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# Mechanics of Supercooled Liquids

*Pure substances can often be cooled below their melting points and still remain in the liquid state. For some supercooled liquids, as further cooling slows down viscous flow greatly, but does not slow down self-diffusion as much. We formulate a continuum theory that regards viscous flow and self-diffusion as concurrent, but distinct, processes. We generalize Newton's law of viscosity to relate stress, rate of deformation, and chemical potential. The self-diffusion flux is taken to be proportional to the gradient of chemical potential. The relative rate of viscous flow and self-diffusion defines a length, which, for some supercooled liquids, is much larger than the molecular dimension. A thermodynamic consideration leads to boundary conditions for a surface of liquid under the influence of applied traction and surface energy. We apply the theory to a cavity in a supercooled liquid and identify a transition. A large cavity shrinks by viscous flow, and a small cavity shrinks by self-diffusion. [DOI: 10.1115/1.4028587]*

## 1 Introduction

In a pure liquid, molecules of the same species touch one another but frequently change neighbors. The rate of change has long been studied in experiments of two types. External forces cause the liquid to deform by viscous flow, characterized by the viscosity [1]. Thermal agitation causes individual molecules to migrate by self-diffusion, characterized by the self-diffusivity [2–4]. In a given liquid, the two kinetic processes often obey the Stokes–Einstein relation

$$\frac{\eta D}{kT} = \frac{1}{Cb} \quad (1)$$

where  $\eta$  is the viscosity,  $D$  the self-diffusivity,  $T$  the temperature,  $k$  the Boltzmann constant,  $b$  a length characteristic of the size of the molecules, and  $C$  a dimensionless number depending on the shape of the molecules. As the temperature drops, the liquid increases its viscosity and decreases its self-diffusivity, but the Stokes–Einstein relation predicts that the ratio  $\eta D/kT$  is a constant independent of temperature. In his original paper [5], Einstein did not analyze self-diffusion; rather, he analyzed a macroscopic particle diffusing in a liquid using the continuum theory of Stokes flow. Nonetheless the Stokes–Einstein relation holds for particles down to the molecular scale [6]. In particular, the

relation holds for self-diffusion for many liquids over wide ranges of temperature [7].

The Stokes–Einstein relation even holds for some supercooled liquids, substances that remain in the liquid state and do not crystallize when the temperature drops below their melting points [8]. For instance, water has a melting point of 0 °C, but can remain in the liquid state down to  $-37.5$  °C [9]. As the temperature drops, a supercooled liquid like silica increases its viscosity and reduces its self-diffusivity, both by orders of magnitude, but keeps the ratio  $\eta D/kT$  independent of temperature (Fig. 1).

In recent decades, however, evidence has accumulated that some supercooled liquids violate the Stokes–Einstein relation [10–16]. When the temperature drops, such a supercooled liquid increases its viscosity steeply (Fig. 1(a)), but does not reduce its self-diffusivity as steeply (Fig. 1(b)), such that the ratio  $\eta D/kT$  increases often by orders of magnitude (Fig. 1(c)).

For a liquid that obeys the Stokes–Einstein relation, viscous flow and self-diffusion proceed through a single rate-limiting process: molecules change neighbors. By contrast, for a liquid that violates the Stokes–Einstein relation, viscous flow and self-diffusion proceed by distinct rate-limiting processes. In many cases, a supercooled liquid forms a dynamic structure that consists of regions larger than individual molecules [17–30]. Molecular rearrangement is much slower in some regions than others (Fig. 2). Viscous flow proceeds by disrupting the dynamic structure, but self-diffusion proceeds by the migration of individual molecules through the fast regions. Consequently, as the temperature drops, the dynamic structure increasingly jams viscous flow, but does not retard self-diffusion as much.

As molecular dynamic simulations and microscopic experiments continue to shed light on the physics and chemistry of supercooled

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# Supercooled Liquids

**J Rink**



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