Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Cooperative Activated Transport of Dilute Penetrants in Viscous Molecular and Polymer Liquids KENNETH SCHWEIZER, RUI ZHANG, University of Illinois at Urbana-Champaign — We generalize the force-level Elastically Collective Nonlinear Langevin Equation theory of activated relaxation in one-component supercooled liquids to treat the hopping transport of a dilute penetrant in a dense hard sphere fluid. The new idea is to explicitly account for the coupling between penetrant displacement and a local matrix cage re-arrangement which facilitates its hopping. A temporal casuality condition is employed to self-consistently determine a dimensionless degree of matrix distortion relative to the penetrant jump distance using the dynamic free energy concept. Penetrant diffusion becomes increasingly coupled to the correlated matrix displacements for larger penetrant to matrix particle size ratio (R) and/or attraction strength (physical bonds), but depends weakly on matrix packing fraction. In the absence of attractions, a nearly exponential dependence of penetrant diffusivity on R is predicted in the intermediate range of 0.2<R<0.8, and the various high packing fraction results collapse well onto a master curve if R is scaled by the matrix transient localization length. Calculations are performed for real thermal liquids based on an a priori mapping to a reference hard sphere mixture.

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Date submitted: 08 Nov 2016 Electronic form version 1.4