Gulliver
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Nonlinear response functions in glassy fluids: What do they probe?

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Scientific context: Dense liquids gradually solidify at low temperature via a physical process called the glass transition. While well-known and understood at the macrosopic scale, very little is known from direct experimental measurements about the dynamics of molecules in the vicinity of the glass transition. A microscopic understanding of this important process remains a lively research problem, which has resisted several decades of careful studies [1].

In the last 10 years, non-linear dielectric measurements have been performed in molecular systems, that are interpreted as a unique way to probe emerging glassy correlations in these systems [2]. In parallel, modern computer simulations techniques were recently developed to study the motion of molecular models at very long times [3], revealing the existence of a complex hierarchy of correlated molecular motion in these dense glassy fluids.

While the emergence of non-trivial correlations in liquids near a glass transition is well accepted, there is no consensus and no theoretical understanding of their precise nature. Are these correlations related to an underlying phase transition accompanied by structural correlations, or instead are these correlations purely dynamic in nature, characterising the correlated motion of molecules with no relation to any static phase transition?

Description of the thesis: In this project we propose to use the newly developed numerical approaches to systematically measure non-linear response functions similar to the ones determined experimentally, while resolving in space and time the molecular dynamics that give rise to this signal. By measuring systematically static and dynamic correlations, and their relation to non-linear response functions, a microscopic interpretation of the physical content of non-linear response functions will be available. Ultimately, this work will provide the missing link between the competing theoretical approaches and the available experimental observations.

Supervision: The project will take place at Gulliver (ESPCI UMR 7083 CNRS) and be supervised by L. Berthier (DR, CNRS). The lab has developed well-known expertise for the study of disordered and non-equilibrium materials, and has a large experience regarding the numerical simulations of these systems [4]. The work will also be performed in close collaborarion with: C. Scalliet (LPENS, UMR 8023) who has developed a numerical expertise in advanced simulation techniques for supercooled liquids [3]; J-P. Bouchaud (CFM), who first suggested to measure non-linear response functions in supercooled liquids [4], and F.

Ladieu (CEA Saclay) who has measured experimentally non-linear response functions in several molecular liquids [2].

Références :

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