Anomalous dynamical light scattering in soft glassy gels

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Abstract. We compute the dynamical structure factor $S(q, \tau)$ of an elastic medium where force dipoles appear at random in space and in time, due to "micro-collapses" of the structure. Various regimes are found, depending on the wave vector q and the collapse time θ. In an early time regime, the logarithm of the structure factor behaves as $(q\tau)^{3/2}$, as predicted in (L. Cipelletti *et al.*, Phys. Rev Lett. **84**, 2275 (2000)) using heuristic arguments. However, in an intermediate-time regime we rather obtain a $(q\tau)^{5/4}$ behaviour. Finally, the asymptotic long-time regime is found to behave as $q^{3/2}\tau$. We also give a plausible scenario for aging, in terms of a strain-dependent energy barrier for micro-collapses. The relaxation time is found to grow with the age t_w, quasi-exponentially at first, and then as $t_w^{4/5}$ with logarithmic corrections.

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1 Introduction

There is currently a great interest in soft glassy materials, such as pastes, foams or colloidal gels [1]. These systems are often out of equilibrium and exhibit interesting aging effects, typical of other glassy systems such as spin-glasses or polymer glasses [2,3]. For example, their rheological properties are found to depend strongly on the age of the system [4,5]. From a theoretical point of view, aging is expected both in response functions (such as magnetic susceptibilities or elastic moduli) but also in correlation functions, which are notoriously harder to study experimentally. Recently, a new experimental "multi-speckle" technique has allowed one to properly investigate aging in a dynamical correlation function [6]. The difficulty is to make the experimental acquisition time much smaller than the typical age of the system, otherwise the age of the system changes significantly during the experiment and the measured correlation function is meaningless. Using this technique, the dynamical structure factor of a colloidal gel made of aggregating polystyrene particles was measured and revealed several unexpected features [6]. i) For a given waiting time t_w (counted from the moment when the gel is formed), the dynamical structure factor $S(q, \tau, t_w)$ is found to decay as $\exp(-A(q\tau)^{3/2})$. This must be contrasted with the usual diffusive decay, as $\exp(-Dq^2\tau)$: both the q- and τ-dependence are anoma-

lous, and non-intuitive. The exponent 3/2 shows that the time decay is *faster* than an exponential, whereas one would have expected a slow decay in a glassy system. A similar exponent 3/2 was also found in different systems, such as diblock polystyrene/polyisoprene copolymers [7], laponite [8] or other systems [9]. ii) The scaling in $q\tau$ suggests that some kind of convection, rather than diffusion, is present in the system. However, since the system is density matched, this cannot be a global sedimentation effect. iii) When the age of the system increases, the relaxation of the dynamical structure factor becomes slower but retains its shape. In other words, only the coefficient A in the above expression is found to be age dependent. The corresponding relaxation time $\tau_r(q, t_w)$ (defined as $A(q\tau_r)^{3/2} = 1$) is found to increase *exponentially* with t_w at first, and then as t_w^μ with $\mu < 1$ for larger waiting times. Such an exponentially growing relaxation time with age was also reported for laponite [10], but is very unusual. The second regime, however, is typical of most experimental glassy systems [11,12].

A heuristic interpretation of the $(q\tau)^{3/2}$ scaling was proposed in [6]. The basic mechanism is that the gel contracts in a very heterogeneous way: localized "micro-collapses" create a long-range elastic deformation field which is ultimately responsible for the $q^{3/2}$ behaviour. In order to get the correct τ-dependence the strain within these collapsing regions was postulated to be linear in τ. This is however awkward since the instant $\tau = 0$ has no special meaning and a linear growth would eventually lead

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to unbounded strains. Furthermore, this argument does not account for aging.

We show in this paper that an important time scale was left out in the analysis of [6], namely the typical collapse time which we will call θ. We find that for $\tau \ll \theta$, the relaxation of the structure factor for a given t_w does indeed only depend on $q\tau$, but exhibits two distinct regimes: a short-time regime, where we recover the $(q\tau)^{3/2}$ behaviour, and an intermediate-time regime, where the decay is slower, as $\exp(-B(q\tau)^{5/4})$, which might be of experimental relevance. For very large τ's, the $q\tau$ scaling breaks down and a new dynamical regime is found. We also give a simple argument to rationalize the dependence of τ_r on t_w.

