Memory Effects in Granular Material

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We present a combined experimental and theoretical study of memory effects in vibration-induced compaction of granular materials. In particular, the response of the system to an abrupt change in shaking intensity is measured. At short times after the perturbation a granular analog of aging in glasses is observed. Using a simple two-state model, we are able to explain this short-time response. We also discuss the possibility for the system to obey an approximate pseudo-fluctuation-dissipation theorem relationship and relate our work to earlier experimental and theoretical studies of the problem.

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Granular materials comprise an important class of complex systems whose simple fundamental mechanics gives rise to rich macroscopic phenomenology [1]. Recent experiments on granular compaction [2,3] suggest they are an ideal system for studying jamming, a phenomenon lying outside the domain of conventional statistical physics, yet highly reminiscent of glassiness. These studies showed that a loose packing of glass beads subjected to vertical "tapping" slowly compacts, asymptoting to a higher steady state packing fraction. This "equilibrium" packing fraction is somewhat lower than the random close packing limit, $\rho_{\rm rcp} \approx 0.64$, and is a decreasing function of the vibration intensity, typically parameterized by Γ, the peak applied acceleration normalized by gravity, g. The relaxation dynamics are extremely slow, taking many thousands of taps for the packing fraction, ρ, to approach its steady state value. During this evolution, ρ increases logarithmically with the number of taps, t, which is typical for self–inhibiting processes [4]. The average time scale τ of the relaxation decreases with Γ, and in this sense the shaking intensity plays, at least qualitatively, the role of temperature. For small Γ, the relaxation rate becomes so slow that the system cannot reach the steady state density within the experimental time scale. It was also found that compaction can be maximized through an annealing procedure. This process involves a slow "cooling" of the system starting from a high shaking intensity Γ. Another qualitative similarity to glasses is observable in the density fluctuation spectrum of the granular system near its steady-state density. The spectrum was found to be strongly non-Lorentzian [3], revealing the existence of multiple time scales in the system. The shortest and the longest relaxation timescales differ by as much as three order of magnitude, and the behavior of the spectrum for the intermediate frequencies is highly non-trivial; in certain regimes it can be fitted with a power law.

These previous experimental observations are suggestive of glassy behavior and this connection has been explored in recent models of compaction using ideas from magnetic systems [5]. However, a more direct test of the glassy nature of granular compaction comes from measurements of the response of the system to sudden perturbations of the effective temperature, given by Γ. This idea originates from classical experiments for the study of aging in glasses [6] and has recently been explored using computer simulations [7]. In this letter we present direct experimental observations of memory effects in a vibrated granular system obtained by measuring the short-time response to an instantaneous change in tapping acceleration Γ and propose a simple theoretical framework.

We used the experimental set-up described in refs. [2,3]: 1 mm–diameter glass beads were vertically shaken in a tall, evacuated, 19 mm-diameter glass tube, and the packing density of the beads was measured using capacitors mounted at four heights along the column.

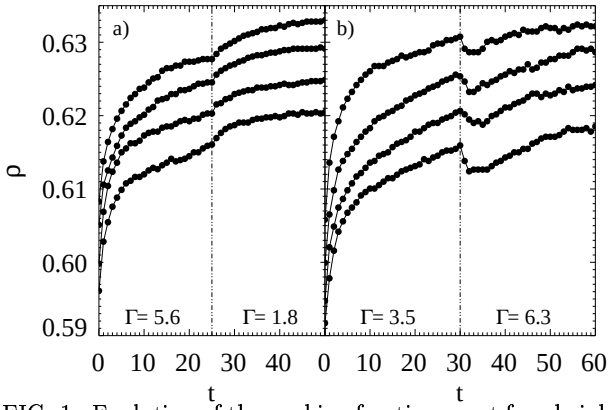

FIG. 1. Evolution of the packing fraction, ρ, at four heights in the column, as a function of tap number, t. Two different single-switch experiments: (a) Γ was lowered from 5.6 to 1.8 at $t_0 = 25$; and (b) Γ was increased from 3.5 to 6.3 at $t_0 = 30$. Curves are shifted vertically for clarity. Each curve is an average over 4 runs, and the measurement uncertainty in ρ is 4×10^{-4}.

