Observation of strongly heterogeneous dynamics at the depinning transition in a colloidal glass

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We study experimentally the origin of heterogeneous dynamics in strongly driven glass-forming systems. Thereto, we apply a well defined force with a laser line trap on individual colloidal polystyrene probe particles seeded in an emulsion glass composed of droplets of the same size. Fluid and glass states can be probed. We monitor the trajectories of the probe and measure displacements and their probability distributions. Our experiments reveal intermittent dynamics and bimodal van-Hove distribution functions around a depinning transition at a threshold force. For smaller forces, linear response connects mean displacement and quiescent mean squared displacement. Mode coupling theory calculations rationalize the observations. Our findings highlight the important differences between quiescent and driven motion in crowded environments.

Experiment.– We study experimentally the active microrheology of uniform oil-in-water emulsion droplets, mean diameter d = 2.01 µm, that show nearly hard-sphere behavior with an experimentally confirmed glass and a jamming transition at volume fractions of 0 < φ < 0.59 and 0.64, respectively. For the volume fractions considered, 0.53 < φ < 0.61, the hard-sphere like droplets are far from touching and there is no stress bearing network of contact points as it is the case for jammed emulsion. The solvent and the emulsion droplets are refractive index and buoyancy matched and a small amount of added polystyrene probe particles of the same size provide optical contrast for laser trapping, see also the Supplemental Material (SI). For each packing fraction, the probe particle mean square displacement (MSD) is first monitored in the quiescent state, without applying any force for 1200 seconds. We find the MSD of particles for φ above it, that show nearly hard-sphere behavior with an experimentally confirmed glass and a jamming transition at volume fractions of 0 < φ < 0.59 and 0.64, respectively. For the volume fractions considered, 0.53 < φ < 0.61, the hard-sphere like droplets are far from touching and there is no stress bearing network of contact points as it is the case for jammed emulsion. The solvent and the emulsion droplets are refractive index and buoyancy matched and a small amount of added polystyrene probe particles of the same size provide optical contrast for laser trapping, see also the Supplemental Material (SI). For each packing fraction, the probe particle mean square displacement (MSD) is first monitored in the quiescent state, without applying any force for 1200 seconds. We find the well known slowing down of the long-time diffusion approaching the colloidal glass transition and the caging of particles for φ above it. The results are in quantitative agreement with previous experiments on similar systems and with calculations from mode coupling theory (see Supplemental Material (SI)).
FIG. 1. Motion of a polystyrene probe particle seeded in a glassy emulsion at $\phi = 0.601$. The diameter of probe particle and the emulsion droplets $d = 2\mu m$ are the same. The optical force is applied in x-direction and the results for two different laser power settings are shown. a) $P_l = 4.5$ mW and b) $P_l = 21.5$ mW. Probe position shown right before (top) and 20 min (bottom) after the constant force line trap has been activated. The lower panels show a map of x-y positions of the probe particle over the whole duration of the experiment. Inset: Enlarged view of the probe particle trajectory. Corresponding movies (accelerated 10×) are included in the Supplemental Material [30].

latter comparison confirms the mapping of density and length scale between measurements and theory. We position the probe particle in a gradient intensity line trap such that a constant force $F_x$ is created along the scan direction, while in the two perpendicular directions the particle motion is strongly confined, Figure 1 [33].

From reference measurements in a simple viscous liquid we find that the force is constant $\pm 2\%$ over a range of $25\mu m$, corresponding to more than 12 particle diameters. The magnitude of the force can be adjusted by tuning the power of the laser $P_l$ and the forces that can be generated are of the order of several hundred femtonewtons. In the experiment the probe particle is first captured at a depth $z \simeq 2 - 6d$ using a single-beam point trap. Larger depths are not accessible due to residual scattering and optical aberrations. The finite depth might induce a small numerical shift of the results due to wall effects, similar to the case of simple fluids [34], but we do not expect significant qualitative changes of the dynamics. Subsequently we align the optical tweezer and the probe particle position and at $t = t_0$ switch the optical configuration to apply a constant force in the $x-$direction parallel to the surface of the sample. The accuracy of tracking the probe particle is approximately $\pm 30\mu m$ [27] and the uncertainty with respect to $t_0$ is set by the acquisition rate of the camera to $\pm 0.1$ sec. To account for this we introduce a small adjustable offset $\delta x$ between $0.01d$ to $0.1d$ to bring out short-time diffusion more clearly. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes. Data points for larger displacements and longer times are not affected due to the logarithmic scales on both axes.
When applying a relatively small force the mean displacement (MD) of the probe should obey the linear response relation:

