Translational and Rotational Dynamical Heterogeneities in Granular Systems

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(Received 6 March 2018; published 6 July 2018)

We use x-ray tomography to investigate the translational and rotational dynamical heterogeneities of a three dimensional hard ellipsoid granular packing driven by oscillatory shear. We find that particles which translate quickly form clusters with a size distribution given by a power law with an exponent that is independent of the strain amplitude. Identical behavior is found for particles that are translating slowly, rotating quickly, or rotating slowly. The geometrical properties of these four different types of clusters are the same as those of random clusters. Different cluster types are considerably correlated or anticorrelated, indicating a significant coupling between translational and rotational degrees of freedom. Surprisingly, these clusters are formed already at time scales that are much shorter than the α-relaxation time, in stark contrast to the behavior found in glass-forming systems.

DOI: 10.1103/PhysRevLett.121.018002

The relaxation dynamics of most disordered materials, such as glass-forming liquids, polymers, foams, and granular materials, differs significantly from the Debye behavior found in simple liquids in that it shows a marked nonexponential time dependence [1,2]. Although several mechanisms can give rise to this type of time dependence, it is often the local disorder of the particle arrangement that is the origin for this behavior [3–5]; i.e., each particle has a different local environment, and as a result, the relaxation dynamics depends strongly on the particle considered. Previous studies have shown that the slowly (or quickly) relaxing particles are not distributed uniformly in space but instead form clusters. This effect, named "dynamical heterogeneity" (DH) is believed to be a key ingredient to understanding the α-process of glass-forming systems, and hence the phenomenon of the glass transition. As a consequence, many studies have been carried out to study the nature of the DHs [3–12].

Usually DH are associated with the translational degrees of freedom (TDOF) of the particles. The particles in molecular systems and granular materials have, however, also rotational degrees of freedom (RDOF). Since these are coupled with the TDOF it is evident that they will be important for the relaxation dynamics of the system as well [13–17]. However, in practice it is difficult to probe the RDOF in molecular systems since experiments do not allow us to track directly the orientation of individual particles. As a consequence, only indirect experimental probing of the RDOF has been possible so far [7,13] and most of our current knowledge of them comes from computer simulations [18–20]. (Plastic crystals are an exception since they have no disorder in the TDOF [21].) The situation is not much better for the case of granular materials since these are usually opaque and hence it is very challenging to probe in three dimensions (3D) the displacement and reorientation of the particles [22–28]. Because of the nonspherical shape of the particles and the presence of friction, there is often a strong coupling between the TDOF and RDOF, making the experimental study of the DH for both TDOF and RDOF indispensable for understanding of the relaxation dynamics [29]. In the present work we thus use x-ray tomography to investigate these DH in a *three dimensional* granular packing driven by oscillatory shear, making it, to the best of our knowledge, the first experimental investigation to probe the DH of the TDOF *as well as* the ones for the RDOF.

Our system consists of 4100 hard prolate ellipsoids made of polyvinyl chloride with an aspect ratio of 1.5 (polydispersity 0.9%), i.e., a shape that makes the crystallization of the system difficult [30] and allows for a rather strong *T-R* coupling. The dimension of the minor axis of the particles is $2b = 12.7$ mm, and in the following we will use b as the unit of length. The particles are in a rectangular box of dimension $40.2b \times 43b \times 22.6b$ that can be sheared in the y direction. More details of the setup are given in [31]. We drive the system to a steady state by cycling it many times at all strain amplitude γ investigated ($\gamma = 0.26$, 0.19, 0.10, and 0.07, see Supplemental Material [32] for details).