The simplest form of this experiment consists of a single instantaneous change of vibration intensity from Γ_1 to Γ_2 after t_0 taps. For $\Gamma_2 < \Gamma_1$ (Fig. 1a) we found that on short time scales the compaction rate increases. This is in sharp contrast to what one may expect from the

long-time behavior found in previous experiments where the relaxation is slower for smaller vibration accelerations. For $\Gamma_2 > \Gamma_1$ (Fig. 1b) we found that the system dilates immediately following t_0. These results too, are opposite from the long-time behavior seen in previous experiments where the compaction rate increased: not only does the compaction rate decrease, it becomes negative (i.e. the system dilates). Note that after several taps the "anomalous" dilation ceases and there is a crossover to the "normal" behavior, with the relaxation rate becoming the same as in constant–Γ mode.

These data constitute a short-term memory effect: the future evolution of ρ after time t_0 depends not only on $\rho(t_0)$, but also on information about the previous tapping history, contained in other "hidden" variables. In order to demonstrate this in a more explicit manner, we modified the above experiment. In this second set of three experiments the systems was driven to *the same density* ρ_0 with three different accelerations Γ_0, Γ_1, and Γ_2. After ρ_0 was achieved at time t_0, the system was tapped with *the same intensity* Γ_0 for all three experiments. As seen in Figure 2, the evolution for $t > t_0$ strongly depends on the pre-history. The need for extra state variables in the problem is consistent with strongly non-Lorentzian behavior of the fluctuation spectrum, observed in earlier experiments [3].

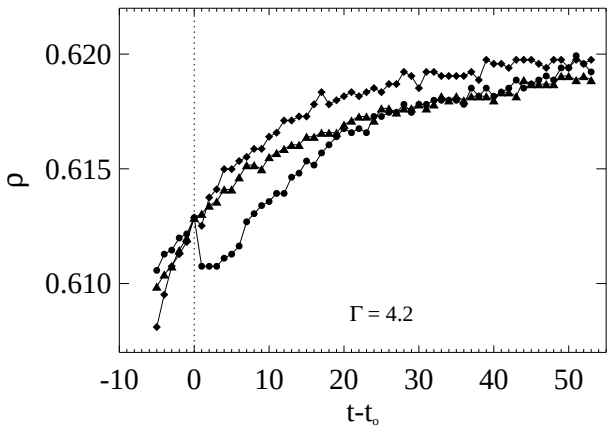

FIG. 2. The time evolution of packing fraction ρ for a system which was compacted to $\rho_0 = 0.613$ at time t_0 using three different accelerations: $\Gamma_1 = 1.8$ (circles), $\Gamma_0 = 4.2$ (triangles), and $\Gamma_2 = 6.3$ (diamonds). After the density ρ_0 was achieved, the system was vibrated at acceleration Γ_0. The evolution for $t > t_0$ depended strongly on the pre-history. Each curve is an average over four experimental runs.

To give a theoretical interpretation of the above results, we view the problem as an evolution in the space of discrete "microscopic" states corresponding to different realizations of the packing topology (in addition to the topological changes, there are continuous deformations of the network, which we assume to relax on the time scale of a single tap [8]). For each tap there is a possibility for a transition from one microscopic state to another. Since the dynamics is dissipative and the system is under external gravity, a transition to a denser configuration is typically more probable than the reverse one. We now introduce the concept of a *Baseline Configuration* (BC), which plays the role of a local free energy minima for our non–thermal system. Namely, a BC may be defined as a state where any transition to a different configuration has a lower probability than the reverse one. Hence, there is a mesoscopic time scale on which the system gets trapped in the vicinity of a given BC, and its evolution is dominated by a number of flip–flop modes, i.e. local "excitations" of the baseline structure, any of which would normally relax back to the same BC.

Neglecting the coupling between individual flip–flop modes, we may replace the complicated configuration space with a set of independent two-state systems, each of which is characterized by two transition rates, $\kappa_{e\to g} > \kappa_{g\to e}$. $\kappa_{e\to g}/\kappa_{g\to e}$ gives the ratio of the equilibrium probabilities of populating each state: "ground" and "excited" (with BC corresponding to all modes at their ground state). As we have argued, the higher probability ground state is typically the one with higher density, i.e. the volume change v between the ground and the excited states is normally positive. Our no-coupling approximation is close in its spirit to a number of two-state models recently proposed by several reserch groups [9].

