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# Ultra-long-range dynamic correlations in a microscopic model for aging gels

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We use large-scale computer simulations to explore the nonequilibrium aging dynamics in a microscopic model for colloidal gels. We find that gelation resulting from a kinetically-arrested phase separation is accompanied by ‘anomalous’ particle dynamics revealed by superdiffusive particle motion and compressed exponential relaxation of time correlation functions. Spatio-temporal analysis of the dynamics reveals intermittent heterogeneities producing spatial correlations over extremely large length scales. Our study is the first microscopically-resolved model reproducing all features of the spontaneous aging dynamics observed experimentally in soft materials.

Understanding the complex mechanisms underlying the formation and stability of colloidal gels remains a challenge, despite the diversity of existing applications exploiting their mechanical properties. Gels are low-density structures forming percolating networks, with bonds that are either permanent (chemical gels) or transient (physical gels) [1–6]. A prevalent method to form physical gels follows a nonequilibrium route by quenching a homogeneous fluid into a phase coexistence region [1, 7–12], which generates bicontinuous structures. When the dense phase forms an amorphous solid, the phase separation is kinetically hindered [13–16] and a gel forms. The microscopic dynamical and structural properties of these nonequilibrium gels evolve slowly with time.

This aging dynamics has been the subject of many experimental studies, which established that aging in colloidal gels is ‘anomalous’ [17, 18], i.e. it differs qualitatively from the aging observed in conventional glassy materials, such as polymer and colloidal glasses [19, 20]. Scattering experiments [21–25] report that time correlation functions are described by *compressed exponential* relaxations. Such behaviour differs from the (exponential) diffusive dynamics in simple liquids or the (stretched exponential) decay observed in glassy fluids [26]. In addition, compressed exponentials are seen to emerge only for *large enough displacements*, with the relaxation timescale  $\tau(q)$  crossing over from  $\tau \sim q^{-2}$ , characteristics of diffusion, to  $\tau \sim q^{-1}$ , characteristic of ballistic dynamics, with decreasing the scattering wavevector  $q$ . Finally, spatially-resolved dynamic measurements revealed the existence of *long-ranged correlations* extending up to the system size [27, 28], again contrasting with the much smaller-ranged dynamic heterogeneity observed in glassy materials [29]. Such peculiar behaviour is hypothesized to follow from the infrequent release of ‘internal stresses’ that relax the fractal network [18], but this interpretation remains to be confirmed by direct observation. This overall phenomenology has been reported for laponite, carbon black, micellar polycrystals, multilamellar vesicles, implying it is generic to a large class of soft materials [18].

A microscopic perspective via theoretical modeling is also missing. To study this problem, one needs a model

with a realistic gel structure, studied for large enough timescales to observe the slow dynamics, and over large enough lengthscales to detect long-ranged correlations. Our work successfully addresses these three challenges. There have been extensive studies of aging effects in particle-based computer simulations of dense glassy systems (e.g., [30, 31]), but none of the above anomalous signatures are observed. For nonequilibrium gelation, the focus has been mainly on the formation of arrested fractal structures via quenches into the phase-coexistence region, with little insight into the aging dynamics. Compressed exponentials in time correlation functions were reported in equilibrium gels but they result from inertial effects at short time and length scales [32, 33]. Other attempts produced compressed relaxations in gel-like structures either by heating out-of-equilibrium initial states [34] or by driving the material mechanically [35]. Compressed relaxations were also reported for mechanically induced coarsening at zero-temperature [36], but the lengthscale dependence and spatial correlations were not analysed. A more recent work on a low density gel reported instead subdiffusive behaviour [37]. At a more coarse-grained level, it was suggested that localised bond-breaking events in gels may result in compressed exponentials [38], a mean-field scenario that was recently revisited using a mesoscopic elasto-plastic model for generic glassy materials [39], in which the gel structure however plays no direct role.

