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Edwards' approach to horizontal and vertical segregation in a mixture of hard spheres under gravity

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Abstract

We study the phenomenon of size segregation, observed in models of vibrated granular mixtures such as powders or sand. This consists of the de-mixing of the different components of the system under shaking. Several mechanisms have been proposed to explain this phenomenon. However, the criteria for predicting segregation in a mixture, an issue of great practical importance, are largely unknown. In the present paper we study a binary hard-sphere mixture under gravity on a three-dimensional lattice using Monte Carlo simulations. The vertical and horizontal segregation observed during the tap dynamics is interpreted in the framework of a statistical mechanics approach to granular media in the manner of Edwards. A phase diagram for the vertical segregation is derived, and compared with the simulation data.

1. Introduction

In the present paper we study the phenomenon of size segregation in models of granular media using a statistical mechanics approach in the manner of Edwards [1]. Segregation is a ubiquitous and intriguing phenomenon observed in vibrated granular mixtures such as powders or sand: in the presence of shaking the system is not randomized, but its components tend to separate [2, 3]. Recently it has become clear that powders not only segregate but can also spontaneously form patterns in the process (see for instance [2–7]). Several mechanisms have been proposed to explain these phenomena [7–15], but the criteria for predicting segregation in a mixture, an issue of great practical importance, are still largely unknown [2]. In the present paper we show that the segregation process observed using Monte Carlo simulations in a lattice model of a hard-sphere binary mixture under gravity subject to 'taps' can be well understood in Edwards' scenario.

Edwards' original hypothesis [1] is that, by gently shaking a granular system under the constraint of fixed volume V, the distribution over the mechanically stable states where the system is found at rest is uniform [16–20]. In other words, the macroscopic stationary state reached by gently shaking the system can be characterized by a thermodynamic parameter,

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the Edwards compactivity, $X^{-1} = \frac{\partial \ln \Omega}{\partial V}$, where Ω is the number of mechanically stable states corresponding to the fixed volume V. This suggested the possibility of defining a configurational entropy (similar to that of glassy systems)[1], $S = \ln \Omega$, for granular media, and to treat non-thermal systems (such as granular media in their mechanically stable states and glassy systems blocked at $T = 0$ in their inherent structures [21]) in the framework of a unified statistical mechanics approach [16–20]. More generally, it was shown [18–20] that more than one 'thermodynamic' parameter may be required to describe the stationary state, even though the criteria for determining these parameters *a priori* may not be easily accessible.

In previous papers [20] we have checked the validity of Edwards' approach in a simple lattice model of hard spheres under gravity. We found that the 'inherent states' (defined as the stable configurations in the potential energy landscape) explored at stationarity, by shaking the system using sequences of taps, are distributed according a generalized Gibbs measure obtained by maximizing the configurational entropy under suitable constraints, such as a given average energy. The macroscopic state reached by the system can therefore be completely characterized by the configurational temperature[2], $T_{conf}^{-1} \equiv \beta_{conf} = \frac{\partial \ln \Omega}{\partial E}$, the thermodynamic parameter conjugate to the energy.

More specifically [20], in a system made of particles of two different sizes under gravity, two configurational temperatures must be introduced. Here we present the results obtained by studying the system using Monte Carlo simulations and the consequent interpretation of the segregation phenomena observed.

In section 2, the hard-sphere binary mixture and its description in statistical mechanics terms are presented. In sections 3 and 4, the results for vertical and horizontal size segregation obtained using Monte Carlo simulations are interpreted in terms of such a statistical mechanics approach. The correspondence with coarsening phenomena [7, 22][3] is also discussed in some detail.

2. The model

We study a binary system of hard spheres under gravity; the system is made up of two species, 1 (small) and 2 (large), with grain diameters a_0 and $\sqrt{2}a_0$, on a cubic lattice of spacing $a_0 = 1$. We set the units such that the two kinds of grain have masses $m_1 = 1$ and $m_2 = 2$, and the gravitational acceleration is $g = 1$. The hard core potential, $\mathcal{H}_{hc}$, is such that two large nearest neighbour particles cannot overlap. This implies that only pairs of small particles can be nearest neighbours on the lattice. The overall Hamiltonian of the system is:

$$\mathcal{H} = \mathcal{H}_{hc} + m_1 g H_1 + m_2 g H_2, \tag{1}$$

where $H_1 = \sum_i^{(1)} z_i$, $H_2 = \sum_i^{(2)} z_i$, and the height of ith particle is z_i. The two sums are over all particles of species 1 and 2 respectively. The gravitational energies are $E_1 = H_1$ and $E_2 = 2H_2$.