Obviously, the experimentally–observed density is different from that of the current BC, ρ_b, due to a non-zero fraction of excited modes:

$$\rho = \rho_b(t)\left(1 - \frac{1}{V}\sum_n v^{(n)}\left(1 + \frac{\kappa^{(n)}_{e\to g}}{\kappa^{(n)}_{g\to e}}\right)^{-1}\right). \qquad (1)$$

The summation here is performed over all the flip–flop modes of a given BC, V is the total volume, and $v^{(n)}$ is the volume difference between the excited and the ground states of the n-th mode. Assuming that the vibration intensity Γ is a qualitative analog of temperature, we expect the population of the excited states, $P(\Gamma) = (1 + \kappa^{(n)}_{g\to e}/\kappa^{(n)}_{e\to g})^{-1}$, to grow with Γ, starting from zero at $\Gamma = 0$. Hence, for a given ρ_b, the total density ρ will be lower at higher acceleration. This explains the observed anomalous compaction following an abrupt change of Γ. After a switch from Γ_1 to Γ_2 at time $t_0 = 0$, the flip-flop mode contribution to the total density, $G_{\Gamma_1,\Gamma_2}(t)$, would relax to its new equilibrium value in the following way:

$$G_{\Gamma_1,\Gamma_2}(t) = \rho_b \int_0^{\kappa_{\max}} F_{\Gamma_1,\Gamma_2}(v,\kappa)\,(1 - \exp(-\kappa t))\,dv d\kappa \qquad (2)$$

Here κ is the relaxation rate of an individual mode, and the distribution function $F_{\Gamma_1,\Gamma_2}(v,\kappa)$ is introduced as follows:

$$F_{\Gamma_1,\Gamma_2}(v,\kappa) \equiv \frac{1}{V}\sum_n \left(P^{(n)}(\Gamma_2) - P^{(n)}(\Gamma_1)\right)\delta(v - v^{(n)})$$

$$\delta\left(\kappa - \kappa^{(n)}_{g\to e}(\Gamma_2) - \kappa^{(n)}_{e\to g}(\Gamma_2)\right). \quad (3)$$

The distribution function is normalized so that $\int F_{\Gamma_1,\Gamma_2}(v,\kappa))dvd\kappa = \rho^*(\Gamma_2) - \rho^*(\Gamma_1)$, where $\rho^*(\Gamma)$ is the equilibrium number density of the excited modes at given Γ. One can see from Eq. (1) that $\rho = \rho_b(1-\langle v\rangle\rho^*)$, i.e. since $\langle v\rangle$ is expected to be of the order of a single particle volume, ρ^* is of the order of the flip-flop correction to the total density. The observed amplitude of the density changes in our experiments imply that ρ^* is normally less than 1% of the particle density, and thus one can estimate the typical separation between neighboring flip-flop systems as $(\rho/\rho^*)^{1/3} \sim 5$ particle sizes, which is a good justification for our no-coupling approximation. According to Eq. (2), if F_{Γ_1,Γ_2} does not vanish in the limit $\kappa \to 0$, the late stage of the relaxation of $G_{\Gamma_1,\Gamma_2}(t)$ is given by the power law:

$$G_{\Gamma_1,\Gamma_2}(t) = G_{\Gamma_1,\Gamma_2}(\infty) - \frac{\text{const}}{t}. \quad (4)$$

Note that ρ_b is also dependent on time: although this cannot be described within our two-state approximation, the collection of elementary modes slowly evolves. Thus, one can observe two different processes: on short (in fact, mesoscopic) time scales, a fast relaxation due to the flip–flop modes is dominant, while over the long times, the dynamics are determined by the logarithmically slow evolution of the baseline density $\rho_b(t)$. The crossover between the two regimes is particularly obvious in Fig. 1b, where it results in a non-monotonic evolution. Such dynamics is unusual in spin glasses, but has been observed for conventional glasses [10]. For experiments performed at sufficiently late stages of the density relaxation, the dynamics of the baseline density could be neglected compared to the contribution of the flip-flop modes (note that what we call a late-stage relaxation corresponds in fact to mesoscopic time scales which are always shorter than the relaxation time for ρ_b). It has to be emphasized that the described experiments provide us with a tool for study of the response of the system, which is *not limited* to the nearly–equilibrium regime.

One can use our simple model to predict the response of the system to a more complicated pattern of changes of Γ. First, we reach, using annealing dynamics, a "quasi-steady" state at amplitude Γ_0, so that one can consider ρ_b constant later on. Let us switch the shaking acceleration from Γ_0 to Γ_1 for a finite number of taps δt, and then switch it back to Γ_0. During the intermediate Γ_1–stage, the system does not have enough time to completely relax to its new equilibrium. In our two-state model, the modes whose relaxation rate (at Γ_1) is below δt^{-1} remain unrelaxed. Assuming that the slow modes at Γ_1 are mostly the same as at Γ_0, we can calculate the backward density relaxation similarly to Eq. (4), with $F(v,\kappa)$ effectively depleted below a minimal rate, κ_0. This cut-off frequency, κ_0, is expected to decrease monotonically with increasing perturbation duration δt. In the spirit of spin glass theories, we can characterize the density relaxation after returning to Γ_0 by the "aging" response function which now depends both on t and waiting time δt. Eq. (2) gives the following form for its late–stage behavior

$$G_{\Gamma_1,\Gamma_0}(t,\delta t) = G_{\Gamma_1,\Gamma_0}(\infty) - \text{const}\left(\kappa_0 + \frac{\exp(-\kappa_0 t)}{t}\right). \quad (5)$$

