Aging in Out-of-Equilibrium Dynamics of Models for Granular Media

Mario Nicodemi and Antonio Coniglio
Dipartimento di Fisica, Universitá di Napoli "Federico II," Unità INFM and INFN Napoli, Mostra d'Oltremare, Padiglione 19, 80125 Napoli, Italy
(Received 11 March 1998)

In the framework of recently introduced frustrated lattice gas models, we study the out-of-equilibrium dynamical processes during the compaction in granular media. We find irreversible-reversible cycles in agreement with recent experimental observations, along which we can individuate an equivalent "glass transition," Γ_g. In analogy with the phenomenology of glassy systems we find aging effects during the compaction process. In particular, we find that the two-time density correlation function $C(t,t')$ asymptotically scales as a function of the single variable $\ln(t')/\ln(t)$. [S0031-9007(98)08224-6]

PACS numbers: 45.70.Mg, 05.50.+q, 75.10.Nr, 81.40.Cd

The experimental study of dynamic processes in granular media [1] has recently revealed the presence of interesting behaviors. Under tapping, dry granular media reach very slowly a more compact state which is well fitted by a logarithmic relaxation [2]. More recently, Novak *et al.* [3] have also shown that such materials exhibit nontrivial irreversible-reversible cycles. These phenomena stem from slow relaxation processes due to large "cooperative rearrangements" of many particles. In such a perspective, granular materials share features of thermal systems, such as glasses or spin glasses, which are also characterized by extremely long relaxation times and the presence of irreversible-reversible cycles [4,5].

In this paper, in the framework of recently introduced microscopic models [6–8], we reproduce the irreversible-reversible cycles of Novak *et al.* and investigate the effect of the "cooling" rate on the compaction process. We find a behavior which is strongly reminiscent of the phenomenology of the glass transition. Finally, we study the nonequilibrium time-dependent density-density autocorrelation function and find aging effects typical of glassy systems. These results suggest that similar effects could also be found in real experiments.

In dynamical processes of granular media a crucial role is played by geometric frustration (originated by steric hindrance between interlocked neighboring grains) which induces the necessity of large scale cooperative rearrangements for relaxation. Based on these concepts, two types of frustrated lattice gas models were introduced [6–8]. Both models showed logarithmic compaction, segregation, and other phenomena typical of granular media under shaking.

These models consist of a system of particles which occupy the sites of a square lattice tilted by 45° (see Fig. 1). Particles are characterized by an internal degree of freedom, $S_i = \pm 1$, corresponding, for instance, to two typical orientations of grains on the lattice. Two nearest-neighbor sites can be occupied only if the particles have the right reciprocal orientation, so that they do not overlap, otherwise, due to excluded volume, they have to move away. In the absence of vibrations the particles are subject only to gravity and as they move downwards they always fulfill the nonoverlap constraint. The effect of vibration is introduced by allowing the particles to diffuse with a probability $p_{\rm up}$ upwards and a probability $p_{\rm down} = 1 - p_{\rm up}$ downwards. An important parameter governing the dynamics is the adimensional parameter $\Gamma \equiv -1/\ln(x_0)$, with $x_0 = p_{\rm up}/p_{\rm down}$, which is related (see below) to the effective temperature of the system and, consequently, plays the same role as the amplitude of the vibration intensity in the experiment of Novak *et al.* [3].

Such models can be described in terms of the following lattice gas Hamiltonian (see [7,8]) in the limit $J \to \infty$:

$$H = J \sum_{\langle ij \rangle} f_{ij}(S_i, S_j) n_i n_j . \quad (1)$$

Here $n_i = 0, 1$ are occupancy variables, $S_i = \pm 1$ are spin variables associated with the two orientations of the particles, J represents the infinite repulsion felt by the particles when they have the wrong orientations, and $f_{ij}(S_i, S_j) = 0$ or 1 depending whether the configuration S_i, S_j is right (allowed) or wrong (not allowed).