Using large-scale numerical simulations of a particle-based model for gel formation [13, 14], we show that the spontaneous microscopic aging dynamics during gelation possesses all anomalous signatures reported experimentally. We find a subdiffusive aging dynamics at short lengthscales, corresponding to caged particle motion inside the gel strands, crossing over to superdiffusive relaxation at larger lengthscales triggered by intermittent snapping of the fractal network. These relaxation events result in compressed exponential relaxations at small enough scattering vectors and correspond also to dynamic correlation lengthscales that are much larger than the typical pore size of the gel. These results thus provide a coherent microscopic picture of the anomalous

aging dynamics observed experimentally.

Following previous work [13, 14], we study the properties of a binary Lennard-Jones mixture after it is suddenly quenched into its liquid-gas phase coexistence region. To access large-enough system sizes, we work in two spatial dimensions. We consider a 65:35 A-B Lennard-Jones mixture with model parameters as in Ref. [40]. Because of its glass-forming ability, phase separation is kinetically hindered, which results in gelation. We perform molecular dynamics simulations using LAMMPS [41]. In our simulations, energies and lengths are expressed in Lennard-Jones units of interaction energy  $\epsilon_{AA}$  and diameter  $\sigma_{AA}$  for the majority specie. Particles have equal masses,  $m$ , and the time unit is  $\sqrt{m\sigma_{AA}^2/\epsilon_{AA}}$ .

In these reduced units, we observe incomplete phase separation of the binary mixture when temperature is lower than  $T \approx 0.3$ , and a bicontinuous fractal structure is obtained for number densities in the range  $0.25 < \rho < 1.1$ . Our strategy is to first equilibrate the system at high temperature in the homogeneous region,  $T = 3.0$ , for different densities  $\rho = 0.4, 0.5, 0.6, 0.7, 0.8, 1.0$ . Then, at time  $t = 0$ , we instantaneously quench the system to low temperatures in the phase coexistence region,  $T = 0.05, 0.10, 0.15, 0.20, 0.30, 0.40$ , and  $0.50$ . Thereafter, we follow the dynamical properties of the system at these different state points. In order to explore spatial correlations we study large systems ranging from  $N = 10^4$  to  $2 \times 10^6$  particles. A majority of the results shown below correspond to  $N = 5 \times 10^5$ . All data are averaged over at least 4 different initial conditions. During the dynamics, the temperature control is done via dissipative particle dynamics (DPD) thermostat [42], using a friction coefficient value of  $\xi = 1$ ; tests performed with larger damping  $\xi = 3, 10$  showed identical results.

The nearly-arrested bicontinuous structures formed at large times for  $T = 0.15$  and various densities are shown in Fig. 1, for a system with  $N = 2 \times 10^6$ . These images reveal the coexistence of a connected network of dense amorphous domains and void-like regions. For  $\rho = 0.3, 0.5$ , the denser regions form a fractal network with tenuous links, having a morphology reminiscent of those formed during nonequilibrium gelation in experiments. At higher densities,  $\rho = 0.8, 1.0$ , the voids shrink and the dense domains become more compact. At higher temperatures,  $T > 0.3$ , these structures would rapidly coarsen until phase separation is complete. On the other hand at very low temperatures, the gelation process is essentially arrested, because the particle bonds become nearly permanent. A detailed characterization of the gel structure can be found in [14]. Below we explore the intermediate, experimentally relevant regime where the gel evolves very slowly with time, focusing on the temperature  $T = 0.15$ .

