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Abstract. – We use time-resolved dynamic light scattering to investigate the slow dynamics of a colloidal gel. The final decay of the average intensity autocorrelation function is well described by $g_2(q,\tau) - 1 \sim \exp[-(\tau/\tau_\mathrm{f})^p]$, with $\tau_\mathrm{f} \sim q^{-1}$ and p decreasing from 1.5 to 1 with increasing q. We show that the dynamics is not due to a continuous ballistic process, as proposed in previous works, but rather to rare, intermittent rearrangements. We quantify the dynamical fluctuations resulting from intermittency by means of the variance $\chi(\tau,q)$ of the instantaneous autocorrelation function, the analogous of the dynamical susceptibility χ_4 studied in glass formers. The amplitude of χ is found to grow linearly with q. We propose a simple –yet general– model of intermittent dynamics that accounts for the q dependence of both the average correlation functions and χ.

Soft matter systems where the constituents are packed at high volume fraction or interact strongly have a dynamical behavior reminiscent of that of molecular glasses [1]: they exhibit very slow relaxations, non-exponential response or correlation functions, history-dependent dynamics, and dynamical heterogeneity [2]. However, soft glassy systems may also exhibit peculiar dynamical features not found in hard glasses. An example is provided by low volume fraction colloidal gels resulting from the aggregation of strongly attractive particles. In these gels, the decay of the intensity correlation function $g_2(q,\tau) - 1$ measured by dynamic light scattering is steeper than exponential and the relaxation time, τ_f, has an anomalous q^{-1} dependence on the scattering vector [3]. This "compressed" exponential, ballistic-like dynamics has to be contrasted with the stretched exponential relaxations and the diffusive behavior ($\tau_\mathrm{f} \sim q^{-2}$) usually found in molecular systems [1]. Quite intriguingly, this unusual dynamics is not restricted to dilute colloidal gels, but has been observed recently in a large variety of soft systems with both attractive and repulsive interactions [4–10].

For colloidal gels, it has been proposed that the dynamics be due to the continuous evolution of strain fields set by dipolar sources of internal stress [3, 5, 11]. At a microscopic level, stress is presumably accumulated by changes in the local structure driven by the formation of new bonds and/or bond breaking, which constitute elementary steps in the direction of a more stable, compact structure. More generally, a similar continuous evolution of dipolar

strain fields has been invoked to explain the ballistic-like dynamics of other systems exhibiting a compressed exponential relaxation of g_2 [4–10].

Time-resolved light scattering experiments performed on similar gels [12] have suggested that the dynamics be temporally heterogeneous, thus challenging the arguments of refs. [3,5,11] that are based on a continuous dynamics [13]. However, these experiments were performed on samples at much higher concentration ($\varphi \sim 0.1$ as opposed to $10^{-3} - 10^{-4}$) and in the multiple scattering regime, where the dynamics in probed on much shorter length scales (less than a particle size as opposed to hundreds of particle sizes). Whether their outcome might be extrapolated to single scattering experiments on diluted gels at low q is thus still unclear, especially since time-resolved light scattering measurements in the same low-q range as that of ref. [3] failed to evidence any significant temporal heterogeneity [14]. Clarifying the nature and the physical origin of the dynamics of the gels is particularly important, given the widespread occurrence of similar dynamics in soft glassy materials.

In this Letter, we tackle these issues by studying the dynamics of strongly attractive colloidal gels in a range of q vectors about one decade higher than in [3] and using time-resolved dynamic light scattering. We find that at all q the final relaxation of the average intensity correlation function $g_2(q,\tau) - 1$ is well fitted by a compressed exponential with the same q^{-1} dependence of $\tau_{\rm f}$ as observed previously [3], and with a compressing exponent $p > 1$. While at the lowest q $p \approx 1.5$, the exponent unexpectedly decreases with increasing q, eventually approaching one. We demonstrate that the dynamics is intermittent and quantify the resulting fluctuations of g_2 by means of a "multipoint" correlation function χ analogous to the dynamical susceptibility χ_4 introduced in simulations of glass formers [15–17]. We find that χ increases linearly with q and introduce a general model of intermittent dynamics that accounts for both the average dynamics and its temporal fluctuations.

