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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

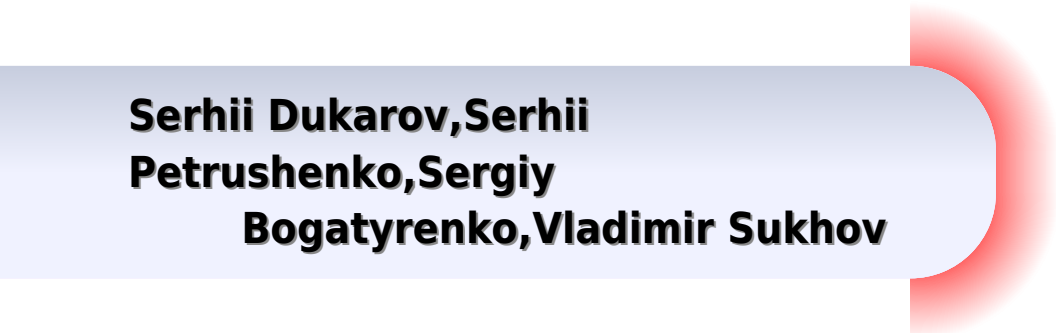
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Supercooled Liquids

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Petrushenko, Sergiy
Bogatyrenko, Vladimir Sukhov**



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

Metastable Liquids Pablo G. Debenedetti, 2020-06-16 *Metastable Liquids* provides a comprehensive treatment of the properties of liquids under conditions where the stable state is a vapor a solid or a liquid mixture of different composition It examines the fundamental principles that govern the equilibrium properties stability relaxation mechanisms and relaxation rates of metastable liquids Building on the interplay of kinetics and thermodynamics that determines the thermophysical properties and structural relaxation of metastable liquids it offers an in depth treatment of thermodynamic stability theory the statistical mechanics of metastability nucleation spinodal decomposition supercooled liquids and the glass transition Both traditional topics such as stability theory and modern developments including modern theories of nucleation and the properties of supercooled and glassy water are treated in detail An introductory chapter illustrates with numerous examples the importance and ubiquity of metastable liquids Examples include the ascent of sap in plants the strategies adopted by many living organisms to survive prolonged exposure to sub freezing conditions the behavior of proteins at low temperatures metastability in mineral inclusions ozone depletion the preservation and storage of labile biochemicals and the prevention of natural gas clathrate hydrate formation All mathematical symbols are defined in the text and key equations are clearly explained More complex mathematical explanations are available in the appendixes

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

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

Non-equilibrium Phenomena In Supercooled Fluids, Glasses And Amorphous Materials - Proceedings Of The Workshop Mario P Tosi,Dino Leporini,Marco Giordano,1996-09-20 This volume contains the Proceedings of the International Workshop on Non Equilibrium Phenomena in Supercooled Fluids Glasses and Amorphous Materials held in Pisa in the early fall of 1995 as a joint initiative of the University of Pisa and of the Scuola Normale Superiore The goal was to bring together liquid state physicists chemists and engineers to review current developments and comparatively discuss experimental facts and theoretical predictions in this vast scientific area The core of the Workshop was a set of general lectures followed by more specific presentations on current issues in the main areas of the field This structure has been maintained in this volume in which a set of five overviews is followed by topically grouped contributions in the five areas of ionic glasses and glassy materials the glass transition viscous flow and microscopic relaxation complex fluids and polymers The volume also preserves a record of the many short contributions given to the Workshop through posters which are grouped in it under the subjects of inorganic glasses organic glasses and complex fluids polymers and theoretical aspects

