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Free Volume Model of Enhanced Mobility at a Free Surface¹

NICHOLAS B. TITO, JANE E. G. LIPSON, Dartmouth College, SCOTT T. MILNER, The Pennsylvania State University — Experiments on polymer thin films during the past two decades have revealed a number of intriguing properties as they approach the glass transition. In addition to dynamic heterogeneity, which is also characteristic of the bulk, there is a substantial body of evidence for enhanced mobility at and near a free surface, leading to local suppression of the glass transition temperature. We have developed a simple kinetic lattice model of free volume and mobility transport in a near-glassy fluid. The model qualitatively exhibits hallmarks of the glass transition in bulk fluids, e.g. power-law growth of the cooperative length scale of glassified material, and slowing global dynamics on approach to a “kinetic arrest” transition. In this talk we discuss how introducing a free surface into the model yields a gradient of mobility, the depth of which depends on proximity to the bulk glass transition.

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