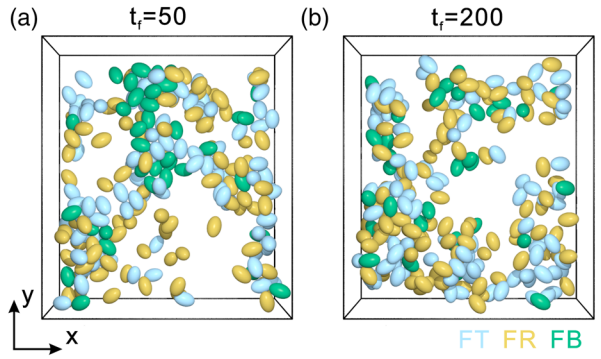

FIG. 1. Snapshots showing the fast translating (FT, blue) and fast rotating (FR, yellow) particles [$\gamma = 0.10$(L), $t = 1000$]. Also shown are the particles that are FT as well as FR (FB, green). Note that both types of particles form clusters and that these clusters overlap significantly. Panels (a) and (b) are for the filter time $t_f = 50$ and $t_f = 200$, respectively.

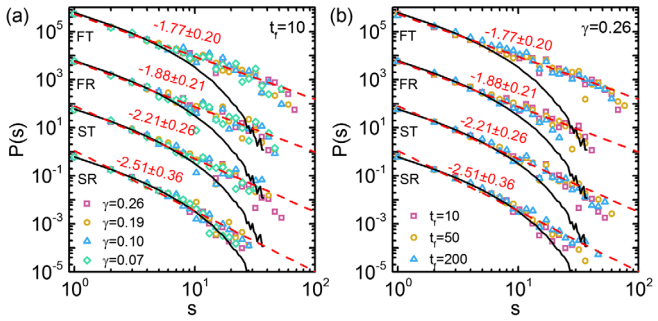

FIG. 2. Cluster size distribution $P(s)$ for FT, FR, ST, and SR. (The first three sets have been shifted vertically.) The solid black lines are the distributions obtained by randomly picking particles in the system. The dashed red lines are fits to the data with a power law with exponents stated next to these lines. (a) Different strain amplitude γ. (b) Different filter times t_f.

Subsequently, the position and orientation of all particles are determined by x-ray tomography scans. Scans were made after each complete cycle, thus giving a stroboscopic view of the dynamics with the time unit of one shear cycle, and in the Supplemental Material [32] we show the mean squared displacement for the TDOF and the RDOF, which allows us to get an idea on the relevant time and length scales in the system.

To probe the DH we have tracked the particles for a "filter time" t_f and determined the distribution of their T and R displacements (see Supplemental Material [32]). Fast (slow) translation particles are defined as the 10% fastest (slowest) particles in this distribution and we denote these particles as FT and ST. The same was done for the RDOF, thus allowing us to define the fast (slow) rotating particles, FR and SR.

Figure 1 shows typical snapshots of the FT and FR particles (blue and yellow, respectively) for two values of the filter time t_f. One recognizes that both populations form clusters demonstrating that the T and R dynamics are spatially very heterogeneous. A significant part of the FT particles are also FR (FB, green), indicating that the translational and rotational motion are significantly coupled. Similar conclusions are reached for the slowly moving particles (see Supplemental Material [32]). In the following, we will make a quantitative characterization of these DHs.

To determine the cluster size distribution $P(s)$ of the four populations of particles we define two particles to be neighbors if their Voronoi cells have a common face and use this to construct connected clusters. Figure 2(a) shows $P(s)$ for the four populations at different strain amplitudes γ and one recognizes that within the accuracy of the data the curves for different γ coincide; i.e., $P(s)$ is independent of γ. This somewhat surprising result is likely related to the fact that for granular systems the details of the relaxation dynamics are universal and independent of γ due to the particular manner by which such systems explore their phase space [31].