The gels are made of polystyrene particles of radius 10 nm suspended in a buoyancy matching mixture of H_2O and D_2O (45/55 by volume). Particles are mixed with a $MgCl_2$ solution in order to induce aggregation in the DLCA regime [18]. The final particle volume fraction and salt concentration are $\varphi = 6\times10^{-4}$ and 10 mM, respectively. A gelled structure is obtained after about 2 hours, formed by interconnected fractal clusters of radius $R_{\rm c} \approx 10$ μm, as revealed by static light scattering [3,18]. The dynamics of the gel slows down with age [3]. To prevent significant aging during the experiment, we focus on a time window of duration $T_{\rm exp} = 20000$ sec starting at $t_{\rm w} = 280\,000$ sec, where $t_{\rm w} = 0$ is the time when the gel is formed.

The gel dynamics is measured by using a charge-coupled device (CCD) camera-based light scattering apparatus similar to that described in [19], slightly modified to access larger q vectors. The dynamics is measured simultaneously at several q's (0.4 $\mu{\rm m}^{-1} \leq q \leq 5.5$ $\mu{\rm m}^{-1}$), corresponding to length scales intermediate between the particle and the cluster size. In order to access both the average dynamics and its temporal fluctuations, we use the time-resolved correlation scheme [12, 20]. The degree of correlation between pairs of images of the speckle pattern scattered at time $t_{\rm w}$ and $t_{\rm w} + \tau$ is calculated according to $c_I(t_{\rm w},\tau,q) = G_2(t_{\rm w},\tau)/\left(\langle I_p(t_{\rm w})\rangle_p \langle I_p(t_{\rm w}+\tau)\rangle_p\right) - 1$, where $G_2(t_{\rm w},\tau) = \langle I_p(t_{\rm w}) I_p(t_{\rm w}+\tau)\rangle_p$ and $I_p(t)$ is the scattered intensity at pixel p and time t. $\langle\cdots\rangle_p$ is an average over pixels corresponding to the same magnitude of $\mathbf{q}$ but different azimuthal orientations. The intensity autocorrelation function is $g_2(q,\tau) - 1 = \overline{c_I(t_{\rm w},\tau,q)}$, where $\overline{\cdots}$ indicates a time average over $T_{\rm exp}$. Dynamical fluctuations are quantified by the temporal variance of c_I at fixed τ and q [12,20], as discussed in more detail in the following.

The average dynamics is shown in Fig. 1a) for several q's. At all scattering vectors, $g_2(q,\tau) - 1$ exhibits an initial decay, followed by a slightly tilted plateau and a final relaxation. The initial decay is barely observable due to the limited frame rate of the CCD camera; it is

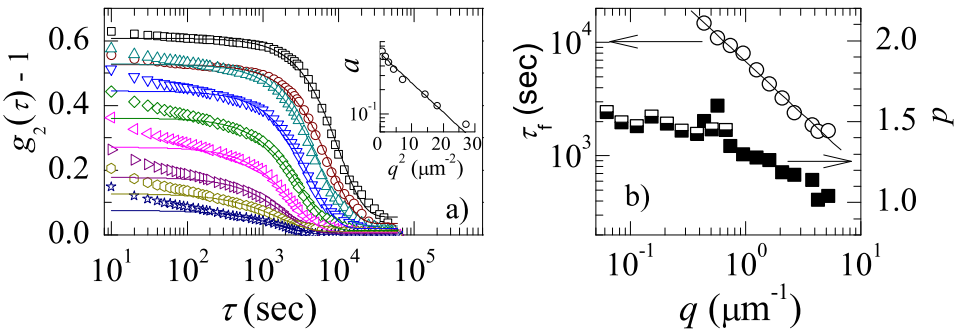

Fig. 1 – a): $g_2(q,\tau)-1$ for a gel at $\varphi = 6\times10^{-4}$. From top to bottom, q varies from 0.74 to 5.22 μm^{-1}. The lines are compressed exponential fits to the final relaxation of g_2-1. Inset: semilogarithmic plot of the plateau height a *vs.* q^2. The line is an exponential fit. b): q dependence of the relaxation time τ_f (open circles, left axis) and of the compressing exponent p (right axis, solid squares: this work; semiopen squares: ref. [3]). The line is a power law fit to τ_f yielding an exponent -0.94 ± 0.03.

