Aging and response properties in the parking-lot model

J. Talbot[1,2], G. Tarjus[2], and P. Viot[2,3,a]

[1] Department of Chemistry and Biochemistry, Duquesne University, Pittsburgh, PA 15282-1530, USA
[2] Laboratoire de Physique Théorique des Liquides[b], Université Pierre et Marie Curie, 4 place Jussieu, 75252 Paris Cedex 05, France
[3] Laboratoire de Physique Théorique[c], bâtiment 210, Université de Paris-Sud, 91405 Orsay Cedex, France

Abstract. An adsorption-desorption (or parking-lot) model can reproduce qualitatively the densification kinetics and other features of a weakly vibrated granular material. Here we study the the two-time correlation and response functions of the model and demonstrate that their behavior is consistent with recently observed memory effects in granular materials. Although the densification kinetics and hysteresis are robust properties, we show that the aging behavior of the adsorption-desorption model is different from other models of granular compaction. We propose an experimental test to distinguish the possible aging behaviors.

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1 Introduction

The vibratory compaction of granular materials has long been of importance in technological applications, but it is only recently that physicists have started to investigate the process from a fundamental perspective. In particular, Knight *et al.* performed experiments [1] in which a column containing monodisperse spherical beads was subject to a long sequence of taps with an intensity characterized by $\Gamma = A/g$ where A is the maximum acceleration and g is the gravitational constant. These experiments showed that the density of the beads increases monotonically and surprisingly slowly with the number of taps, n_t, for various intensities of tapping. More specifically, the density approaches the steady state value as $1/\ln(n_t)$.

In two other studies, Nowak *et al.* [2,3] investigated the effect of cycling Γ, *i.e.* vibrating the column for a fixed number of taps with a sequence of Γ values. They found that as this parameter increases, the density of the granular material increases. When Γ is subsequently reduced by reversing the initial sequence, the density continues to *increase*. This second branch is reversible in that it is retraced if the increasing sequence of Γ is repeated. In the same experiments, Nowak *et al.* monitored the power spectrum of the density fluctuations near the steady state for different values of Γ. The spectrum is distinctly non-Lorentzian, with the highest and lowest characteristic

frequencies being separated by a non-trivial power-law-like regime.

How can these effects be understood? The energy required to displace a granular particle through a distance equal to its diameter is typically much larger than $k_{\mathrm{B}}T$, so granular materials are essentially athermal systems and their properties are dominated by geometrical frustration effects. When the system is already quite dense, even a small additional increase in density requires a large scale cooperative rearrangement of the particles. The logarithmic compaction, the irreversible/reversible cycles and the non-Lorentzian power spectrum of the density fluctuations are essentially the result of this effect.

The experimental results have stimulated a few Monte Carlo studies [4] and a plethora of simple models that incorporate geometrical frustration or quenched disorder (see Ref. [5] and references therein). The latter include the (off-lattice) adsorption-desorption or parking lot model (PLM), Frustrated Lattice Gas (FLG) models, including Tetris, and one-dimensional lattice models with short range dynamical constraints [6]. All these are capable of reproducing qualitatively the experimentally observed kinetics.

Different Frustrated Lattice Gas (FLG) models [7–11] have been proposed in which particles occupy the sites of a square lattice tilted by 45°. Each particle has two internal degrees of freedom which, in the Tetris realization [7], correspond to two possible orientations at a site. Neighboring sites can only be occupied if both particles have a favorable orientation. Vibration is introduced by allowing the particles to move upward with a probability p_{up}.

The connection to the compaction experiments is provided by the parameter $\gamma = -1/\ln(p_{\mathrm{up}}/p_{\mathrm{down}})$ which plays the same role as Γ. The main virtue of this description is that it accounts for the layer structure of the granular material. In the absence of quenched disorder, the system evolves to a compact steady state according to a $1/\ln(t)$-law. To mimic the geometric frustration of the packing, kinetic constraints [7,10] can also be replaced by the introduction of quenched disorder [8].

