Collective motion of macroscopic spheres floating on capillary ripples: Dynamic heterogeneity and dynamic criticality

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When a dense monolayer of macroscopic spheres floats on chaotic capillary Faraday waves, a coexistence of large scale convective motion and caging dynamics typical for jammed systems is observed. We subtract the convective mean flow using a homogenization or coarse graining method and reveal subdiffusion for the caging time scales followed by a diffusive regime at later times. We apply the methods of dynamic heterogeneity and show that the typical time and length scales of the fluctuations due to rearrangements of observed particle groups significantly increase when the system approaches its densest experimentally accessible concentration. To connect the system to the dynamic criticality literature we fit power laws to our results. The resultant critical exponents are consistent with those found in dense suspensions of colloids indicating universal stochastic dynamics.

Small-scale events can dominate statistical systems to such an extent that one observes phenomena on a global scale. From the classical to the quantum limit, microscopic fluctuations may even change the phase of matter when appropriate control parameters are tuned to critical values 1,2. Even if their origin and nature is not always understood, these spatiotemporal microscopic fluctuations can drive common observable behavior near to such a phase transition. For classical particulate systems, a vast range of materials exhibits a sudden change to a rigid state called a glass or jamming transition. Thermal systems, e.g., supercooled liquids at a critical temperature, or emulsions and colloidal suspensions at a critical packing fraction 3–6, exhibit a glass transition. Furthermore, athermal systems such as foams and granulates experience a jamming transition, also at a critical packing fraction 7–11. In all these systems, transient spatial fluctuations lead to a large scale cooperative motion of their constituents near the transition 3,12,13.

In this Letter, we investigate the dynamics of the collective events near the jamming transition in an alternative experiment: Macroscopic spheres floating on the surface of capillary Faraday waves. Our control parameter is the floating sphere concentration \( \phi \) on the surface which is varied from a moderate value to the maximum value attainable experimentally. Erratic forces due to the surface waves 20 and the attractive capillary interaction among the spheres 21,22 make our system markedly different from the previously studied ones 14,15,17,23,24: A distinct feature is a large scale convection of the spheres on the wave which (for all \( \phi \)) forms naturally and strongly affects the visible dynamics. We aim to understand to what extent concepts from the glass and jamming literature—such as dynamical heterogeneity (DH) and dynamical criticality (DC)—still hold in this convective system. To do so we subtract the convective mean flow using a coarse graining (CG) method and analyze the features of our system both before and after this procedure.

Dynamical heterogeneity (DH) investigates the relation between the local dynamic events on the particle scale and the resultant large-scale cooperative motion 4,6,25–28. For its quantification two observables are introduced: The dynamic susceptibility, a measure of to what extent the dynamics of the system is heterogeneous in space and irregular in time, and the four-point correlation function, a measure of how often and from how far two arbitrarily chosen locally heterogeneous events correlate to one another in time. Both quantities were calculated for colloids 3,12,13, driven hard granulates 14–16,24,25,31, and foams 19. For all cases, the length scales, time scales, and the number of collective events, such as rearrangements of particle groups, dramatically increase near the transition.

The common nature of the behavior of classical particulate systems near transitions encourages to ask whether there is universality. This has led to the concept of dynamical criticality (DC) 32,33. Briefly, DC postulates a power-law relation between the (diverging) length and time scales close to the phase transition. The uniqueness of this—and other—exponents in different systems would then support the existence of universality 32,33. There is evidence pointing to universality in the above sense in various systems 15,17,24. However, investigation is ongoing 25,26,37 and increasing the number of systems either obeying or disobeying the universality is key to reaching a more complete understanding. We will therefore analyze our system in the light of both DH and DC.

A schematic illustration of the experimental setup is shown in Fig. 3. A rectangular container [Fig. 1(a)] is attached to a shaker providing a vertical sinusoidal oscillation such that the vertical position of the container varies as a function of time \( t \) as \( a_0 \sin(2\pi f_0 t) \), where \( a_0 \) is the shaking amplitude and \( f_0 \) is the shaking frequency.
Here, both $a_0$ and $f_0$ are fixed to 0.1 mm and 250 Hz, respectively. This combination is chosen to create capillary ripples on the water surface with a wavelength in the order of the floater diameter ($\approx 0.62$ mm). The container is filled with purified water (Millipore water with a resistivity $> 18 \text{ MΩ-cm}$) such that the water level is perfectly matched with the container edge as shown in Fig. 1(g) to create the brim-full boundary condition [38]. Spherical hydrophilic polystyrene floaters [Fig. 1(d)], contact angle 74° and density 1050 kg/m$^3$, with an average radius $R$ of 0.31 mm are carefully distributed over the water surface to make a monolayer of floaters [39]. The polydispersity of the floaters is approximately 14% and assumed to be just wide enough to avoid crystallization [40]. To avoid any surfactant effects, both the container and the floaters are cleaned by performing the cleaning protocol as described in Ref. [41].

