Experimental observations of oscillations and segregation in a binary granular mixture

Sparisoma Viridi, Malte Schmick, and Mario Marku[s*](#page-0-0)

Max-Planck-Institut für molekulare Physiologie, Postfach 500247, 44202 Dortmund, Germany (Received 14 September 2005; revised manuscript received 8 August 2006; published 3 October 2006)

We present experimental evidence for a "granular clock." We use a setup in which vertical vibrations (10-20 Hz) of a bidisperse granular medium cause horizontal oscillations (periods in the order of minutes) between two connected compartments. Moreover, we present the first experimental evidence for full segregation of smaller particles into one compartment, leaving the larger ones in the other compartment. Simulations, using a simple model, describe phenomenologically the observed oscillations.

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: $45.70.Mg$, $05.45.-a$, $05.65.+b$, $47.54.-r$

Granular media are in a state that behaves similar to a solid or a liquid or a gas; moreover, these media have their own, distinct behavior as well. An example is clustering in a granular gas. This phenomenon is due to a larger dissipation of energy due to inelastic collisions between the particles. Since dissipation is larger in dense regions, these regions become even denser. In the present work we will show experimentally and in simulations that clustering can alternate periodically between two connected compartments. The energy losses by dissipation are compensated in our setup by vertical vibrations of the bottom of the compartments. This phenomenon has been predicted by simulations $[1,2]$ $[1,2]$ $[1,2]$ $[1,2]$ and has been called "granular clock" $\lceil 2 \rceil$ $\lceil 2 \rceil$ $\lceil 2 \rceil$.

A different type of oscillations has been reported in the literature for granular media; it is associated with the tilting occurring at the boundary between two types of vibrated particles $[3]$ $[3]$ $[3]$. Still another type of reported oscillations is that happening in the (vertical) "ticking hour glass" [[4](#page-3-3)], which also occurs in flows out of vertical containers $\lceil 5 \rceil$ $\lceil 5 \rceil$ $\lceil 5 \rceil$.

In addition to the "granular clock," we will show that for appropriate parameters the smaller particles leave the mixture and cluster irreversibly in one compartment while the larger ones remain in the other compartment. Other forms of particle segregation have been investigated, e.g., in rotating drums or boxes, horizontal or vertical shaking and chute flows $[6–8]$ $[6–8]$ $[6–8]$ $[6–8]$. However, in such devices the direct contact of layers with different particles make a final separation difficult if optimal segregation is desired, e.g., in mineral processing $[9,10]$ $[9,10]$ $[9,10]$ $[9,10]$ or in resource recovery from waste $[11]$ $[11]$ $[11]$. Therefore, segregation into separate compartments, as investigated here, is a promising task.

A number of findings in experiments and simulations has hitherto been reported for shaken granular media in compartmentalized systems $\left[1,2,12-18\right]$ $\left[1,2,12-18\right]$ $\left[1,2,12-18\right]$ $\left[1,2,12-18\right]$ $\left[1,2,12-18\right]$ $\left[1,2,12-18\right]$. It was shown for monodisperse systems that particles accumulate preferentially in one of two compartments $[12,13]$ $[12,13]$ $[12,13]$ $[12,13]$. Related investigations were afterwards performed for more than two compartments [[14](#page-3-13)[,15](#page-3-14)]. For bidisperse systems and two compartments, it was shown experimentally that all particles cluster in one compartment; the compartment at which this occurs depends on the initial distributions of particle sizes in the box $[16]$ $[16]$ $[16]$. Later on, simulations showed that in shaken bidisperse system, partial segregation can take place $\lceil 17 \rceil$ $\lceil 17 \rceil$ $\lceil 17 \rceil$. If the shaking is sufficiently strong, a particle mixture is distributed symmetrically among the compartments $[16,18]$ $[16,18]$ $[16,18]$ $[16,18]$. Neither oscillations nor a stationary segregation of a bidisperse system into monodisperse subsets, distributed in different compartments, have hitherto been reported for any experimental setup.

