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Dynamic Heterogeneity and Relaxation Time Very Close to Dynamic Arrest

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In supercooled molecular fluids or concentrated colloids and grains, the dynamics slow down markedly with no distinct structural changes as the glass and jamming transitions are approached. There is now ample evidence that structural relaxation in glassy systems can only occur through correlated rearrangements of particles, leading to dynamics that are heterogeneous both in time and in space. On approaching these transitions, the size of these rearranging domains grows in glass-formers, colloids, and driven granular materials alike, providing a possible explanation for kinetic slowing. However, little is known yet on the behavior of dynamical heterogeneity and relaxation time very close to dynamical arrest. In this talk, I'll present recent results from our group for a simple model system –colloidal hard spheres–, as well as for other glassy and jammed soft materials. By extending previous data [1] by at least 2 orders of magnitude in time, we established that the volume fraction dependence of relaxation time and dynamic heterogeneity in colloidal hard spheres follow mode coupling theory (MCT) [2] predictions only in a restricted density range [3]. Unlike previous studies, we provide equilibrium measurements above the MCT critical packing fraction, thereby proving unambiguously that in our samples the algebraic divergence is absent at the predicted packing fraction. The behavior of dynamical heterogeneity is even more surprising. While in supercooled hard spheres the size of domains undergoing cooperative rearrangements is limited to a few particles at most, closer to jamming the correlation length of the dynamics increases dramatically, approaching the system size in a variety of systems [4,5]. In this regime, spatial and temporal fluctuations of the dynamics may decouple, as observed for near hard sphere particles: while the range of spatial correlations continuously increase on approaching jamming, the temporal fluctuations of the dynamics initially increase with particle volume fraction, but drop markedly very close to jamming [5], unveiling a richer-than-expected scenario.

[1] W. van Meegen et al., Phys. Rev. E 58, 6073 (1998);

[2] W. Götze, J. Phys. Condens. Matter 11, A1 (1999);

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[4] A. Duri et al., “Resolving long-range spatial correlations in jammed colloidal systems using photon correlation imaging,” Phys. Rev. Lett. 102 085702 (2009);

[5] Pierre Ballesta, Agnès Duri, & Luca Cipelletti, “Unexpected drop of dynamical heterogeneities in colloidal suspensions approaching the jamming transition,” Nature Physics 4, 550 - 554 (2008)