Dynamic Heterogeneity and Correlation in Liquids and Polymers

C.M. Roland
Naval Research Laboratory

The origin of the dramatic behavior of liquids and polymers as they approach their vitreous state – small variations in temperature or pressure can change the time scale for molecular motions from nanoseconds to a duration exceeding the human lifespan – is a major unsolved problem in condensed matter. These changes are accompanied by greater dynamic heterogeneity, which refers to both spatial variation and spatial correlation of molecular mobilities. The question is whether the changing dynamics is coupled to this heterogeneity; that is, does the latter cause the former? To address this we carried out nonlinear dielectric experiments that enable both the timescale and degree of cooperativity of molecular motions to be quantified. We find that the dynamic correlation volume depends primarily on the relaxation time, sensibly independent of temperature and pressure. We support this result by molecular dynamic simulations showing that the four-point dynamic susceptibility of density fluctuations is essentially constant along an isochrone. Our findings are consistent with dynamic heterogeneity serving as the principal control parameter for the slowing down of molecular motions in supercooled materials.