

Review

Reactive Molecular Dynamics in Ionic Liquids: A Review of Simulation Techniques and Applications

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Abstract: Ionic liquids exhibit distinctive solvation and reactive properties, making them highly relevant for applications in energy storage, catalysis, and CO₂ capture. However, their complex molecular interactions, including proton transfer and physisorption/chemisorption, necessitate advanced computational efforts to model them at the atomic scale. This review examines key molecular dynamics approaches for simulating ionic liquid reactivity, including quantum-mechanical methods, conventional reactive force fields such as ReaxFF, and fractional force fields employed in PROTEX. The strengths and limitations of each method are assessed within the context of ionic liquid simulations. While quantum-mechanical simulations provide detailed electronic insights, their high computational cost restricts system size and simulation timescales. Reactive force fields enable bond breaking and formation in larger systems but require extensive parameterization. These approaches are well suited for investigating reaction pathways influenced by the local environment, which can also be partially addressed using multiscale simulations. Fractional force fields offer an efficient alternative for simulating significantly larger reactive systems over extended timescales. Instead of resolving individual reaction mechanisms in full detail, they incorporate reaction probabilities to model complex coupled reactions. This approach enables the study of macroscopic properties, such as conductivity and viscosity, as well as proton transport mechanisms like the Grotthuß process—phenomena that remain inaccessible to other computational methods.

Keywords: proton transfer; reactive force field; molecular dynamics; polarizable force field



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1. Introduction

Solvents are often considered secondary players in many chemical reactions [1], primarily contributing solvation energy to the overall free energy of the system. However, their direct interaction with reactants can significantly impact reaction pathways by lowering activation energies or generating electric fields that alter the potential energy surface. Among solvents, ionic liquids (ILs) are a fascinating class of charged, dipolar systems capable of modulating and accelerating various chemical reactions [2,3].

1.1. Proton Transfer Reactions in Ionic Liquids

Protic ILs [4–7] have garnered significant attention due to their dual functionality as solvents and reactants, particularly in proton transfer reactions exemplified by the equilibrium:



Dynamics Of Molecular Liquids

Gerald Reinhard Kneller, Alfons Geiger



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Dynamics of Molecular Liquids Walter G. Rothschild, 1984-01-20 Good No Highlights No Markup all pages are intact Slight Shelfwear may have the corners slightly dented may have slight color changes slightly damaged spine

Molecular Liquids A.J. Barnes, W.J. Orville-Thomas, J. Yarwood, 2012-12-06 This ASI was planned to make a major contribution to the teaching of the principles and methods used in liquid phase research and to encourage the setting up of collaborative projects as advocated by the European Molecular Liquids Group secretary Dr J Yarwood University of Durham U K During the past five years considerable progress has been made in studying molecular liquids The undoubted advantages of international collaboration led to the formation of the European Molecular Liquids Group EMLG in July 1981 The activities of the EMLG were widely disseminated in a special session of the European Congress on Molecular Spectroscopy EUCMOS held in September 1981 for details see *J Mol Structure* 80 1982 375 421 Following the success of this meeting it was thought that the aims and objectives of the E G would be best served by the organisation of a broader based gathering designed to attract those interested in the study of the structure dynamics and interactions in the liquid state Thanks to the generous support by the Scientific Affairs Division of NATO it was possible to hold a NATO ASI on Molecular Liquids at the Italian Centre of Stanford University Florence Italy during June July 1983 This book is based on the lectures presented at that meeting The contents of this volume cover the three broad areas of current liquid phase research a Analytical theory

Scaling of structure and dynamics in molecular liquids, **The Structure and Dynamics of Molecular Liquids** R. W. K. Impey, 1982 [Molecular Liquids](#), 1984 **Dynamics of Molecular Liquids Under Conditions of High Pressure and Geometric Confinement** Scott L. Wallen, 1994 Raman spectroscopy is utilized to further the understanding of the molecular dynamics of liquids The fluids are studied under conditions of high pressure and confined to spatial geometries whose dimensions approach the molecular diameters of the fluids The dynamic processes studied include intermolecular and intramolecular energy transfer vibrational and reorientational relaxation mechanisms and the influence of attractive and repulsive molecular interactions Typically when one changes the temperature of a system the kinetic energy of the system is altered however equally important changes in the molecular packing or volume occur High pressure techniques can be utilized to separate effects on the fluid molecular dynamics due to changes in the packing and those due to changes in the kinetic energy This is accomplished by examining trends of the molecular parameters as temperature is varied at constant density and as density is varied under isothermal conditions The liquid systems examined in this manner are furan propylene carbonate chloroethylene carbonate and dichloroethylene carbonate In the first system a previously reported nonmonotonic temperature dependence of the bandwidth is shown to be due to density changes Also the results of frequency and bandwidth changes as a function of density and temperature are interpreted in terms of available theoretical models In the carbonate systems the noncoincidence effect the peak frequency and the isotropic bandwidths are investigated as a function

of density and temperature Another area investigated is the fluid molecular dynamics when the fluid's motion is restricted to spaces approaching its own molecular diameter This is accomplished by studying the fluid imbibed in porous sol gel prepared xerogels with narrow pore size distributions The local order of the confined systems are very different from those in the bulk fluid as are various relaxation mechanisms that the molecules undergo The observed differences have a significant impact on the understanding of chromatographic and membrane separation processes and the mechanisms of reaction in confined media The effect of geometric confinement on the symmetric stretching modes of methyl iodide is investigated with particular emphasis placed on the resonant intermolecular vibrational coupling of the ν_{22} vibrational mode The results indicate that the liquid has greater orientational order and slower resonant energy transfer when confined to the pores of the xerogel hosts The noncoincidence effect of confined acetone N N dimethylformamide and carbon disulfide is investigated and it is determined that confinement does not alter the noncoincidence value but specific interactions between the confined liquid and the silanol groups on the xerogel surface cause a decrease in the noncoincidence value The effect of confinement on the liquid molecular dynamics of carbon disulfide are examined The reorientational correlation times are observed to increase significantly upon confinement and the results are interpreted in terms of a model which predicts a steric hindrance of molecular motions at and near the surface The vibrational dephasing times are shown to decrease as a function of decreasing pore size while the intramolecular Fermi resonance coupling strength decreases with decreasing pore size as does the anharmonic force constant Lastly the gauche anti population ratio of 1,2 dichloroethane is investigated and the results show that specific surface interactions between the liquid and the xerogel's silanol groups cause a change in the population ratio however geometric confinement has no effect within the experimental error Abstract shortened by UMI