Here we focus on the adsorption-desorption model in which hard rods are placed on a line at randomly selected positions with a constant rate k_+. If the trial particle does not overlap any rod already on the line, it is accepted. In addition, all adsorbed rods are subject to removal (desorption) at random with a constant rate k_-. The properties of the model depend only on the ratio $K = k_+/k_-$, with large values of K corresponding to small desorption rates. (Pioneering work was done in the limit of $K \to \infty$ [12,13].) The connection to the granular compaction experiment is made by regarding the particles on the line as a layer in the vibrating column. The effect of a tap is to eject particles from the layer (desorption), which is followed by the replacement of the particles, generally in different positions, and possibly the incorporation of additional particles. $1/K$ plays a role similar to that of Γ. In this approach, mechanical stability is implicitly included by the absence of motion of the particles when the desorption is switch off. This microscopic and off-lattice description of granular compaction is suitable for describing all previous experiments and, as we see below, is in fair agreement with experimental results.

The kinetics and equilibrium properties of the adsorption-desorption model have been previously investigated [3,14–17]: in the limit of small desorption rate (that is appropriate to describe granular compaction), the density increases very slowly as $1/\ln(t)$ until it nears the steady state value, at which point the kinetics cross over to an exponential form. The characteristic relaxation time τ_{eq} of this final regime behaves like $K^2/\ln(K)^3$ [15,16]. Moreover, the density-density correlation function exhibits two characteristic time-scales: the shortest one is proportional to $\ln(K)$ and is associated with a local rearrangement of particles, whereas the longest one, τ_{eq}, corresponds to a collective rearrangement permitting an increase of the density. The ratio of these time scales increases rapidly as K becomes larger, and the intermediate timescale region is characterized by a power law behavior.

Here we show that the irreversible/reversible cycles [2,3] and the recently demonstrated presence of memory effects [18] in the compaction of glass beads can be reproduced by the adsorption-model. All the salient features of the phenomenology of vibrated granular materials are thus qualitatively described by the model. We also show that, because of the very slow relaxation to equilibrium (due to cooperative rearrangements of the particles), the model displays out-of-equilibrium dynamical effects such as aging [19], and we study these properties both in the response and the correlation functions. Interestingly, the aging behavior of the two-time correlation function of the density

fluctuations is found to be different from that observed for the FLG and Tetris models [8,10]. This leads us to suggest an experimental way of discriminating among the different models applied to vibrated granular materials.

2 Model and simulation

If time is expressed in units of k_+, the kinetics is given by

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \Phi(t) - \frac{\rho}{K}, \tag{1}$$

where $\Phi(t)$, the insertion probability at time t (or density ρ), is the fraction of the line that is available for the insertion of a new particle. The presence of a relaxation mechanism, implies that the system eventually reaches a steady state that corresponds to an equilibrium configuration of hard particles with $\rho_{\mathrm{eq}} = K\Phi_{\mathrm{eq}}(\rho_{\mathrm{eq}})$, where ρ_{eq} denotes the equilibrium density. At equilibrium, the insertion probability is given exactly by

$$\Phi_{\mathrm{eq}}(\rho) = (1 - \rho)\exp(-\rho/(1 - \rho)). \tag{2}$$

The kinetics of the model were simulated using the event-driven algorithm described previously [17].

3 Densification branches and memory effects

Following the experimental procedure of compaction cycles [3,2], we have performed simulations in which the rate of desorption, $1/K$, is first increased at a given rate and then cycled down and up, the simulation being stopped after the same time $t = 40\,000$. (Recall that $1/K$ plays the same role as the tapping intensity and that, when expressed in units of k_+, time is assumed to measure the number of taps; this leads to the reasonable behavior that the number of desorption events increases as the tapping strength increases.) Figure 1a displays the density as $1/K$ varies between 10^{-4} and 10^{-3}, and is similar to the behavior observed experimentally and numerically in the FLG models [8,10]. Along the first (irreversible) branch the density increases rapidly and then passes through a maximum as $1/K$ increases. When the initial sequence of $1/K$ is reversed ($1/K$ decreasing), the density increases monotonically. When the initial (increasing) sequence of $1/K$ is repeated, the density now decreases monotonically, nearly retracing the second branch. The residual hysteresis observed between the second and third branches is also present in other models [5], and it diminishes as one considers larger values of K and larger time intervals between changes of desorption rate. Note that the densities attained in the present adsorption-desorption model are typical of a one-dimensional system and that more realistic values would be obtained by employing the two-dimensional version of the model.