A continuous white fiber light source (Schott) is used to illuminate the floaters from far away as shown in Fig. 1(b). The positions of the floaters are recorded with a high-speed camera (Photron Fastcam SA.1) at 60-500 frames per second. The lens (Carl Zeiss 60mm) is adjusted such that it focuses on the floaters at the non-deformed water surface. Here, we use the random capillary Faraday waves to just agitate the dense floaters so that there is no macroscopic apparent amplitude observed. The wave amplitude is always considerably smaller than the floater radius ($\approx 0.31$ mm).

The resultant capillary ripples on the water surface in the container, made from transparent hydrophilic glass with 10 mm height and a $81 \times 45$ mm$^2$ rectangular cross section, are shown in Fig. 1(h). To eliminate the boundary effects due to the sharp corners of the container, an elliptic rim made from plastic is used to contain the particles. Each image taken with the high speed camera is $512 \times 640$ px$^2$ ($36 \times 28$ mm$^2$), where px means pixel, as shown by the yellow rectangle (size ratios are preserved). The horizontal field of view is $\sim 35\%$ of the total area of the ellipse. Due to the asymmetric surface deformation around each hydrophilic heavy sphere, there is an attraction $[21, 22]$ between the spheres so that the floaters are cohesive $[42]$. For moderate $\phi$, the monolayer can be considered two-dimensional [Fig. 1(i)]. In the dense regime however [Fig. 1(j)], particles are so close that the layer may (locally) buckle and have three-dimensional aspects $[43]$.

The control parameter of the experiment is the floater concentration $\phi$, which (ignoring buckling) is measured by determining the area fraction covered by the floaters in the area of interest [Fig. 1(h)]. In this study, $\phi$ is increased from moderate to dense concentrations, $\phi = 0.65 - 0.77$.

Under the influence of the erratic capillary waves and the attractive capillary interaction, a large scale convective motion is observed with a typical length scale $\sim 60$ times larger than the floater diameter, which is $\sim 1/2$ of the system size, and a typical time scale $\sim 250$ times longer than that of the capillary Faraday waves. To focus on microscopic fluctuations, we subtract the displacement due to the large scale convective motion from our experimental data. At first, we define the velocity field by the coarse graining (CG) method [44, 46] as

$$u(x,t) = \frac{\sum_i \psi_i(t) \psi_d(|x - x_i(t)|)}{\sum_i \psi_d(|x - x_i(t)|)}$$

with the position $x_i(t)$ and the velocity $v_i(t)$ of the $i$-th floater, where we adopt both Gaussian, $e^{-\langle x/d \rangle^2}$, and Heaviside, $\Theta_d(x) = 1 (x \leq d)$ and zero otherwise, as CG kernel functions $\psi_d(x)$. Here, $d$ is a length scale of the order of the particle diameter. Subsequently, we subtract the displacement $l_i(t) = \int_0^t u(x_i(s), s) ds$ due to this macroscopic flow from the position as $x_i(t) \equiv x_i(t) - l_i(t)$ and define an actual displacement during the time interval $\tau$ as $D_i(t, \tau) \equiv |x_i(t + \tau) - x_i(t)|$.

First, we look at single particle dynamics. Approaching the maximal density, particles experience a cage effect, i.e., they are locally trapped by the nearest neighbors, a cage from which, in the presence of fluctuations, sudden escape jumps may occur [3]. The caging and the jumps leave their tracks in the mean square displacement of individual particles as a subdiffusion regime for short times and ordinary diffusion at later times. For our system, we find very similar behavior.

Fig. 2(a) shows the mean square displacement (MSD) of the floaters $\Delta(t) = \langle \sum_i D_i^2(t, \tau)/N \rangle$, where the brackets $\langle \ldots \rangle_i$ represent an average over time $t$ [47] and
$N$ is the number of floaters in the sample. In our experiment, the floaters are transported by the large scale convection, and thus, the resultant motion is always ballistic. Therefore, when we do not subtract the displacement $I_1(t)$ from the experimental data, the MSD quadratically increases with time with a slope 2 in the log-log plot [open squares in Fig. 2(a)]. However, when we do subtract the additional displacement due to the convection for a suitable value of $d$ [48], both the initial subdiffusive and the later diffusive regimes are found.

