Abstract Submitted
for the MAR11 Meeting of
The American Physical Society

Evidence for Two Simultaneous Mechanisms Causing Tg Reductions in High Molecular-Weight Free-Standing Films Observed as Dual Glass Transitions More Than 30 K Apart in a Single Film JUSTIN PYE, CONNIE ROTH, Emory University — Glass transition temperature (Tg) changes seen in nanoconfined polymer films have been well documented over the past 15+ years. Supported films exhibit a molecular-weight (MW) independent Tg reduction that manifests itself as a gradient in dynamics emanating from the free surface. Low MW free-standing films show qualitatively the same Tg reduction as supported films, but with the presence of two free surfaces resulting in a Tg reduction that is twice as large for a given film thickness. In contrast, high MW free-standing films exhibit a qualitatively different behavior with a linear reduction in Tg that is MW dependent, potentially described by de Gennes' sliding mode theory. These observations suggest that there may exist two separate mechanisms which can propagate enhanced mobility from the free surface into the film. With ellipsometry measurements over an extended temperature range, we have observed two reduced Tgs more than 30 K apart in individual high MW free-standing polystyrene films suggesting that both mechanisms act simultaneously within a film. These results may explain recent studies on high MW free-standing films using different experimental techniques that contradict the original literature.

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Date submitted: 30 Nov 2010 Electronic form version 1.4