The choice of $f_{ij}(S_i, S_j)$ depends on the particular model. The Tetris [8] model is made of elongated particles (see Fig. 1), which may point in two directions coinciding with the two lattice bond orientations. In this case

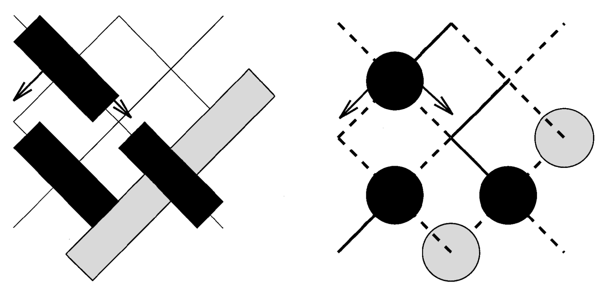

FIG. 1. A schematic picture of the two types of frustrated lattice gas models described in the text. Left: The Tetris model. Right: The Ising frustrated lattice gas. Solid and dashed lines represent the two types of interactions $\epsilon_{ij} = \pm 1$. Filled circles are the present particles with "orientation" $S_i = \pm 1$ (black/white).

 0031-9007/99/82(5)/916(4)$15.00

$f_{ij}(S_i, S_j)$ is given by $f_{ij}(S_i, S_j) = 1/2[S_i S_j - \epsilon_{ij}(S_i + S_j) + 1]$, where $\epsilon_{ij} = +1$ for bonds along one direction of the lattice and $\epsilon_{ij} = -1$ for bonds on the other. This Hamiltonian model has an ordered "antiferromagnetic" ground state, and its dynamics has the crucial constraint that particles can flip their "spin" only if three of their own neighbors are empty.

A real granular system may contain more disorder due to a wider shape distribution or to the absence of a lattice. Therefore in a more realistic model the number of internal states is $q > 2$ ($S_i = 1, 2, \ldots, q$) and the function $f_{ij}(S_i, S_j)$ is zero only for allowed nearest-neighbor configurations.

However, to simplify the model, the number of states was still kept at $q = 2$ and the randomness was taken into account by introducing random quenched variables corresponding to the freezing of some degree of freedom in the high density regime. Thus an Ising frustrated lattice gas model (IFLG) was proposed [7] in which $f_{ij}(S_i, S_j)$ was given by $f_{ij}(S_i, S_j) = 1/2(\epsilon_{ij} S_i S_j - 1)$, and $\epsilon_{ij} = \pm 1$ are quenched random interactions associated with the bonds of the lattice.

The Hamiltonian (1) is without gravity. In the presence of gravity there is an extra term $g \sum_i n_i y_i$, where g is the gravity and y_i is the ordinate of the particle i. The temperature T is related to the ratio $x_0 = p_{\mathrm{up}}/p_{\mathrm{down}}$ via $e^{-2g/T} = x_0$ (notice, $\Gamma = T/2g$).

Interesting enough, the two extreme models, the Tetris and the IFLG, show similar behavior. This suggests that the results found are rather robust and will not depend much on the details of the model. In particular, under tapping they reproduce the logarithmic behavior in agreement with the experimental results of Knight *et al.* [2]. Experimentally a "tap" is the shaking of the container of the grains by vibrations of given duration and amplitude. In our Monte Carlo simulations each single tap is realized by letting the particle diffuse under the gravity by keeping $\Gamma =$ const during the time interval of a tap τ_0 and then switching off the vibration by setting $\Gamma = 0$ until the system reaches a static configuration. Time t is measured in such a way that one unit corresponds to one single average update of all particles and spins of the lattice.

In a previous paper [7] we have performed on the IFLG a particular cycle sequence of taps to show the presence in granular media of this hysteresis effect. Recently, real experiments were also performed on density relaxation in granular media under cyclic tapping [3], which showed indeed the presence of such a hysteresis effect. In order to compare better with the experimental data, we have simulated the same cycle of Novak *et al.* on the IFLG model. We have considered a system of size 30×60 (our data are robust to size changes), with periodic boundary conditions along the x axis and rigid walls at the bottom and top. Our data are averaged over 8 different lattice realizations, each averaged over 30 different noise realizations.

