Granular Dynamics in Compaction and Stress Relaxation

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Elastic and dissipative properties of granular assemblies under uniaxial compression are studied both experimentally and by numerical simulations. Following a novel compaction procedure at varying oscillatory pressures, the stress response to a step strain reveals an exponential relaxation followed by a slow logarithmic decay. Simulations indicate that the latter arises from the coupling between damping and collective grain motion predominantly through sliding. We characterize an analogous ''glass transition'' for packed grains, below which the system shows aging in time-dependent sliding correlation functions.

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Mechanically agitated granular materials are characterized by slow relaxation dynamics, arising from the rearrangement of the constituent grains within the volume in which they are confined [1]. This leads to a slow compaction of the system volume, which follows a logarithmic decay in time, as seen in several experiments and predicted by theoretical models of granular compaction [2,3]. This property has prompted analogies between the physics of athermal granular materials and thermal glasses, the theory of which is better understood at the fundamental level [1]. The field of granular matter therefore benefits from such parallels, as new ways of investigating the system's complex properties are discovered. An advantage of using granular materials over glasses is that it facilitates a much easier exploration of the microstructure through grain-grain interactions.

Logarithmic relaxation and rate-dependent strengthening have also been observed in *compressed* granular matter [4], although the underlying mechanism is still under much debate. The collective rearrangement of the grains in the bulk could be responsible for the slow relaxation, as suggested by experiments on slowly sheared granular materials by Hartley and Behringer [4]. On the other hand, it is also known that aging occurs at the contacts between the particles [5], which manifests itself as a logarithmic increase of the friction coefficient between grains as a function of time. It could also be responsible for the observed slow dynamics, as has been suggested in experiments by Ovarlez *et al.* and Nasuno *et al.* [6,7], which show rate dependence and slow strengthening characteristics of aging at the interparticle contacts, respectively.

The goal of this Letter is to demonstrate the existence of slow relaxation in the response of dense granular matter to infinitesimal strain perturbations and to elaborate on the origin of the dynamics. The experiments reveal a very slow stress relaxation under a constant applied differential strain. This behavior is well characterized by a two-step relaxation dynamics, analogous to the slow relaxation in ''glassy'' systems [1].

We investigate this dynamics via computer simulations, which employ various dissipative processes into the system in order to compare their relative effects. The results show that the main process responsible for the logarithmic stress relaxation is the collective particle motion and rearrangements of grains, predominantly through sliding. We compute the fraction of sliding particles and find that when the damping in the system exceeds a critical value, a slow increase in the number of sliding particles as a function of time is observed. This slow strengthening leads to the logarithmic stress relaxation. Moreover, the system shows the hallmark of glassy behavior: aging in the sliding correlation function.

Experimental arrangement.—We use an INSTRON press to measure the mechanical properties of granular assemblies confined in a cylindrical cup and piston. The machine was strain controlled and enabled oscillatory, step, and ramp compressional tests up to a maximum limit of 300 kN load. The cylinder cup had the following dimensions: diameter $d = 5.08$ cm, height $h = 7.62$ cm, and wall thickness $l = 0.85$ cm. These dimensions were chosen to achieve a good statistical ensemble for nearly monodisperse glass beads of diameter (355 ± 5) μm. We perform two tests: first, we measure the strain response (compaction) under large stress oscillations, and then we measure the stress response under an infinitesimal step-strain perturbation.

Compaction experiments (stress-controlled).—Before performing the stress relaxation experiments it was necessary to develop a method of reaching the jammed state, ensuring that reproducible experiments can be performed.