The blocked configurations are visited by using tap dynamics, where each tap consists of raising the Monte Carlo bath temperature from zero to a value T_Γ (defined as the 'tap

[1] $\Omega(E, V, \ldots)$ is the number of inherent states corresponding to a fixed set of thermodynamic quantities, such as energy E, volume V, etc.

[2] β_{conf} coincides with the Edwards compactivity in the case where the density is uniform in the bulk and the energy is given only by the gravitational term.

[3] The presence of coarsening phenomena in granular compaction was discussed in [22]. The possibility of describing the size segregation patterns observed in binary mixtures of particles under gravity as a critical phenomenon was suggested in [7]. A horizontally shaken shallow layer of a binary mixture of dry particles was experimentally studied. Stripes orthogonal to the direction of vibration appeared and their average width increased as a function of time as a power law, $t^{0.25}$. The existence of a critical event associated with the onset of the segregation pattern was suggested.

amplitude') and, after a lapse of time τ_0 (the 'tap duration'), quenching it back to zero. By cyclically repeating the process the system explores the space of the inherent states (see [20] for details). In [20], we found that the stationary state reached by the system in this way can be described by a generalized Gibbs measure obtained by maximizing the configurational entropy under the constraint that the energies of the two species are independently fixed. Specifically, the probability distribution, P_r, of observing the system in the generic inherent state r corresponding to an energy E_{1r} for the small particles and E_{2r} for the large particles is given by:

$$P_r = \frac{\mathrm{e}^{-\beta_1 E_{1r} - \beta_2 E_{2r}}}{Z}, \tag{2}$$

where β_1 and β_2 are the thermodynamic parameters, called 'inverse configurational temperatures', conjugate respectively to the small particle energy and to the large particle one.

In the whole configurational space, the system can thus be described by an effective Hamiltonian

$$\mathcal{H}_{eff} = \mathcal{H}_{hc} + \beta_1 H_1 + 2\beta_2 H_2 + \delta, \tag{3}$$

where the term δ is zero if the configuration is an inherent state and infinite otherwise. Consequently, the study of the model, equation (1), can be accomplished in the context of statistical mechanics by solving the problem connected to the effective Hamiltonian, $\mathcal{H}_{eff}$. In this way the properties (such as size segregation patterns, 'Brazil nut' and 'reverse Brazil nut' effects) of the model, equation (1), under the tap dynamics [2–7, 10, 11, 15] could be predicted.

3. Vertical segregation

In figure 1 we show the diagram for the vertical segregation obtained by simulating the effective Hamiltonian, equation (3), using Monte Carlo techniques [17]. The full circles in the figure correspond to pairs (T_1, T_2) in which difference between the average heights of small and large grains, $\Delta h = h_1 - h_2$ ($h_1 = H_1/N_1$ and $h_2 = H_2/N_2$), is equal to zero. By decreasing T_2 at a fixed value of T_1, a crossover at $T_2 = T_2^*(T_1)$, from a region where large particles are on average above (the 'Brazil nut' effect) to a region where they are below (the 'reverse Brazil nut' effect [12]) small particles, is observed. Solving the model, equation (3), in mean field approximation [20, 23], we have found that this crossover corresponds to the drift of the minimum of the free energy, $F = -\ln Z$, in the inherent space from negative to positive values of the vertical segregation parameter, $\Delta h = h_1 - h_2$.