Also included in Fig. 2(a) is a fit to the data with a power law, a dependence that has been observed in previous experiments probing the DH [35]. The exponent is small for the FT clusters and larger for the SR ones (see legend), which shows that the particles with a slow dynamics belong on average to smaller clusters than the FT particles. (The average cluster sizes are 3.92, 3.16, 2.46, and 2.04 for FT, FR, ST, and SR, respectively.) In the Supplemental Material [32] we show that the average volume of the Voronoi cell of slow particles is smaller than the one of the fast particles; i.e., the ST and SR clusters are a bit more densely packed than the ones for FT and FR. This shows that fast particles prefer to form extended loose clusters which will allow for cooperative fast motion, similar to the case of glass-forming liquids [10]. Also included in the figure are the $P(s)$ obtained if one picks 10% of particles in a random manner; i.e., one does not select the fast or slow ones. The so obtained average cluster size is 1.94 and the corresponding $P(s)$ shows at intermediate and large s the expected exponential decay, i.e., a s dependence that is very different from the power law that we find for the fast or slow particles [36].

Figure 2(b) presents $P(s)$ for $\gamma = 0.26$ and different values for the filter time t_f demonstrating that the distribution is also independent of t_f. Therefore, Fig. 2 demonstrates that the DH are independent of the time scale considered, i.e., t_f, and of the manner the system is driven, γ. This is in contrast to the findings for thermal glass-forming systems in which the DH are found to depend on the time scale considered and also on the temperature, i.e., a parameter that is somewhat analogous to our driving strength γ. The surprising fact that clusters are present already at very small t_f, also confirmed by the observation that the non-Gaussian parameter is basically independent of t (Fig. SI-8 in the Supplemental Material [32]), shows that the DH are *not* related to the α-relaxation process, in

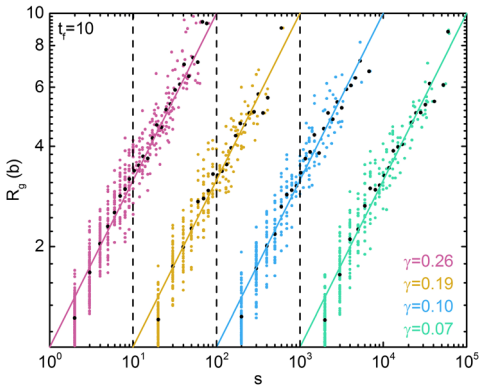

FIG. 3. Radius of gyration of the FT clusters vs s for different values of γ. Black dots are the average at fixed s. The solid lines are a guide to the eye with slope 0.5. For the sake of clarity the data for $\gamma < 0.2$ have been shifted to the right by 10, 10^2, and 10^3.

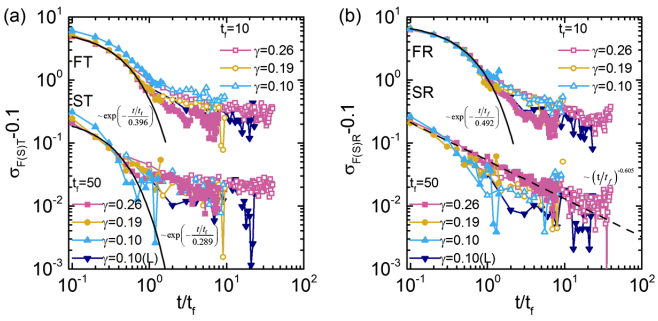

FIG. 4. Probability that a particle that was fast or slow at $t = 0$ is still fast or slow at time t. Black solid lines are exponential fits to the data at short times. Panels (a) and (b) are for the TDOF and RDOF, respectively. The data for SR are fitted with a power law (black dashed line). The FR and FR data have been shifted vertically.

contrast to the findings for thermal glass formers [10,20]. Instead, as argued below, we conjecture that it is the surface roughness of the particles that is the source of disorder which leads to the DH, i.e., the same mechanism that gives rise to the universal relaxation dynamics documented in Ref. [31].

