MAR12-2011-008278

Abstract for an Invited Paper
for the MAR12 Meeting of
the American Physical Society

Theoretical prospects for understanding the glass transition in polymer thin films

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Observations of confinement effects on the glass transition in thin polymer films remain vexing to theorists. Many experiments at sufficiently low frequencies consistently find that the free surfaces of many polymer films, whether freely suspended or supported, have anomalously low glass transition temperatures. This surface effect appears to be independent of molecular weight, and persists 10-20nm into the film. In contrast, freely suspended films have been observed by ellipsometry and other bulk-sensitive techniques to have anomalously low film-averaged T_g values, depending strongly on both film thickness and molecular weight for Mw above about 4×10^5g/mol. The dependence on Mw persists to astonishingly large values ($\sim 10^7$g/mol). Recent work of Pye and Roth strongly suggests the presence of two distinct mechanisms in these films, one operative at the free surface and Mw-independent, one depending on Mw and giving a weak second glass transition at considerably lower temperatures. In this talk, I will review the status of various theoretical proposals we have explored for explaining some of these disparate phenomena, including the effects of interrupted percolation at the free surface and the "sliding mechanism" originally suggested by de Gennes for Mw-dependent lowering of T_g. I will describe the present prospects for a successful theory, as well as suggest some potentially helpful experiments and analysis.