2 The model

Following [6], we assume that the dominant mechanism is the random appearance of micro-collapses: since the micro-particles forming the gel attract each other rather strongly, the gel tends to restructure locally so as to create dense regions of particles. (*Post mortem* analysis indeed reveals that particles actually tend to *fuse* together.) Since the collapsing particles belong to a gel network, their motion will induce a certain strain field around them; other particles therefore move and dynamical light scattering probes this motion. Assume for a moment that the gel is a one-dimensional chain of beads and springs. When two particles decide to leave their equilibrium position to stick together, the left particle imposes a positive force $+f_0$ on the left part of the chain, while the right particle imposes an equal opposite force $-f_0$ on the right part of the chain. In other words, this creates a *dipole* of forces. This is also true in three dimensions, where the collapse of particles will result in a force dipole of intensity P_0 in a certain direction that we call $\mathbf{n}$. When the dipole is formed at point $\mathbf{r}_0$, the elastic strain field $\mathbf{u}$ at point $\mathbf{r}$ can be computed assuming a simple central force elasticity:

$$K\Delta\mathbf{u} = -\mathbf{f}(\mathbf{r}), \qquad (1)$$

where K is a compression modulus and $\mathbf{f}(\mathbf{r})$ is a local force dipole of the form:

$$\mathbf{f}(\mathbf{r}) = f_0\left[\epsilon \cdot \nabla\delta(\mathbf{r} - \mathbf{r}_0)\right]\mathbf{n}, \qquad (2)$$

where we will assume that $\epsilon = \epsilon\mathbf{n}$, *i.e.*, the mean displacement of the particles creates a force in the same direction. This dipolar force can be simply expressed by its Fourier transform:

$$\mathbf{f}_k = iP_0\left(\mathbf{k}\cdot\mathbf{n}\right)\mathbf{n}\exp(-i\mathbf{k}\cdot\mathbf{r}_0) \qquad (3)$$

with $P_0 = f_0\epsilon$.

The solution of equation (1) is of course

$$\mathbf{u}(\mathbf{r}) = -\frac{P_0}{4\pi K}\frac{(\mathbf{r} - \mathbf{r}_0)\cdot\mathbf{n}}{|r - r_0|^3}\mathbf{n}. \qquad (4)$$

The r^{-2}-dependence of the strain field has an immediate consequence which will be of importance in the following:

if there is a finite density of force dipoles randomly scattered in space, the probability that the stress at a given point has an amplitude u decays for large u as $u^{-5/2}$, which has a diverging variance [13]. (This divergence is however cut off if the finite size of the dipoles is taken into account.) This property of the distribution of displacements and stresses will be responsible for the unusual q-dependence of the structure factor. Note also that from dimensional arguments, $P_0/4\pi K = v_0$ is a volume given by $\xi^3\lambda^2$, where ξ is the typical size of the collapsing region, and $\lambda < 1$ is the contraction ratio (that we will assume to be of order 1 in the following).

Now, let us assume that the micro-collapses are not instantaneous but take place progressively, over a time scale θ. A given event j starts at time t_j and is completed at time $t_j + \theta$; the dipole intensity $P(t)$ at time t is a certain function of $(t - t_j)/\theta$. We will assume that this function is approximately linear, and simply write $P_j(t) = P_0(t - t_j)/\theta$, which saturates at P_0 when the collapse is completed. (Note that a micro-collapse could itself be the result of many successive events.) The dynamics of the individual particles is presumably dominated by viscous friction, therefore we write the following equation of motion for the strain field $\mathbf{u}$:

$$\gamma\frac{\partial\mathbf{u}(\mathbf{r}, t)}{\partial t} = K\Delta\mathbf{u} + \sum_j \mathbf{f}_j(\mathbf{r}, t) + \eta(\mathbf{r}, t), \qquad (5)$$

where the Fourier transform of the dipolar force is

$$\mathbf{f}_j(\mathbf{k}, t) = iP_j(t)\left(\mathbf{k}\cdot\mathbf{n}_j\right)\mathbf{n}_j\exp(-i\mathbf{k}\cdot\mathbf{r}_j) \qquad (6)$$

and γ is a friction coefficient, and η is the thermal random force due to the viscous bath, uncorrelated in time and in direction.

We have taken into account the fact that many micro-collapses take place, at different times t_j and different positions $\mathbf{r}_j$, with different orientations $\mathbf{n}_j$ of the dipoles. Actually, we will assume in the following that these events occur randomly in space and time, with a certain rate ρ per unit volume and unit time. The quantity K/γ is a diffusion constant that we will call D. Equation (5) defines the model that we want to study and from which we will compute the dynamical structure factor $S(q, \tau)$, defined as

$$S(q, \tau) = \langle\exp\left[i\mathbf{q}\cdot(\mathbf{u}(\mathbf{r}, t + \tau) - \mathbf{u}(\mathbf{r}, t))\right]\rangle, \qquad (7)$$

where the brackets refer to a spatial average over $\mathbf{r}$ or, equivalently, over the random location and time of the micro-collapse events.