A starting particle configuration is prepared by randomly inserting particles into the box from its top and then letting them fall down, with the dynamics described above, until the box is filled. We performed cycles of taps in which the vibration amplitude Γ is varied at fixed amplitude increment $\gamma = \Delta\Gamma/\tau_0$ holding constant their duration τ_0. More precisely we performed a sequence of $\mathcal{N}$ taps, of amplitude $\Gamma_1, \ldots, \Gamma_n, \ldots, \Gamma_{\mathcal{N}}$, from an initial amplitude $\Gamma_1 = 0$ to a maximal amplitude $\Gamma_{\max} = 15$, back to $\Gamma = 0$, and then up again to $\Gamma_{\mathcal{N}} = \Gamma_{\max}$. After each tap we measured the static bulk density of the system $\rho(\Gamma_n)$ (n is the nth tap number).

Our results are qualitatively very similar to those reported in real experiments on dry granular packs [3]. We find that, when the system is successively shaken at increasing vibration amplitudes, the bulk density of the system typically grows and then decreases as shown in Fig. 2. However, when the amplitude of shaking decreases back, the density follows the same path up to only some value of Γ and then deviates from it; in fact, it does not bend down and it keeps growing. As in the experimental data the second part of the shaking cycle is approximately reversible (see Fig. 2). For this reason, these processes are called "irreversible-reversible" cycles [3]. Interestingly, the reversible cycle is a monotonic function of the shaking amplitude (see Fig. 3).

To study the dependence on the cooling rate γ, we repeated the tapping sequence for different values of the

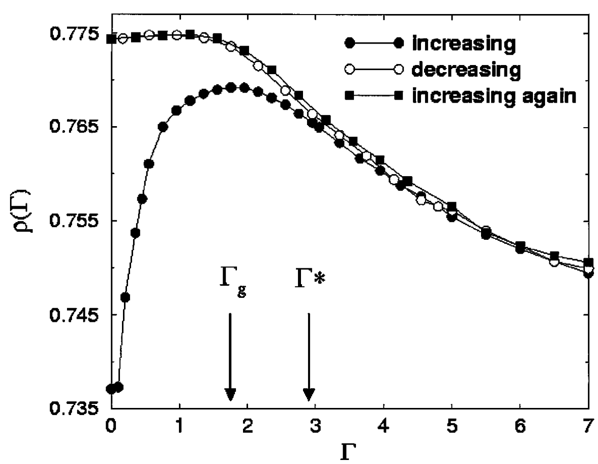

FIG. 2. The static bulk density $\rho(\Gamma)$ of the IFLG model (the Tetris model gives analogous results) as a function of the vibration amplitude Γ in cyclic vibration sequences. The system is shaken with an amplitude Γ which at first is increased (filled circles), then is decreased (empty circles), and, finally, increased again (filled squares) with a given "annealing-cooling" velocity $\gamma \equiv \Delta\Gamma/\tau_0$ (at each value of Γ the system has undergone a tap of duration $\tau_0 = 10^3$). Here we fixed $\gamma = 1.25\ 10^{-3}$. The upper part of the cycle is approximately reversible (i.e., empty circles and filled squares fall roughly on the same curve). The data compare rather well with the experimental data of Novak *et al.* Γ^* is approximately the point where the irreversible and reversible branches meet. Γ_g signals the location of a glass transition (see Fig. 3).

