Static and dynamic lengthscales in glassforming liquids

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Abstract:

Among the mysteries of dynamical arrest is solidification without significant change in structure. In recent years, higher-order structural motifs have been identified in a number of model glassformers, with the geometry specific to the particular system. [1-3] While concurrent growth of lengthscales associated with these structural motifs with dynamic lengthscales would provide strong evidence for a structural mechanism for vitrification, in the dynamical regime accessible to colloid experiment and computer simulation, such concurrent scaling is controversial, with the dynamic lengthcsales often showing rather greater growth with supercooling [2-4].

A second mystery lies in the so-called isoconfigurational ensemble, in which many dynamical realizations starting from the same configuration are realized. Propensity, the ensemble-averaged mobility, is highly nonuniform indicating that a particle's likelihood of moving is somehow encoded in the initial configuration, yet no direct link with structure has yet been established [5].

Here we address both of these challenges, by identifying a propensity-based dynamical lengthscale. This dynamic lengthscale scales in a comparable way to structural lengthscales. Moreover, being a single-particle quantity, it can directly reveal local heterogeneities in dynamic lengthscales similar to dynamical heterogeneity in structural relaxation times. Finally, by correlating our analysis with the topological cluster classification, we are able to identify those local structures prone to relax (high propensity) with those that are long-lived (low propensity). Thus we resolve the conundrum relating structure and propensity, and introduce a local dynamic lengthscale which scales in a similar way to structural lengthscales.

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