To account for experimental findings, we need to study the microscopic dynamics as a function of the time  $t_w$  spent since gelation started. Such  $t_w$ -dependence is demonstrated in Fig. 2(a) for the mean-squared displace-

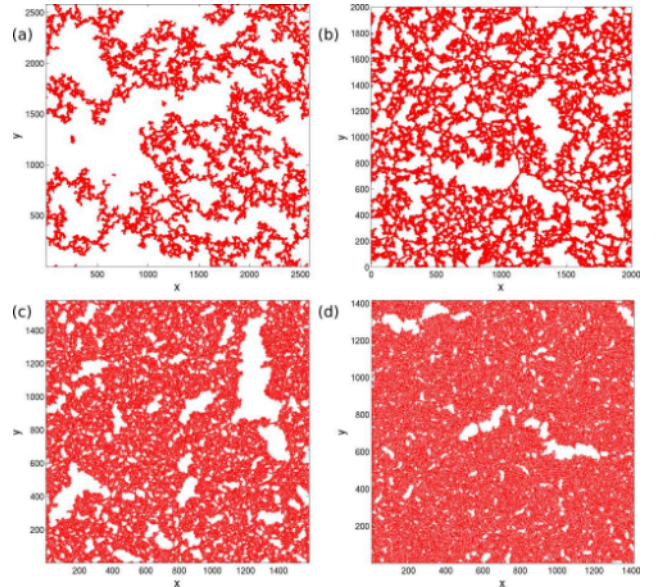


FIG. 1. Snapshots of the two-dimensional binary Lennard-Jones mixture at large time,  $t = 2.7 \times 10^4$ , following a quench from  $T = 3.0$  to  $T = 0.15$  at various densities (a)  $\rho = 0.3$ , (b)  $\rho = 0.5$ , (c)  $\rho = 0.8$ , (d)  $\rho = 1.0$ .

ments,  $\Delta^2(t, t_w) = \frac{1}{N_A} \langle |\mathbf{r}_i(t) - \mathbf{r}_i(t_w)|^2 \rangle$ , for the majority specie (type-A particles), where  $\mathbf{r}_i(t)$  represents the position of particle  $i$  at time  $t$ , and the brackets represent an average over initial conditions. The data in Fig. 2(a) reveal that particle motion depends on  $t_w$ , the dynamics being slower for ‘older’ systems, a feature commonly observed in glassy materials. The details of the particle dynamics are however intriguing. Except for the very short waiting times where the gel structure is barely formed, four dynamic regimes are observed. For  $t_w = 10^2$  and  $10^3$ , for instance, the initial ballistic regime is followed by subdiffusive motion, corresponding to particle caging inside the dense glassy domains. Whereas the cage escape is strongly subdiffusive in aging glasses, here we observe instead a remarkable *superdiffusive* particle motion, with  $\Delta^2(t, t_w) \approx (t - t_w)^{1.6}$ , eventually crossing over to a diffusive regime at very large times. This superdiffusive regime is insensitive to the choice of damping in the DPD thermostat, demonstrating that it is a robust physical feature of the aging process.

To get closer to experimental observations, we spectrally resolve the dynamics and consider the self-part of the intermediate scattering function  $F_s(q, t, t_w) = \frac{1}{N_A} \sum \langle \exp \{ i\mathbf{q} \cdot [\mathbf{r}_i(t) - \mathbf{r}_i(t_w)] \} \rangle$  for a given scattering vector  $\mathbf{q}$ . The data in Fig. 2(b) are for  $|\mathbf{q}| = 1$ , thus probing the dynamics over a ‘mesoscopic’ lengthscale of about 5 particle diameters. The data again display a strong dependence on  $t_w$ , with the relaxation slowing down with  $t_w$ . A striking observation is that the time decay of these correlations can be fitted with a *compressed exponential* form,  $F_s(q, t, t_w) \approx A \exp[-(\frac{t-t_w}{\tau})^\beta]$  with fitting param-