$$\langle \Delta x(t) \rangle = \frac{(\Delta x^2(t))}{2k_BT} F,$$

(1)

which identifies the equilibrium 1D-MSD divided by $2k_BT$ as time-integrated mobility. Equation (1) predicts that the ratio $\langle \Delta x(t) \rangle / F$ collapses onto the MSD (in units of $2k_BT$) for times and forces where nonlinear effects are negligible. Interestingly, to our best knowledge, this law has never been tested experimentally for strongly correlated colloidal liquids. Figure 2 (a) shows that for small forces the linear response relation holds in the supercooled state for a wide window in time where the probe explores the glassy cage and it’s slow relaxation. Finding the best match between the experimental data and Eq. (1), at short times or over the full range (where it applies), thus provides a very accurate measurement of the applied force, which will be used in the following to convert the laser power setting $P_l$ (in mW) to forces using a fixed conversion factor for each sample (see also Supplemental Material [30]). Taking the force scale from experiment, the MCT calculations also find that the motion within the cage obeys Eq. (1) for small forces. At longer times, MCT predicts a force-induced speed-up of the motion setting in at earlier times for increasing $F$, as seen in simulations [10, 12, 21]. It indicates the force-induced mobilization of the probe and breaking of cages. The experimental results confirm this scenario as shown in Fig. 2 (a). Increasing the laser power and employing forces of order $100k_BT/d$, the measured MD indicate that probes are quickly pulled out of their surrounding cages. The force-induced escape from cages dominates relative to the one by thermal fluctuations in the viscoelastic fluid state. MCT supports this conclusion; quantitative differences exist in the magnitude of the effect. We tentatively attribute the measured stronger localization in the experiment to the use of a line-trap with a confining lateral potential. A build-up of bath particles in front of the probe may slow it down compared to when the perpendicular motion can fluctuate freely.

**Depinning and intermittent dynamics in the glass.–** Linear response also can be observed in glass states for times up to a force-dependent limit. Figure 2(b) gives the measured MD data in a glass and includes the measured force-free MSD scaled onto the MD of the lowest force. MCT calculations for the given forces and densities are included, where the MSD is scaled onto the MD of the lowest two forces. At the lowest force, the small MD are close to the limit of experimental resolution. For higher forces the growth of the MD initially follows the MSD according to Eq. (1) and exceeds it during the cage-breaking process and for longer times. The qualitative behavior is recovered by MCT which (again) overestimates the displacements quantitatively. MCT predicts a threshold force of $F_c d/k_BT = 34.4$ in the glass where probes are pulled free from the cages [12, 25], which is well within the experimentally accessible range. It should be noted that this transition is quite sharp in theory, i.e. a small variation in the force causes a large variation in the behavior of the mean displacement and thus a phase diagram separating delocalized and localized regimes can be established as shown in [12]. In the experiments and previous simulations [12] this phenomenon appears over a broader range of forces. This makes it more difficult to find the threshold force in the experiment. From our data, we estimate it to be $37 < F_c d/k_BT < 182$, which is larger than the MCT prediction, but compatible with the simulation results [12].

**FIG. 3.** Strong forces induce intermittent displacements. We compare experiments (histograms, 40 experiments for $\phi = 0.601$, 10-15 for $\phi = 0.535$) and theory (solid lines) at times, where the mean displacement (dotted vertical line) is the same. The left panels show the van-Hove function in the liquid ($\phi = 0.535$), while the right panels show it in the glass ($\phi = 0.601$) for a small force (top row) at $\langle \Delta x \rangle = 0.4d$ and a larger force (bottom row) at $\langle \Delta x \rangle = 1.7d$. The forces in theory are chosen such that the similarity (as introduced in [35]) between the histogram and PDF is maximized. For comparison, we plot a Gaussian (dashed red line) with the same mean and variance given by the quiescent MSD at the same time. The times corresponding to these displacements are indicated in Fig. 2 with black open diamonds.