We tested the above predictions by performing this three stage experiment, varying the duration, δt, of the perturbation(Γ_1) stage (Fig. 3). As predicted, the time needed to recover the steady-state density increases with the number of taps δt spent in the "hot" regime $\Gamma_1 > \Gamma_0$. In the coordinates chosen, the relaxation curves should follow the $\delta t = \infty$ dynamics until the saturation at the cut-off time, $\kappa_0^{-1}(\delta t)$. We approximate the distribution function F by a constant above this low frequency cut-off at $\kappa_0^{-1}(\delta t)$, up to a high-frequency cut-off, $\kappa_{\max} \simeq 1$. This eliminates the unphysical low-t divergence in Eq. (5). Figure 3 shows fits of the data to Eq. (2), where $\kappa_0(\delta t)$ is determined from the fit. The best-fit is achieved at $\kappa_{\max} = 0.4$, and the variation of this parameter would result in a simple rescaling of the time axis.

Figure 3 demonstrates good agreement between model and experiment, with some systematic error at the earliest relaxation stage (which is an expected result of our oversimplified description of the short time dynamics). For the late stage relaxation, we conclude that (i) within our experimental precision, the $\delta t = \infty$ relaxation is consistent with the predicted $1/t$ law; (ii) finite–δt relaxation curves can be parameterized by a low frequency cut-off, κ_0; and (iii) κ_0 is a decreasing function of the waiting time δt, shown in the insert of the Figure 3. As discussed earlier, the wide range of relaxation times reveals itself both in our response measurements and in the the fluctuation spectra of the density. It is tempting to relate these two kinds of data through an analog of a Fluctuation-Dissipation Theorem (FDT). Of course, there is no fundamental reason for FDT to be applicable to the granular system [11]. Even though the above two-state model could be mapped onto a thermal system (in which FDT is expected to work), the thermodynamic variable conjugate to density in the context of such a mapping has no clear physical meaning. Nevertheless, we can obtain a pseudo-FDT relationship for the granular system if we neglect the correlation between the volume change v and the life time κ^{-1} of an individual mode, i.e. assume $F_{0,\Gamma}(v,\kappa) = f(\kappa)g(v)$. Then the density autocorrelation function can be written as follows:

$$\langle\delta\rho(0)\delta\rho(t)\rangle_\Gamma \simeq \frac{\rho^2}{2V}\int\langle v^2\rangle\exp(-\kappa t)f(\kappa)d\kappa = \frac{\rho\langle v^2\rangle}{2V\langle v\rangle}\left(G_{0,\Gamma}(t) - G_{0,\Gamma}(\infty)\right). \quad (6)$$

Thus, the density correlator is simply proportional to the response function corresponding to the switch between a very low acceleration (at which virtually all the modes are in their ground states) and the given one, Γ. An experimental check of this relationship requires further high-precision studies of both the relaxation dynamics and the fluctuation spectrum.

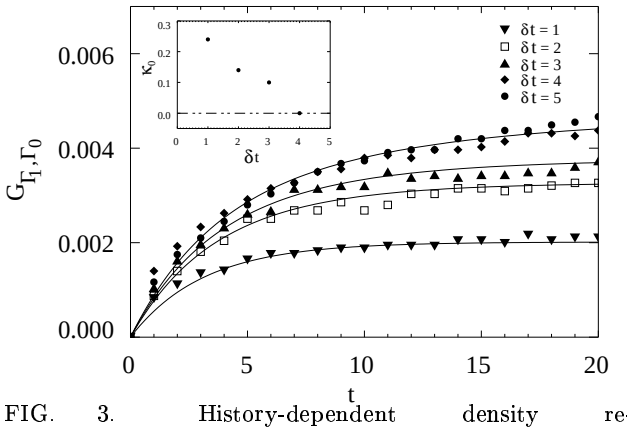

FIG. 3. History-dependent density relaxation ($G_{\Gamma_1,\Gamma_0}(t,\delta t)$) of the system, prepared by tapping for a long time at $\Gamma_0 = 1.8$ and then tapping for a variable number, δt, of taps at a "hotter" intensity $\Gamma_1 = 4.2$ before being returned to Γ_0 at time t_1. The solid lines represent the theoretical curves, with appropriate values of the parameter κ_0. The dependence of the cut-off rate κ_0 on the waiting time δt is shown at the insert for $\delta t \leq 4$ taps. We do not show the value for $\delta t = 8$ since we found it null within the error bar, as for $\delta t = 4$. Each experimental graph is an average of 12 runs.