We have checked this prediction by simulating the model, equation (1), using Monte Carlo techniques. N_1 small particles and N_2 large particles[4] are confined in a box of height L_z and linear size in the horizontal directions L (in the horizontal directions periodic boundary conditions are considered). At $t = 0$ the system is prepared in a random loose stable packing; then it evolves under the tap dynamics. At stationarity the average heights of small and large grains, h_1 and h_2, are measured. In figure 2, the differences between average heights of small and large grain, $\Delta h = h_1 - h_2$, are plotted as a function of the tap amplitude, T_Γ, for the tap duration $\tau_0 = 10$ MCS[5]. As we can see, at high T_Γ, the larger grains are found above the smaller ones as in the 'Brazil nut' effect, whereas at low T_Γ, the opposite is found (the 'reverse

[4] We consider a fixed density $\rho_1 = N_1/L_z * L^2 = 0.15$ and $\rho_2 = N_2/L_z * L^2 = 0.05$, and box height $L_z = 4$, and four different values of the horizontal linear size L, to study the dependence of the cluster properties on the cluster size.

[5] τ_0 is measured in Monte Carlo steps (MCS), where 1 MCS corresponds to N attempts to move a particle randomly chosen, and $N = L_z * L^2$ is the site number of the box.

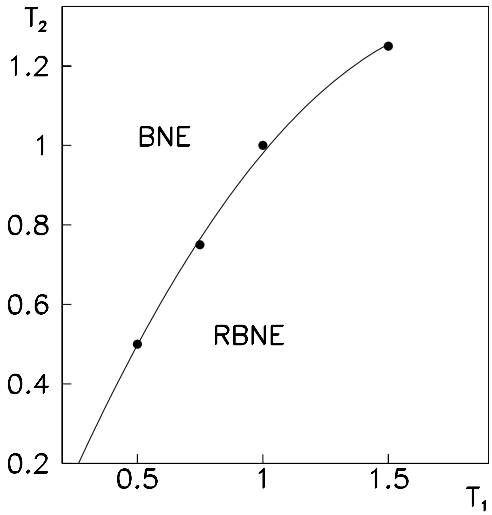

Figure 1. The diagram for vertical segregation (obtained by Monte Carlo simulations of the effective Hamiltonian, equation (3)) in the plane of the configurational temperatures (T_1, T_2) associated with the two species in the mixture. The region marked 'BNE' corresponds to the 'Brazil nut effect', where larger particles are on average on the top of the system. The 'RBNE' region corresponds to the reverse situation. The system contains $N_1 = 120$ small particles and $N_2 = 40$ large particles confined in a three-dimensional box of horizontal linear size $L = 20$ and height $L_z = 4$. The continuous curve is a guide for the eye.

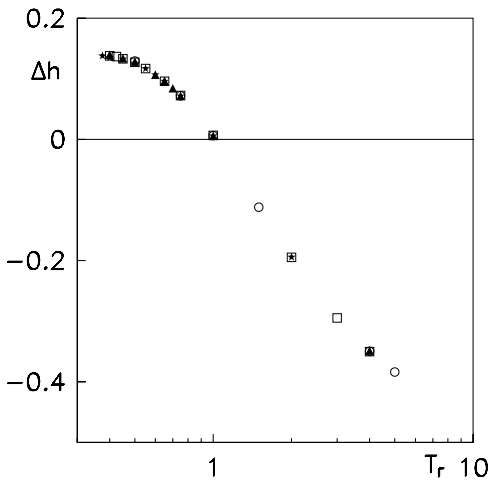

Figure 2. The differences between average heights of small and large grains, $\Delta h = h_1 - h_2$, measured during the tap dynamics, in the hard-sphere binary mixture under gravity, as a function of the tap amplitude T_Γ (in units $m_1 g a_0$), for the tap duration $\tau_0 = 10$ MCS, and different values of the linear size $L = 20, 40, 60, 80$.

Brazil nut' effect). In this scenario the system (evolving under the tap dynamics with fixed duration, τ_0) can be thought to be following a given path, $T_2 = T_2(T_1)$ (where $T_1 = T_1(T_\Gamma, \tau_0)$), in the ($T_1$, T_2) plane, crossing from the 'Brazil nut' effect region to the 'reverse Brazil nut' effect region.

