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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, a 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 η 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

John T. Fourkas



Supercooled Liquids:

Structural Glasses and Supercooled Liquids Peter G. Wolynes, Vassiliy Lubchenko, 2012-03-12 With contributions from 24 global experts in diverse fields and edited by world recognized leaders in physical chemistry chemical physics and biophysics **Structural Glasses and Supercooled Liquids Theory Experiment and Applications** presents a modern complete survey of glassy phenomena in many systems based on firmly established characteristics of the underlying molecular motions as deduced by first principle theoretical calculations or with direct single molecule experimental techniques A well rounded view of a variety of disordered systems where cooperative phenomena which are epitomized by supercooled liquids take place is provided These systems include structural glasses and supercooled liquids polymers complex liquids protein conformational dynamics and strongly interacting electron systems with quenched self generated disorder Detailed calculations and reasoned arguments closely corresponding with experimental data are included making the book accessible to an educated non expert reader

Functional Organic Liquids Takashi Nakanishi, 2019-03-12 The first book to comprehensively cover the burgeoning new class of soft materials known as functional organic liquids Functional organic liquids a new concept in soft matter materials science exhibit favorable properties compared to amorphous polymers and ionic liquids They are composed of a functional core unit and a side chain which induces fluidity even at room temperature Due to their fluidity functional organic liquids can adopt any shape and geometry and fulfill their function in stretchable and bendable devices for applications in photovoltaics organic electronics biomedicine and biochemistry Presented in five parts this book starts with an overview of the design methods and properties of functional organic liquids The next three parts focus on the applications of this exciting new class of soft materials in the fields of energy conversion nanotechnology and biomaterials They study the liquids for energy conversion those containing inorganic nanoclusters and solvent free soft biomaterials Functional Organic Liquids concludes with a comparison in terms of properties and application potential between functional organic liquids and more conventional soft matter such as ionic liquids and liquid metals Examines the current state of science and technology for functional organic liquids Focuses on potential and already realized applications such as functional organic liquids for energy conversion Stimulates researchers to move forward on future development and applications Functional Organic Liquids is an excellent book for materials scientists polymer chemists organic chemists physical chemists surface chemists and surface physicists

Dynamics of Supercooled Liquids Albert Chun Pan, 2005

The Dynamic Properties of Supercooled Liquids Gilroy Harrison, 1976 Summary 2000 feature film directed by Tom Tykwer This film follows Sissi who is a nurse in a psychiatric hospital On her way to the bank to do a favour for a friend she is run over by a truck A young drifter Bodo Riemer saves her life by performing a DIY tracheotomy and then vanishes as he is wanted by the police for robbery Sissi makes a full recovery and becomes determined to find her saviour In German with English subtitles

Statistical Study of Freezing in Supercooled Liquids David Arthur Tansey, 1958 **Supercooled**

Liquids, Glass Transition and Bulk Metallic Glasses: Volume 754 Takeshi Egami, 2003-07-16 There has been a renaissance in glass science brought about by the development of concepts such as fragility index and the energy landscape with megabasins Research on bulk metallic glasses has been explosive since their advent when MRS offered its first book on the topic In 2000 a second book broadened the scope to include supercooled liquid bulk glassy and nanocrystalline states This book enhances the scope to include glass transition in diverse materials such as water silicate and polymeric melts Bringing these threads together in an interdisciplinary manner was fruitful and offers proof that while there is much common ground gaps between various approaches to the glassy state remain to be bridged Subjects include the supercooled liquid glass formability structural relaxation and dynamics structure determination and modeling processing and applications of bulk metallic glasses mechanical properties mechanical properties composites crystallization electronic and magnetic structure and properties and nanoparticles and nonmetallic glasses

Apparatus and Method for Measuring Concentrations of Supercooled Liquid Water Geoffrey E. Hill, 1984 [A Study of Orientational Dynamics in Supercooled Liquids](#) Rick Stanley Francis, 1999 **The Freezing of Supercooled Liquids** Charles Alfred Knight, 1967

Freezing of Supercooled Water N. Ernest Dorsey, 1948 Detailed discussion of experiments made since 1936

Supercooled Liquid, Bulk Glassy and Nanocrystalline States of Alloys: Volume 644 Akihisa Inoue, 2001-10-05 The MRS Symposium Proceeding series is an internationally recognised reference suitable for researchers and practitioners

An Introduction to the Principles of Physical Chemistry from the Standpoint of Modern Atomistics and Thermo-dynamics Edward Wight Washburn, 1915

Supercooled Liquids John T. Fourkas, 1997 This book presents some of the most exciting recent work on supercooled liquids Topics include domain models of supercooled liquids inhomogeneity and polymorphism and Mode Coupling Theory and its applications It provides in depth coverage of supercooled water and of the connections between supercooled liquids and the conformational dynamics of proteins The introduction includes a detailed discussion of terminology major problems and leading theoretical and experimental approaches

Concepts of Supercooling Lars Grund, 2015-02-10 This book brings a comprehensive elaboration on the fundamentals of supercooling phenomenon The existence of supercooled liquids can be traced in the atmosphere various creatures metallurgy and diverse industrial processes For centuries maintaining the stability of the metastable supercooled liquid and inspiring the associated process of nucleation have both been topics of great scientific interest This text is valuable as a resource of beginning guidance for professionals keen on exploring supercooling of water and water based solutions in biology and industry This book also elucidates the modeling and the configuration of subsequent dendritic development of supercooled solutions and glass transitions and interface balance

Precipitation Enhancement Roscoe Braham, 2015-03-30 This book is the outcome of a workshop held at Park City Utah 23 25 May 1984 It is a collection of papers focusing focuses on physics of precipitation formation in clouds and the response of clouds to glaciogenic seeding This book documents the debates and discussions that

surrounded the topic of glaciogenic seeding during the time of the workshop It is interesting as a historical evidence of the scientific progress of that time **Physics Briefs** ,1992 **Journal of the Chemical Society** Chemical Society (Great Britain),1900 Titles of chemical papers in British and foreign journals included in Quarterly journal v 1 12 Metastable, Mechanically Alloyed and Nanocrystalline Materials 1995 Robert Schulz,1996-07-01 Proceedings of the International Symposium on Metastable Mechanically Alloyed and Nanocrystalline Materials ISMANAM 95 held in Qu bec Canada July 1995 The London, Edinburgh and Dublin Philosophical Magazine and Journal of Science ,1900 Bulletin of the Chemical Society of Japan Nihon Kagakkai,1985

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