CORRELATED HETEROGENEOUS DYNAMICS IN GLASS-FORMING POLYMERS

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Although glasses are very common in everyday life, the transition from a supercooled liquid to the glassy state is one of the mysteries in condensed matter physics [1]. Upon approaching the glass transition, the sample dynamics slows down by orders of magnitude while the sample structure remains almost unchanged. In addition, relaxation phenomena become non-exponential pointing to a broad distribution of relaxation times and the existence of dynamical heterogeneities that are closely connected to spatial heterogeneities [1,2]. Such heterogeneities can be accessed by higher-order correlation functions, as demonstrated in simulations [3] and recently for coherent X-ray scattering experiments [4].

Here, we study the dynamics of polypropylene-glycol (PPG) with the help of colloidal silica tracer particles by means of X-ray photon correlation spectroscopy (XPCS) [5]. A temperature range from room temperature to the glass transition temperature at $T_g = 205$ K was investigated. Three temperature regimes are identified: At high temperatures, Brownian motion of the tracer particles is observed. Near T_g, the dynamics is hyperdiffusive and ballistic. Around 1.12 T_g we observe an intermediate regime. By analyzing higher-order correlations in the scattering data we find that dynamical heterogeneities dramatically increase in this intermediate temperature regime. This leads to two effects: a) increasing heterogeneous dynamics and b) correlated motion at temperatures close to and below 1.12 T_g.

References

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