due to overdamped, thermally activated fluctuations of the gel strands [21]. The height a of the plateau is related to the average amplitude, δ_p, of these fluctuations by $a \sim \exp(-q^2\delta_\mathrm{p}^2/3)$ [21]. By fitting $a(q)$ to this Gaussian form, we find $\delta_\mathrm{p} = 500\pm150$ nm (inset of Fig. 1a). The final relaxation is well fitted by a compressed exponential decay: $g_2(q,\tau) - 1 = a\exp[-(\tau/\tau_\mathrm{f})^p]$, where a, τ_f, and p depend on q. Figure 1b) shows the q dependence of p (solid and semiopen symbols) and τ_f (open circles). We find $\tau_\mathrm{f} \sim q^{-0.94\pm0.03}$, consistently with previous measurements at lower q [3]. This behavior rules out diffusive motion and indicates that, on average, the particle displacement increases linearly with time. The compressing exponent p is approximately 1.5 at the smallest q probed in this work, in agreement with ref. [3] (semiopen squares) and in analogy with refs [4, 5, 7, 8, 10]. Surprisingly, however, at larger scattering vectors p decreases with q, finally approaching $p = 1$. A similar trend has been reported very recently for other systems with analogous dynamics [9, 10], although its origin remained unclear. We stress that this behavior is incompatible with the arguments proposed in refs. [3, 5], which predict $p = 1.5$ regardless of q. The model by Bouchaud and Pitard predicts $p = 1.5$ for $q \to 0$ and $p = 1.25$ for $q \to \infty$ [11]. However, in the intermediate q regime $g_2 - 1$ should deviate from a compressed exponential, contrary to our observations.

To better understand the origin of the gel dynamics, we investigate time-resolved quantities. Figure 2a) shows the time dependence of the instantaneous degree of correlation c_I for $q = 2.07$ μm^{-1} and for several time lags τ (similar results are obtained for all q's). The top curve shows data for $\tau = 250$ sec, a time lag much smaller than the relaxation time $\tau_\mathrm{f} = 3220$ sec; the data are correct for the contribution of measurement noise as described in Sec. IV c of ref. [20]. Sharp drops of c_I departing from a high plateau are visible, indicating sudden changes in the speckle pattern caused by discrete rearrangement events in the gel. The typical width of the downward spikes correspond essentially to τ, indicating that the rearrangements are shorter than the temporal resolution of the experiment (10 sec) [22]. We locate the time of the events by analyzing the drops of c_I for $\tau = 250$ sec and by verifying that similar drops are observed at the same time for all q's. The vertical bars in Fig. 2a) indicate the occurrence of the rearrangements thus identified. We count 23 events during the time $T_\mathrm{exp} = 20000$ sec, implying that the average time between events is $\Delta t = T_\mathrm{exp}/23 = 870$ sec. For $\tau = 3000$ sec $\approx \tau_\mathrm{f}$, much larger fluctuations of c_I are observed, because the number of events occurring between two speckles images separated by 3000 sec may significantly vary, due to the random nature

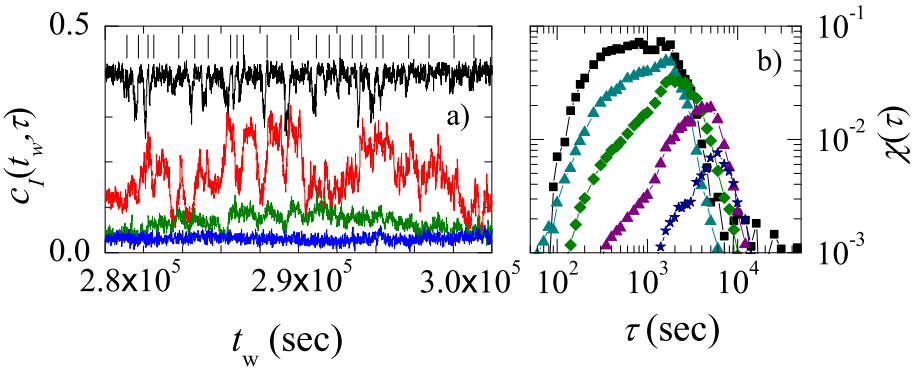

Fig. 2 – a): temporal fluctuations of the instantaneous degree of correlation c_I for $q = 2.07\ \mu\mathrm{m}^{-1}$. From top to bottom, $\tau = 250, 3000, 8000,$ and 20000 sec. The vertical bars above the top curve indicate the time of individual rearrangement events. b): dynamical susceptibility $\chi(\tau, q)$ (normalized variance of c_I). From bottom to top, $q = 0.74,\ 1.24,\ 2.07,\ 3.78,$ and $5.22\ \mu\mathrm{m}^{-1}$ (same symbols as in Fig. 1a). For the sake of clarity, not all the available curves have been plotted.

