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Impact of Gradient in Dynamics at the Free Surface on the Physical Aging of Polystyrene Films and Its Connection to the Glass Transition Temperature Reductions JUSTIN PYE, KATE ROHALD, ELIZABETH BAKER, CONNIE ROTH, Dept of Physics, Emory University — The glass transition and physical aging in nanoconfined polymer films have been heavily studied. Recently there have been an increasing number of studies pointing towards a gradient in mobility emanating from the free surface in these films. Using a new streamlined ellipsometry technique, we have measured the temperature dependence of the physical aging rate β for both bulk (2430 nm) and thin (29 nm) polystyrene (PS) films supported on silicon. We find that the thinner films have reduced physical aging rates at all temperatures that are inconsistent with a simple shift in the temperature dependence of β corresponding to the shift in T_g observed in these films. The reduced β values measured at all physical aging temperatures are consistent with a gradient in dynamics originating from the free surface of the film. Our data is well fit by a simple two-layer model that has been previously employed to explain the T_g reductions in PS thin films, suggesting that the enhanced dynamics present at the free surface are responsible for both effects. The surface layer thickness of this two-layer model, which increases with decreasing temperature, characterizes the depth to which the enhanced mobility at the free surface propagates into the film.

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