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Energy Landscape–Based Study of Atomic Displacements in Glass Forming Materials

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1. Introduction

In this work [1-2] we investigate the role of inherent structures (ISs), i.e., minima on the potential energy hypersurface with respect to all microscopic degrees of freedom, in the dynamics of glass forming materials by studying the mean square atomic displacement (MSD), which is related to the self-diffusivity (D_s) via the Einstein equation ($6D_s = \lim_{t\to\infty} \partial \left\langle r^2 \right\rangle / \partial t$).

2. Method and Results

For temperatures below the glass temperature (T_g) we have developed a model that describes the dynamics of the atomistic system as a series of transitions, following first-order kinetics and Poisson statistics, between basins surrounding the ISs. Our validation of this model is based on hazard analysis of the motion out of individual basins, as sampled by atomistic molecular dynamics (MD) simulations, and on its ability to reproduce the MSD resulting from full-blown MD trajectories. Our results for a model two-component Lennard-Jones mixture [3] clearly show [1] that our Poisson process approach is able to reproduce the IS dynamics obtained directly from MD over a wide range of sub-T_g temperatures, from 9 K to 38 K. At higher temperatures, the long-time diffusive motion of atoms in the system can be captured simply by tracking the MSD between IS. We utilize and extend this methodology in order fully to reconstruct the system dynamics in the form of the MSD as a function of time at finite temperatures. We consider the succession of transitions in a network of basins and evaluate the contribution of vibrational motion around each IS through short MD runs artificially trapped within each one of the basins. We provide [2] the mathematical formulation for "lifting" the coarse-grained Poisson process model of transitions between states back to the atomistic level. Our approach is in excellent agreement with the full MD for temperatures around and below the T_g of our system (see Figure 1), where the Poisson approximation is valid. In order to apply our proposed methodology to temperatures around and above T_g we need to sample several hundreds of minima, monitoring rare events not between basins, but between collections of basins, also known as metabasins. To accomplish this, we developed two approaches: **a)** distribution of the computational load in a parallel procedure that demands the same computational cost as the corresponding conventional MD run but affords obtaining results more than two orders of magnitude faster on the real time scale; **b)** improvement of the dynamical sampling via a temperature-biased technique that can access transitions to neighboring metabasins at small computational

cost. The first approach is necessary for the implementation of the second. Applying the first approach, we have quantified the Speed-up factor (S_p), i.e. how faster is a parallel algorithm than the corresponding sequential one. As one can see in Figure 2, our approach has great efficiency even for 250 processors.

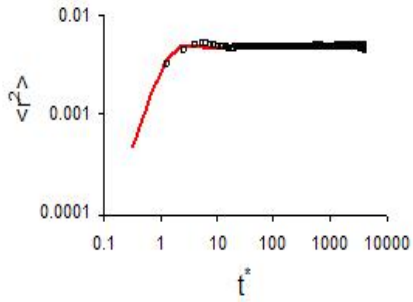

Fig. 1: Reconstruction of the MSD at 38 K by the proposed method (line) compared to MD data(□). Units adapted from [3].

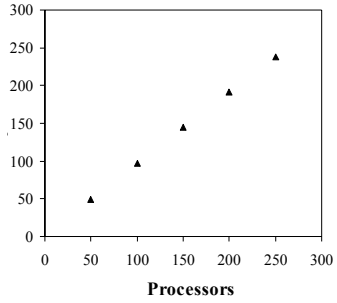

Fig. 2: Speed-up factor (S_p) as a function of the number of processors used.

3. Conclusions

The dynamical transitions between basins surrounding ISs can be well described by a first order kinetic scheme at low temperatures. We provide the mathematical formulation for "lifting" the coarse-grained Poisson process model of a succession of interbasin or intermetabasin transitions and reproducing the full dynamics of the atomistic system.

4. Acknowledgments

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