MAR13-2012-020465

Abstract for an Invited Paper
for the MAR13 Meeting of
the American Physical Society

Reversible and Irreversible Behavior of Glass-forming Materials from the Standpoint of Hierarchical Dynamical Facilitation

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Using molecular simulation and coarse-grained lattice models, we study the dynamics of glass-forming liquids above and below the glass transition temperature. In the supercooled regime, we study the structure, statistics, and dynamics of excitations responsible for structural relaxation for several atomistic models of glass-formers. Excitations (or soft spots) are detected in terms of persistent particle displacements. At supercooled conditions, we find that excitations are associated with correlated particle motions that are sparse and localized, and the statistics and dynamics of these excitations are facilitated and hierarchical. Excitations at one point in space facilitate the birth and death of excitations at neighboring locations, and space-time excitation structures are microcosms of heterogeneous dynamics at larger scales. Excitation-energy scales grow logarithmically with the characteristic size of the excitation, giving structural-relaxation times that can be predicted quantitatively from dynamics at short time scales. We demonstrate that these same physical principles govern the dynamics of glass-forming systems driven out-of-equilibrium by time-dependent protocols. For a system cooled and re-heated through the glass transition, non-equilibrium response functions, such as heat capacities, are notably asymmetric in time, and the response to melting a glass depends markedly on the cooling protocol by which the glass was formed. We introduce a quantitative description of this behavior based on the East model, with parameters determined from reversible transport data, that agrees well with irreversible differential scanning calorimetry. We find that the observed hysteresis and asymmetric response is a signature of an underlying dynamical transition between equilibrium melts with no trivial spatial correlations and non-equilibrium glasses with correlation lengths that are both large and dependent upon the rate at which the glass is prepared. The correlation length corresponds to the size of amorphous domains bounded by excitations that remain frozen on the observation time scale, thus forming stripes when viewed in space and time. We elucidate properties of the striped phase and show that glasses of this type, traditionally prepared through cooling, can be considered a finite-size realization of the inactive phase formed by the s-ensemble in the space-time thermodynamic limit.