The cluster size distribution $P(s)$ is only related to the number of particles in a cluster but contains no information about its geometry. Therefore, we determine the radius of gyration R_g of a cluster via

$$R_g^2 = \frac{1}{s}\sum_{i=1}^{s}(\mathbf{r}_i - \bar{\mathbf{R}})^2, \tag{1}$$

where $\mathbf{r}_i$ is the position of particle i and $\bar{\mathbf{R}}$ is the center of the cluster. The s dependence of R_g for the FT clusters is presented in Fig. 3, showing that it is given by a power law with an exponent of 0.5, i.e., a mass fractal exponent of 2.0 [37]. This value is independent of the type of particle we consider (FT, FR,...), see Supplemental Material [32], indicating that the geometry of the clusters is independent of the type of motion considered, in contrast to thermal glass-forming systems for which one finds that the clusters with FT are more open than the ones for ST [38]. Figure 3 also demonstrates that these values are again independent of γ. If we select the particles randomly the resulting mass fractal exponent is around 1.9, i.e., a value that is very close to the one we find for the DH clusters, indicating that the geometry of the DH clusters is very similar to the one of a random cluster, but that they have an enhanced probability to be large.

The nature of the particles, FT, FR,..., will change with time and hence it is of interest to probe how long a particle keeps this property since this time can be expected to be related to the lifetime of the clusters. Therefore, we define the quantity $\sigma_\alpha(t)$ as the probability that a particle which at time $t = 0$ had a property $\alpha \in \{\mathrm{FT}, \mathrm{ST}, \mathrm{FR}, \mathrm{SR}\}$ has the same property at time t. Figure 4 shows that the t dependence of σ_α is independent of γ, i.e., the curves for the different strain amplitudes fall on top of each other. This result is surprising since naively one might have expected that a larger strain would lead to a faster loss of memory, because for a given fixed time t the mean squared displacements of the particles increase quickly with γ (see Fig. SI-4 in the Supplemental Material) [39]. The figure also demonstrates that the master curve does not depend on the filter time t_f if one plots the data as a function of the reduced time t/t_f. This independence shows that the details of the relaxation dynamics do not depend on the time scale considered; i.e., there is a scale invariance of the dynamics in the time window we probe, suggesting that the configuration space explored by the system has a fractal-like nature.

Also included in the figure are fits with an exponential function (black solid lines). These fits describe the data well for $t/t_f \lesssim 1$ indicating that at short times the particle changes its nature via a simple stochastic escape process. However, for longer times one observes a slower decay: For FR and SR we find a power law with an exponent around 0.6, whereas for FT and ST the data seem to go to a finite value given by 0.13 and 0.12, respectively; i.e., the memory does not vanish within the time scale of our experiment. The persistence of this memory regarding the population type of the particle is likely related to the result discussed in Ref. [31] where it was found that the TDOF do show a noticeable memory effect in their motion [40]. From panel (b) we recognize that $\sigma_\alpha(t)$ for the RDOF shows at intermediate time a power law with an exponent around 0.6; i.e., there is no plateau, a result that is coherent with the findings that the RDOF show only a rather weak memory in their dynamics [41]. From a physical point of view the absence of a plateau is reasonable since the cycling motion and the presence of friction will always lead to a slow rotation of the particles, thus permitting them to change their nature (FR or SR).

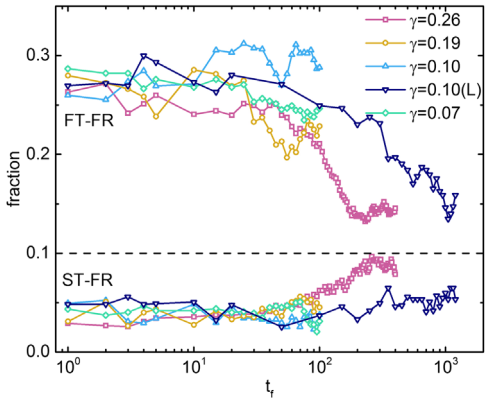

FIG. 5. Probability that a particle is undergoing a FT as well as FR (ST and FR) as a function of the filter time t_f. The different symbols correspond to different strain amplitudes. The dashed line shows the probability at long times.