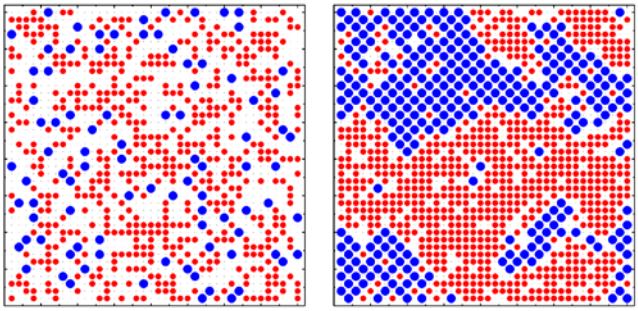

Figure 3. Particle configurations on the bottom layer during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 0.35$, at times $t = 0$ (left) and 40 000 (right).
(This figure is in colour only in the electronic version)

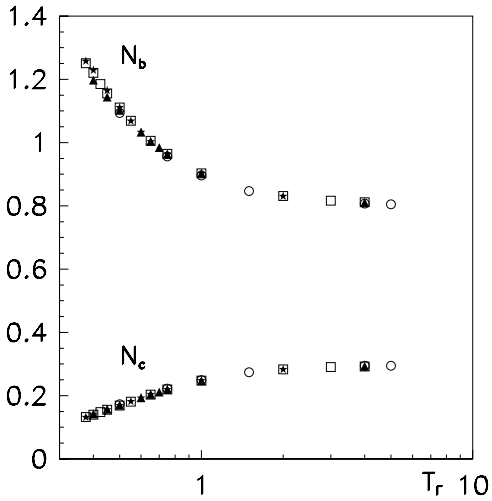

Figure 4. The contact numbers, N_b, and the cluster numbers, N_c (per particle), of the small particles plotted as a function of the tap amplitude T_Γ (in units $m_1 g a_0$), for a tap duration $\tau_0 = 10$ MCS, and different values of the linear size $L = 20, 40, 60, 80$.

4. Horizontal segregation

At low tap amplitudes, T_Γ, the formation of clusters made up of one type of particle is observed during the dynamics. In figure 3, particle configurations on the bottom layer are shown at different times during the tap dynamics.

In the model studied here, a cluster of small particles is a set of nearest neighbours, whereas a cluster of large particles is a set of next neighbours (since two large particles cannot be nearest neighbours). We have studied the properties of such clusters at stationarity. In figures 4 and 5 the contact numbers, N_b, between one type of particle and the cluster numbers, N_c (per particle), are shown respectively for small and large particles. For both large and small particles, we observe an increase in the contact number N_b, and a decrease in the cluster number N_c, as the tap amplitude T_Γ decreases. It is clear that as T_Γ is lower, the stationary state reached by the system under shaking is characterized by larger clusters, each one made up of one type of particle (see also figure 3).

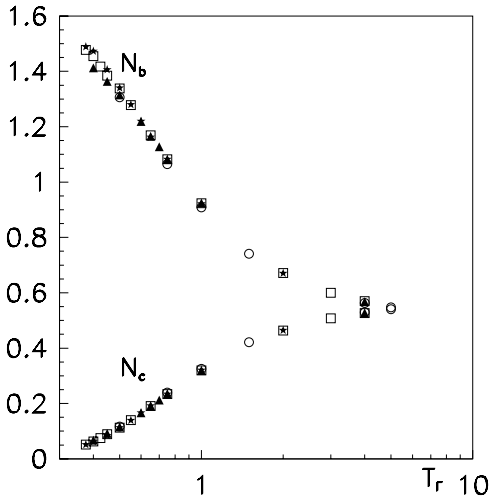

Figure 5. The contact numbers, N_b, and the cluster numbers, N_c, (per particle), of the large particles plotted as a function of the tap amplitude T_Γ (in units $m_1 g a_0$), for a tap duration $\tau_0 = 10$ MCS, and different values of the linear size $L = 20, 40, 60, 80$.