Structure and Motion in Molecular Liquids, 1978 **Scaling of Structure and Dynamics in Molecular Liquids** Erik Hillo Lørup, 2024 **Novel Approaches to the Structure and Dynamics of Liquids: Experiments, Theories and Simulations** Jannis Samios, Vladimir A. Durov, 2004-05-31 Proceedings of the NATO Advanced Study Institute Rhodes Greece from 6 to 15 September 2002 **Molecular Liquids: New Perspectives in Physics and Chemistry** José Teixeira, 2012-12-06 In its combination of an advanced teaching standpoint with an emphasis on new perspectives and recent advances in the study of liquids formed by simple molecules *Molecular Liquids: New Perspectives in Physics and Chemistry* provides a clear understandable guide through the complexities of the subject A wide range of topics is covered in the areas of intermolecular forces statistical mechanics the microscopic dynamics of simple liquids thermodynamics of solutions nonequilibrium molecular dynamics molecular models for transport and relaxation in fluids liquid simulations statistical band shape theories conformational studies fast exchange dynamics and hydrogen bonding The experimental techniques covered include neutron scattering X ray diffraction IR Raman NMR quasielastic neutron scattering and high precision time resolved coherent Raman spectroscopy **Ultrafast Dynamics of Molecular Liquids Investigated by Femtosecond Light**

Scattering Bern Kohler,1990

Structure and Dynamics of Molecular Liquids Maximilian Rudolf Becker,2025

Dynamics at the Onset of Self-diffusion in Molecular Liquids Humphrey Morhenn,2013 The Structure of Molecular

Liquids Laurent Bianchi,2000

DENSITY-FUNCTIONAL MOLECULAR DYNAMICS SIMULATIONS OF SHOCKED MOLECULAR LIQUIDS. ,2001

Molecular dynamics MD simulations have been performed for highly compressed fluid deuterium nitrogen and oxygen in the density and temperature regime of shock compression experiments using density functional DF electronic structure techniques to describe the interatomic forces The Hugoniot derived from the calculated equation of state for deuterium does not exhibit the large compression predicted by the recently reported laser driven experiments However the Hugoniot derived for nitrogen and oxygen agree well with explosively driven and gas gun experiments The nature of the fluid along the Hugoniot as calculated with DF MD has been analyzed All three species D₂ N₂ and O₂ undergo a continuous transition from a molecular to a partially dissociated fluid containing a mixture of atoms and molecules *Dynamics of Complex Molecular Liquids - Computer Simulations and Experiments* Gerald Reinhard

Kneller,Alfons Geiger,2000

Microscopic Structure and Dynamics of Liquids J. Dupuy,2012-12-06

What did we have in mind when in May 1976 we Professor de Gennes Dr Tourand and ourselves thought of a Summer School in the field of liquids First we wanted to present and discuss the new results that have been obtained recently in particular at the high flux reactor of the Institut Laue Langevin in Grenoble since it became operational in 1972 In order to achieve this goal the major part of this Summer School was devoted to an extensive presentation of the general concepts and methods of studying this state of matter time dependent correlation functions molecular dynamics intermolecular forces spectroscopic techniques and concentrated on a few specific systems which have seen significant development in the last few years both theoretically and experimentally These systems are the different classes of simple liquids metallic liquids ionic liquids simple molecular liquids and the new field of superionic conductors solid electrolytes Furthermore we wanted to put some emphasis on a particular research area in the field of liquids namely critical phenomena in fluids This was chosen both because of our personal interest in this field and the major theoretical advances which have occurred in the last ten years We also wished that some new powerful techniques or new theoretical approaches be presented at this School Thus picosecond laser techniques theoretical calculations on dipolar fluids and angular correlations in molecular liquids were the subject of specific seminars

Molecular Liquids: New Perspectives in Physics and Chemistry ,1992-09-30

Proceedings of the NATO Advanced Study Institute Luso Portugal September 22 October 3 1991

Molecular Liquids Christopher Hohman Marshall,1988

Exploring Life Phenomena with Statistical Mechanics of Molecular Liquids Fumio Hirata,2020-02-12

In a living body a variety of molecules are working in a concerted manner to maintain its life and to carry forward the genetic information from generation to generation A key word to understand such processes is water which plays an essential role in life phenomena This book sheds light on life phenomena which are woven by biomolecules as warp and water as weft by means of statistical

mechanics of molecular liquids the RISM and 3D RISM theories both in equilibrium and non equilibrium A considerable number of pages are devoted to basics of mathematics and physics so that students who have not majored in physics may be able to study the book by themselves The book will also be helpful to those scientists seeking better tools for the computer aided drug discovery Explains basics of the statistical mechanics of molecular liquids or RISM and 3D RISM theories and its application to water Provides outline of the generalized Langevin theory and the linear response theory and its application to dynamics of water Applies the theories to functions of biomolecular systems Applies the theories to the computer aided drug design Provides a perspective for future development of the method

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