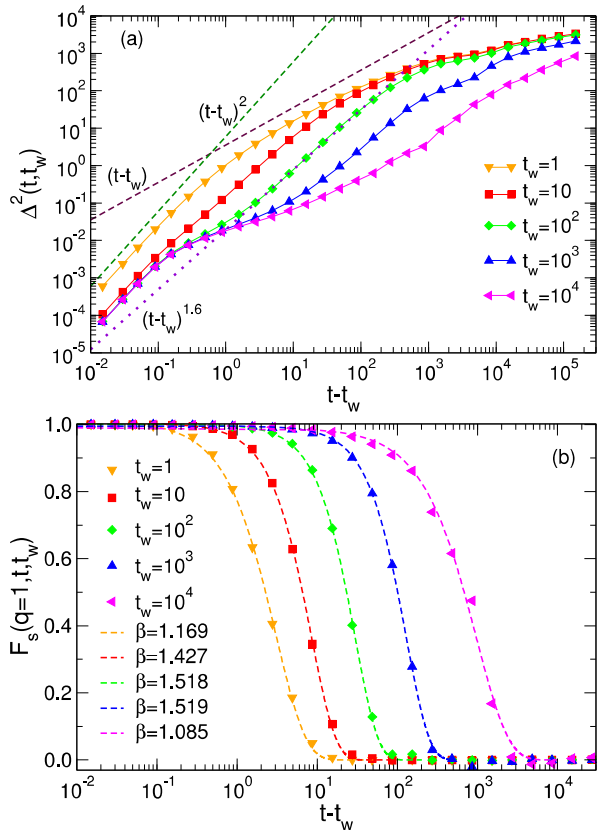


FIG. 2.  $\rho = 0.5$ ,  $T = 0.15$ . (a) Mean-squared displacements,  $\Delta^2(t, t_w)$ , measured for different ages,  $t_w$ . A succession of ballistic, subdiffusive caging, superdiffusive cage escape and long-time diffusion are observed, with various time dependences indicated via dashed lines. (b) Corresponding self-intermediate scattering function,  $F_s(q, t, t_w)$ , measured for  $q = 1$ . The dashed lines correspond to fits with compressed exponentials with the corresponding  $\beta$  values indicated.

ters ( $A, \beta, \tau$ ) which depend both on the age  $t_w$  and on the wavevector  $q$ . The fitted  $\beta$ -values in Fig. 2(b) indicate that  $\beta > 1$ . This implies that there is a fast relaxation process emerging at mesoscopic length scales in the aging gel. This corresponds also to the superdiffusive regime in Fig. 2(a). We suspect that finite-size effects affect data for the largest  $t_w$  shown.

We also investigate how the relaxation varies across lengthscales, by studying the variation of  $F_s(q, t, t_w)$  with  $q$  in Fig. 3(a) for a given age,  $t_w = 10^2$ . The corresponding values for the relaxation time  $\tau$  and the exponent  $\beta$  are shown in Figs. 3(b,c) for two waiting times. In both cases, a complex behaviour is observed. At large  $q$ , corresponding to in-cage motion, we find diffusive dynamics  $\tau \sim q^{-2}$  accompanied by stretched exponential relaxations,  $\beta < 1$ . At larger lengthscales, a crossover to nearly ballistic dynamics  $\tau \sim q^{-1}$  is accompanied by compressed exponential  $\beta > 1$  with a peak value near  $\beta \approx 1.5$  as frequently reported in experiments [23, 27].