The persistent rotational motion will induce also a translational motion of the neighboring particles because of steric effects and hence we do expect that friction leads to an enhancement of the rotational-translational coupling [29] which in turn might lead to correlations between the DH of the TDOF with the ones of the RDOF. To study this correlation we determine the overlap between the different clusters, i.e., the probability that a particle simultaneously belongs to two different populations. Figure 5 shows this probability for the combinations FT-FR and ST-FR as a function of t_f [42]. We see that for small and intermediate t_f the overlap for FT-FR is around 0.27, i.e., 2.7 higher than the trivial value of 0.1 expected for random clusters, and that this enhancement is independent of γ. Hence, there is a significant probability that a particle which is translating quickly is also rotating quickly. For $t_f \gtrsim 10^2$ the overlap starts to decay. From the mean squared angular displacement (see Fig. SI-4 [32]) one recognizes that this time scale is related to the onset of a significant rotational dynamics; i.e., the particles have rotated far enough that the nature of their rotational dynamics has been randomized, thus leading to a decrease of the overlap. For the ST-FR pair the overlap is two times lower than the trivial value; i.e., slowly translating particles have a significantly reduced probability to rotate quickly. This overlap starts to approach the equilibrium value 0.1 for times that are again on the order of 10^2 cycles, i.e., when the particle has rotated by a significant amount (about 0.5 rad^2, see Fig. SI-4 [32]). In Fig. SI-7 we show that also the pair ST-SR has an enhanced overlap and the pair FT-SR has a decreased overlap. None of these overlaps depend on γ if t_f is not too small and only the decay to the trivial value depends on the driving strength, indicating that the decay is indeed related to the randomization of the RDOF.

The presented results show that our granular system has DH for the TDOF that are qualitatively similar to the ones found in simple glass-forming liquids. Having access for the first time to the RDOF in a 3D experimental system, we have probed also the dynamics of the RDOF and find that also they do show DH with cluster sizes that are just a bit smaller than the ones for the TDOF. The cluster size distribution of all four populations of particles can be described well by a power law, thus a distribution that is very different from the one of random clusters. Remarkably, these distributions are independent of the strain amplitude, indicating that the dynamics is system universal [31]. The strong correlation and anticorrelation between different types of clusters shows that in this system the translational and rotational degrees of freedom are strongly coupled. This coupling is likely not only caused by the aspect ratio of the particles but also by the presence of friction, an effect that in molecular systems is replaced by microscopic forces.

In Ref. [31] we argued that 3D granular systems show a relaxation dynamics that is very different from the one of thermal glass formers (e.g., there is no cage effect). Although we now find that the DH are qualitatively similar to the ones of thermal glass formers, we emphasize that the DH we observe here occur on the time scale that is significantly shorter than the α-relaxation time, i.e., the time scale at which the particles leave their neighborhood. This surprising observation shows that the energy landscape of granular materials has a structure that is very different from the one of thermal glass formers since it has a roughness on a length scale that is much smaller than the size of the particles. It can be expected that it is this particle inherent disorder that gives rise to the DH, in contrast to the case of thermal systems in which the variations of the local packing are the cause for the DH.

In summary, we conclude that the presence of DH is not a feature that is unique to thermal glass formers, but instead can be found in other disordered systems as well and hence is more universal than expected. The mechanisms leading to these DH are, however, strongly dependent on the system considered. In recent years it has been recognized that one possible approach to learn more about the relaxation dynamics of systems with glassy dynamics is to probe multipoint correlation functions [43,44]. Although with our setup such measurements are not feasible because of the lack of statistics, in the future it should be possible to do such type of experiments. This will then allow us to gain a more detailed insight into the relaxation dynamics of 3D granular systems.

Some of the preliminary experiments were carried out at BL13W1 beam line of Shanghai Synchrotron Radiation Facility. The work is supported by the National Natural Science Foundation of China (No. 11175121, No. 11675110, and No. U1432111), Specialized Research Fund for the Doctoral Program of Higher Education of China (Grant No. 20110073120073), and ANR-15-CE30-0003-02.

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