3 Results and comparison with experiments

A first step is to calculate the Fourier transform of the *time derivative* of the displacement field $\mathbf{u}(\mathbf{r}, t)$ created by a single dipole located at $\mathbf{r}_j$, in direction $\mathbf{n}_j$, that we denote $\mathbf{v}(\mathbf{k}, t|\mathbf{r}_j, \mathbf{n}_j, t_j)$. One finds

$$\mathbf{v}(\mathbf{k}, t|\mathbf{r}_j, t_j) = -i\exp(-i\mathbf{k}\cdot\mathbf{r}_j)\frac{P_0\mathbf{n}_j}{\theta}\frac{\mathbf{n}_j\cdot\mathbf{k}}{Kk^2}\exp(-Dk^2t)$$
$$\times\left[\exp(Dk^2t_j) - \exp(Dk^2\min(t_j + \theta, t))\right]. \quad (8)$$

The displacement difference between t and $t+\tau$ can therefore be expressed as

$$\mathbf{u}(\mathbf{r}, t+\tau) - \mathbf{u}(\mathbf{r}, t) =$$
$$\int_t^{t+\tau} dt' \sum_{j/t_j < t'} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \exp(i\mathbf{k}\cdot\mathbf{r})\mathbf{v}(\mathbf{k}, t|\mathbf{r}_j, t_j). \quad (9)$$

The analysis of this expression reveals that there are *a priori* many different cases to consider for the relative position of the time t_j when the j-th micro-collapse takes place and the other relevant times.

A new q-dependent time τ_q can be introduced,

$$\tau_q \equiv \frac{D\theta}{qv_0}\theta, \quad (10)$$

such that, depending on the ratio τ/τ_q, the dominant contribution to the decay of $S(q, \tau)$ comes from different regions of the t'-t_j plane. For $\tau \ll \tau_q$ and $\tau \ll \theta$ the final result reads (details of the calculation can be found in [14])

$$S(q, \tau) = \exp\left[-\frac{16\sqrt{2\pi^3}}{75}\rho\theta(D\theta)^{3/2}\left(\frac{\tau}{\tau_q}\right)^{3/2}\right], \quad (\tau \ll \tau_q), \quad (11)$$

which has the form suggested by the arguments of [6], in particular, it indeed only depends on $(q\tau)^{3/2}$. Note that the combination $\hat{\rho} \equiv \rho\theta(D\theta)^{3/2}$ is adimensional and represents the average number of events taking place within a time interval θ and within a diffusion volume $(D\theta)^{3/2}$.

For the regime $\tau_q \ll \tau \ll \theta$,

$$S(q, \tau) = \exp\left[-\frac{2^{10}\pi^{3/8}\Gamma(-\frac{5}{4})\Gamma(\frac{17}{8})\sin\frac{\pi}{8}}{3^4 5^{17/8}}\hat{\rho}\left(\frac{\tau}{\tau_q}\right)^{5/4}\right],$$
$$(\tau_q \ll \tau \ll \theta). \quad (12)$$

Therefore, we find that in this regime the $q\tau$ scaling still holds, but the power $3/2$ is replaced by $5/4$.

Let us now study the regime where $\tau \gg \theta$, corresponding to "fast" micro-collapses. (Note that the fact that θ is small does not mean that the events are frequent: this is described by the nucleation parameter ρ.) The calculations are very similar to the above case. The relevant time scale which now appears naturally is

$$\tilde{\tau}_q = qv_0/D = \theta^2/\tau_q. \quad (13)$$

When $\tau \ll \tilde{\tau}_q$, we find exactly the same decay as equation (12) above: note indeed that θ actually drops out of this expression. However, when $\tau \gg \tilde{\tau}_q$, the dynamical structure factor reads

$$S(q, \tau) = \exp\left[-\frac{16\sqrt{2\pi^3}}{75}\rho v_0^{3/2}q^{3/2}\tau\right], \quad (\tau \gg \tilde{\tau}_q), \quad (14)$$

which is independent of the diffusion constant D. Therefore, the asymptotic decay of $S(q, \tau)$ is a pure exponential, with an anomalous decay time $\propto q^{-3/2}$. This is the result one obtains if all retardation effects are neglected (*i.e.*

$D \to \infty$): the factor $\rho\tau$ simply counts the average number of events between t and $t+\tau$, and $q^{3/2}$ reflects the fact that the distribution of local displacements u decays as $u^{-5/2}$ and has a diverging variance (see, *e.g.* [13]). For a distribution with a finite variance, one would obtain the usual q^2-dependence.

