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Consequences of Residual Stresses in Thin Polymer Films

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In our quest for making functional devices smaller, the thickness of polymer films has reached values even smaller than the diameter of the unperturbed molecule. However, despite enormous efforts over the last decade, our understanding of the origin of some puzzling properties of such thin films is still not satisfactory and several peculiar observations remain rather mysterious. In this context, we explore the consequences of the transition from a dilute polymer solution to the glassy state with respect to the properties of polymers in thin films. This transition is likely to result in residual stresses, arising from out-of-equilibrium chain conformations due to rapid solvent loss. Consequently, depending on thermal history and ageing time, such films exhibit significant changes even in the glassy state which we quantify by performing detailed studies of viscoelastic dewetting of thin polystyrene films on solid substrates. We explored relaxation times, residual stresses, and temporal changes of the stability of non-equilibrated thin films as they progress toward stable equilibrium behaviors. To do so, we have focused primarily on times shorter than the reptation time of the polymer. The number of spontaneously nucleated holes per unit area is seen to decrease as the films were aged below the glass transition, showing the meta-stability of the system. The ratio of stress over elastic modulus was found to increase strongly with decreasing film thickness and increasing chain length. Full equilibration of chain conformations required long times comparable to bulk reptation times. However, for chains longer than about 3000 monomers, the residual stress relaxed faster, at a rate independent of chain length. We present some tentative ideas on the relation between these observed atypical mechanical and relaxational behaviors and meta-stable states introduced by sample preparation.