Here we introduce an alternative method to the one of Knight *et al.* [2] of compaction based on oscillatory pressure of varying amplitude to generate reversible jammed states. The compaction procedure consists of the stages depicted in Fig. 1, where we show the displacement of the piston Δh during the application of the stress σ. The material is first compressed with a constant, slow velocity of 0.1 mm/min to a target stress of 0.5 MPa (from A to B

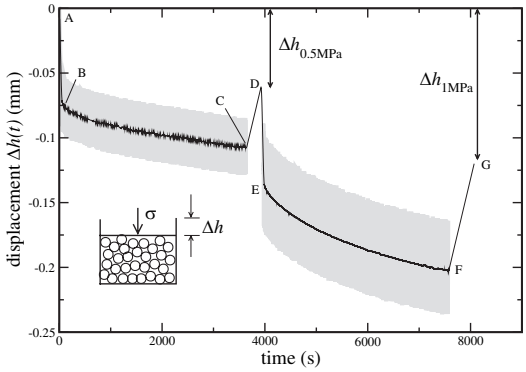

FIG. 1. Compaction procedure: Sinusoidal displacement Δh of the system height in response to an oscillatory stress around the mean value of 0.5 MPa (from B to C) and 1 MPa (from E to F). The solid black line is the window average displacement of the height and shows the logarithmic behavior of the strain under large stress amplitude according to Eq. (1) for the later stages of the time evolution. The shaded gray area symbolizes the amplitude of the oscillatory strain response. The inset shows a schematic of the experimental arrangement.

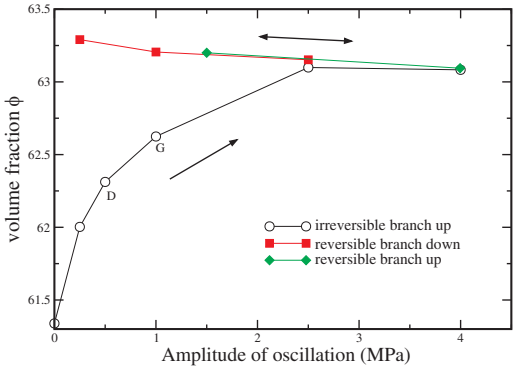

FIG. 2 (color online). Volume fractions obtained by oscillatory compression experiments at varying oscillatory stress amplitudes. The final heights at D and G in Fig. 1 are used to calculate the volume fractions shown in this figure.

in Fig. 1, which shows only the strain response), then oscillated between zero and double the mean value with a frequency of 1 Hz (from B to C, the amplitudes of oscillation are always taken as equal to the mean value). Each compaction procedure consists of 3600 cycles (until C), after which the material is again slowly released to its uncompressed state (from Point C at $\sigma = 0.5$ MPa to D at $\sigma = 0$). The final height of the material after the compaction cycle is calculated at Point D, $\Delta h_{0.5\ \mathrm{MPa}}$, and used to obtain the volume fraction at the given stress amplitude. The sequence is then repeated for increasing values of the mean stress at 1 MPa ($D \rightarrow E \rightarrow F \rightarrow G$) and the correspondingly larger amplitudes of oscillation lead to a new height and packing density $\Delta h_{1\ \mathrm{MPa}}$ for the new oscillatory stress.

The dynamics of compaction is well described by a logarithmic dependence of $\Delta h(t)$ for each compaction cycle valid for long times:

$$\Delta h(t) \sim -\ln(t). \tag{1}$$

At short times, it crosses over to a faster decay. This implies that the density behaves as $\sim 1/\ln(t)$ at long times in agreement with previous experiments [2]. This is shown in Fig. 1 as the solid line from B to C and from E to F. The relaxation is so slow that one could argue that the final steady-state density has not been achieved on the time scale of the experiment.

The volume fraction after each compaction cycle is plotted against the amplitude of oscillatory stress in Fig. 2. The protocol involves a stepwise ramp of compaction cycles from zero stress amplitude to 4 MPa, then back to zero, and back again to 4 MPa. The figure shows an irreversible branch as the mean stress is first increased, until a plateau in the volume fraction is reached. As the stress is then reduced the system enters into the reversible branch of compaction. By ramping up and down in oscillatory stress we find that the system reproduces the same curve. These are the reversible jammed states that we use as the reference states in the stress relaxation experiments.

