Brownian motion of ellipsoidal particles on a granular magnetic bath

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We study the Brownian motion of ellipsoidal particles lying on an agitated granular bath composed of magnetic particles. As we decrease the magnetic forcing, the bath takes characteristics of a molecular system that is cooled towards the glass transition. We quantify the mobility of different floating ellipsoidal particles using the mean square displacement and the mean square angular displacement, and relate the diffusion coefficients to the bath particle motion. The ratio of translational and rotational diffusion constants for the floating particles is forcing-independent, although the particle shape matters (with longer floating particles rotating slower than shorter). Unusual aspects of the floating particle motion include non-Gaussian statistics for their displacements, and a shape-dependent and forcing-dependent anisotropy of translational diffusion coefficients parallel and perpendicular to the ellipsoid long axis.

I. INTRODUCTION

Brownian motion (diffusion) occurs in a wide variety of processes in nature. While initially observed in regular liquids, its theory has been applied not only in physics, but in biology, chemistry, and even economics. In macroscopic granular systems, external forcing can effectively "thermalize" the system and result in diffusive motion \cite{1–5}. In super-cooled liquids, diffusion slows greatly, but with rotational diffusion of non-spherical probes slowing even more than translational diffusion \cite{12–16}. Some granular experiments \cite{5–8} and simulations \cite{9, 10} showed that granular systems can exhibit glassy behavior under some conditions, with diffusive motion greatly slowed. However, rotational diffusion has not been studied in these granular experiments.

In this manuscript, we study the translational and rotational behavior of floating ellipsoidal particles on a magnetic granular bath, and examine the influence of the bath particle intensity on the motion of the floating particles. By varying the forcing applied to the magnetic bath particles, we can vary the diffusivity of the floating particles by nearly two orders of magnitude. For low magnetic forcing, the bath particles behave like a supercooled fluid. Across the range of bath particle conditions, we find that the floating particles always have rotational diffusion and translational diffusion coupled, that is, their ratio is constant, unlike what has been seen in prior glassy experiments mentioned above. While the coupling of the diffusion constants is to be expected for particles in a “normal” fluid, we find other unusual aspects of the floating particle motion such as non-Gaussian statistics and a forcing-dependent anisotropy of translational diffusion coefficients parallel and perpendicular to the ellipsoid long axis.

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FIG. 1: The experimental setup consists of magnetic particles in a horizontal container (diameter 132 mm) in the middle of Helmholtz coils (inner diameter 152 mm, outer diameter 179 mm). A sinusoidal signal fed into a power amplifier produces an alternating magnetic field. The spherical bath particles have permanent magnetic dipoles so the changing field causes the particles to roll on the surface in random directions. A flat particle “floats” on the bath particles and moves due to the random kicks it receives from the underlying bath particles.

II. EXPERIMENTAL SETUP

The experimental configuration consists of a flat particle floating on a forced granular medium; see Fig. 1. This granular medium is composed of a 2D ensemble of stainless steel spherical particles of diameter 1 mm, mass 4.2 mg, and area fraction 0.23. The system is placed in a time-dependent magnetic field. The steel particles have been previously magnetized so that they possess a permanent magnetic dipole. The magnetic field is generated by a pair of Helmholtz coils fed by a power amplifier which receives a sinusoidal signal. This alternating field causes the particles to move randomly: each steel particle tries to align its magnetic dipole moment with the
instantaneous field (pointing up or down), causing the particle to rotate in a random direction to implement the reorientation. When the field reverses, the particle tries to reorient, subject to the rotational inertia it already possesses. The result is that the steel particle motion is quite random and plays the role of a thermal bath. Decreasing the magnitude of the magnetic field results in an effectively “cooler” granular bath. Although this system is highly dissipative, it reaches a stationary state which fits the necessary conditions to be considered as an Ornstein-Uhlenbeck stochastic process [19]. The dynamics of a system similar to these bath particles has already been studied, and it has been demonstrated that for certain conditions the magnetic field is proportional to an effective temperature [20]. The intensity of the bath particle motion is varied with the magnitude of the oscillating magnetic field: $B_0 = 66, 60, 54, 48, 42, 36$ and $30 \text{ G}$, all of them with a frequency of $10 \text{ Hz}$. In this system, for high intensities of the field, the bath particles show the dynamics similar to that of a Brownian gas, and the velocity distributions are Maxwell-Boltzmann distributions [21]. For low intensities, the motion of the particles decreases, showing freezing-like effects resembling the dynamics near the glass transition [8].

