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

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When a densely packed monolayer of macroscopic spheres floats on chaotic capillary Faraday waves, a coexistence of large scale convective motion and caging dynamics typical for glassy systems is observed. We subtract the convective mean flow using a coarse graining (homogenization) method and reveal subdiffusion for the caging time scales followed by a diffusive regime at later times. We apply the methods developed to study 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 largest experimentally accessible packing 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 densely packed suspensions of colloids.

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I. INTRODUCTION

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 are 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 observable behavior near to such a transition temperature, and emulsions and colloidal suspensions at a critical packing fraction [5,6,8], exhibit a glass transition. Furthermore, athermal systems such as foams and glassy systems near transitions encourage to ask whether there is universality. This has led to the concept of dynamic criticality [17–20], which 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 dynamic heterogeneity summarized in Ref. [28].

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 dynamic criticality [17–19,37–40], which 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 dynamic heterogeneity.
universality [17–19,37–40]. There is evidence pointing to universality in the above sense in various systems [17–19,27]. However, investigation is ongoing [32,41–43]. Furthermore, increasing the number of systems either obeying or disobeying the universality in the above sense in various systems [17–19,27]. There is evidence pointing to universality [17–19,37–40].

We will therefore analyze our system in the light of both dynamic heterogeneity and dynamic criticality.

II. EXPERIMENT

A schematic illustration of the experimental setup, which is the same one having been already used in our earlier study [25], is shown in Fig. 1. 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 (\( \sim 0.62 \) mm).

The container is filled with purified water (Millipore water medium) with an average radius \( R \) of 0.31 mm 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 [45]. The polydispersity of the floaters is approximately 14% and assumed to be just wide enough to avoid crystallization [46]. To avoid any surfactant effects, both the container and the floaters are cleaned by performing the cleaning protocol as described in Ref. [47].

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 control parameter of the experiment is the floater (packing) 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 large 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.

III. DYNAMIC HETEROGENEITY AND DYNAMIC CRITICALITY

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 method [50–52] as

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

(1)

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

First, we look at single particle dynamics. Approaching high packing fractions, the particles experience caging [33], i.e., a cage composed of a group of particles locally trapped by their neighbors. The particles in the cages are immobile. However, in the presence of fluctuations near the critical packing density, escape jumps occurring at a certain relaxation time create heterogeneous flow, i.e., fast and slow flow regions appear simultaneously [3].

The caging and the successive jumps leave their tracks in the mean-square displacement of individual particles: It has been first theoretically suggested as a measure of the relaxation time for densely packed suspensions exhibiting jamming [39] and then also experimentally confirmed in both a jammed driven granular system [28,39] and colloidal glasses [53]. Unlike glassy systems, in jammed systems there are no jumps and the particles only do jittering motion in the cages [28,39]. Therefore, one can observe a very long plateau in the temporal evolution of the mean-square displacement near the jamming packing fraction [28] (or the plateau persists as long as the simulation runs [39]). On the other hand, the plateau in glasses either does not persist very long [5,53] or may not even exist at all near the critical packing fraction (or the glass transition temperature). Moreover, two diffusive regimes are observed [5,53]. For our system, we find very similar behavior to a glass transition: A subdiffusion for short times and an ordinary diffusion at later times. However, instead of observing the finite plateau, we observe an intermediate (a transient) regime to separate the two diffusions.

Figure 2(a) shows the mean-square displacement of the floaters $\Delta (r) = \langle \sum D_i^2(t,\tau)/N \rangle$, where the brackets $\langle \cdots \rangle_t$ represent an average over time $t$ and $N$ is the number of floaters in the sample [54]. In our experiment, the floaters are transported by the large scale convection, and thus, the resultant motion is always ballistic. Consequently, when we do not subtract the displacement $l_i(t)$ from the experimental data, $\Delta (\tau)$ 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 $d$, both the initial subdiffusive and the later diffusive regimes are found [55].