To further compare the model predictions to the available experimental data on vibrated granular materials, we
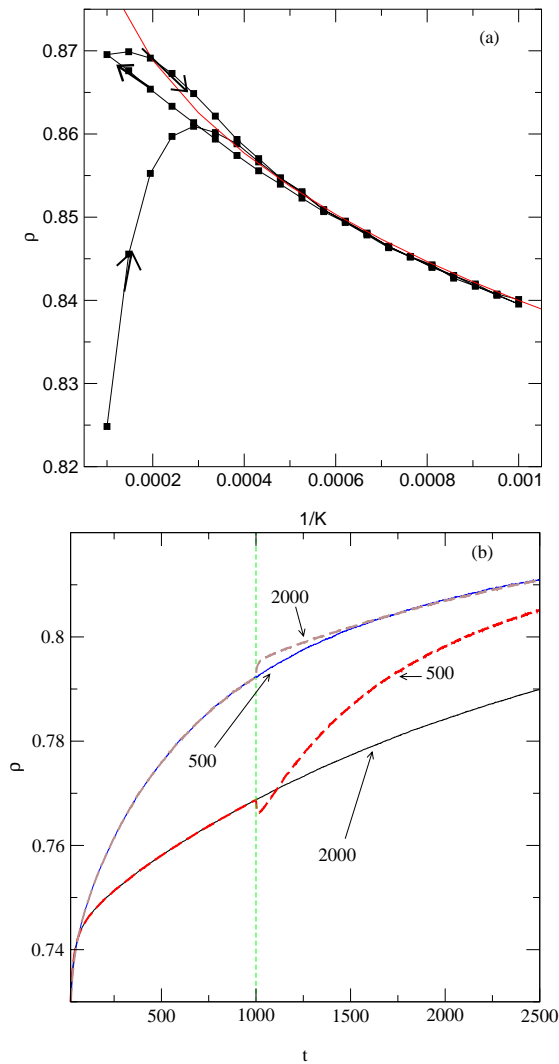
Fig. 1. (a) Irreversible and reversible densification branches. Starting from a "loose" packed state, the process consists of a sequence of decreasing values of K. For each K, the duration is 40 000. The arrows indicate the way of cycle compaction. The dotted line is the equilibrium curve. (b) Memory effect at short times after a rapid change of the desorption rate: the full curves correspond to a process with a constant K, whereas the dashed curves show the kinetics of a process where K is switched from 500 to 2000 and *vice versa* at $t = 1000$.

have studied the effect of an abrupt change in the desorption rate $1/K$ on the densification kinetics. This is illustrated in Figure 1b in which K is switched from 500 to 2000 and *vice versa*. The same "anomalous" behavior as observed experimentally by Josserand *et al.* [18], *i.e.* an acceleration in the densification process when $1/K$ is suddenly lowered and the reverse phenomenon when $1/K$ is increased, is found in the adsorption-desorption model.

4 Response functions and aging

Compaction cycles and memory effects characterize the response of the system to a moderate or large change of

the tapping strength. We consider below another facet of the out-of-equilibrium dynamical behavior of the system by examining the response to a small perturbation and the associated two-time correlation function.

Non-equilibrium studies have been performed on FLG models: Nicodemi [9] demonstrated that these models exhibit a negative response and Nicodemi and Coniglio [8] showed that the aging regime of this model is characterized by $C(t, t_w) \sim \ln(t_w)/\ln(t)$. Barrat and Loreto [10] performed extensive simulations on the Random Tetris model and clearly showed that the aging behavior is the same.

We calculate the response function as follows. Starting from an empty line, the system evolves at a fixed desorption rate. At the end of a fixed waiting period, t_w, two clones of the system are made. The original systems continues to evolve with the same desorption rate K, and the two others with $K \pm \delta K$. The response then corresponds to the difference in density of the copy and the original system at a later time t. In order to obtain reasonable statistics, it is necessary to average over many independent runs (typically 10^4). Since the model at equilibrium is equivalent to a grand canonical ensemble, the variable conjugate to the density is $\beta\mu$ where $\beta = 1/k_BT$ and μ is the chemical potential. From equation (1) at equilibrium we have that $\beta\mu = \ln(K)$ and the response function to a change in $\ln(K)$ is therefore defined as

$$R(t, t_w) = \frac{\partial \rho(t)}{\partial \ln(K(t_w))} \; . \tag{3}$$

To be more precise, this is an integrated response function since the perturbation is applied over an extended time. Note that when t becomes larger than the ergodic time τ_{eq}, the response function attains the positive equilibrium value $R_{eq} = \rho(1-\rho)^2$. The fluctuation-dissipation theorem applies when $t_w > t_{eq}$ and we recover the time translation invariance $R(t, t_w) = R(\tau = t - t_w)$ in which case the response function associated with changes in the external field $\beta\mu = \ln(K)$ (coupled to the density fluctuations) and the density-density correlation function are related:

$$R(\tau) = \tilde{C}(0) - \tilde{C}(\tau) \tag{4}$$

where $\tilde{C}(\tau) = \langle \delta\rho(\tau)\delta\rho(0) \rangle$. (Note that as usual for hard objects, the temperature is irrelevant and does not explicitly enter in equation (4) because it is included in $\ln(K) = \beta\mu$.) From our previous study of the fluctuations around equilibrium [17] we derive the analytical short time form which is exact for $K \to \infty$:

$$R(\tau) = \frac{L_W(K)^2}{1 + L_W(K)} \left(1 - \frac{L_W(K)}{\tau + L_W(K)} e^{-\tau/K} \right) + O(1/K) \tag{5}$$

where L_W is the Lambert-W function and for long times the equilibrium value is attained exponentially with a characteristic time of order $K^2/\ln(K)^3$. It is worth stressing that with the definition adopted above of a response to $\ln(K)$, a positive response means that an increase of
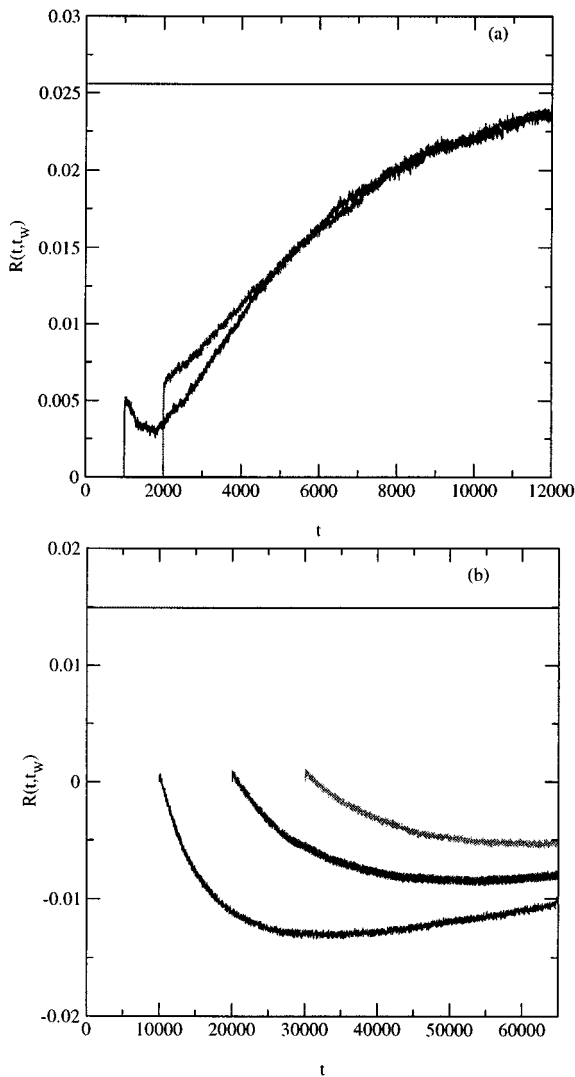
Fig. 3. Two-time density-density correlator $C(t, t_w)$ as a function of time for various waiting times t_w ($t_w = 50, 100, 500, 1000, 5000$ left to right).

Fig. 2. Two-time response function $R(t, t_w)$ as a function of time t for (a) $K = 500$ with $t_w = 1000, 2000$ and for (b) $K = 5000$ with $t_w = 10\,000, 20\,000, 30\,000$. The horizontal lines correspond to the equilibrium value, $R_{eq} = \rho(1 - \rho)^2$.

K (*i.e.*, a decrease in tapping strength) increases the density, whereas a negative response corresponds to a density decrease.