We turn now to our main aim, namely to characterize the dynamics at the depinning transition at $F_c$. To answer the question whether the force-induced motion differs qualitatively from the (intrinsic) thermally induced particle motion, we consider the probability distribution function (PDF) of displacements in force direc-
tion, viz. the van Hove function $G^s(\Delta x, t)$. Figure 5 shows histograms of the PDF at fixed average displacement $\langle \Delta x \rangle$ comparing data at (almost) the same forces from fluid (left) and glass (right panels) samples. The chosen distances $\langle \Delta x \rangle = 0.4d$ (upper) and $1.7d$ (lower panels) correspond to the largest mean displacements available in this setup, before the first particle reaches the end of the line trap. We compare forces below and above the depinning transition for the glass and choose similar forces for the liquid. To illustrate the non-Gaussian behavior, a Gaussian distribution with the same average displacement (viz. MD from Fig. 4) and quiescent variance (viz. MSD at the same time) is compared to the data. Also MCT calculations for the same MD values are included; because of the force mismatch in the theory, MCT-forces are fitted to the histograms optimizing the similarity (over a range of mean displacements) following Ref. [35]. In the fluid state for the lower force, the PDF of the probe still resembles the Gaussian solution of the drift-diffusion equation expected in dilute systems [3]. In the glass at this force, however, the PDF extends to far larger displacements than the shifted Gaussian even though it has the same average $\langle \Delta x \rangle = 0.4d$. While some probes remain localized within their cages, some other probes could escape their neighborhood and reach displacements comparable to the bath particle size. This reveals the heterogeneity in the cage strength and the collective origin of the force pinning the probe. MCT predicts the appearance of an exponential tail when approaching $F_c$ [12] which is compatible with the histogram. Even stronger heterogeneity in the probe motion is visible at the larger force. The PDF develops a bimodal shape in the glass consisting of one sub-population of pinned and another sub-population of mobilized particles. In the fluid state, the additional bath motion somewhat narrows the PDF as cages open more uniformly by thermal fluctuations. Bimodal PDF arise in the MCT calculations in a range of forces below and close to the glass transition (not shown) which implies that a characteristic force remains meaningful also in fluid states; it separates intrinsic from force-induced cage breaking processes.

In Figure 4 we show several trajectories $\Delta x(t)$ of the probe particle for an emulsion volume fraction of $\phi = 0.601$ (glass) at a force $F > F_c$ close or slightly above the depinning transition. For comparison, we include trajectories in the fluid ($\phi = 0.535$) for a similar force. Also the complete histograms for large median displacements $\langle \Delta x \rangle = 5.7d$ are compared for fluid and glass sample. Clearly, the motion is far more intermittent in the glass than in the fluid state and the PDF is far broader. MCT predicts bimodal shapes of pinned and mobilized sub-populations close to the depinning force $F_c$. A PDF at $F_c$ and identical median displacement is added for comparison in Fig. 4(c). It correlates well with the sampled histograms. In summary, the intermittent motion is caused by the depinning transition and is more easily observed in the glass than in fluid where it can only be seen for times short compared to intrinsic bath motion (see also Supplemental Material (SI) [30] for more data sets).

Discussion and conclusion.— In summary, we have shown that force-induced intermittent motion can be observed and quantified in glass-forming dispersions, tracking colloidal probes manipulated in an optical line-trap. Linear response rationalizes the behavior for small forces of the order of $O(10k_BT/d)$ and for not too long times. Force-dominated motion sets in at longer times, including in glass states where a force threshold $F_c$ needs to be overcome. Depinning and cage-breaking is characterized by intermittent probe motion and anomalous broaden-
n of the displacement probability distribution. Theory rationalizes the observations and predicts bimodal distributions, where a sub-population of particles remains trapped while another subpopulation moves far. Intermittent motion arises in undercooled fluid states and gets stronger when approaching the glass transition, as correlates with the growth of dynamically heterogeneous regions seen in quiescent dispersions [36]. Yet, it is strongest in glass where only smaller cooperative clusters were observed without force. A comparison with mode coupling theory is possible. In the experiment, anomalous dynamics is observed over a broader range of forces than predicted theoretically.

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