We use floating particles with aspect ratios $\alpha = 1.00, 1.44, 1.48,$ and $2.20$, as shown in Fig. 2. Further details of these particles are listed in Table I. The floating particles are non-magnetic, and do not interact with the magnetic field. Rather, they sit atop the magnetic bath particles, and move due to the random kicks of the bath particles.

We obtain three series of three-minute videos for each floating particle and magnetic forcing amplitude. From each video we track the translational and rotational motion of the floating particle, along with the bath particle motion [21]. Our temporal resolution is $1/60 \text{ s}$, our spatial resolution is better than $0.07 \text{ mm}$ ($x$ and $y$ position), and our angular resolution is better than $0.006 \text{ rad}$ ($\approx 0.4^\circ$).

### III. RESULTS

To characterize the motion of the bath particles, we calculate the mean square displacement (MSD) for each magnetic field amplitude we consider. The results are shown in Fig. 3. It is interesting to note that for all of the data, at short $\Delta t$, the MSD of the bath particles is superdiffusive with $\langle \Delta r^2 \rangle \sim \Delta t^{1.7}$. This implies that over a short time scale particles are more likely to move in straight lines, rather than being purely diffusive random walks [22]. At high magnetic field intensities, the motion of the bath particles is diffusive (top curves of Fig. 3). As the magnetic field intensity decreases, bath particle

<table>
<thead>
<tr>
<th>aspect ratio $\alpha$</th>
<th>$2r_{\text{major}}$ (mm)</th>
<th>$2r_{\text{minor}}$ (mm)</th>
<th>mass (mg)</th>
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TABLE I: Details of the floating particles, including semi-major diameter and semi-minor diameter.

FIG. 2: Images showing floating particles with aspect ratio (a) $\alpha = 1.00$, (b) $\alpha = 1.44$, (c) $\alpha = 1.48$, and (d) $\alpha = 2.20$. The bath particles are the smaller spheres. Note that the image in (a) is viewed with reflected light, while the other are viewed with transmitted light, leading to the different appearance of (a) from the other panels. The circular particle in (a) has a white ellipse drawn on it to facilitate rotational tracking of this otherwise circularly symmetric particle.
motion is slower. At the lowest forcing, the MSD has a plateau at long time scales (bottom curve of Fig. 3). This resembles a supercooled fluid close to the glass transition

We likewise consider the floating particle motion, which has both translational and rotational components. Example translational and rotational trajectories of a floating particle with aspect ratio $\alpha = 2.20$ for a high and a low excitation case are shown in Fig. 4. Both the rotational orientation and the position in $x$ show a greater variation for higher excitations than for the lower ones. The full trajectories for high excitations expand noticeably over a larger area compared with the trajectories at low excitations (insets of Fig. 4).

Figure 4 shows the MSD $\langle \Delta r^2 \rangle$ and the mean square angular displacement $\langle \Delta \theta^2 \rangle$ for the floating particle with aspect ratio $\alpha = 2.20$. The vertical dotted line corresponds to the period of the magnetic forcing, and coincides with a crossover region from short time scale behavior to long time scale behavior. An interesting feature that stands out in these graphs is the diffusive behavior at large time scales for both MSD and MSAD, regardless of the aspect ratio of the bath. This result is interesting, since it indicates that, although the bath particles go from diffusive to glassy states as we decrease the magnetic forcing (Fig. 4), the floating particles maintain their diffusive behavior. Intriguingly, the bath particle MSDs all have a constant initial power law growth (Fig. 4) constant initial slope on the log-log plot) whereas the floating particle MSDs exhibit an initial slope dependent on the forcing. Thus, within the time scales $\Delta t < 0.1$ s (the forcing period), the coupling between the bath particles and floating particles is nontrivial. The $\Delta t \to \infty$ behavior is also interesting: for the bath particles, the slope of the MSD changes with the magnetic forcing intensity, whereas for the floating particles their behavior is always diffusive at long time scales (slope = 1 on the log-log plot).

