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Theory of the effect of deformation on the relaxation and mechanical properties of polymer glasses KANG CHEN, KENNETH SCHWEIZER, University of Illinois at Urbana-Champaign — Recently Saltzman and Schweizer have developed a statistical dynamical theory of segmental relaxation in deeply supercooled polymer melts by combining and extending methods of mode coupling, dynamic density functional and activated barrier hopping theories. The approach is built on the concept of a nanometer scale nonequilibrium free energy which quantifies dynamic localization due to interchain forces. We have now generalized this approach to treat quiescent relaxation, physical aging, and nonlinear mechanical properties in the nonequilibrium glass state. Applied stress weakens dynamical constraints in the effective free energy which accelerates alpha relaxation and softens the elastic modulus. A constitutive equation has been constructed which allows the prediction of dynamic yielding and mechanical response under constant strain rate, constant stress (creep), and other modes of deformation. Multiple predictions that fundamentally differ from the phenomenological Eyring model are made for the temperature and strain rate dependence of the dynamic yield stress and the deformation modified segmental relaxation time. Comparison of the theoretical results with experiments on PMMA reveals good agreement. The coupling of stress and aging, the strain softening effect, and large deformation strain hardening, can also be treated within our approach.

Kang Chen
University of Illinois at Urbana-Champaign

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