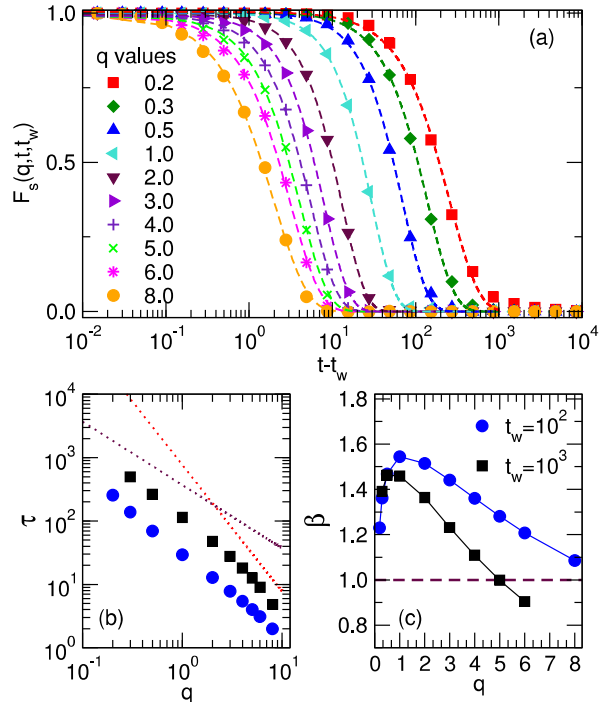


FIG. 3. (a)  $F_s(q, t, t_w)$ , for different wavevectors  $q$ , for  $t_w = 10^2$ ,  $T = 0.15$  and  $\rho = 0.5$ . The dashed lines correspond to compressed exponential fits with fitting parameters reported in (b, c) for both  $t_w = 10^2$  (circles) and  $10^3$  (squares).

Eventually, diffusive dynamics and exponential relaxation should be recovered at large enough lengthscales, but these are difficult to access within our numerical simulations.

To visualise the spatio-temporal evolution of the relaxation, we resolve the dynamics for each particle using a mobility function  $q_i(t, t_w) = 1 - \exp[-\Delta_i^2(t, t_w)/2a^2]$ , such that  $q_i(t, t_w) \approx 1$  when the particle's squared displacement  $\Delta_i^2(t, t_w) \gg a$ , and  $q_i(t, t_w) \approx 0$  otherwise. We then construct a map of the mobility field  $q(\mathbf{r}, t, t_w)$  by creating a spatial grid of mesh size 1.5, and averaging  $q_i$  over each grid for a given pair  $(t, t_w)$ . Varying the probe length  $a$  allows us to independently consider the various regimes of single-particle dynamics. Our most striking finding is reported in Fig. 4, where we consider the mobility field for  $a = 6$  corresponding to the superdiffusive regime and compressed exponential dynamics. These maps reveal that, during the aging process, the local relaxation dynamics is highly heterogeneous and becomes spatially correlated over *ultra-long lengthscales* which are much larger than the typical domain size characterizing the gel. We find no such large dynamic correlation length when the probe length  $a$  is smaller, since then only the caged glassy dynamics is probed. Such features are again in excellent agreement with experiments.

To quantify the size of these dynamic domains, we measure the spatial correlation function  $G_4(r, t, t_w) =$

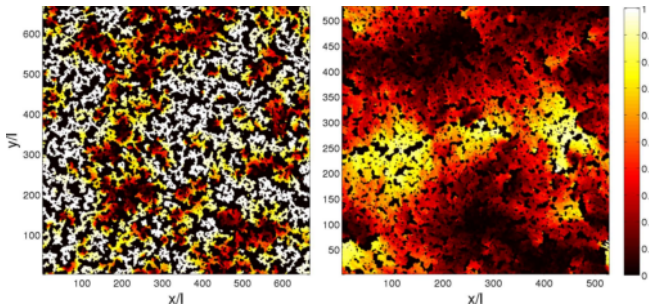


FIG. 4. For  $\rho = 0.5$  (left) and  $0.8$  (right), spatial map of the mobility field,  $q(x, y)$ , at  $t - t_w = 482.74$ , for  $t_w = 10^2$ .

$\langle \delta q(\mathbf{r}', t, t_w) \delta q(\mathbf{r} + \mathbf{r}', t, t_w) \rangle / \langle \delta q^2(\mathbf{r}', t, t_w) \rangle$ , where  $\delta q = q - \bar{q}$ , with  $\bar{\dots}$  corresponding to a spatial average. Representative data for  $G_4(r, t, t_w)$  are shown in Fig. 5. We first show in Figs. 5(a,b) that the size of correlated domains increases with the time  $t - t_w$  indicating that large-scale correlations develop as the structure reorganises. In agreement with Fig. 4, the correlations seem longer for the larger density. Such large length scales (of more than 100 particle diameters) measured in our simulations are reminiscent of the ultra-long correlation lengths measured in experiments using spatially resolved light scattering techniques [21].