The differing behavior at long time scales between the bath particles and the floating particles suggests that the direct influence of the bath is only at shorter time scales; that the diffusive behavior of the floating particles is emergent from the random influence of the bath at shorter time scales. To investigate this, we fit the $\Delta t \to \infty$ portion of the floating particle MSD and MSAD curves to determine the diffusion coefficients through $\langle \Delta r^2 \rangle = 4D_T \Delta t$, $\langle \Delta \theta^2 \rangle = 2D_R \Delta t$. We then examine the dependence of the measured diffusion coefficients on various quantities related to the bath particles. In particular we use the bath particle motions (quantified by the MSD) using the path integral formalism for the time scale, $\Delta t = 0.1$ s (the period of the magnetic forcing). We define a length scale $L = \sqrt{\langle \Delta r^2 \rangle}$ using displacements over this time scale. Figure 5 shows a linear relationship between $L$ and the logarithms of all of the diffusion coefficients. This relationship, $\ln(D) \sim L/L_1$, holds for all our results with a common length scale $L_1 = 0.22$ mm; this is clear from the least square fit lines in Fig. 5 where the fit is constrained to have a common slope and differing intercepts. Defin-
FIG. 6: (a) Translational diffusion coefficients $D_T$ as a function of the forcing $L$. The different symbols are for different aspect ratio floating particles as indicated. (b) Rotational diffusion coefficients $D_R$ as a function of the forcing $L$. The symbols are the same as (a). For both panels, all fit lines have a constant slope: $\ln(D) \sim L/L_1$ with $L_1 = 0.22$ mm. The uncertainty in $D$ is the symbol size.

FIG. 7: Angell plot of the floating particles with different aspect ratios for (a) translational diffusion and (b) rotational diffusion. Straight lines suggest an activated process, with the forcing $L$ playing the role of a temperature; see text for details. The symbols correspond to particles with aspect ratios as labeled in (a). The dashed lines are the same fitting shown in Fig. 6. Note that in the original paper by Angell (Ref. [27]) the horizontal axis is non-dimensionalized by the glass transition temperature. Here we do not identify a glass transition for the floating particles, so we do not attempt to non-dimensionalize the horizontal axis. The uncertainty is the symbol size.

We note that fitting the diffusion coefficients to $\ln(D) \sim L^2/L_2^2$ works nearly as well, with $L_2 = 0.78$ mm. The least square error is slightly larger, but the overall appearance is similar to the results shown in Fig. 6. The two fitting constants $L_1$ and $L_2$ are both similar in order of magnitude to the bath particle radius 0.5 mm. The dependence of $\ln(D)$ on $L$ or $L^2$ suggests something like an activated process, although it is not apparent why this would be the case.

An alternate way of thinking of an activated process is inspired by an analysis technique introduced by C. A. Angell for analyzing glassy data [27, 28]. He argued that viscosity $\eta$ (at high temperature) is an Arrhenius process with some activation energy $E_a$, so $\eta \sim \exp(E_a/k_B T)$, with $k_B$ being Boltzmann’s constant and $T$ being the absolute temperature. By plotting $\ln(\eta)$ as a function of $1/T$, a straight line will be found (with slope $E_a/k_B$).

We do not measure viscosity in our experiment – in fact, it may not be a relevant concept for our particles floating on the granular bath. Nonetheless, in analogy with diffusion coefficients for fluids, we note for fluids $\eta \sim T/D$ (for both translational and rotational diffusion). If our $L$ acts like a temperature, then plotting $\ln(L/D)$ as a function of $1/L$ should give a straight line. We show this in Fig. 7 here the linear dependence works plausibly well for the “hot” data (left symbols). The linear fit does not pass through the points corresponding to the lowest forcing (right-most points) and in fact we do not include those points in the least square fit. The lines have a variety of slopes, which is plausible as the “activation energy” would be expected to depend on the floating particle shape, size, and motion (translational or rotational).

That being said, the fitting shown in Fig. 6 has fewer free parameters and seems more compelling, albeit less physically motivated. The fitting of Fig. 6 is shown as the dashed lines in Fig. 7.

