Abstract Submitted
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Glassy dynamics of intermediate-chain-stiffness crystallizable polymer melts[1] HONG NGUYEN, ROBERT HOY, University of South Florida — We contrast the dynamics in model unentangled polymer melts of chains of three different stiffnesses: flexible, intermediate, and rodlike. Flexible and rodlike chains, which readily solidify into close-packed crystals (respectively with randomly oriented and nematically aligned chains), display simple melt dynamics with Arrhenius temperature dependence and a discontinuous change upon solidification. Intermediate-stiffness chains, however, are fragile glass-formers displaying Vogel-Fulcher dynamical arrest, despite the fact that they also possess a nematic-close-packed crystalline ground state. No clear static-structural cause of this dynamical arrest is found. However, we find that the intermediate-stiffness chains display qualitatively different cooperative dynamics. Specifically, their stringlike motion (cooperative rearrangement) is correlated along chain backbones in a way not found for either flexible or rodlike chains. This activated "crawling" motion is clearly associated with the dynamical arrest observed in these systems, and illustrates one way in which factors controlling the crystallization vs. glass formation competition in polymers can depend nonmonotonically on chain stiffness.

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