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Perturbation of Glassy Dynamics in Thin Polymer Films due to Interfaces

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Many confinement studies have focused on free-standing polymer films, having been historically billed as the simplest system, containing only two symmetric air-polymer interfaces with no substrate interactions. However, free-standing films have instead exhibited some of the most complex molecular weight (MW)-dependent average film glass transition temperature Tg(h) behavior with decreasing film thickness. We have previously demonstrated that high MW free-standing polystyrene (PS) films can exhibit two distinct transitions in thermal expansion on cooling, with qualitatively different MW dependences suggesting that two separate mechanisms are acting simultaneously to propagate enhanced mobility from the free surface deeper into the material. To investigate the nature of these transitions, we present physical aging measurements below and in-between these two transitions. How the presence of a free surface alters the Tg in confined systems is complicated by the ill-defined magnitude of the perturbation in local mobility. Thus, we have recently begun investigating polymer-polymer interfaces where the magnitude of the Tg perturbation at the interface can be readily determined. Such studies provide a more well-defined system for investigating the length scales over which such perturbations are propagated, which we believe are associated with the glass transition.