Our model also gives a simple interpretation to the decreasing dependence of the steady-state density on Γ: it can be attributed to the growth of the population of the excited states, $P(\Gamma)$. Indeed, the corresponding correction to the total density is about 1%, i.e. of the same order as the variation of the equilibrium packing fraction with Γ [3]. The slow dynamics associated with the evolution of the baseline density can also be addressed within our approach. To do so we need to consider the excitation–assisted transitions between different BC (which in turn results in the change of the set of available flip-flop modes). In introducing the coupling between individual modes, it is a reasonable assumption that the relaxation of one mode to its ground state may frustrate such a transition for some of its neighbors (e.g. in 3D the most compact local cluster can be created only at the expense of less dense neighboring regions). Thus, we arrive at an effective anti-ferromagnetic (AF) coupling (of an infinite strength) between the flip-flop modes.

This extension of our model makes it remarkably similar to the so-called reversible Parking Lot Model (PLM) [12], which has been successful in describing many aspects of granular compaction experiments [2,3]. A mutual frustration of individual modes is also a key ingredient of the "tetris model" (TM), another fruitful approach for modelling the dynamics of the system [6,5,7]. PLM, TM and our flip-flop model with AF coupling all appear to belong to the same *generic* class of frustrated spin systems. Indeed, numerical simulations we performed on the PLM display the same memory effects discussed above, and similar behavior is also observed in the TM [13].

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[1] H. Jaeger, S. Nagel and R. Behringer, *Rev. Mod. Phys.* **68**, 1259 (1996).
[2] J.B. Knight, C.G. Fandrich, C.N. Lau, H.M. Jaeger and S.R. Nagel, *Phys. Rev. E* **51**, 3957 (1995).
[3] E.R. Nowak, J.B. Knight, E. Ben-Naim, H.M. Jaeger and S.R. Nagel,*Phys. Rev. E* **57**, 1971 (1998).
[4] T. Boutreux and P. G. de Gennes, *Physics A* **244**, 59 (1997).
[5] E. Caglioti, V. Loreto, H. J. Herrmann, and M. Nicodemi, *Phys. Rev. Lett.* **79**, 1575 (1997).
[6] E. Vincent, J. Hammann and M. Ocio, in *Recent progress in random magnets*, World Scientific (1992); J.-P. Bouchaud, L.F. Cugliandolo, J. Kurchan and M. Mézard, in *Spin Glasses and Random Field*, World Scientific (1997).
[7] M. Nicodemi, *Phys. Rev. Lett.* **82** 3734 (1999); A. Barrat and V. Loreto, J. Phys. A *in press* (2000).
[8] G.C. Barker and A. Mehta, *Phys. Rev. A* **45**, 3435 (1992).
[9] D. Grinev and S.F. Edwards, *Phys. Rev. E* **58**,4758 (1998); P.G. de Gennes, preprint(1999); A. Mehta, and G. C. Barker, *Europys. Lett.* **27** 501 (1994); S. Krishnamurthy, V. Loreto and S. Roux, *Phys. Rev. Lett.* **84** 1039 (2000).
[10] F. Alberici-Kious, J.P. Bouchaud, L.F. Cugliandolo, P. Doussineau and A. Levelut, *Phys. Rev. Lett.* **81** 4987 (1998).
[11] There are, however, several recent approaches that formulate FDT relationships for non-equilibrium glassy systems: L. F.Cugliandolo and J. Kurchan *Phys. Rev. Lett.* **71**, 173 (1993).
[12] P.L. Krapivsky and E. Ben-Naim, *J. Chem. Phys.* **100**, 6778 (1994); A. L. Kolan, E. R. Nowak, and A. V. Tkachenko, *Phys. Rev. E* **59**, 3094 (1999).
[13] A. Tkachenko and C. Josserand, (in preparation); A. Barrat, private communication.
[14] M. Nicolos, P. Duru and O. Pouliquen, preprint **cond-mat/0006252**.