It is useful to summarize our results in a schematic way, in terms of the behaviour of $\Psi \equiv -\log S(q, \tau)$. The two physical cases depend on the relative position of τ_q and $\tilde{\tau}_q$, or equivalently on the ratio $D\theta/qv_0$. For $D\theta \ll qv_0$ one finds $\tau_q \ll \theta \ll \tilde{\tau}_q$ and

$$\begin{aligned}
\Psi &\sim (q\tau)^{3/2}, & (\tau \ll \tau_q), \\
\Psi &\sim (q\tau)^{5/4}, & (\tau_q \ll \tau \ll \tilde{\tau}_q), \\
\Psi &\sim q^{3/2}\tau, & (\tau \gg \tilde{\tau}_q),
\end{aligned} \quad (15)$$

whereas for $D\theta \gg qv_0$, the $(q\tau)^{5/4}$ regime is squeezed out and the results are simply

$$\Psi \sim (q\tau)^{3/2}, \quad (\tau \ll \theta); \qquad \Psi \sim q^{3/2}\tau, \quad (\tau \gg \theta). \quad (16)$$

To which of these regimes does the experiment correspond? First, a rather good $q\tau$ scaling is observed across most of the time regime ($\tau \leq 10^4$ seconds, or 3 hours). This means that the asymptotic regime is not observed, and therefore that $\max(\theta, \tilde{\tau}_q) > 10^4$ s. Let us first suppose that the experiments are in the regime $D\theta \ll qv_0$, where $\tau_q \ll \theta \ll \tilde{\tau}_q$. According to our calculations, a $(q\tau)^{5/4}$ behaviour should be observed for $\tau_q \ll \tau \ll \tilde{\tau}_q$. We have checked that such a possibility is indeed compatible with the experimental data, although the data is by no means compelling. Under this assumption, we find that $q\tau_q = D\theta^2/v_0$, is of order $3 \cdot 10^7$ cm^{-1} s. It is reasonable to assume that the volume of the collapsing region $v_0 \sim \xi^3$ is of the order of the cluster size at gelation determined in [6], *i.e.* $3 \cdot 10^{-8}$ cm^3 ($\xi \simeq 30 \ \mu$m). The elastic diffusion constant can be estimated from the elastic modulus of the structure [6] ($G' \simeq 10^{-3}$ dyn/cm^2), the value of ξ and of the viscosity $\eta \simeq 10^{-2}$ P: $D = K/\gamma \simeq G'v_0/\eta\xi \simeq 10^{-6}$ cm^2/s. Therefore, one gets θ on the order of 1000 seconds. For the maximum experimental value of $q \sim 6000$ cm^{-1}, one finds $\tau_q \sim 5000$ seconds, which is a factor five larger than θ, in contradiction with our hypothesis. However this factor five is marginal in view of the roughness of our estimates. For example, the collapsing volume v_0 could be somewhat larger and numerical factors could help. Another possibility is that the $(q\tau)^{5/4}$ regime is in fact not seen experimentally, *i.e.* $D\theta \gg qv_0$. This however requires that θ is actually quite large, at least 10^4 seconds. It would be interesting to determine ρ or θ from an independent experiment in order to clarify this question.

4 The aging phenomenon

We finally discuss an important aspect of the experiments that we left out up to now, namely the fact that the dynamical structure factor is age dependent: the relaxation time grows with the age t_w of the sample. This

growth is even unusually (exponentially) fast in the initial stages [6,10].

The basic mechanism for aging is that micro-collapses induce a tensile strain on the non-collapsed regions. It is plausible that the energy barrier for a rearrangement bringing the particles closer together in such a strained region will increase, and these regions find it harder to collapse subsequently.

We assume that the volume density of dipoles is $\phi(t_w)$. Therefore $\ell^* = \phi^{-1/3}$ is the typical distance between two dipoles. Let us assume also that micro-collapse events are thermally activated, so that their rate $\rho(t_w)$ can be expressed as

$$\rho(t_w) = \rho_0 \exp\left(-\frac{\Delta E(t_w)}{k_B T}\right), \qquad (17)$$

