Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Theory of Cooperative Activated Structural Relaxation in Polymer Nanocomposites Composed of Small and Sticky Particles SHIJIE XIE, KENNETH SCHWEIZER, University of Illinois at Urbana-Champaign — Recently, Cheng, Sokolov and coworkers have discovered qualitatively new dynamic behavior (exceptionally large Tg and fragility increases, unusual thermal and viscoelastic responses) in polymer nanocomposites composed of nanoparticles comparable in size to a polymer segment which form physical bonds with both themselves and segments. We generalize the Elastically Collective Nonlinear Langevin Equation theory of deeply supercooled molecular and polymer liquids to study the cooperative activated hopping dynamics of this system based on the dynamic free energy surface concept. The theoretical calculations are consistent with segmental relaxation time measurements as a function of temperature and nanoparticle volume fraction, and also the nearly linear growth of Tg with NP loading; predictions are made for the influence of nonuniversal chemical effects. The theory suggests the alpha process involves strongly coupled activated motion of segments and nanoparticles, consistent with the observed negligible change of the heat capacity jump with filler loading. Based on cohesive energy calculations and transient network ideas, full structural relaxation is suggested to involve a second, slower bond dissociation process with distinctive features and implications.

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Date submitted: 07 Nov 2016 Electronic form version 1.4