In our experiments, we used spheres of soda lime glass (Sigmund Lindner, Germany). If not stated otherwise, we put T_S =138 small spheres (diameter d_S : 2 mm) and T_L =27 large spheres (diameter d_L : 4 mm) in a polystyrene box (height: 7.7 cm, base: 0.73 cm \times 5 cm). This box was partially divided into two equal compartments by a vertical polystyrene barrier (height *h*: 3 cm; thickness: 0.8 mm; see Figs. [1](#page-1-0) and [2](#page-1-1)). Note that for easier visualization the setup is quasi-twodimensional, its depth being less than twice the largest particle diameter. It is not obvious that the observed phenomena would occur in a fully three-dimensional system. The box was closed on the top and mounted on a sinusodial shaker with adjustable frequency *f*. For a given *f*, we set the amplitude *a* as follows. A control knob in our setup allowed us to change $\Gamma = (2\pi f)^2 a/g$, i.e., the maximum acceleration relative to g . The value of Γ was monitored by an analog accelerometer (Silicon Designs, Inc.; Model 1221L-010) until the desired amplitude *a* was attained. Previous investigations (e.g., Refs. $[14-16]$ $[14-16]$ $[14-16]$) indicate that the relevant control parameter for the driving of the system is $v = af$. As initial condition, we mixed the particles and poured all of them into one of the compartments. At this point we observed electrostatic charging, as some of the smaller glass spheres more or less "levitated" over the mixture. We counteracted this by directing a faint stream of air with water vapor (temperature \approx 35 °C) onto the device; the spheres then settled down after a few seconds and we did not observe any signs of electrostatic charging within the next hour, which is longer than the duration of any of our experiments. The experiments were monitored with a Basler A602f video camera at a rate of 250 frames per second.

Altogether, we observed four regimes in the experiments: (i) mixing with symmetrical distribution among the two compartments; (ii) oscillations [Fig. [1](#page-1-0) and Figs. $3(a)-3(c)$ $3(a)-3(c)$]; (iii) full segregation of large and small particles (Fig. [2](#page-1-1)); and (iv) the particles remain as in Fig. $1(a)$ $1(a)$ or $2(a)$ $2(a)$ i.e., in the initial mixture within one compartment. The transition between regimes (i) (symmetry) to (ii) (oscillations) was ob-

^{*}Electronic address: markus@mpi-dortmund.mpg.de; URL: www.mpi-dortmund.mpg.de/markus

FIG. 1. Experimentally observed oscillations. (a) Initial configuration consisting of a mixture of small and large particles in the right compartment. (b) Only the small particles get sufficiently fast to overcome the barrier. (c) Segregation: the small particles are at the left and the large ones are at the right. (d) Due to reduced dissipation, the large particles now gain sufficient energy to get over the barrier. (e) The right compartment has become emptied of large particles; the configuration is nearly symmetrical to that in the left-most picture and the process starts in the opposite direction. $a=6$ mm, $f=20$ Hz. Times: (a) 0, (b) 3.1, (c) 58.3, (d) 66.2, (e) 103.2 s.

served at $v \approx 0.17$ m/s. The transition between regimes (ii) and (iii) occurred at $v \approx 0.11$ m/s, and the transition between regimes (iii) and (iv) occurred at $v \approx 0.08$. Figures [3](#page-2-0)(a)-3(c) were obtained by counting (in the video recordings) the numbers of particles that exchange compartments and then adding to or subtracting from the total numbers before. The ex-perimentally observed oscillations (as shown in Fig. [1](#page-1-0)) of the bulk of spheres between compartments in regime (ii) occur with a oscillation-period *T*, which depends on the driving parameter $v = af$. This dependence is shown in Fig. [4.](#page-2-1) *T* was determined as the average over at least 6 periods, each period being defined by the distance between successive lower intersections of the curve *n* vs *t* with the curve *m* vs *t*. The oscillations can be considered periodic, because the fluctuations of T are negligible ($\leq 9\%$ within each time series). Note in Fig. [4](#page-2-1) that the period tends to diverge to infinity slightly below $v \approx 0.11$ m/s; at lower values of *v*, full segregation occurs.

Figure [5](#page-2-2) shows how the particle sizes, for which full segregation occurs, can be regulated by the frequency *f*. In other words, one can "sieve" a certain range of particle sizes by adjusting the shaking frequency. For the experiments in Fig. 5 we tested, at each frequency f , all pairs of diameters (in mm) within the set {0.8,1,1.6,2,2.4,2.5,3,3.2,4,4.5,4.8, 5,5.6,6.