Stress relaxation experiments (strain-controlled).—It is important to distinguish between processes related to large scale deformations in granular compaction studied above and infinitesimal perturbations related to supporting the stress once the volumetric conditions have been satisfied. In the latter, an application of an external stress will result in dissipation mechanisms quite different from the compaction process. Therefore, we next probe the mechanisms of energy dissipation of the fully compacted system by performing infinitesimal step compression experiments and observing the resulting response in the stress.

We perform uniaxial compression tests with the plate-cup configuration used above. We contract the system by applying a step strain $\Delta\epsilon$ in the range $(1\text{–}3) \times 10^{-3}$ to the glass bead sample at a given confining pressure. Meanwhile, we monitor the temporal evolution of the differential stress, $\Delta\sigma(t)$. It is defined as the difference between the stress at time t measured after the strain is applied and the stress before the perturbation is applied. It follows that $\Delta\sigma(0)$ is the change in stress between the baseline (stress before perturbation) and just after the perturbation is applied [8].

Figure 3 shows the resulting relaxation at different confining pressures from 0.1 to 5 MPa. At the slowest strain rate of $\Delta\dot{\epsilon} = 6.6 \times 10^{-5}\ \mathrm{s}^{-1}$ we find a relaxation which is logarithmic in time, at long times. When we repeat the experiment at the maximum speed allowed by the press, $\Delta\dot{\epsilon} = 9.8 \times 10^{-3}\ \mathrm{s}^{-1}$, we are able to observe the "instantaneous" stress response in addition to the subsequent relaxation. In this case we find a two-step relaxation, which is well approximated with the following equation, plotted in Fig. 3 for the 4 MPa stress relaxation:

$$\Delta\sigma(t)/\Delta\sigma(0) = A + Be^{-t/\tau_1} - C\ln(t), \tag{2}$$

where $\tau_1 = 1.4$ s is the fast relaxation time and $A = 0.9$, $B = 0.09$ are constants, and $C = 2 \times 10^{-3}$ sets the rate of

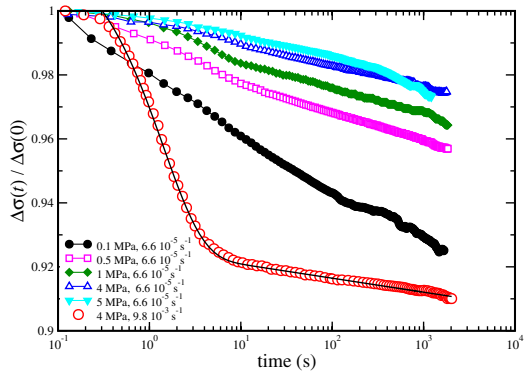

FIG. 3 (color online). Experiments: stress relaxations at different confining pressures. Results are shown for two strain rates. The fastest one reveals the two-step relaxation.

the slow relaxation. Since the slow strain rate corresponds to a straining time of $T = 21.6$ s and the fast rate to $T = 0.18$ s for the case of the 4 MPa sample, the fast relaxation is only observed in the latter experiment, as the relaxation time τ_1 is slower than the application of the strain.

We argue, by analogy with glassy dynamics, that the fast relaxation is a single particle relaxation mechanism whereas the slow is representative of a collective rearrangement of many grains via sliding and their ensuing "aging" properties. We provide supporting evidence for this claim by means of molecular dynamic simulations.

Computer simulations.—In order to decipher the main microscopic mechanism, we perform a numerical study based on molecular dynamics of elasto-frictional spherical particles.

Interparticle forces are computed using the principles of contact mechanics and consider normal Hertz forces F_n, tangential Mindlin forces F_t, and dry Coulomb friction $F_t \leq \mu F_n$, with μ the friction coefficient. Full details are given in [9].