![FIG. 2. (Color online) (a) Mean square displacement (MSD) of the floaters for $\phi = 0.755$. The open squares are obtained without subtracting the displacement $I_1(t)$ due to the large scale convection. The red circles and open triangles are the closed circles and squares in Fig. 2(a)]. Here, $\sigma$ is the floater diameter. The solid line represents $\phi_G$ Gaussian, respectively. Here, $1/s$ is the number of floaters in the sample. In our experimental average at which we were able to measure. By fitting a power law $\tau_c \sim (\phi_j - \phi)^\alpha$ to our data [54], we find that $\phi_j \approx 0.82$, which is consistent with the above and leads us to conclude that $\phi_j = 0.82 \pm 0.02$. Note that this value is considerably larger than the suggested static buckling density of the attractive monodisperse spheres [51], namely $\phi_c \approx 0.71$. Next, we use this fixed value for $\phi_j$ in our power-law fit to obtain the exponent $\alpha \approx -3.9 \pm 0.9$, which is consistent with the exponent $\alpha \approx -4.0 \pm 0.6$ found in an earlier experiment [44].

To quantify the heterogenous dynamics of the floaters, we introduce the self-overlap order parameter $q_c(t, \tau) = \sum w_a(D_i(t, \tau))/N$ and the dynamic susceptibility $\chi_a(\tau) = N \left[\langle q_a^2(t, \tau) \rangle_t - \langle q_a(t, \tau) \rangle^2_t \right]$. Here, $w_a(x)$ is the overlap function (OF) defined as a Gaussian $e^{-(x/a)^2}$ or a Heaviside step function $\Theta_a(x)$ as defined previously (1 for $x \leq a$ and 0 otherwise). The width of the OF, $a$, is a measure for the typical distance over which a single floater can move within time $\tau$. To disregard the motion of the floaters in the cage, $a$ is chosen to be larger than their typical displacement inside a cage and also chosen to maximize the extremal value of the dynamic susceptibility as shown in the inset of Fig. 2(b) [17].

The various coarse graining (CG) functions and overlap functions (OF) in total give us six different manners of analyzing the data, if we also include the possibility of not subtracting the displacement due to the large scale convection before the DH analysis. These are summarized in Table I together with the optimal values of $d$ and $a$ obtained as described above [52]. When we plot the dynamic susceptibility $\chi_a(\tau)$ we obtain similar results in all six cases [the inset of Fig. 3(b)]. In particular the location of the peak in $\chi_a(\tau)$ provides us with an estimate of the typical time scale $\tau^*$ of the dynamic heterogeneity, which are plotted for all six cases as functions of $\phi$ in Fig. 3(b). To investigate the dynamic correlation length of the floaters, we apply the four-point correlation function [53]

$$g_a(r, \tau) = \frac{1}{2\pi r N} \left\langle \sum_{i,j} \delta(r - r_{ij}(t)) c_{ij}(t, \tau) \right\rangle_t - \rho \langle g_a(t, \tau) \rangle^2_t$$

satisfying $\chi_a(\tau) = 2\pi \int r g_a(r, \tau) dr$, where $\rho \equiv N/S$ and $S$ are the number density of the floaters and the area of interest, respectively. $N$ is the number of floaters as introduced previously. In addition, we define $r_{ij}(t) = |r_i(t) - r_j(t)|$ and $c_{ij}(t, \tau) \equiv w_a(D_i(t, \tau))w_a(D_j(t, \tau))$. Furthermore, we assume the Ornstein-Zernike form of the four-point correlation function [53], in which the dynamic correlation length $\xi^*$ is obtained considering the scaling $g_a(r, \tau^*) = A(r/\xi^*)^{-\beta} e^{-r/\xi^*}$ for some amplitude $A$ and exponent $\beta$, where $\tau^*$ is the time scale obtained from $\chi_a(\tau)$.

![FIG. 3. (Color online) (a) Dynamic susceptibility $\chi_a(\tau)$ for $\phi = 0.755$. The open squares are obtained without subtracting the displacement $I_1(t)$ due to the large scale convection. The red circles and open triangles are the closed circles and squares in Fig. 3(a)]. Here, $\sigma$ is the floater diameter. The solid line represents $\phi_G$ Gaussian, respectively. Here, $1/s$ is the number of floaters in the sample. In our experimental average at which we were able to measure.

| CG function | OF | $d/\sigma$ | $a/\sigma$ |
|-------------|----|------------|-------------|
| (i) None    | Gaussian | – | 0.49 |
| (ii) None   | Heaviside | – | 0.52 |
| (iii) Gaussian | Gaussian | 1 | 0.038 |
| (iv) Gaussian | Heaviside | 1 | 0.042 |
| (v) Heaviside | Gaussian | 1.6 | 0.042 |
| (vi) Heaviside | Heaviside | 1.6 | 0.046 |
resultant $G_a(r/\xi^*)$ successfully collapses onto a single master curve $e^{-r/\xi^*}$ for each $\phi$ except for the tails. This procedure is repeated for each condition in Table I. Remarkably, we find that neither the value of the exponent nor the master curve presents any significant difference.