In addition, we also observe that the measured correlation lengths, for any  $t - t_w$ , depend upon the system size. This is seen in Fig. 5(c), where we plot  $G_4(r, t, t_w)$  for various system sizes at a given state point. Clearly, the correlation functions decay more slowly for larger systems. If we rescale distances by the linear size of the simulation box,  $L$ , there is an approximate collapse of  $G_4(r, t, t_w)$ ; see Fig. 5(d). Such a dependence of the correlations with  $L$  suggests that the system size is the only characteristic lengthscale over which the relaxation is spatially correlated, at least for the system sizes that we can access numerically (our largest  $L$  is about 2000 particle diameters). Direct visualization indicates that such large scale correlations arise due to the spontaneous elastic recoiling of the network over large distances after local bonds intermittently break due to thermal activation.

Overall, our large-scale simulations reproduce all anomalous features observed in scattering experiments performed in aging gels, most notably subdiffusive caged dynamics crossing over to superdiffusive, compressed exponential dynamics at large enough lengthscales characterized by ultra-long-ranged dynamic correlations. In our model, this arises spontaneously because rare localised relaxation events affect the dynamics of the tenuous gel structure over large lengthscales. Such aging dynamics is not observed in dense glasses. Although our observations are for a two-dimensional system, the underlying mechanism would also apply for three-dimensional experimental systems, and future numerical work would explore that. Studying the mechanical response of such

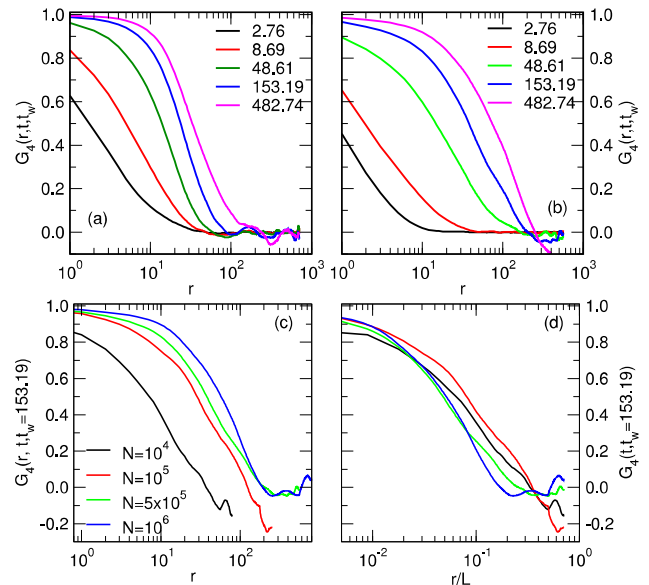


FIG. 5. Spatial correlations of mobility,  $G_4(r, t, t_w)$ , for different  $t - t_w$  (as marked), corresponding to  $t_w = 10^2$ , at  $\rho = 0.5$  (a),  $0.8$  (b), and (c) for different system sizes ( $N$ ), as marked, at  $\rho = 0.8, t_w = 153.19$ . (d) Variation of  $G_4(r, t, t_w)$  corresponding to (c) with rescaled distances  $r/L$ .

aging nonequilibrium gels would also be interesting, with a possible interplay of intrinsic and extrinsic timescales. Also, the potential connection to the observation of sudden collapse in many gels after an initial lag time [43] needs exploration. Whether the mechanisms discussed in the context of such low density materials can be extended to understand similar fast relaxations reported for denser aging glasses [44–46] also requires more investigations.

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