We next wish to compare $D_T$ to $D_R$. In a 3D Newtonian fluid, the rotational motion of ellipsoids follows $D_T \sim 1/r_{\text{major}}$, $D_R \sim 1/r_{\text{major}}^3$, in part because the two diffusion coefficients have different units. Accordingly, we consider the non-dimensional ratio $r_{\text{major}}^2 D_R/D_T$, plotted as a function of $L$ in Fig. 8. Given that $D_T$ and $D_R$ have the same functional dependence on $L$ (Fig. 6), it must be true that their ratio is a constant, as Fig. 8 shows (within the uncertainty). The largest value of
The ratio of diffusion coefficients for floating particles with aspect ratios as indicated, as a function of the forcing intensity \( L \). The ratio is nondimensionalized by multiplying by \( r_{\text{major}}^2 \), the square of the semi-major axis. Representative error bars are shown for two data sets; the uncertainties of the other two data sets are identical. The horizontal lines indicate the mean value of the ratio for each particle type: from top to bottom, these values are 3.2, 2.0, 1.5, and 1.0 rad\(^2\), corresponding to \( \alpha = 1.00, 1.48, 1.44, \) and 2.20.

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r_{\text{major}}^2 D_R/D_T \text{ appears for the floating particle with aspect ratio } \alpha = 1.00, \text{ with the non-dimensional ratio being } 3.2. \text{ For spheres diffusing in 3D, this ratio would be } 3/4 \text{ [29–31], so a somewhat similar order of magnitude. For our larger aspect ratio floating particles the ratio } r_{\text{major}}^2 D_R/D_T \text{ decreases. This decreasing trend with increasing aspect ratio disagrees with numerical results for 3D rod diffusion [32–33]. This disagreement suggests, not surprisingly, that the mechanism leading to diffusion in our 2D granular experiment are qualitatively different from normal 3D diffusion in liquids.}

According to the classic Stokes-Einstein-Sutherland and Stokes-Einstein-Delye equations for \( D_T \) and \( D_R \) respectively, the ratio \( D_T/D_R \) should be a constant independent of temperature and viscosity; the exact constant will depend on the floating particle shape. In glass-forming systems, prior observations found “decoupling” of translational and rotational diffusion, such that \( D_R/D_T \) is not constant but rather decreases as the glass transition is approached. This has been seen in experiments with small molecule glass-formers [34–35], colloidal glass experiments [13, 16, 57], and simulations [38–41]. Our bath particles become somewhat glassy as the magnetic forcing is decreased, as seen in Fig. 8. Despite our bath particles becoming glassy, in our experiments we do not observe decoupling of the translational and rotational diffusion: there is no systematic dependence of the data in Fig. 8. This is further evidence that the long-time behavior of the bath particles does not connect with the long-time behavior of the floating particles.

To understand the influence of the anisotropic shape of our floating particles, we examine motion along the instantaneous axes parallel and perpendicular to the ellipsoid. We determined the displacements between a given frame and the next, with the axis defined at the first frame [17]. We use this short time motion to define \( D_{\text{par}} \) in the direction parallel to the ellipsoid long axis, and \( D_{\text{perp}} \) perpendicular to the long axis. (That is, \( D_{\text{par}} = (\Delta r_{\text{par}}^2)/\Delta t \) with \( \Delta t = 1/60 \) s, and similarly for \( D_{\text{perp}} \). We use this short time interval so that the tracer barely rotates, and thus the axes stay well defined.) Figure 9 shows \( D_{\text{par}}/D_{\text{perp}} \) for the four tracers as a function of the forcing. As one would expect for ellipsoids in 2D or 3D [17, 18], the parallel motion is faster than perpendicular motion and as aspect ratio increases, this ratio is larger.

A surprising observation is that for the circular floating particles (\( \alpha = 1.00 \), violet diamonds in Fig. 9) the ratio is not identically 1. For these tracers, the orientation is indicated by a mark on the circular particle [see Fig. 2(a)]. It is likely that the particle is somehow asymmetric, for example due to the ink printing the mark, or the particle having deformed slightly due to collisions with the bath particles. Note that we initially define directions parallel and perpendicular to the mark on the circular particle, but then find the axes relative to that mark that maximize the anisotropy; it is this coordinate system that leads to the data shown in Fig. 9.