In order to gain information about the time evolution of the various length scales present in the system, we have also measured the dynamic structure factor, $S_p(\boldsymbol{k}, t)$, independently for the small and the large particles on the bottom layer (corresponding respectively to $p = 1$ and 2). For continuous systems, the dynamic structure factor is defined as the Fourier transform of the density–density correlation function; for the discrete system we have studied, it is

$$S_p(\boldsymbol{k}, t) \equiv \frac{1}{N_p^b}\left\langle \sum_{ij}^{(p)} \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j)} \right\rangle, \tag{4}$$

where the sum $\sum_{ij}^{(p)}$ is done over all particles of type p on the bottom layer, N_p^b is the number of particles of type p on the bottom layer at time t, $\boldsymbol{k} = 2\pi \boldsymbol{n}/L$, $\boldsymbol{n} \equiv (n_x, n_y)$ and $|\boldsymbol{n}| = 1, 2, \ldots, L/2$ (the linear size of the box is fixed $L = 40$, and the ensemble average $\langle \cdots \rangle$ is done over 40 independent realizations). We have always measured the average quantity $S_p(k, t) = (S_p(\boldsymbol{k_x}, t) + S_p(\boldsymbol{k_y}, t))/2$, where $\boldsymbol{k_x} \equiv (k, 0)$ and $\boldsymbol{k_y} \equiv (0, k)$.

For high tap amplitudes a stationary state is reached during the tap dynamics, and the equilibrium dynamic structure factor is measured:

$$S_p(k) = \overline{S_p(k, t)}, \tag{5}$$

where again $p = 1$ or 2, and $\overline{\cdots}$ is the time average at stationarity.

In figure 6, the equilibrium structure factor, $S_1(k)$, for the small particles on the bottom layer (a similar behaviour is observed for the large particles) is plotted for several values of the tap amplitude T_Γ. As we can see in the figure, $S_1(k)$ has a maximum for $k = k_{min}$ ($k_{min} = 2\pi/L$), and this maximum increases as the tap amplitude T_Γ decreases. We have fitted $S_1(k_{min})$ against T_Γ by a power law, $(T_\Gamma - T_\Gamma^*)^{-\gamma}$, where $\gamma = 0.52 \pm 0.20$ and $T_\Gamma^* = 0.43 \pm 0.05$ (see figure 7). Because the system does not reach a stationary state for T_Γ very close to T_Γ^*, our data cannot establish whether T_Γ^* corresponds to a real critical point or the divergence is only apparent.

In figures 8 and 9, the dynamic structure factor, $S_p(k, t)$, for the small and large particles on the bottom layer, are respectively shown at a low tap amplitude, $T_\Gamma =$

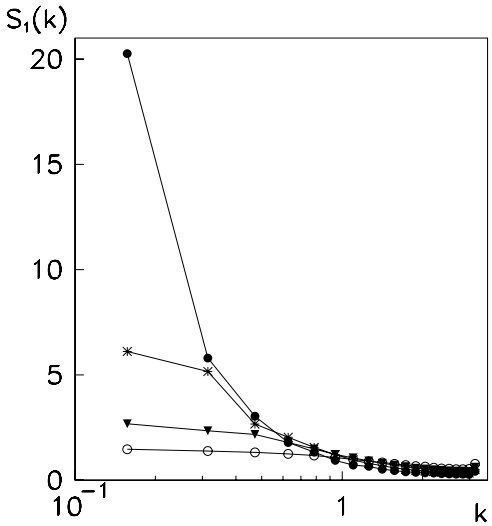

Figure 6. The equilibrium structure factor, $S_1(k)$, for the small particles on the bottom layer as a function of the wavevector k during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 5, 1, 0.60, 0.45$ (from bottom to top).

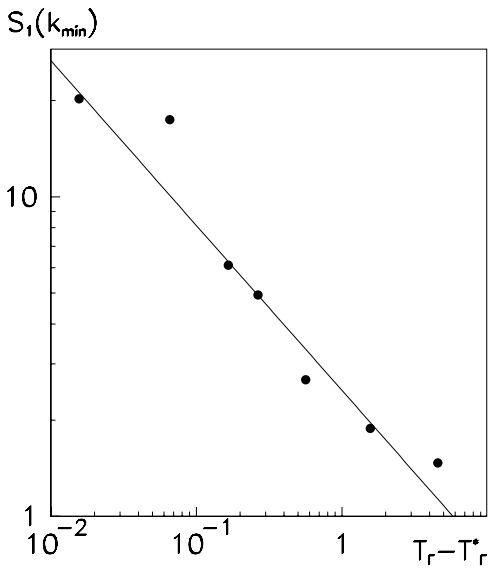

Figure 7. The maximum of the equilibrium structure factor for the small particles on the bottom layer, $S_1(k_{min})$, as a function of the tap amplitude T_Γ can be fitted by a power law (the full line in the figure) diverging at a tap amplitude $T_\Gamma^* = 0.43 \pm 0.05$. In the figure, $S_1(k_{min})$ is plotted as a function of $T_\Gamma - T_\Gamma^*$.