Figure 2a displays two response functions for a system with $K = 500$. For a waiting time ($t_w = 2000$) larger than the ergodic time [19], the system is already at equilibrium and the response is obviously monotonically increasing and in agreement with the analytical result, equation (5). At short times, the behavior of both curves is quite similar, but for $t_W = 1000$ the response then decreases and attains a minimum before increasing again towards the equilibrium value (as expected in a system with interrupted aging). Figure 2b shows the response functions in the aging regime for a larger value of $K = 5000$ for three different waiting times. The shape of the curves is similar to Figure 2a, but the amplitude of the initial increase of $R(t, t_w)$ is smaller than for $K = 500$. The negative mini-
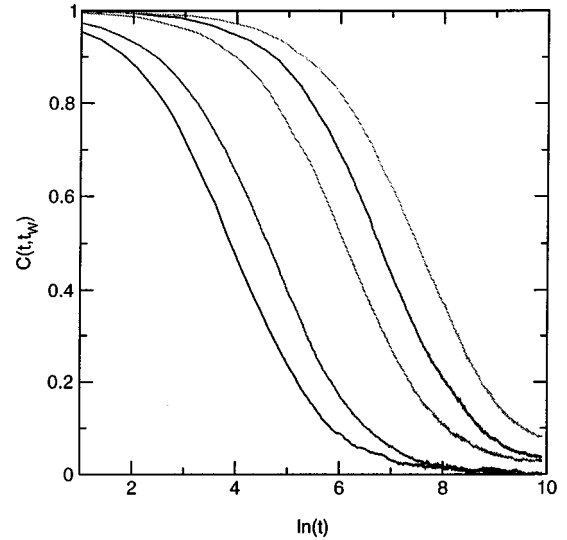
mum, that results from a competition between local rearrangements (leading to short-time "anomalous" response discussed above) and global rearrangements (allowing for the densification), is less pronounced as t_w increases. For long times, which are not accessible in the simulations, the response becomes positive as it approaches the equilibrium value.

In order to investigate the out-of-equilibrium evolution of the density-density correlation function, we consider the adsorption-desorption model in the limit where the desorption is infinitely small [12]. In this case ($K \to \infty$), the relaxation time τ_{eq} [19] is infinite and the system always evolves with out-of-equilibrium dynamics. For K large, but finite (5000), the out of equilibrium and aging behavior is interrupted since t may become larger than τ_{eq}. We have however checked that the aging behavior when $t < \tau_{eq}$ is similar to that of the $K \to \infty$ case.

The normalized two-time density-density correlation function is defined as follows:

$$C(t, t_w) = \frac{\langle \rho(t)\rho(t_w) \rangle - \langle \rho(t) \rangle \langle \rho(t_w) \rangle}{\langle \rho(t_w)^2 \rangle - \langle \rho(t_w) \rangle^2} \tag{6}$$

where the angular brackets denote an average over independent runs and $t \geq t_w$.

Figure 3 shows $C(t, t_w)$ as a function of t for different values of the waiting time t_w. The system exhibits aging: the larger t_w, the longer the memory of the initial configuration persists. This aging regime is characterized by a violation of the fluctuation-dissipation theorem and the breakdown of time-translational invariance [19].

When t and t_w are large enough (but still smaller than the equilibration time), the aging behavior is usually described by a non-trivial form involving a single scaling variable that is the ratio of a function of t (or of the elapsed time $t - t_w$) divided by the same function of t_w.
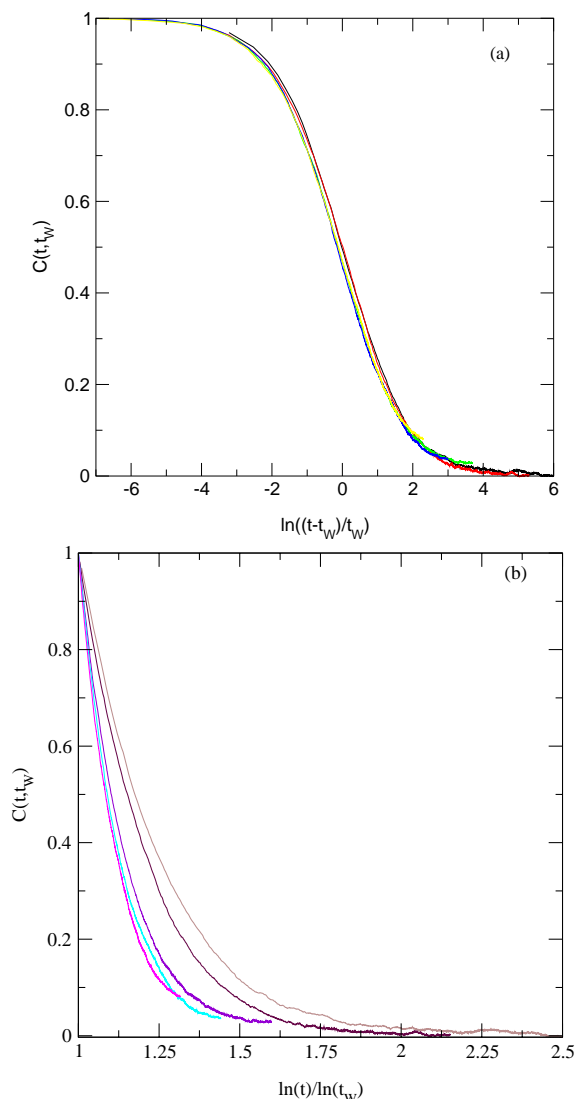
This finding suggests an experimental test for deciding which phenomenological model, each built on different assumptions, geometrical frustration in the adsorption-desorption model, quenched disorder or kinetic constraints and layer structure in FLG models, is more relevant to describe the physics of vibrated granular materials. Since all predict the logarithmic compaction, the irreversible/reversible cycles, the non-Lorentzian power spectrum of the density fluctuations and the memory effects observed experimentally, a promising way to validate or invalidate them as a minimal theoretical approach would be to study experimentally the two-time correlation function of the density fluctuations in the course of the densification process.