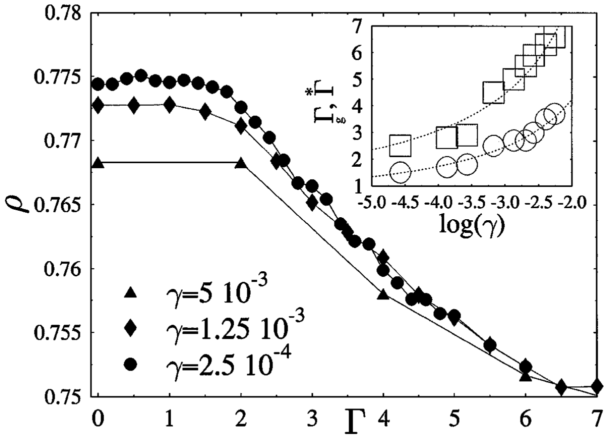

FIG. 3. Main frame: As in Fig. 2, we report the density $\rho(\Gamma)$ as a function of the vibration amplitude Γ for three different values of the cooling velocity γ. For the sake of clarity, we plot here only the descending reversible parts of the cycle. As in experiments on glasses, a too fast cooling drives the system out of equilibrium. The position of the shoulder, $\Gamma_g(\gamma)$, in these curves schematically individuates a glass transition. Inset: Our numerical estimate, in the IFLG model (the Tetris model gives analogous results), of the dependence of $\Gamma_g(\gamma)$ (circles) and $\Gamma^*(\gamma)$ (squares) on the cooling rate γ. Superimposed are approximate fits with the logarithmic law of the text, in analogy to the fit for the glass transition temperature, $T_g(\gamma)$, in glasses [4]. This result apparently shows the dependence of the approximately "reversible branch" on γ.

tap amplitude increment $\Delta\Gamma$ with a fixed tap duration τ_0 (see Fig. 3). We find that the reversible branches have a common part for high values of Γ while, for small Γ, they split into different curves depending on the cooling rate γ. The slower the cooling rate the higher the final density observed at the end of the descending part of the cycle. One can schematically define the point $\Gamma_g(\gamma)$, where the system freezes and goes out of equilibrium as the location of the "shoulder" in these "reversible" branches (see Figs. 2 and 3). Thus, $\Gamma_g(\gamma)$, which depends on γ, corresponds to a "glass transition." Notice that $\Gamma_g(\gamma)$ is usually different from the point $\Gamma^*(\gamma)$, where the "irreversible" and reversible branches meet (see Fig. 2). However, as γ gets smaller, Γ_g and Γ^* become closer and they may coincide in the limit $\gamma \rightarrow 0$.

As in glasses the system gets out of equilibrium due to the fact that the characteristic times of relaxation are much larger than the time τ_0 involved in the experiment, but, for the same reason, the location of the path depends on the rate γ.

The limit of γ going to zero defines an ideal glass transition amplitude Γ_0. Using the analogy with the glass transition we expect a slow logarithm dependence of $\Gamma_g(\gamma)$ on γ. In the inset of Fig. 3 we show our numerical estimate of the dependence of $\Gamma_g(\gamma)$ and $\Gamma^*(\gamma)$ on the cooling rate γ. We also show a possible approximate fit of the data for Γ_g based on the following phenomenological formula [very close to the typical fit of the glass transition temperature $T_g(\gamma)$ in glasses [4]]:

$$\Gamma_g(\gamma) \simeq \Gamma_0 - \Delta\Gamma_g / \ln(\gamma/c_g), \tag{2}$$

where Γ_0, $\Delta\Gamma_g$, and c_g are fit parameters [Γ_0 is the quoted limit value of $\Gamma_g(\gamma)$ when $\gamma \rightarrow 0$, and c_g is a quantity dimensionally corresponding to a characteristic cooling rate]. The above fit is compatible with $\Gamma_0 = 0.0 \pm 0.5$, results stating that in our model the "ideal" transition should be located at $\Gamma = 0$ ($\Delta\Gamma_g = 13 \pm 2$, $c_g = 0.24 \pm 0.05$). The fit for $\Gamma^*(\gamma)$ is analogous, with again $\Gamma_0^* = 0.0 \pm 1$, $\Delta\Gamma^* = 23 \pm 2$, and $c^* = 0.16 \pm 0.05$. Experimental results in all of these directions will be also very interesting.

