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Locally Favoured Structures and Dynamic Arrest

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The mechanism by which a liquid may become arrested, forming a glass or gel, is a long standing problem of condensed matter physics. While possible dynamic mechanisms have received considerable attention, the prevailing view is that “the arrangement of atoms and molecules in glass is indistinguishable from that of a liquid.” On the contrary, here we present direct experimental evidence of a structural mechanism for dynamical arrest. In particular, long-lived (energetically) locally favoured structures (LFS), whose geometry may prevent the system relaxing to its equilibrium state, have long been thought to play a key role in dynamical arrest. Here we propose a general definition of LFS which we identify with a novel topological method and combine these with experiments on colloidal liquid-gel, and glass-liquid-glass transitions. In these systems, the equilibrium state is crystal-fluid coexistence, and a crystal respectively: in both colloidal gels and glasses, suppression of crystallisation forms a key part of dynamical arrest. The population of LFS is a strong function of (effective) temperature in the ergodic liquid phase, rising sharply approaching dynamical arrest. We show that the “arms” of the colloidal gel are entirely comprised of LFS, which we argue form on shorter timescales than crystallisation. Gelation and dynamical arrest are identified with the formation of a percolating network of LFS, which we show are intimately related to dynamical heterogeneities. We seek to demonstrate that LFS can provide a structural order parameter for dynamical arrest. In the case of the glass-liquid-glass transition exhibited by “sticky spheres” at high density, like the colloidal gel, the LFS provide a clear structural signature of dynamical arrest.