As shown in Fig. 2(b), the crossover time $\tau_d$ between these subdiffusive and diffusive regimes, rapidly increases with $\phi$. Since the subdiffusion represents the cage effect of the floaters described above, it is plausible that the crossover time diverges when the system is jammed, where no floater can ever escape from the cage [28,39]. On physical grounds, the jamming happens at a critical density $\phi_c$, e.g., if the system does not have temperature, $\phi_c$ is nearly equal to the random close packing of polydisperse disks, $\phi_{RCP} \approx 0.84$, while the temperature or external driving force slightly increases $\phi_c$ [40]. However, the capillary action keeps floaters at a distance, which might compete against the increase of $\phi_c$ due to the
as described above [59]. When we plot the Table I, together with the optimal values of displacement due to the large scale convection before the if we also include the possibility of in total give us six different manners of analyzing the data, (c) The dynamic correlation length = external driving [56]. In addition, \( \phi \) needs to be considerably larger than \( \phi_{\text{exp}} = 0.77 \), the largest experimental average at which we are able to measure, so that we can assume the critical density is in the range \( \phi_{\text{exp}} = 0.77 < \phi_c < \phi_{\text{RCP}} \).

By fitting a power law \( \tau \sim (\phi_c - \phi)^\alpha \) to our data [57] we find that \( \alpha \approx 0.82 \), which is consistent with the above and leads us to conclude that \( \phi_c = 0.82 \pm 0.02 \). It is worth mentioning that previous experiments found a similar divergence of the relaxation time (associated with the contact number) near the critical density 0.8151 [28,33]. Note that this value is considerably larger than the suggested static buckling density of the attractive monodisperse spheres [58], namely \( \phi_0 \approx 0.71 \). Next, we use this fixed value for \( \phi_c \) 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 [27].

To quantify the heterogenous dynamics of the floaters, we introduce the self-overlap order parameter \( q_{\alpha}(t, \tau) = \frac{\sum w_a[D_i(t, \tau)]}{N} \) and the four-point dynamic susceptibility \( \chi_4(t) = \left( \sum_{i,j} |q_{ij}(t, \tau)| \right) - \left( q_{ij}(t, \tau) \right)^2 \). Here \( w_a(x) \) is the overlap function defined as a Gaussian \( e^{-\frac{(x-a)^2}{2}} \) or a Heaviside step function \( \Theta_a(x) \) as defined previously (1 for \( x \leq a \) and 0, otherwise). The width of the overlap function \( a \) 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 cages, \( a \) is chosen to be larger than their typical displacement inside a cage and also chosen to maximize [19] the extremal value of the \( \chi_4(t) \) as shown in the inset of Fig. 3(b).

The various coarse graining functions and overlap functions 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 dynamic heterogeneity analysis. These are summarized in Table I, together with the optimal values of \( d \) and \( a \) obtained as described above [59]. When we plot the \( \chi_4(t) \) we obtain similar results in all six cases [case (i) in Table I is shown in the inset of Fig. 3(b)]. In particular the location of the peak in \( \chi_4(t) \) 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 [60]

\[
g_4(r, \tau) = \frac{1}{2\pi r N} \left( \sum_{i,j} \delta[r - r_{ij}(t)]c_{ij}(t, \tau) - \rho(q_{\alpha}(t, \tau))^2 \right)
\]

satisfying \( \chi_4(t) = 2\pi \int_0^\infty r g_4(r, \tau) dr \), where \( \rho = N/S \) 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 \( r_{ij}(t) \equiv |r_i(t) - r_j(t)| \) 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 [60], in which the dynamic correlation length \( \xi^* \) is obtained considering the scaling \( g_4(r, \tau^*) = A(r/\xi^*)^{-\beta} e^{-r/\xi^*} \) for some amplitude \( A \) and exponent \( \beta \), where \( \tau^* \) is the time scale obtained from \( \chi_4(t) \).

Figure 3(a) shows the function \( G_4(r/\xi^*) = (r/\xi^*)^6 g_4(r, \tau^*)/A \), where the (very weak) exponent \( \beta = 0.01 \) is taken to be independent of \( \phi \). The resultant \( G_4(r/\xi^*) \) 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.

Figure 3 displays the time scale of the dynamic heterogeneity \( \tau^* \) [Fig. 3(b)] and the dynamic correlation length

![Figure 3](image-url)

### Table I. Analysis methods: The set of the coarse graining functions and the overlap functions, with \( d \) and \( a \) as described in the main text, both in terms of the floater diameter \( \sigma \).