The analogy with the glass transition suggests, furthermore, the presence of aging phenomena. Below we try to further quantitatively characterize the out-of-equilibrium dynamics in granular matter and make quantitative predictions. In analogy with glassy systems we introduce a two-time density-density correlation function ($t \geq t'$):

$$C(t,t') = \frac{\langle \rho(t)\rho(t')\rangle - \langle \rho(t)\rangle\langle \rho(t')\rangle}{\langle \rho(t')^2\rangle - \langle \rho(t')\rangle^2}, \tag{3}$$

where $\rho(t)$ is the bulk density of the system at time t. In out of equilibrium, $C(t,t')$ is a function of both times t and t' (at equilibrium, just of $t - t'$). The aging properties of the system are characterized by the specific scaling properties of $C(t,t')$.

In order to study the system in a well-defined configuration of its parameters, we evaluate $C(t,t')$ during a "single tap": We prepare the system at $t = 0$ by randomly pouring grains in the box from above as described before, then we start to shake it continuously and indefinitely with a given (small) amplitude Γ. We expect very similar results by considering, instead of a long tap, a series of short taps which is experimentally more convenient (as in Ref. [2]). The data we present here on $C(t,t')$ are averaged over at least 8 different lattice and 512 different noise realizations.

At low Γ, a good fit for the two-time correlation function $C(t,t')$, on the whole five decades in time explored, is given by the following:

$$C(t,t') = (1 - c_\infty)\frac{\ln[(t' + t_s)/\tau]}{\ln[(t + t_s)/\tau]} + c_\infty, \tag{4}$$

where τ, t_s, and c_∞ are fit parameters. Very interesting is the fact that the above behavior is found in both of our models (Tetris and IFLG). The data for the two models, for several values of Γ, rescaled on a single universal master function, are plotted in Fig. 4. In particular, in the explored range of $\Gamma \in [0.11, 0.43]$ (i.e., $x_0 \in [10^{-4}, 10^{-1}]$), we found $\tau \sim e^{z/\Gamma}$ (with $z \sim 2$ in both the IFLG and the Tetris), the mark of activated dynamics [$t_s(\Gamma)$ behaves approximately as τ as a function of Γ]. The asymptotic value c_∞ is difficult to determine with some precision: In the above Γ range we evaluated

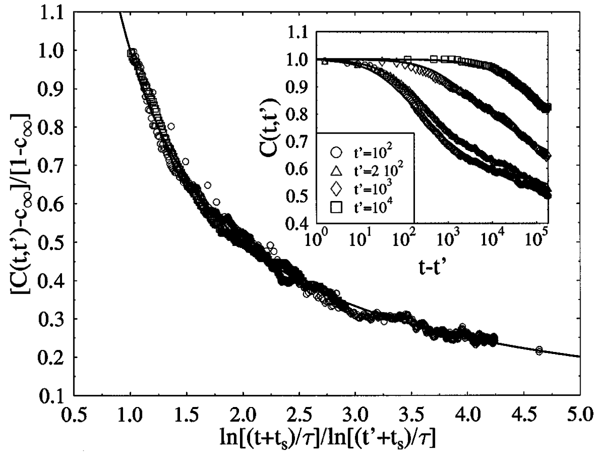

FIG. 4. The two-time density-density correlation function, $[C(t,t') - c_\infty]/(1 - c_\infty)$, as a function of the scaling variable $\alpha = \ln[(t + t_s)/\tau]/\ln[(t' + t_s)/\tau]$. Scaled on the same master function are data from both models considered in the present paper (Tetris, squares; IFLG, circles) and for vibration amplitudes $\Gamma = -1/\ln(x_0)$ with $x_0 \in [10^{-4}, 10^{-1}]$. The master function is $1/\alpha$. Inset: The correlation $C(t,t')$ for the Tetris at $\Gamma = 0.22$ (or $x_0 = 0.01$) as a function of $t - t'$ for four values of $t' = 10^2$, $2\ 10^2$, 10^3, and 10^4.

approximately $c_\infty = 0.2$–0.3 for the IFLG model and $c_\infty = 0.0$–0.2 for the Tetris model.

