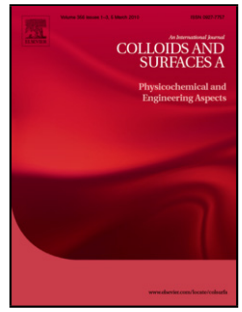
Accepted Manuscript



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PII: S0927-7757(17)30581-2
DOI: http://dx.doi.org/doi:10.1016/j.colsurfa.2017.06.019
Reference: COLSUA 21706

To appear in: *Colloids and Surfaces A: Physicochem. Eng. Aspects*

Received date: 28-2-2017
Revised date: 26-5-2017
Accepted date: 8-6-2017

Many facets of intermittent dynamics in colloidal and molecular glasses

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Abstract

A popular picture of the intermittent dynamics of glassy materials depicts each particle as localized in a temporary cage formed by its neighbors, and seldom jumping to another cage. Drawing on experiments on hard sphere colloidal glasses and molecular dynamic simulations of supercooled liquids, here we show how this simplified scenario leads to quantitative relations between single particle motion and macroscopic properties, within a Continuous Time Random Walk (CTRW) framework. For example, the diffusivity and the relaxation time at different length scales can be directly predicted from the jump properties. In the accessible range of temperatures and volume fractions, these predictions work well, although the CTRW formalism neglects the spatially heterogeneous dynamics of glassy systems. In this respect, we show how these dynamic heterogeneities can be highlighted from a study of the correlations between jumps. Finally, the intermittent motion is exploited to infer correlations between local structure and dynamics.

Keywords: Colloidal suspensions, Structural glasses, Supercooled liquids, Dynamics, Relaxation

1. Introduction

Intermittent single-particle dynamics is widespread in soft materials and may lead to very peculiar phenomena, especially, but not only [1], when the relaxation process is sluggish and heterogeneous as in glass-forming systems. Indeed, such a dynamic signature is a well known property of molecular supercooled liquids, including water [2] and polymer melts [3], and of dense suspensions of hard-sphere colloidal particles [4]. In addition, a similar behaviour is observed in a wide variety of complex fluids, whose dynamics is dominated by particle crowding. These include foams, emulsions, and granular materials [5] as well as biological systems, such as epithelial cell tissues and micro-swimmers [6].

In glass-forming liquids, intermittent dynamics is explained assuming each particle to rattle for a long time in the cage formed by its neighbors and to suddenly jump to another cage [7, 8]. This simple cage-jump picture has been invoked to indirectly explain some distinctive features of the macroscopic glassy dynamics, like the intermediate time plateau of the mean square displacement and of the dynamic correlation functions, as well as rheological properties, such as yielding and flow of sheared colloidal glasses [9]. This suggests that quantitative investigations of the cage-jump motion might allow relating the structural relaxation and the single particle dynamics. The simplest approach [10, 11, 12, 13, 14] to quantify the intermittent single-particle motion involves the analysis of real-space trajectories, so as to identify the intervals particles stay localized (the cages), and those in which fast irreversible rearrangements occur (the jumps). Once trajectories are properly segmented in cages and jumps, the resulting statistical properties can be connected to the macroscopic dynamics. Commonly, this is done assuming the particles to perform a Continuous Time Random Walk (CTRW) [15]. The use of the CTRW formalism to describe the dynamics involves two strong assumptions, whose validity is not obvious, especially in deeply supercooled liquids. First of all, one assumes that jumps are the only relaxation mechanism, whereas other non-intermittent relaxation channels, such as slow and continuous rearrangements, may be in principle relevant. Secondly, spatial correlations between cage-jumps are not explicitly considered. Thus, the CTRW approach essentially neglects Dynamic Heterogeneity (DH), a well–known feature of glass-formers [16]. Indeed, on approaching the glass transition, extended re-

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