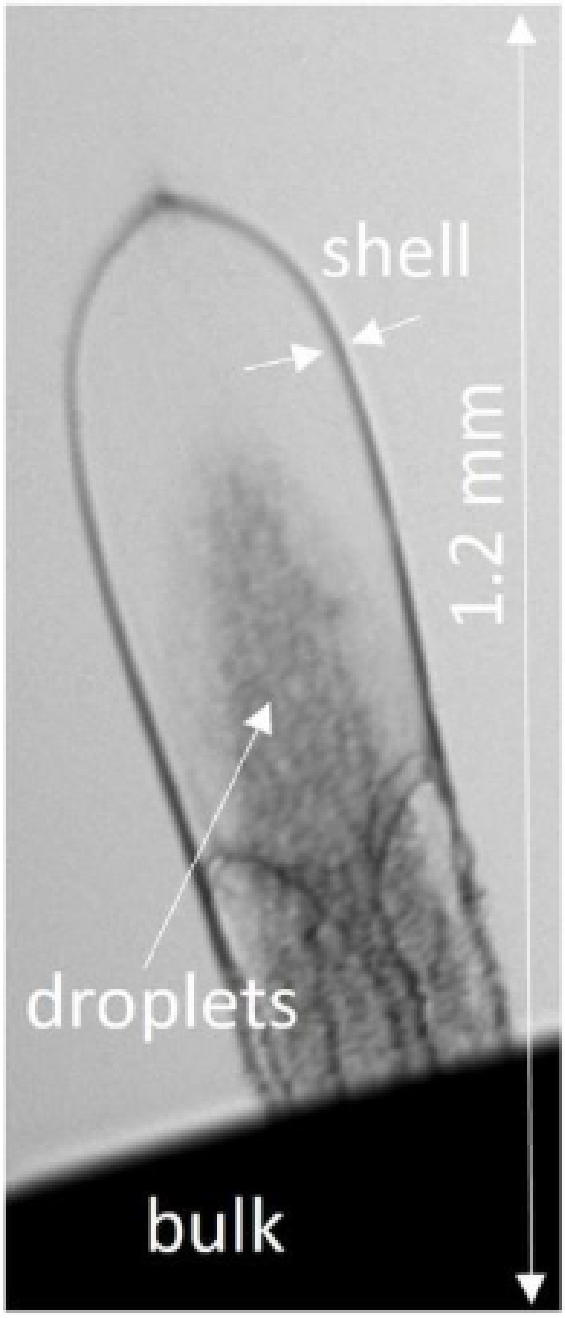

Viscosity characterizes the ability of a fluid to flow. The energy required to deform a viscous liquid to flow is dissipated as heat. In an elastic solid, the deformation energy is stored and can be recovered. Although it is a good approximation to describe perfect crystals as elastic and mobile fluids as viscous, in general it is a matter of the time scales of the experiment to emphasize different degrees of viscous or elastic behavior. Newtonian fluids, such as small-molecule liquids like water and glycerol are examples of almost pure viscous behavior since the relevant inter-molecular relaxation times of stored elastic energy are well below the nanosecond. This shear elasticity of liquid glycerol can be quantified in a linear small strain regime by e.g. acoustic wave measurements and other high-frequency approaches or, remarkably, at very low frequencies [2] with rheological techniques by identifying minuscule elastic contributions. Here, we present evidence of a rubber-like elastic large response in a Newtonian liquid at surprisingly large strains. The elasticity -not viscosity- fully dominates the free-surface flow dynamics of this laser-driven glycerol plume at strain rates as high as 1e6/s. The elasticity persists for microseconds and hence several orders of magnitude longer than the relaxation time t of e.g. a Maxwell-Debye model. This persistence cannot arise from the tail of a distribution or spectrum of relaxation times around t but presumably appears by somehow frustrating the fast single-molecule dissipation. Dissipation is closely related to the Stokes-Einstein diffusion coefficient, a fundamental concept of viscous fluids. For times considerably longer than the existence of this shear elasticity, it requires long-range correlations typical for solids and incompatible with the short-range interaction associated with Newtonian fluids —this poses a challenge for our present understanding of the liquid state.

The elastic shell created by photomechanical spallation (Fig. 1) is only a few micrometers thick. While this does not constitute "nano" confinement, where bulk properties are largely lost, it nevertheless considerably confines the thickness compared to the lateral dimensions of the bubble. This would find support from a theory by Zaccone and Trachenko of viscoelasticity of confined liquids stressing that confinement increases liquid shear elasticity by suppressing long-wavelength shear waves responsible for dissipation [3].

References

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[3] A. Zaccone et al., Proc. Natl. Acad. Sci. U. S. A. 117, 19353 (2020).