When the particles were mixed, we generally observed a phenomenon that is reminiscent of the so-called reverse brazil nut (RBN) effect $[19,20]$ $[19,20]$ $[19,20]$ $[19,20]$, meaning that larger particles go to the bottom, contrary to the brazil nut (BN) effect. There are simulations $[19]$ $[19]$ $[19]$ and observations $[20]$ $[20]$ $[20]$ of the RBN, but there are also doubts on its existence $[21]$ $[21]$ $[21]$. We can vaguely relate our own observations to the RBN in the sense that the large particles are trapped (mixed with small ones) in the bulk, while small ones appear without large ones in a thin granular gas on the top [Figs. $1(a)$ $1(a)$, $1(b)$, $1(e)$, $2(a)$ $2(a)$, and $2(b)$].

This configuration causes an outflow of the small particles from the compartment [Figs. $1(b)$ $1(b)$, $1(e)$, and $2(b)$ $2(b)$]. While this outflow occurs, there is a steady supply of small particles drifting from the lower mixture to the thin granular gas at the top.

In addition to these experimental observations, we tried a theoretical approach, as well. Mikkelsen *et al.* [[16](#page-3-15)] developed a model for a vertically shaken, bidisperse granular medium; it contains the rough approximation that a granular temperature, which is the same for both species, can be defined. This model describes well the clustering of particles into one compartment, but (as far as we examined it) does not render oscillations. However, oscillations, as well as segregation and symmetrical distribution of particles, similar to our experimental results, were predicted in calculations by Costantini *et al.* [[1](#page-3-0)] and by Lambiotte *et al.* [[2](#page-3-1)]. Both groups of authors performed simulations using molecular dynamics (MD), as well as using ordinary differential equations. However, when comparing their simulations with our experiments, one encounters difficulties for the following reasons: (a) the simulations by Costantini et al. were done for particles with equal diameters (unlike our setup) and different masses; (b) the MD simulations by Lambiotte et al. were done for different diameters (as in our setup) but with equal masses (unlike our setup); (c) the simulations by Costantini *et al.* and the MD simulations by Lambiotte *et al.* were done for a 2D system, while our setup has a finite extension into the third dimension; (d) the simulations by Lambiotte et al. yield the BN effect (the larger particles are on top), contrary to our observations, in which the smaller particles are (mainly) on top (RBN). As a result of the BN effect in Ref. [[2](#page-3-1)], the oscillations of the larger particles are (unlike our experiments) slightly phase shifted to earlier times with respect to the oscillations of the smaller particles. Note that the appearance of the RBN or the BN effect is sensitive to the

FIG. 2. Experimentally observed full segregation. $a=4.7$ mm, $f=20$ Hz. Times: (a) 0, (b) 43.3, (c) 392.2, (d) 401.5, (e) 931.9 s. The system, as shown here, gets stuck in the configuration shown in Fig. $1(c)$ $1(c)$. The reason for this is that the large particles do not get enough energy to overcome the barrier, even after all the small particles have left the right compartment, and have thus reduced the dissipation.

FIG. 3. Typical experimental time series of oscillations of the fraction of large *m*, dashed curve) and small $(n,$ continuous curve) spheres in one compartment. $f=20$ Hz. (a) $a=5.6$ mm. (b) $a=6$ mm. (c) $a=6.5$ mm. (d) Simulated oscillations $(a=6.5 \text{ mm}, \alpha=47.8 \text{ s}^2 \text{ m}^{-2}, \lambda=0.01, R$ $= 1.5$; abscissa: arbitrary units).

system parameters in a complicated way and there is as yet no conclusive knowledge on their influence (see outliers in Ref. $[20]$ $[20]$ $[20]$ and irreproducibilities $[21]$ $[21]$ $[21]$).

In order to compare simulations with experiments, we aimed to adapt one of the existing models yielding oscillations. A promising model is the detailed, quantitative approach (yielding the RBN effect, as in our experiments) described by six mean-field differential equations in the work of Costantini *et al.* However, we decided to leave an adaptation of this model to the future, because of its complexity. Instead, we favored minimality, adapting the four differential equations by Lambiotte *et al.*, which we write here as follows:

$$
\frac{\partial n}{\partial t} = -nF(n,m)P(S_L) + (1 - n)F(1 - n, 1 - m)P(S_R), (1)
$$

$$
\frac{\partial m}{\partial t} = -mF(n,m)Q(S_L) + (1-m)F(1-n,1-m)Q(S_R),
$$
\n(2)