0.35, where a stationary state is not reached during the tap dynamics. As we can see in the figures, the behaviour observed strongly resembles that of the phase separation process [24]. During spinodal decomposition the position of the peak, k_m, moves to a smaller value of k. Thus k_m is typically measured as a function of time during the dynamics.

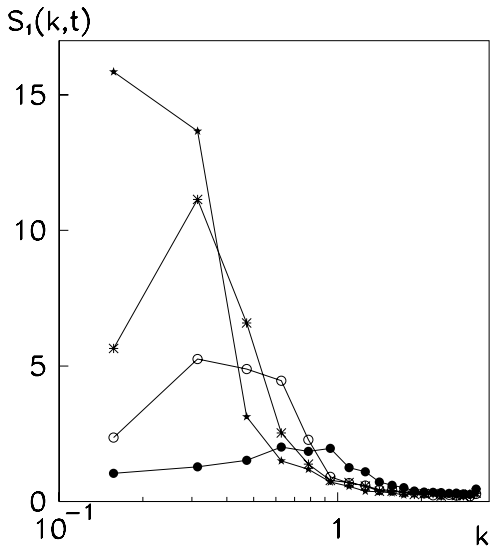

Figure 8. The dynamic structure factor, $S_1(k, t)$, for the small particles on the bottom layer during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 0.35$ as a function of the wavevector k, for $t = 100$, 2000, 10 000, 40 000 (from bottom to top).

We calculate an equivalent quantity, the first moment of $S_p(k, t)$:

$$k_1^p(t) \equiv \frac{\sum_k k(t) S_p(k, t)}{\sum_k S_p(k, t)}. \tag{6}$$

Because this quantity is calculated using data acquired over the entire range of wavevectors, $k_1^p(t)$ can be calculated more accurately than the peak position $k_m^p(t)$. The two quantities k_1^p and k_m^p should scale in the same way with time, so that either quantity is an acceptable measure of the characteristic length in the system. Figure 10 shows the first moment $k_1^1(t)$ of $S_1(k, t)$ plotted against time. As in the phase separation process, these data can be fitted by a power law, $t^{-\alpha}$; however, the exponent ($\alpha \simeq 0.13$) obtained is not consistent with the value $1/3$ expected for late-stage growth in a system with a conserved order parameter [25]. This may be due to the effects of gravity or to the fact that during the dynamics the numbers of small and large particles on the bottom layer are not conserved (as we can see in figure 11, where the fractions of small particles and large particles on the bottom layer are plotted against time t).

The tap amplitude T_Γ^* is the value at which the maximum of the equilibrium structure factor seems to diverge, and below which a slowing down of the dynamics is observed (below T_Γ^* the system does not reach a stationary state, and it shows properties strongly resembling those of a phase separation process). In the framework of the statistical mechanics approach to granular media, these results suggest the presence of a critical point in the effective Hamiltonian, equation (3). This may be verified by a direct study of the effective Hamiltonian, using both mean field approximations and Monte Carlo simulations [23].

It should be noted that our data cannot establish whether the system encounters a real critical point, as the tap amplitude decreases, or whether it crosses the coexistence line near a critical point. Other simulations with a different tap duration, τ_0, are necessary to clarify this question [23].