Figure 9 shows that for all floating particles the ratio depends on the forcing. The larger the force is, the lower the ratio \( D_{\text{par}}/D_{\text{perp}} \) is, in contrast to ellipsoids in regular fluids where this ratio should be constant, independent of the fluid viscosity [17]. In all cases, we observe \( D_{\text{par}}/D_{\text{perp}} < 2 \); 2 is the limiting value for a long ellipsoid diffusing in 3D [18].

To better understand particle motion (both bath and floating particles), we show representative probability distribution functions of displacements in Fig. 10. In each panel the blue data are for the lowest excitation and are the narrow distributions; the red data are for the
FIG. 10: Probability distribution functions $P(\Delta x)$ for (a) bath particles, (b) floating particles with aspect ratio $\alpha = 1.00$, and (c) floating particles with aspect ratio $\alpha = 2.20$. In each case the blue data correspond to $L = 0.90$ mm (the lowest excitation) and the red data correspond to $L = 1.82$ mm (the highest excitation). $\Delta t = 0.1$ s is used to define the displacements. The dashed and dotted lines are Gaussian fits with the same standard deviation as the experimental data. The non-Gaussian parameter $\alpha_2$ has the values (a) 1.4, 0.1; (b) 6.2, 0.5; (c) 2.4, 0.6 for small and large $L$.

highest excitation and are the broad distributions. The dotted and dashed black lines are Gaussian fits, and it is immediately apparent that the distributions are broader than a Gaussian. That is, large displacements are rare, but more common than would be expected if the distribution was a Gaussian of the same standard deviation. Non-Gaussian distributions are often seen in supercooled systems [11, 23, 24], so the result is plausible for the bath particles. It is intriguing that the distribution functions are non-Gaussian for the floating particles even though these particles are diffusive at long time scales (Fig. 5). That is, often strong non-Gaussian behavior is associated with a more pronounced plateau in the mean square displacements; Fig. 5 shows a weak plateau, which is most obvious for the lowest forcing data. In all cases, the distributions are close to Gaussian for higher excitation. The same qualitative results are found for the rotational displacements (data not shown).

The relative width of the displacement probability distributions can be quantified with the non-Gaussian parameter $\alpha_2(\Delta t)$ [42],

$$\alpha_2(\Delta t) = \frac{\langle (\Delta r^4) \rangle (\Delta t)}{2[\langle (\Delta r^2) \rangle (\Delta t) ]^2} - 1. \quad (1)$$

This quantity is zero for a Gaussian distribution, and positive for distributions with broad tails such as those shown in Fig. 10; the specific values of $\alpha_2$ are given in the caption to the figure.

The non-Gaussian parameter is shown in Fig. 11(a) for translational motion, and (b) for rotational motion, as a function of the forcing strength $L$. For these results, the time scale $\Delta t = 0.1$ s is used to define displacements, although the qualitative behavior is not too sensitive to this choice. For low forcing, the non-Gaussian parameter is largest, reflecting the extreme width of the blue data in Fig. 10 as compared to the Gaussian fit. $\alpha_2$ drops for larger $L$ in Fig. 11. In this respect the behavior is similar to glassy liquids, which are more non-Gaussian close to the glass transition, and more Gaussian when well away from the glass transition [23, 43, 44].

IV. CONCLUDING REMARKS

In our experiments the granular bath particles become glassy at the lowest forcing $L$, while the floating particles are still diffusive. We conjecture this is because the
floating particle does not interact with the bath particles directly. Typically one thinks of glassy behavior as arriving due to crowding of molecules or particles. In our experiment, no matter how the bath particles are crowded or interact with one another, the floating particle sits atop them and does not experience the crowding. It is interesting that the floating particle’s long-time motion continues being diffusive even though the particles in the bath are not, and even though the floating particles’ displacement probability distributions are non-Gaussian. As described above, the bath particles are forced periodically with a period of 0.1 s, and this time scale appears to play a key role in determining the floating particle motion. It suggests that in our experiment we have a novel source of random energy acting like some sort of effective temperature. This may provide insight into other situations with non-thermal “effective temperature” baths.

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