Fig. 3 displays the time scales of the dynamic heterogeneity, $\tau^*$ [Fig. 3(b)], and the dynamic correlation length, $\xi^*$ [Fig. 3(c)], plotted versus $\phi$, where both increase strongly with $\phi$. One can introduce the power law fits [21, 32, 34]

$$\tau^* = C(\phi_J - \phi)^\eta,$$

$$\xi^* = D(\phi_J - \phi)^\lambda.$$  

Both the time exponent $\eta$ and the length exponent $\lambda$ are calculated considering all conditions reported in Table I. Fitting to the data we obtain $\eta \simeq -3.9 \pm 0.4$ and $\lambda \simeq -1.4 \pm 0.4$ for each condition in Table I where we again used $\phi_J \simeq 0.82 \pm 0.02$. Finally, combining Eqs. (3) and (4) in the light of dynamical criticality [22, 33], namely $\tau^* \sim \xi^{\nu}$, we quantify the relation between $\eta$ and $\lambda$ as $\nu = \eta/\lambda \simeq 2.7 \pm 1.2$.

Summarizing, by eliminating the naturally occurring large-scale convection of the floaters, we find from the mean square displacement that the single floater dynamics resembles the caging observed in glassy liquids, first with subdiffusion followed by normal diffusion for later times. The crossover time $\tau_\phi(\phi)$ between the two regimes [Fig. 2(b)] diverges near the estimated jamming point $\phi_J$, which can be fitted by a power law $(\phi_J - \phi)^\alpha$ with exponent $\alpha \simeq -3.9$. A second time scale is that of the dynamic heterogeneity $\tau^*$ [Fig. 3(b)], which can also be fitted by a power law which, remarkably but consistently, has the very same exponent $\eta \simeq -3.9$. The typical distance between two correlated, successive events, the dynamic correlation length $\xi^*$ [Fig. 3(c)] obtained from the four-point correlation function [Fig. 3(a)], presents a power law scaling with exponent $\lambda \simeq -1.4$. Both of our dynamical exponents, $\eta$ and $\lambda$, are in a good agreement with the previous experiments on sheared microgel spheres by Nordstrom et al. [24], where the critical exponents of the time and length scales were found to be $-4$ and $-4/3$, respectively.

The coarse graining procedure allows us to successfully remove the convective flow. While this is indeed necessary to study micro-fluctuation driven by diffusion, paradoxically, one of our main results is that it is not necessary to remove this mean flow for the dynamic susceptibility and the four-point correlation function. These results only depend insignificantly on whether the mean flow is subtracted or not. In addition, results do hardly depend on the choice of coarse graining and overlap functions, as long as the length scales in these test functions are optimized. In fact, from Table I it can be appreciated that $a$ must be an order of magnitude lower with mean flow subtraction ($a \simeq 0.04\sigma$) than without ($a \simeq 0.5\sigma$). Finally, we determine from fits that $\phi_J \simeq 0.82$, which is considerably larger than the suggested critical density for static monodisperse floaters ($\phi_0 = 0.71$), and also than the largest concentration that we could reach experimentally, namely $\phi_{\text{max}} \simeq 0.77$. For larger $\phi$, our layer of floating spheres is not stable under driving. Understanding the difference between $\phi_{\text{max}}$ and $\phi_J$ requires further study and lies beyond the scope of this paper.

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[21] The procedure in the ensemble average is to calculate $|r_i(t + \tau) - r_i(t)|$ using arbitrary starting times $t$ and averaging over $\tau$. A similar procedure is followed in calculating ensemble averages in the self-overlap order parameter, dynamic susceptibility and four-point correlation function.
[22] The optimal values for $d$ were obtained as follows: When looking at the subtraction procedure as a function of $d$ we find that the displacement rises steeply from zero for $d < \sigma$, $\sigma$ is the floater diameter, into a plateau from which it continues to rise. A value in the center of the plateau is chosen, which happens to correspond roughly to the particle diameter.
[23] From the (two-dimensional) pair correlation function $g(r)$ we observe no evidence for significant crystallization which may cause an increase of this upper limit for $\phi_d$. Secondly, although buckling may be a significant factor, it does not lead to a broadening of the first peak in $g(r)$ that one would expect to be present if particles start to overlap for increasing $\phi$. And, finally, the homogenized local packing fraction shows a sharp cut-off at $\phi \approx 0.84$. 

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These facts together suggest that $\phi_J < 0.84$.

[50] Due to their limited range other functional forms could possibly also fit our dynamic time and length scales. However, we restrict ourselves to power-law fits in order to compare our results to those in the literature.

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