Structural Details of Aging in Colloidal Glasses

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Are glasses merely extremely slow liquids, and if so, why are they many orders of magnitude slower than conventional liquids? We study concentrated colloidal suspensions, a model system which has a glass transition. We view the motion of these colloidal particles in three dimensions by using an optical confocal microscope. This allows us to directly study the microscopic behavior responsible for the macroscopic viscosity divergence of glasses.

In particular, we are interested in aging, the dependence of physical properties on the time elapsed since the creation of a glassy sample. This is exemplified by the mean square displacement, as shown in Fig. 1. Initially after creating the sample, particles are able to move reasonable distances on relatively short time scales. As the sample ages, it takes long times to move comparable distances.

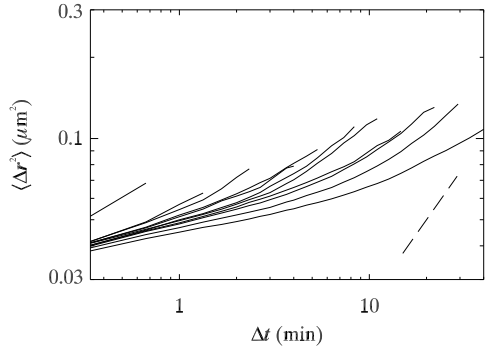

Figure 1: A log-log plot of the mean-square displacement vs lag time. The different curves have different ages; from left to right, the ages are 3.0, 4.2, 5.9, 8.2, 12, 16, 23, 32, 44, 62, 87 min. The dashed line has a slope of 1.

In earlier work we studied the approach of the glass transition, as the colloidal particle concentration was increased. We found that particle dynamics are heterogeneous in both space and time: particle motion occurs via cooperative groups of particles [1]. Furthermore, the size of these groups grows as the glass transition is approached, while their frequency decreases. This then suggests a possible mechanism for aging, that it could also occur via cooperative groups, and that in an older sample, the size of the groups may be larger, thus explaining the slowing of the dynamics such as seen in Fig. 1.

We do find that aging occurs via cooperative groups of rearranging particles [2]. Suprisingly, the size of these groups seems not to relate to the age of the sample. Furthermore, after the sample has aged appreciably, even on short time scales cooperative groups of particles are seen (although in this case they do not necessarily cause rearrangements).

To understand the origin of these rearrangement events, we investigate the non-equilibrium behavior in terms of colloidal packing. Tetrahedra, or triangular based pyramids, represent the ideal packing of 4 spheres in three dimensions, however they do not tile 3-D space. This frustration between local and global packing optimization has been invoked as a possible origin for the glass transition. We therefore study how these tetrahedra evolve as the sample ages. We find that static properties of the distribution of tetrahedra do not change with the age of the sample. However, the shape of tetrahedra does influence the mobility of the particles comprising the tetrahedra. For example, particles in the most irregular tetrahedra tend to be more mobile, and it is precisely these mobile particles which rearrange to result in an "older" sample.

References

[1] E. R. Weeks, J. C. Crocker, A. C. Levitt, A. Schofield, and D. A. Weitz, *Science* **287** (2000) 627.

[2] R. E. Courtland and E. R. Weeks, *J. Phys. Cond. Mat.* **15** (2003) S359.