of the events. This has to be contrasted with the case $\tau = 250$ sec $<< \Delta t$, where at most one event occurs between two images, yielding the discrete drops discussed above. For $\tau >> \Delta t$ (two bottom curves in Fig. 2a), the fluctuations are again reduced, because on this time scale many events occur and hence their (relative) number fluctuations are smaller, by virtue of the Central Limit theorem.

We quantify the fluctuations of the dynamics resulting from the intermittent rearrangements by calculating $\chi(\tau, q)$, the variance of c_I corrected for the measurement noise contribution [20]. As discussed in [20], this quantity is the analogous in light scattering experiments of the dynamical susceptibility χ_4 much studied in theoretical and numerical works on glass formers and gels [15–17, 24]. In order to compare data taken at different q vectors, we focus on the relative fluctuations by normalizing the variance of c_I with respect to the amplitude of the final decay of $g_2 - 1$: $\chi(\tau, q) = a(q)^{-2}\left[\overline{c_I(t_\mathrm{w}, \tau, q)^2} - \overline{c_I(t_\mathrm{w}, \tau, q)}^2\right]$. Results for some representative q vectors are shown in Fig. 2b): for all q, χ has a peaked shape, the peak position corresponding approximately to the decay time of the average correlation function, where the fluctuations of c_I are largest as observed in Fig. 2a). This peaked shape is analogous to that reported in previous simulations and experiments on other glassy systems [16, 20, 23–25]. In these systems, fluctuations usually arise from *spatial* correlations of the dynamics that reduce the number of statistically independent regions in the system. By contrast, for the gels χ is dictated by the *temporal* intermittency of the rearrangement events, since they affect simultaneously the whole scattering volume, as found by directly measuring the spatial correlation of the slow dynamics [26]. The length scale dependence of the fluctuations, quantified by the height of the peak, $\chi^*(q)$, has a very surprising behavior. As shown in Fig. 3b), we find that χ^* increases strongly with q ($\chi^* \sim q^{1.13\pm0.11}$). To our knowledge, no other experimental data on 3D systems are available; however we point out that the gel behavior is in contrast with measurements on a 2D granular system, where the peak of χ_4 showed virtually no q dependence [25].

The intermittent dynamics shown here is in stark contrast with the continuous ballistic motion assumed in [3,5,11]. In the following, we develop a simple yet general model of dynamic light scattering from an intermittent dynamical process, aiming at rationalizing both the

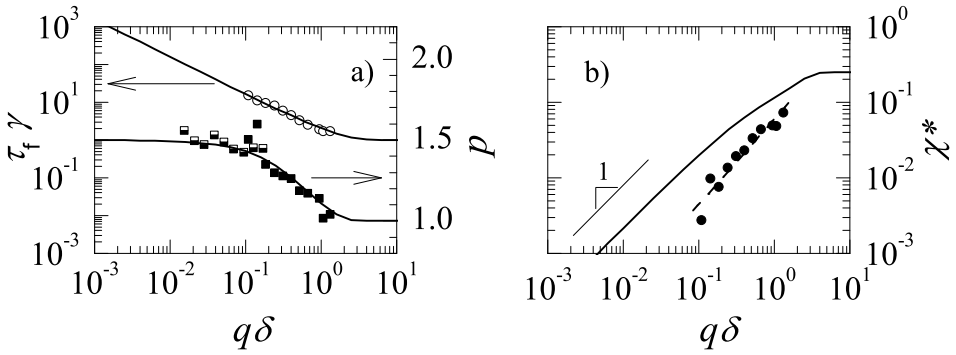

Fig. 3 – a): q dependence of the relaxation time (top line, left axis) and of the compressing exponent (bottom line, right axis) in reduced variables, as obtained from the model described in the text. The symbols are the experimental data shown in Fig. 1b): a very good agreement with the model is obtained for $\delta = 250$ nm and $\gamma^{-1} = 960$ sec. b) peak of the dynamical susceptibility χ^* *vs* $q\delta$ for the model (line) and the experiments (symbols). The dashed line is a power law fit with an exponent 1.13 ± 0.11