FIG. 4. Experimentally observed period of the oscillations vs the driving parameter $v = af$.

$$
\frac{\partial S_L}{\partial t} = \lambda (nm - S_L),\tag{3}
$$

$$
\frac{\partial S_R}{\partial t} = \lambda \left[(1 - n)(1 - m) - S_R \right],\tag{4}
$$

where

$$
F(n,m) = e^{-(n^2 + Rm^2)/A}.
$$
 (5)

 n (m) is the proportion of small (large) particles in the left compartment. We set

$$
A = \alpha(af)^2 \tag{6}
$$

(see Refs. $[14-16]$ $[14-16]$ $[14-16]$); we shall determine the constant α by fitting the model's transitions to the transitions in our experiments. S_L (S_R) is a measure of the vertical segregation at the

FIG. 5. Experimentally determined conditions for full segregation. For each frequency f (abscissa) full segregation occurs if and only if the larger particles have a diameter Φ_L above the upper point (\blacksquare) and are mixed with smaller particles with a diameter Φ_S below the lower point (\bullet) . $a = 4.7$ mm.

left (right) compartment. P and Q describe the influence of the vertical segregation on the fluxes. Lambiotte *et al.* wrote $P(x) = 1 - x$ (*x*: *S_R* or *S_L*) in order to account for the inhibition of the flux of small particles by the large particles above them, due to the BN effect. In addition, they wrote $Q(x)=1$ $+x$ to account for the activation of the flux of large particles by the small particles below them. In order to account for our observed RBN effect, we changed $Q(x)$ by writing

$$
Q(x) = 1 - x.\tag{7}
$$

In analogy to the equations above, we could write $P(x)$ $=1+x$, in order to account for the activation of the flux of small particles by the large particles below them (RBN effect). However, the ansatz $P(x) = 1 + x$ yielded no oscillations. Instead, we did find oscillations if $P(x) \in [0,1]$, just as $Q(x) \in [0,1]$, and thus we set

$$
P(x) = x.\t\t(8)
$$

Equations (1) (1) (1) - (4) (4) (4) , under consideration of Eqs. (5) (5) (5) - (8) (8) (8) , yielded oscillations, segregation, and symmetrical distribution. Contrary to Ref. $[2]$ $[2]$ $[2]$ and due to Eqs. (7) (7) (7) and (8) (8) (8) , which account for our observed RBN effect, the oscillations of the small particles are phase shifted to earlier times with respect to those of the larger particles. This is exemplified in Fig. $3(d)$ $3(d)$ and roughly resembles the experiments, as shown in Figs. $3(a) - 3(c)$ $3(a) - 3(c)$. We obtained transitions between the regimes (i) - (iv) at the following values of $A: 1.24, 0.4$, and 0.175. Considering the experimental values of $v = af$ at these transitions, as given above, we estimate α =47.8±6.1 s² m⁻². In spite of this quantification, we want to stress that the modelling of the present system remains a challenge, since our model is only a rudimentary description of the experiments. In fact, the physical interpretation of S_R and S_L is vague and the influence of these variables on the fluxes is only sketchy.

The oscillations we observed in the experiments, as exemplified in Figs. $3(a)-3(c)$ $3(a)-3(c)$, are periodic. Note that whenever we measured a new time series for a given set of control parameters, we obtained slightly different periods, as shown by the spread of values in Fig. [4;](#page-2-1) however, within each time series, the oscillations were indeed periodic apart from fluctuations of less than 9% of the period. This periodicity is consistent with our simulations, with those by Lambiotte *et al.* [[1](#page-3-0)] and with the mean-field calculations of Costantini *et al.* [[2](#page-3-1)]. In contrast, the MD simulations of Costantini *et al.* yielded aperiodic oscillations with an exponential distribution of the times of residence in one compartment. By virtue of the observed periodicity, our oscillations may be described as those of a "self-resetting hour glass": after one compartment becomes empty, it autonomously refills again, the process repeating periodically. Of course, our "hour glass" is very particular, regarding its quasi-two-dimensional nature; investigations of deeper boxes, i.e., of fully threedimensional systems remains open. The "sieving by shaking," quantified in Fig. [5,](#page-2-2) may well be relevant to the processing of materials $[9-11]$ $[9-11]$ $[9-11]$.

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