We provide two principal mechanisms of dissipation. If the grains are touching, they exert *contact* damping forces, which arise from viscoelastic dissipation between the grains: $f_n^{\text{diss}} = -\gamma_n \xi^{1/2} \dot{\xi}$ and $f_t^{\text{diss}} = -\gamma_t \xi^{1/2} \dot{s}$ [10]. Reference values for the damping constants γ_n and γ_t can be obtained from [11]. Furthermore, the grains are immersed in a viscous fluid, such as air or water, which causes *global* damping according to the classical Rayleigh theory. The drag of a sphere immersed in a viscous fluid is $F^{\text{drag}} = 6\pi\eta R\dot{\mathbf{x}}$, where η is the viscosity of the fluid (an analogous expression holds for torque damping) [11].

The packings were equilibrated at a given pressure of 1 MPa according to the previously established method [9]. We probe the macroscopic mechanical properties by applying an infinitesimal step strain, and we monitor the time dependence of the stress, thus mimicking the experiments.

We find a critical damping (both for global and contact), above which the slow relaxation ensues. The critical values are $\gamma_n^c = 2 \times 10^{-3}$ kg/s $m^{1/2}$, $\gamma_t^c = 2 \times 10^{-4}$ kg/s $m^{1/2}$, while the critical global viscosity is $\eta^c = 1.7 \times 10^{-4}$ Pa s [12]. In Fig. 4(a) we show typical relaxation curves obtained for three systems: with global damping only ($\eta > \eta^c$), or with contact damping only ($\gamma_n > \gamma_n^c$, $\gamma_t > \gamma_t^c$), or with critical contact damping ($\gamma_n = \gamma_n^c$, $\gamma_t = \gamma_t^c$), which dissipates energy mainly via Coulomb friction. We see that the curves with damping larger than the critical value are in good qualitative agreement with those from the experiment, displaying slow stress relaxation at long times, while the curve with mostly frictional dissipation decays very fast. Comparing with real viscoelastic constants for common glass materials [11] and the viscosity of air or water, we find that real viscoelastic damping is almost always above the critical values. Thus, in most experimental situations, except perhaps in vacuum, the system will be overdamped and the slow relaxation will be observed.

Considering that we have the full information on the motion of the particles, we now investigate the microscopic

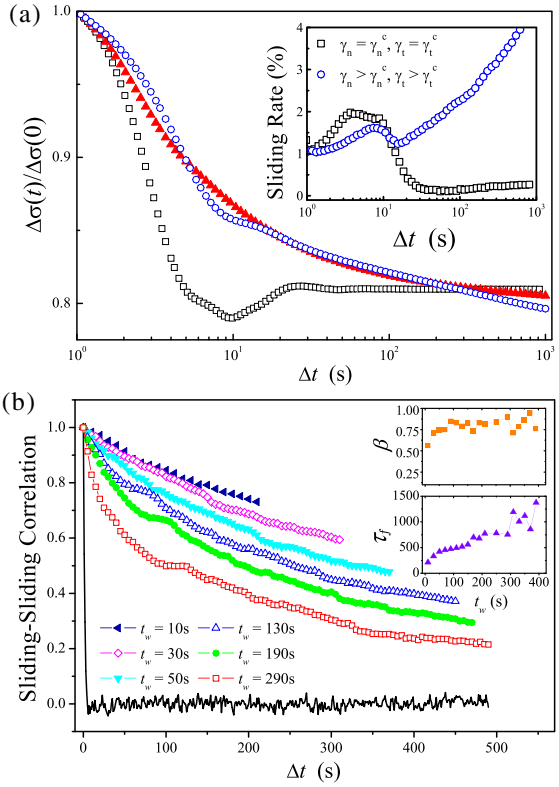

FIG. 4 (color online). Simulations: (a) stress relaxation after an instantaneous step strain at 1 MPa. Solid triangles (red online) correspond to a system with global damping $\eta = 1.7 \times 10^{-3}$ Pa s $> \eta^c$, open circles (blue online, also in the inset) to an overdamped system with contact damping $\gamma_n = 2 \times 10^{-2}$ kg/s $m^{1/2}$, $\gamma_t = 2 \times 10^{-3}$ kg/s $m^{1/2}$, and black open squares (also in the inset) to a system with critical contact damping. The inset shows the behavior of the fraction of the particles sliding in the aging regime and at the critical point. (b) Sliding-sliding correlation function showing the appearance of aging (dependence on t_w) above the critical damping. The black line show the behavior below critical damping. Insets show the stretched exponential exponent β and the characteristic relaxation time τ_f versus t_w.

