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Report:

We have investigated by XPCS the slow dynamics of colloidal gels made of moderately attractive particles of carbon black (CB). The best results have been obtained for gels at 2% volume fraction and two different amounts of dispersant: 0% and 0.2%. The role of the dispersant is to decrease the interaction potential at contact: the smaller the amount of dispersant, the deeper the CB gel is quenched in the jammed phase [1].

We have investigated both the average dynamics and the temporal fluctuations of the dynamics, applying, for the first time, the Time Resolved Correlation (TRC) [2] method to XPCS measurements. All data were obtained using a CCD camera as a detector, in the 15 $\mu m^{-1} < q < 150\ \mu m^{-1}$ scattering vector range. For all samples we have focused on the dynamics a few hours after initializing the sample by sonication. While in the initial regime the dynamics was found to slow down (aging), after a few hours it was found to reach a nearly stationary state.

Average dynamics:

Sample with 0.2% dispersant. The intensity correlation functions g_2-1 measured simultaneously at various q vectors exhibit a 2 step decay, which we fit by the squared sum of two stretched exponential relaxations, with characteristic times τ_1 and τ_2 ($\tau_1 < \tau_2$) and stretching exponents p_1 and p_2, respectively (see Fig. 1). The characteristic time of the initial decay, τ_1, scales roughly as q^{-1} in the intermediate q range and the associated stretching exponent p_1 decreases continuously with q from 1.5 down to 0.4 (data not shown). A possible explanation for the initial relaxation of g_2-1 could be overdamped phonons propagating in the gel network [3]. However, these modes are typically at frequencies much higher than those accessible to a CCD detector. The q-dependence of τ_1 rather hints to rearrangement dynamics driven by the relaxation of internal stress, similar to those observed in many glassy soft materials. This picture is further supported by the observation that, as discussed below, these dynamics are temporally heterogeneous, in analogy with recent measurements of the rearrangement dynamics in strongly attractive colloidal gels [4]. The relaxation time of the final decay is q independent: we attribute it to gel relaxations due to bond breaking and leading to particle displacements larger than $1/q$. (Tests on a Vycor glass ruled out setup instability as a possible source of this decorrelation).

Sample with 0 % dispersant. The dynamics are similar to those for the previous sample (data not shown), although the final relaxation time is much longer (of the order of 1000-2000 sec). Although this is in qualitative agreement with the existence of stronger bonds, we can not rule out artifacts due to setup instabilities on such long time scales; Accordingly, we take this value merely as a lower bound for the final relaxation of the gel.

Temporal fluctuation of the dynamics:

We use the TRC method to analyze the speckle images issued from the XPCS experiment. This allows us to calculate not only the average dynamics (i.e. g_2-1), but also its temporal fluctuations, quantified by χ, the variance of g_2-1 normalized with respect of the amplitude of g_2-1 for τ->0 [4]. This quantity is similar to the dynamical susceptibility χ_4 studied in simulations of glassy systems and has never been measured by XPCS. An example for the gel with 0.2% dispersant is shown in fig. 2. χ has a peaked shape, dynamical fluctuations being largest on the time scale of τ_1, in analogy with numerical findings for glassy systems. We find that (i) χ decreases with increasing q vector; (ii) χ decreases with increasing dispersant content.

(i) is a quite surprising result. There are almost no experimental measurements of χ for 3-D systems, with the exception of low-q data on strongly attractive colloidal gels (light scattering) [4], for which an opposite trend is found (χ scales roughly as q). We are currently working on the interpretation of this surprising result.

(ii) can be understood since, intuitively and in analogy with glass formers, one expects dynamical heterogeneity to be more pronounced for samples closer to a glass/jamming transition.

In the future, it would be interesting to explore systematically the q-dependent amplitude of dynamical fluctuations as a function of theparameters governing the fluid-to-solid transition (e.g. volume fraction and interparticle potential) and for various materials.

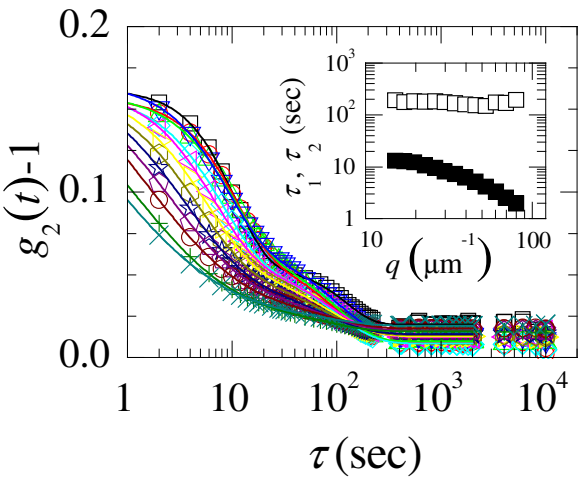

Fig. 1. Symbols, main figure: intensity correlation functions at various q for a CB gel with 0.2% dispersant. From right to left, q ranges from 15 μm^{-1} to 150 μm^{-1} (see label in Fig. 2). The lines are fits to the data as discussed in the text. Inset: fast (τ_1, solid symbols) and slow (τ_2, open symbols) relaxation times obtained from the fit of g_2-1.

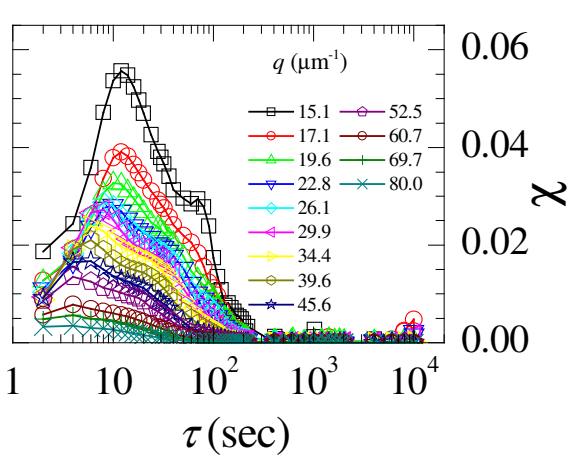

Fig. 2. Dynamical susceptibility χ for the same q vectors as in Fig. 1

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[3] A. H. Krall and D. A. Weitz., *Phys. Rev. Lett.*, **80** 778 (1998).

[4] A. Duri and L. Cipelletti, *Length scale dependence of dynamical heterogeneity in a colloidal fractal gel,* Europhys. Lett. **76**, 972 (2006).