average correlation function and its fluctuations. We assume the slow dynamics to be due to a series of discrete rearrangement events, which we take –for simplicity– to have equal amplitude and to be instantaneous. The degree of correlation is then a function h, to be determined, of q and the number m of events occurring between $t_{\rm w}$ and $t_{\rm w} + \tau$: $c_I(t_{\rm w}, \tau, q) = h[m(t_{\rm w}, \tau), q]$. Within this scenario, the fluctuations of c_I are due to fluctuations of the number of events actually occurring during any given interval $[t_{\rm w}, t_{\rm w} + \tau]$. We calculate the average dynamics and its fluctuations according to

$$g_2(\tau, q) - 1 = \sum_{n=0}^{\infty} P_\tau(n) h(n, q) \tag{1}$$

$$\chi(\tau, q) = \sum_{n=0}^{\infty} P_\tau(n) [h(n, q) - (g_2(\tau, q) - 1)]^2 \,, \tag{2}$$

where $P_\tau(n)$ is the probability that n events affect the scattering volume during a time span τ. In writing Eq. (2), we have taken into account that the dynamics is spatially correlated over a length scale much larger than the size of the scattering volume, so that fluctuations are not reduced by averaging the collected signal over several dynamically independent regions.

In order to calculate g_2 and χ from Eqs. (1,2), we need expressions for P_τ and h. For the former, we choose for simplicity a Poisson law: $P_\tau(n) = \exp(-\gamma\tau)(\gamma\tau)^n/n!$, corresponding to rearrangement events that are random in time and affect the scattering volume at an average rate γ. For the latter, we write $h(n, q)$ in terms of the displacement field generated by n rearrangement events. By introducing $\Delta\mathbf{R}$, the particle displacement due to one single event, one has $h(n, q) =< \exp(-in^\alpha \mathbf{q} \cdot \Delta\mathbf{R}) >$ [27], where the average is taken over all possible orientations of $\mathbf{q}$ and over all particles. The exponent α is expected to be ≤ 1: $\alpha = 0.5$ would correspond to diffusive-like dynamics where the displacement grows as the square root of the number of events, while $\alpha = 1$ would lead, on average, to a ballistic-like motion. For the gels, we expect $\alpha = 1$, because for the average dynamics $q \propto \tau_{\rm f}^{-1}$. The correlation left after n events is then $h(n, q) =< \exp(-in\mathbf{q} \cdot \Delta\mathbf{R}) >= \int PDF(\Delta\mathbf{R}) \exp(in\mathbf{q} \cdot \Delta\mathbf{R}) {\rm d}\Delta\mathbf{R}$, where

$PDF(\Delta\mathbf{R})$ is the probability distribution function (PDF) of the particle displacements [27]. By assuming that the displacement field induced by one single event is that due to the long range elastic deformation of the gel under the action of dipolar stresses [5, 11] and using arguments similar to those developed in ref. [5], one has $h(n,q) = \exp[-(qn\delta)^{\beta}]$. Here δ is the typical displacement of the particles due to one single event and $\beta = 1.5$ corresponds to a PDF with a $\Delta R^{-2.5}$ power law right tail, as predicted for dipolar stress sources randomly scattered in space [5, 11].