origin of the slow relaxation. By monitoring the small displacements of the particles we find that the shear displacements are key to explaining the relaxation mechanism. The inset of Fig. 4(a) shows the fraction of particles that are sliding ($F_t = \mu F_n$) in the system at a given time (for these particles the shear displacement is significant). If the system is underdamped ($\gamma < \gamma^c$, for both normal and shear), this fraction decays rapidly to zero and the stress is quickly relaxed. On the other hand, when the system is overdamped ($\gamma > \gamma^c$) the fraction of sliding particles increases as a function of time, analogous to the strengthening behavior found in previous experiments [7]. This slow dynamical strengthening (which may saturate at larger times) is responsible for the slow stress relaxation.

To further investigate the features of the overdamped state we study the time correlation function of the sliding grains to quantify their motion. At a given time step we construct a state vector $\vec{s}(t)$ with M components (M is the number of contacts in the system), the ith component being 1 or -1 according to whether the ith contact is sliding or not. We consider a time correlation function $C(t, t_w) = \langle \vec{s}(t + t_w)\vec{s}(t_w)\rangle$ with a dependence on the waiting time, t_w, measured from the time when the perturbation is applied. Below the critical damping condition we find [Fig. 4(b)] that the $C(t, t_w)$ decays rapidly with no evidence of t_w dependence. With small damping forces the grains in the system have no time to develop significant shear displacements. The number of sliding particles is very small and the system does not display any slow relaxation.

In contrast, above critical damping we find [Fig. 4(b)] a stretched exponential decay $C(t, t_w) \sim \exp[-t/\tau_f(t_w)]^\beta$, where $\tau_f(t_w)$ is the characteristic time dependent on the waiting time and $\beta \approx 0.8$ is the critical exponent [see insets of Fig. 4(b)]. Large damping sufficiently slows down the system such that the grains have enough time to develop substantial tangential displacements, which in turn generate shear forces large enough to cause sliding of the grains. Interestingly, the same effect has also been found in the history-dependent behavior of packings near the jamming transition, in this case, as a function of the compression rate [13]. The fact that both damping and compression can drive the system into a glassy phase through a well-defined transition, which exhibits aging, carries important implications.

Whereas the transition between an underdamped and overdamped state is not surprising, it is intriguing that the latter state shows signatures of glassy behavior. Conventional glassy systems such as polymer melts undergo the glass transition by fast cooling of the system, while here we show that in granular athermal systems the damping plays a similar role to temperature. A granular ''glass transition'' driven by damping opens interesting unifications between the two types of systems, the details of which we plan to test experimentally in further work. Since damping facilitates dissipation by friction as the time scale of grain contacts is prolonged, we predict that a packing of frictionless droplets in emulsions would not experience the observed slow relaxation and aging dynamics. In fact our simulations indicate that a system with frictionless particles, $\mu = 0$, as well as a system of infinitely rough particles, $\mu \to \infty$, does not display aging, thus confirming that the glassy properties are due to the finite friction coefficient of grains.

In summary, in this work we distinguish between stress relaxation processes related to reaching a granular jammed equilibrium state and the remaining infinitesimal relaxation. Once the system is jammed at a given pressure, the stress relaxation is characterized by a fast exponential relaxation followed by a glassy slow logarithmic decay. The glassy phase is characterized by the aging of the sliding correlation function and the ensuing stretched exponential behavior, which are in turn responsible for the slowdown of the dynamics. It is interesting to note that the amplification of the sliding could be associated with the initiation of an avalanche inside the system.

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