The Relationship of Dynamical Heterogeneity to the Adam-Gibbs and Random First-Order Transition Theories of Glass Formation

FRANCIS STARR, Wesleyan University, Middletown CT, JACK DOUGLAS, NIST, Gaithersburg, MD, SRIKANTH SASTRY, Tata Institute of Fundamental Research, Hyderabad, India — We examine measures of dynamical heterogeneity for a bead-spring polymer melt and test how these scales compare with the scales hypothesized by the Adam and Gibbs (AG) and random first-order transition (RFOT) theories. We show that the time scale of the high-mobility clusters and strings is associated with a diffusive time scale, while the low-mobility particles’ time scale relates to a structural relaxation time. The difference of the characteristic times naturally explains the decoupling of diffusion and structural relaxation time scales. We examine the appropriateness of identifying the size scales of mobile particle clusters or strings with the size of cooperatively rearranging regions (CRR) in the AG and RFOT theories. We find that the string size appears to be the most consistent measure of CRR for both the AG and RFOT models. Identifying strings or clusters with the “mosaic” length of the RFOT model relaxes the conventional assumption that the “entropic droplet” are compact. We also confirm the validity of the entropy formulation of the AG theory, constraining the exponent values of the RFOT theory. This constraint, together with the analysis of size scales, enables us to estimate the characteristic exponents of RFOT.

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