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Two-State "Hopping" Dynamics in Molecular Liquids and Glasses[1] MARCUS CICERONE, QIN ZHONG, MADHUSUDAN TYAGI, JOSEPH CURTIS, National Institute of Standards and Technology, DEVIN AVERETT, University of Wisconsin-Madison, JUAN DE PABLO, University of Chicago — Hopping has long been suspected as an important mode of transport in supercooled liquids at temperatures below T_c. It has been observed in model systems, but until now, has not been directly observed in molecular liquids. We show that incoherent quasi-elastic neutron scattering (QENS) reveals a two-state scenario where, on a 1 ps timescale, molecules are either confined to motion on a lengthscale of 0.05 r_H, or free to undergo motion on a much larger lengthscale of roughly 0.3 r_H, where r_H is the hydrodynamic radius. The motion executed by the less-constrained molecules fits the description of hopping motion observed in model simulations and colloid experiments. The population free to he latter giving rise to hopping at low temperature where the mobile states are long-lived. We show also that this two-state scenario holds well above T_c, where the mobile state lifetime exhibits apparently universal behavior, and transport appears to proceed by both small-step diffusion and larger-step "hopping" processes. Our interpretation of the neutron scattering data is confirmed by atomistic MD simulations, which reveal additional richness, and suggest that this very short-time two-state behavior may be the precursor to dynamic heterogeneity as observed on longer timescales.

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