Fragility and Cooperativity of glass formation

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Abstract:

We briefly review our findings on the relationship between the fragility of glass formation and cooperative motion, focusing on a simple glass-forming polymer melt. We identify string-like cooperative motions with the hypothetical 'cooperatively rearranging regions' of the Adam-Gibbs (AG) description, thereby providing a molecular basis for this interpretation. Moreover, these strings can be described as equilibrium 'living polymers'. Based on this correspondence, we combine the AG model with a theory for living polymerization to provide a more complete theoretical description of the temperature dependence of relaxation, and thus fragility. Accordingly, the variation in the scale of cooperative motion controls changes in fragility. We expect this description is general, including small molecule, non-polymeric glass-forming fluids. We consider the robustness of these ideas by examining fragility changes in polymer-nanoparticle composites and ultra-thin polymer films, systems of practical importance where the fragility can be varied in a controlled way.