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Measuring glassy correlation lengths in ultra-thin polymer films

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We have recently demonstrated that ultra-thin films of organic glass-formers have significantly enhanced dynamics compared to bulk dynamics at temperatures well below the glass transition temperature, Tg. We showed that a sharp glass to liquid transition occurs when the thickness of the layer is decreased below 30 nm. The sharp decrease in fragility in these systems resembles those measured polymeric systems. Here, I discuss how these results can help resolve a long-standing ambiguity in the origin of enhanced dynamics and Tg reduction in thin polymer films. In particular, we show how one can separate "glassy effects" from those induced by the "polymer chain". We show that the free surface and substrate effects present as perturbations in the dynamics that can propagate up to 30 nm into the bulk of the film. These data can explain the extent of Tg reduction and dramatic change in polymer fragility in ultra-thin films in various polymer systems, including homopolymers and miscible blends. For example, we show that in the presence of strong substrate interactions, such as poly (2-vinylpyridine) on silicon, strongly correlated dynamics can result in the emergent of two distinct Tgs, one increasing and the other one increasing upon decreasing film thickness.