Equation (4) essentially states that, for times long enough, the correlation $C(t,t')$ is a function (linear) of the ratio $\ln(t')/\ln(t)$. Such a scaling behavior is known in other disordered systems, such as random ferromagnet or random field models [9], and has been proposed by the Fisher and Huse droplet theory of finite-dimensional spin glasses [10]. However, it seems to be different from other scaling functions proposed to fit experimental data in spin glasses [5]. All of this shows the necessity of experimental confirmation of our results in the framework of granular media.

In conclusion, in the framework of simple frustrated lattice gas models, we have studied the off-equilibrium dynamics of slightly shaken granular materials. These models have been previously shown to share many phenomena characteristics of granular media as logarithmic compaction or segregation. Here, we have studied irreversible-reversible cycles and found good agreement with the experimental data on granular packs [3] and made quantitative the strong analogies with nonequilibrium dynamic properties observed in glassy systems as the importance of "cooling rates" γ. We could properly define an equivalent glass transition point, $\Gamma_g(\gamma) \sim 1/\log(1/\gamma)$, in granular media. To fully characterize the out-of-equilibrium dynamics of our models for granular media during compaction, we evaluated the two-time density correlation function $C(t,t')$, which is found asymptotically to be a function of the single ratio $\alpha = \ln(t')/\ln(t)$. These results observed in two different models are also amenable to an experimental check.

We thank INFM-CINECA for computer time on Cray-T3D/E. This work was partially supported by the TMR Network, Contract No. ERBFMRXCT980183.

[1] H. M. Jaeger and S. R. Nagel, Science **255**, 1523 (1992); H. M. Jaeger, S. R. Nagel, and R. P. Behringer, Rev. Mod. Phys. **68**, 1259 (1996).

[2] J. B. Knight, C. G. Fandrich, C. Ning Lau, H. M. Jaeger, and S. R. Nagel, Phys. Rev. E **51**, 3957 (1995).

[3] E. R. Novak, J. B. Knight, E. Ben-Naim, H. M. Jaeger, and S. R. Nagel, Phys. Rev. E **57**, 1971 (1998); E. R. Nowak, J. B. Knight, M. Povinelli, H. M. Jaeger, and S. R. Nagel, Powder Technol. **94**, 79 (1997).

[4] C. A. Angell, Science **267**, 1924 (1995); M. D. Ediger, C. A. Angell, and S. R. Nagel, J. Phys. Chem. **100**, 13 200 (1996).

[5] J. P. Bouchaud, L. F. Cugliandolo, J. Kurchan, and M. Mezard, in "Spin Glasses and Random Fields," edited by A. P. Young (World Scientific, Singapore, to be published). E. Vincent, J. Hammann, M. Ocio, J.-P. Bouchaud, and L. F. Cugliandolo, *Sitges Conference on Glassy Systems*, edited by M. Rubi (Springer, Berlin, 1997).

[6] A. Coniglio and H. J. Herrmann, Physica (Amsterdam) **225A**, 1 (1996).

[7] M. Nicodemi, A. Coniglio, and H. J. Herrmann, Phys. Rev. E **55**, 3962 (1997); J. Phys. A **30**, L379 (1997); M. Nicodemi, J. Phys. I (France) **7**, 1365 (1997).

[8] E. Caglioti, H. J. Herrmann, V. Loreto, and M. Nicodemi, Phys. Rev. Lett. **79**, 1575 (1997).

[9] A. J. Bray, Adv. Phys. **43**, 357 (1994).

[10] D. S. Fisher and D. A. Huse, Phys. Rev. Lett. **56**, 1601 (1986); Phys. Rev. B **38**, 373 (1988); Phys. Rev. B **38**, 386 (1988).