Fig. 4. (a) Two-time density-density correlator $C(t, t_w)$ as a function of (a) $(t - t_w)/t_w$ and (b) $\ln(t)/\ln(t_W)$ for waiting times $t_w = 50, 100, 500, 1000, 5000$.

References

1. J.B. Knight, C.G. Fandrich, C.N. Lau, H.M. Jaeger, S.R. Nagel, Phys. Rev. E **51**, 3957 (1995).
2. E.R. Nowak, J.B. Knight, M. Povinelli, H.M. Jaeger, S.R. Nagel, Powder Technol. **94**, 79 (1997).
3. E.R. Nowak, J.B. Knight, E. Ben-Naim, H.M. Jaeger, S.R. Nagel, Phys. Rev. E **57**, 1971 (1998).
4. A. Mehta, G.C. Barker, J. Phys. Cond. Matt. **12**, 6619 (2000).
5. D. Head, Phys. Rev. E **62**, 2439 (2000).
6. A. Prados, J.J. Brey, B. Sánchez-Rey, Physica A **284**, 277 (2000).
7. E. Caglioti, V. Loreto, H.J. Herrmann, M. Nicodemi, Phys. Rev. Lett. **79**, 1575 (1997).
8. M. Nicodemi, A. Coniglio, Phys. Rev. Lett. **82**, 916 (1999).
9. M. Nicodemi, Phys. Rev. Lett. **82**, 3734 (1999).
10. A. Barrat, V. Loreto, J. Phys. A **33**, 4401 (2000).
11. A. Barrat, V. Loreto, Europhys. Lett. **53**, 297(2001).
12. X. Jin, G. Tarjus, J. Talbot, J. Phys. A **27**, L195 (1994).
13. P.L. Krapivsky, E. Ben-Naim, J. Chem. Phys. **100**, 6778 (1994).
14. E. Ben-Naim, J.B. Knight, E.R. Nowak, H.M. Jaeger, S.R. Nagel, Physica D **123**, 380 (1998).
15. A.J. Kolan, E.R. Nowak, A.V. Tkachenko, Phys. Rev. E **59**, 3094 (1999).
16. J. Talbot, G. Tarjus, P. Viot, J. Phys. A **32**, 2997 (1999).
17. J. Talbot, G. Tarjus, P. Viot, Phys. Rev. E **61**, 5429 (2000).
18. C. Josserand, A. Tkachenko, D.M. Mueth, H.M. Jaeger, Phys. Rev. Lett. **85**, 3632 (2000).
19. J.-P. Bouchaud, L.F. Cugliandolo, J. Kurchan, M. Mezard, in *Spin Glasses and Random Fields* (World Scientific, Singapore, 1998), p. 161.

In the FLG models, it was found numerically that the appropriate scaling variable is $\ln(t)/\ln(t_w)$ [8]. For the adsorption-desorption model we show in Figure 4a that when the two-time correlation function $C(t, t_w)$ is plotted as a function of $(t - t_w)/t_w$ the data for five waiting times, ranging from 25 to 5000, collapse onto a single curve. This aging behavior, similar to that found in a large variety of systems [19], is thus different from the $\ln(t)/\ln(t_w)$ dependence of the FLG models. Figure 4b clearly demonstrates the absence of a master curve with this scaling. Moreover, no better result is obtained when $\ln(t - t_w)/\ln(t_w)$ is used in place of $\ln(t)/\ln(t_w)$.