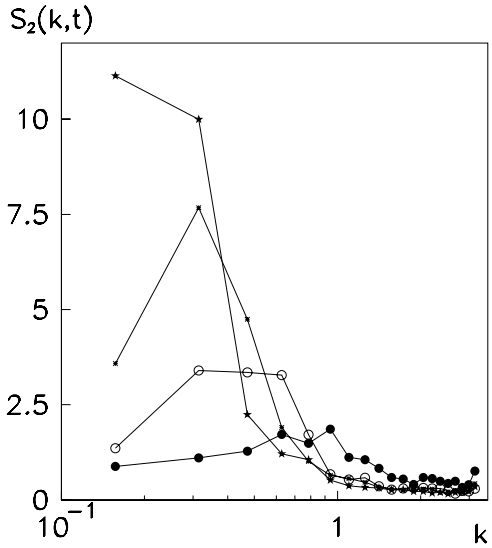

Figure 9. The dynamic structure factor, $S_2(k, t)$, for the large particles on the bottom layer, during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 0.35$, as a function of the wavevector k, for $t = 100$, 2000, 10 000, 40 000 (from bottom to top).

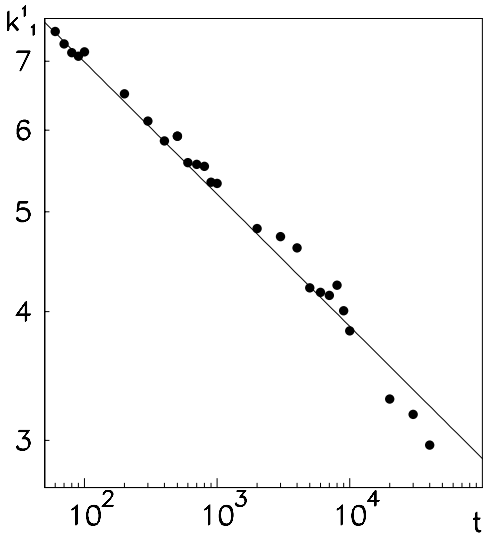

Figure 10. The first moment $k_1^1(t)$ of $S_1(k, t)$, as a function of t, during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 0.35$. The full line is a power law, $t^{-\alpha}$, with an exponent $\alpha \simeq 0.13$. At long times the data can be also fitted by a power law with a higher exponent, $\alpha \simeq 0.18$.

5. Conclusions

In this paper we have shown how a statistical mechanics approach in the manner of Edwards can be used to interpret the behaviour of granular media. In the simple hard-sphere binary mixture studied here, the phenomena of size segregation observed during the tap dynamics can be well explained within such a statistical mechanics framework.

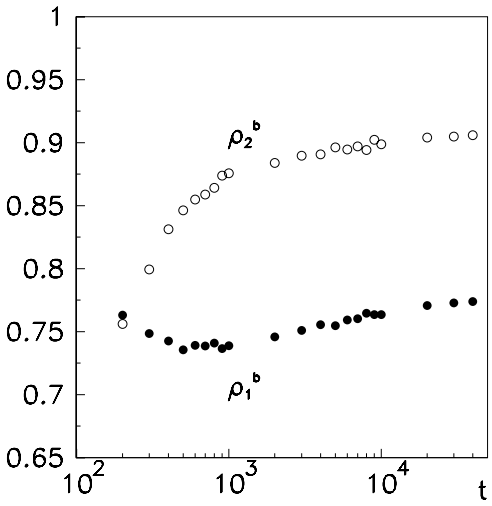

Figure 11. The fractions of the small and large particles on the bottom layer, $\rho_1^b(t)$ and $\rho_2^b(t)$, as a function of time, t, during the tap dynamics with tap duration $\tau_0 = 10$ MCS and tap amplitude $T_\Gamma = 0.35$.

Summarizing, N_1 small particles and N_2 large particles are initially prepared in a random loose stable pack, and then evolved using tap dynamics. Both vertical and horizontal size segregation are observed and, in particular, a crossover from the 'Brazil nut' effect to the 'reverse Brazil nut' effect. As the tap amplitude is reduced, larger clusters made up of one type of particle appear, with properties strongly resembling those of a phase separation process.

By solving the effective Hamiltonian, equation (3), in a mean field approximation [20, 23], the crossover from the 'Brazil nut' effect to the 'reverse Brazil nut' effect is found to correspond to the drift of the minimum of the free energy, $F = -\ln Z$, in the inherent space from negative to positive values of the vertical segregation parameter, Δh.

From the present data, horizontal segregation and its patterns instead seem to be due to the presence of a critical point (see footnote 3) in the effective Hamiltonian and further investigations are under way [23].

Acknowledgments

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