We insert the above expressions for P_τ and h in Eqs. (1,2), using $\alpha = 1, \beta = 1.5$, and calculate both the average dynamics and its fluctuations. The resulting $g_2(\tau,q) - 1$ is very well approximated by a compressed exponential decay, in agreement with the experimental data. We show in Fig. 3a) the compressing exponent p and the dimensionless relaxation time $\gamma\tau_{\rm f}$ issued from the fit of the model, as a function of the dimensionless scattering vector $q\delta$ (lines). For $q\delta \to 0$, we find $p \to \beta = 1.5$. At larger scattering vectors, p decreases approaching one. In all the q regime where $p > 1$, $\gamma\tau_{\rm f} \propto q^{-1}$, a consequence of the choice $\alpha = 1$ that yields, on average, ballistic-like dynamics. Note that the fact that the displacement due to successive events adds up coherently ($\alpha = 1$) suggests that they originate from the same source, i.e. that stress sources are very diluted and their life time exceeds the relaxation time of g_2. Thus, several discrete events are necessary to fully relax a stress source. For $q\delta >> 1$, where p saturates to one, we find $\tau_{\rm f} = \gamma^{-1}$ independently of q. Indeed, when the typical displacement δ is much larger than the length scale $1/q$ probed by light scattering, one single event is sufficient to lead to a complete decorrelation of the scattered light. The only relevant time scale is then γ^{-1}, the average time between events. In this regime $h(n,q) \approx 0$ for $n > 0$: the only non vanishing term in the average correlation function, r.h.s. of Eq. (1), corresponds to $n = 0$ and $g_2(q,\tau) - 1 = \exp(-\gamma\tau)$.

Figure 3a) shows also the experimentally determined p and $\tau_{\rm f}$ plotted using dimensionless variables, with $\delta = 250$ nm and $\gamma = 1.04 \times 10^{-3}$ Hz. With this choice of the parameters, a very good agreement between the model and the experiments is found. Remarkably, δ is of the same order of magnitude of the amplitude of the fluctuations of the gel strands due to thermal motion ($\delta_{\rm p} = 500$ nm). This strongly supports the intuitive picture that the rearrangement events correspond to the formation of new bonds and/or the breaking of the bonds along the gel network. Since both processes are ultimately triggered by thermal energy, they would typically cause particle displacements comparable to those due to the thermal fluctuations of the gel. The average ballistic dynamics is thus the result of a slow compaction of the gel that proceeds by discrete rearrangements, rather than of a continuous process. The event rate γ determined by fitting the model to the experimental average dynamics corresponds to an average time between events of 960 sec, in excellent agreement with $\Delta t = 870$ sec obtained directly from the drops of c_I shown in Fig. 2a). A further test of the model would be the observation of the high q limit where $g_2 - 1 \sim \exp(-\gamma\tau)$. However, for our gels this regime is not experimentally accessible, since as q grows the amplitude a of the slow mode of $g_2 - 1$ decreases as $\exp(-q^2\delta_{\rm p}^2/3)$ (see Fig. 1a), ultimately vanishing for $q\delta_{\rm p} > q\delta >> 1$.

Having fixed the parameters of the model by comparison with the experimental average dynamics, we compare the theoretical predictions for the dynamical susceptibility to the experiments. We find that χ has the same peaked shape as for the experiments; the q dependence of the height of the peak is shown in Fig. 3b) (line) together with the experimental points (filled circles). The model captures correctly the nearly linear growth of $\chi*$ with q and the order of magnitude of the dynamical fluctuations, although it overestimates them by about a factor of two. Given the simplicity of the assumptions and the fact that no adjustable parameter has been introduced specifically for χ, this agreement is quite remarkable, showing that the model captures the essential physics of the slow dynamics of the gel. Interestingly, the $\chi^* \sim q$

growth at low q and the saturation regime at large q are similar to asymptotic predictions for $\chi_4(\tau, q)$ in glass formers proposed very recently by Chandler et al. Indeed, in ref. [28] a saturation regime at large q and a $\chi_4 \sim q^2$ scaling for $q \to 0$ are identified, the q^2 rather that q dependence being the consequence of diffusive rather than ballistic-like dynamics.

In conclusion, we have shown that the slow dynamics of colloidal gels is a highly discontinuous process, due to intermittent, sudden rearrangement events. We have proposed a simple model of intermittent dynamics that captures correctly the q dependence of both the average dynamics and dynamical fluctuations. Although some aspects of our model and their physical interpretation are certainly specific to the dilute colloidal gels studied here, its general features may be relevant also to other soft systems. In particular, more time-resolved experiments will be needed to test whether the underlying picture of intermittent rearrangements may apply also to the numerous soft systems whose average dynamics is similar to that of the gels. This would challenge the currently accepted interpretation in terms of a continuous ballistic process [4–10].

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