Statistical Physics of Liquids at Freezing and Beyond Shankar Prasad Das,2011-06-16 Exploring important theories for understanding freezing and the liquid glass transition this book is useful for graduate students and researchers in soft condensed matter physics chemical physics and materials science It details recent ideas and key developments providing an up to date view of current understanding The standard tools of statistical physics for the dense liquid state are covered The freezing transition is described from the classical density functional approach Classical nucleation theory as well as applications of density functional methods for nucleation of crystals from the melt are discussed and compared to results from computer simulation of simple systems Discussions of supercooled liquids form a major part of the book Theories of slow dynamics and the dynamical heterogeneities of the glassy

state are presented as well as nonequilibrium dynamics and thermodynamic phase transitions at deep supercooling

Mathematical treatments are given in full detail so readers can learn the basic techniques **The Freezing of**

Supercooled Liquids Charles Alfred Knight, 1967 **Theory of Simple Liquids** Jean-Pierre Hansen, I.R.

McDonald, 2006-02-08 The third edition of Theory of Simple Liquids is an updated advanced but self contained introduction to the principles of liquid state theory It presents the modern molecular theory of the structural thermodynamic interfacial and dynamical properties of the liquid phase of materials constituted of atoms small molecules or ions This book leans on concepts and methods from classical Statistical Mechanics in which theoretical predictions are systematically compared with experimental data and results from numerical simulations The overall layout of the book is similar to that of the previous two editions however there are considerable changes in emphasis and several key additions including up to date presentation of modern theories of liquid vapour coexistence and criticality areas of considerable present and future interest such as super cooled liquids and the glass transition the area of liquid metals which has grown into a mature subject area now presented as part of the chapter ionic liquids Provides cutting edge research in the principles of liquid state theory Includes frequent comparisons of theoretical predictions with experimental and simulation data Suitable for researchers and post graduates in the field of condensed matter science Physics Chemistry Material Science biophysics as well as those in the oil industry

Dynamics of Supercooled Liquids Albert Chun Pan, 2005

Formation and Temperature Stability of the Liquid

Phase in Thin-Film Systems Serhii Dukarov, Serhii Petrushenko, Sergiy Bogatyrenko, Vladimir Sukhov, 2023-11-25 This book presents a summary of the topic of supercooling during crystallization in condensed films While recent findings are mainly published in English the foundational classical results were originally published in Russian with limited accessibility to general readers The present work is based on a 2019 Ukrainian monograph Temperature Stability of the Supercooled Liquid Phase in Condensed Films which has been extensively revised and expanded The book includes a detailed analysis of the thermodynamics of supercooled fluids with updated and expanded sections Additionally new results on the supercooling of indium lead In Pb alloys in contact with amorphous molybdenum and fusible metals in contact with nanocrystalline layers are presented These layers occupy a middle ground between amorphous carbon molybdenum as deposited germanium films and polycrystalline copper silver aluminum substrates The book gives particular attention to the peculiarities of contracted geometry conditions which are natural for multilayered structures and can occur through fusible component segregation at grain boundaries The analysis of new data has prompted a rethinking of the role of the more refractory layer s microstructure on the crystallization processes of metastable melts The book includes a thorough discussion of these findings highlighting the crucial role of the microstructure in the crystallization process This book is a valuable resource for researchers and students interested in crystallization in thin film metallic systems This comprehensive study provides a detailed and authoritative analysis of the thermodynamics of supercooled fluids and the impact of microstructure on the

crystallization processes of metastable melts making it an essential addition to any academic library

Decoding **Supercooled Liquids**: Revealing the Captivating Potential of Verbal Expression

In a period characterized by interconnectedness and an insatiable thirst for knowledge, the captivating potential of verbal expression has emerged as a formidable force. Its power to evoke sentiments, stimulate introspection, and incite profound transformations is genuinely awe-inspiring. Within the pages of "**Supercooled Liquids**," a mesmerizing literary creation penned by a celebrated wordsmith, readers embark on an enlightening odyssey, unraveling the intricate significance of language and its enduring impact on our lives. In this appraisal, we shall explore the book's central themes, evaluate its distinctive writing style, and gauge its pervasive influence on the hearts and minds of its readership.

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