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Mechanical heterogeneity in bulk, thin-film, and nanocomposite polymeric glasses

JUAN DE PABLO, University of Wisconsin - Madison

Ultrathin polymer films and nanoscopic polymer structures represent a particularly interesting class of systems for study of the glass transition and cooperative dynamics. Such systems provide a unique test bed to test the limits of validity of traditional continuum mechanics descriptions, and are relevant in technologies being considered for mass-production of next-generation electronic devices. A complete understanding of material properties at nanometer length scales and, in particular, mechanical properties, is crucial for development and optimization of such technologies. Recent experimental data suggest that the thermophysical properties of polymeric materials in small geometries differ from those of the bulk. We use experiments and molecular simulations of polymeric molecules to analyze and explain the origin of such behavior. The overall picture that emerges from our studies is that amorphous, glassy polymeric materials exhibit heterogeneous mechanical properties in nanoscopic structures and in the bulk. Domains measuring several nanometers can have elastic moduli that differ by an order of magnitude. These properties exhibit a Gaussian distribution, whose mean and width vary considerably depending on the geometry of the sample, the presence of surfaces, and the presence (and characteristics) of filler nanoparticles.