<table>
<thead>
<tr>
<th>Coarse graining function</th>
<th>Overlap function</th>
<th>( d/\sigma )</th>
<th>( a/\sigma )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(i) none</td>
<td>Gaussian</td>
<td>–</td>
<td>0.49</td>
</tr>
<tr>
<td>(ii) none</td>
<td>Heaviside</td>
<td>–</td>
<td>0.52</td>
</tr>
<tr>
<td>(iii) Gaussian</td>
<td>Gaussian</td>
<td>1.0</td>
<td>0.038</td>
</tr>
<tr>
<td>(iv) Gaussian</td>
<td>Heaviside</td>
<td>1.0</td>
<td>0.042</td>
</tr>
<tr>
<td>(v) Heaviside</td>
<td>Gaussian</td>
<td>1.6</td>
<td>0.042</td>
</tr>
<tr>
<td>(vi) Heaviside</td>
<td>Heaviside</td>
<td>1.6</td>
<td>0.046</td>
</tr>
</tbody>
</table>
\(\xi^*\) [Fig. 3(c)], where both are plotted versus \(\phi\) and increase strongly with \(\phi\). One can introduce the power-law fits [17–19,27,37–39]

\[
\tau^* = C(\phi_c - \phi)^\eta, \quad (3)
\]

\[
\xi^* = D(\phi_c - \phi)^\lambda. \quad (4)
\]

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 \(\phi_c \simeq 0.82 \pm 0.02\). Finally, combining Eqs. (3) and (4) in the light of dynamic criticality [17–19,37–39], namely \(\tau^* \sim \xi^{1/\omega}\), we quantify the relation between \(\eta\) and \(\lambda\) as \(v = \eta/\lambda \simeq 2.7 \pm 1.2\).

IV. CONCLUSION

Studying agitated floaters by the capillary Faraday waves, after eliminating their naturally occurring large-scale convection, their mean-square displacement dynamics resembles caging as observed in glassy liquids: An initial subdiffusion event, their mean-square displacement dynamics resembles after eliminating their naturally occurring large-scale convection, etc. In fact, from Table I it can be appreciated that the coarse-graining (smoothing) length \(a\) must be an order of magnitude lower with the convective mean flow subtraction \((a \simeq 0.04\sigma)\) than without \((a \simeq 0.5\sigma)\).

Finally, we determine from the fits that \(\phi_c = 0.82\), while close to the two-dimensional random close packing \((\phi_{RCP} \simeq 0.84)\), being considerably larger than the suggested critical density for static monodisperse floaters \((\phi_b = 0.71)\), and also larger than the largest packing density that we could reach experimentally, namely \(\phi_{expt} \simeq 0.77\). For larger \(\phi\), our layer of floating spheres is not stable under driving. Understanding the difference between \(\phi_{expt} \simeq 0.77\) and \(\phi_c\) requires further study. In this respect, investigating the origins of the dynamic heterogeneity, of which it is known that there are many [33], can give further information about our system and help to clarify this difference.

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[45] The particles have been custom made by a collaborating company and are not commercially available.


[48] Even in the dilute case, the distance $r$ between the floaters is much smaller than the capillary length $L_c = \sqrt{\gamma/\rho g}$, with $\gamma$ as the surface tension coefficient of the interface, $\rho$ as the liquid density, and $g$ as the acceleration of gravity. For an air-water interface at $20^\circ C$, $L_c = 2.7 \text{ mm}$.


[54] 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 will be followed in calculating ensemble averages in the self-overlap order parameter, four-point dynamic susceptibility, and four-point correlation function.

[55] 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 $d \ll \sigma$, $\sigma$ 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 floater diameter.

[56] 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 $\phi_c$. Second, 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 cutoff at $\phi_c \approx 0.84$. These facts together suggests that $\phi_c < 0.84$.

[57] 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 to compare our results to those in the literature.


[59] Both $d$ and $a$ are first calculated for each $\phi$ separately, and then appropriate values are obtained by averaging over the determined values.