where $\Delta E(t_w)$ is an energy barrier and T the temperature. The above mechanism means that the barrier height depends on the typical local strain $|\nabla u|_{\text{typ}}$. More precisely one can write that $\Delta E = \frac{K}{2}(\ell_0 + |\nabla u|_{\text{typ}}\ell_0)^2 - \frac{K}{2}\ell_0^2$, where ℓ_0 is the equilibrium length of one elementary elastic unit, supposed here to be a constant. For small enough $|\nabla u|_{\text{typ}}$, one expects a linear relationship: $\Delta E \simeq K\ell_0^2|\nabla u|_{\text{typ}}$, whereas for large $|\nabla u|_{\text{typ}}$ this dependence becomes quadratic: $\Delta E \simeq \frac{K}{2}\ell_0^2|\nabla u|_{\text{typ}}^2$. Obviously, new events will preferentially occur in less strained regions, i.e., far from collapsed regions, inducing some spatial correlations between micro-collapses that we have neglected above; hence, we will assume in the following that the small strain regime $\Delta E \simeq K\ell_0|\nabla u|_{\text{typ}}$ is the dominant one. The typical strain can be estimated as the sum of strains induced by all the dipoles surrounding a given particle in the system. For a unique dipole of orientation $\mathbf{n}$ at a distance r from the particle, one obtains

$$|\nabla u|^2 = \frac{v_0^2}{r^6}\left(3\frac{(\mathbf{r}\cdot\mathbf{n})^2}{r^2} + 1\right). \qquad (18)$$

From this expression, we obtain $|\nabla u|_{\text{typ}}^2 = \langle|\nabla u|^2\rangle$, where the brackets denote the average over all contributions of dipoles at a distance $r \geq \ell^*$ from the particle, the dipoles being distributed with a volume density $\phi = 1/\ell^{*3}$. The result is

$$|\nabla u|_{\text{typ}} = \left(\frac{8\pi}{3}\right)^{1/2} v_0\phi. \qquad (19)$$

Hence, $\Delta E/k_B T = \Gamma\phi$, where $\Gamma \sim \frac{K\ell_0^2}{k_B T}v_0$.

We want now to evaluate the age dependence of $\rho(t_w)$ and $\phi(t_w)$. The time evolution of ϕ is given by

$$\frac{d\phi}{dt_w} = \rho(\phi) = \rho_0 \exp\left(-\frac{\Delta E(\phi)}{k_B T}\right). \qquad (20)$$

The solutions can be written explicitly and one finds that the density of dipoles increases logarithmically with time:

$$\phi(t_w) = \phi(0) + \frac{1}{\Gamma}\log\left(1 + \frac{t_w}{t_0}\right), \qquad (21)$$

where t_0 is the initial activation time $t_0 = [\rho(t=0)\Gamma]^{-1}$. The rate of formation of dipoles (micro-collapses) therefore decreases with time as

$$\rho(t_w) = \frac{1}{\Gamma(t_w + t_0)}. \qquad (22)$$

The above calculations for $S(q,\tau)$ can be simply extended to the case where ρ is not constant, provided ρ does not vary too much on the scale of τ, which is true if t_w is sufficiently large. In order to compare with the experimentally determined relaxation time τ_r, one must solve the equation $S(q,\tau_r,t_w) = e^{-1}$. Assuming that the experiments are in the $(q\tau)^{5/4}$ regime, one finds

$$\tau_r \propto (t_w + t_0)^\mu, \qquad (23)$$

with the exponent $\mu = \frac{4}{5}$. The best agreement with the experimentally determined $\tau_r \sim t_w^{0.9}$ [6] is for $\mu = \frac{4}{5}$.

5 Conclusion

In conclusion, we have computed the dynamical structure factor $S(q,\tau)$ of an elastic medium where force dipoles appear at random in space and time, as "micro-collapses" appear in the structure. The balance between these collapse events and the elastic relaxation of the internal stresses in the medium leads to various regimes, depending on the wave vector q and the collapse time θ. In an early time regime, the logarithm of the structure factor behaves as $(q\tau)^{3/2}$ plus subleading corrections, as anticipated in [6] using heuristic arguments, and observed experimentally. However, in an intermediate time that might be relevant for the experiments of [6], we obtain a $(q\tau)^{5/4}$ behaviour that would have been difficult to guess from simple arguments. Finally, the asymptotic long-time regime is found to behave as $q^{3/2}\tau$, where the $q\tau$ scaling is not obeyed; but this last regime seems not to be observed experimentally and it is likely that before that, a macroscopic collapse occurs as the one observed in colloidal gels [6].

We have also given a plausible scenario for aging, in terms of a strain-dependent energy barrier for micro-collapses. The relaxation time is found to grow with the age t_w, exponentially at first, and then as $t_w^{4/5}$ with logarithmic corrections, which seems to be in good agreement with experiments.

It would be interesting to analyze other experiments where similar effects have been reported [7–9], in particular in micellar crystalline phases and onion phases, along the lines of the present work, and also to understand the link between rheology experiments and light-scattering experiments. In these systems however, motion of defects such as dislocations or grain boundaries may also play a crucial role.

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