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Nanoscale fluctuations and responses in equilibrium and nonequilibrium glassy polymers¹

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Recent developments have made it clear that finding a dynamical correlation length, and its spatio-temporal behavior on approach to the glass transition are the keys to explaining the dramatic growth of viscosity near the glass transition, and testing leading theory candidates. Several lines of indirect evidence (e.g. Berthier et. al. Science 2005) point to a weak but growing temperature-dependence of this length on approach to the glass transition. In this talk I will discuss an approach based on scanning probe microscopy, for probing and imaging spontaneous dipolar noise and dielectric response. We used these methods to study nanoscale spatio-temporal dynamics in polymer glasses and polymer blends. Various space-time correlation functions are analyzed in an effort to search for growing correlation length scales near the glass transition. We are working to extend the instrumental resolution closer to these intrinsic length scales thought to control the glass transition. The ability to quantitatively measure on the nanoscale both fluctuations and responses also allows us to test the validity of the global and local fluctuation-dissipation-relation (FDR) in equilibrium and out-of-equilibrium. Using this approach, we have been able to study, for the first time, FDR violations in the strong aging regime in structural glasses (Oukris, et. al. Nature Physics, 2009). The results give insight into the equilibrium glass order parameter.

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