

Phase transitions and segregation phenomena in vibrated granular systems

Christof A. Kruelle¹, Sébastien Aumaître², Andreas P. J. Breu¹, Andreas Goetzendorfer¹, Tobias Schnautz¹, Rafał Grochowski³, and Peter Walzel³

¹ Experimentalphysik V, Universität Bayreuth, D-95440 Bayreuth, Germany

² Laboratoire de Physique Statistique, ENS, F-75231 Paris, France

³ Mechanische Verfahrenstechnik, Universität Dortmund, D-44227 Dortmund, Germany

Abstract. A summary of results is presented from experiments in granular systems, which are excited by vertical and/or horizontal vibrations. The transitions between different dynamic states depend on internal properties of the granular system like the density of particles as well as external parameters of the driving shakers. Characteristic for granular systems are counterintuitive phenomena like the crystallization by increasing the vibration amplitude and thereby the energy input, or the rise of large particles in a sea of smaller ones (Brazil-nut effect). For horizontal shaking of a binary system the demixing of small and large particles is found to occur at the same critical particle density as the liquid-solid transition, which leads to the conclusion that both phenomena, segregation and phase transition, are closely related.

1 Introduction

An accumulation of macroscopic grains, set in motion by an external driving force, can show a surprising behavior. A peculiar phenomenon – called segregation – occurs as soon as heterogeneous particles are implied, in geophysical rock avalanches as well as in processed powders in the food or chemical industry [1,2]. This demixing of grains, which differ in size, density, or surface properties, has been intensively studied since the 1990s in laboratory experiments, e.g. in rotating drums [3] and under vertical [4] or, more recently, horizontal linear [5] excitation. Another phenomenon frequently encountered when handling granular material is the transition from a disordered phase to a more organized state, when the density of grains is increased beyond a critical value. This can be observed, for instance, in monodisperse particles under vertical vibration without gravity [6] or under horizontal translational excitation [7].

In this brief review we report on experimental investigations of both phase transitions and segregation phenomena in granular systems, which are agitated by three different vibration exciters. When the particles are externally forced to perform stochastic movements a ‘granular temperature’ can be defined as mean kinetic energy in the center-of-mass system. For describing

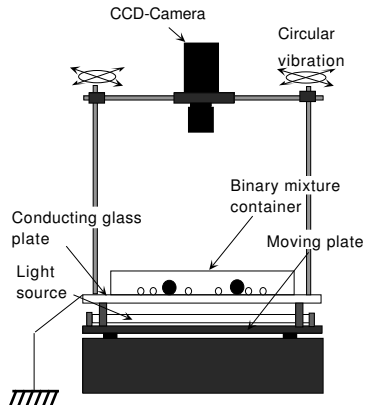


Fig. 1. Sketchy representation of the experimental device for the horizontal circular vibration of a monolayer of glass beads. The diameter of the container is 29 cm. The bottom glass plate is coated with an electrically conducting indium-tin oxide layer to suppress static electricity. From [8].

the physical properties of this ensemble one can ask – in analogy to thermodynamic phase transitions: (i) Are there critical temperatures at which the internal structure undergoes qualitative changes? (ii) Which order parameters characterize the dynamics of the transition? (iii) What are the consequences for granular mixtures?

2 Horizontal circular vibration

Here we present a model system, consisting of two species of glass beads with different size rolling in a horizontal shaker, where both phenomena, segregation and phase transition, are found to be closely related since they occur at about the same granular density [8].

A sketch of our experimental device is shown in Fig. 1. The particles in the dish are excited in a *horizontal circular vibration*, i.e. a circular movement of the entire platform due to the superposition of two sinusoidal vibrations in perpendicular directions. The frequency f of the table motion can be tuned from 0.5 to 2.0 Hz, with a preset amplitude $A = \frac{n}{8} \cdot 2.54$ cm, where $n = 2, \dots, 8$. The granular system is composed of a various number, N , of spheres with diameters $d = 0.4$ or 1.0 cm. To obtain a size-independent control parameter the filling fraction μ is defined as the total cross-sectional area $N \cdot \pi (d/2)^2$ of all spheres divided by the surface $\pi (D/2)^2$ of the cell with diameter $D = 29$ cm. During the motion, the positions of all particles are captured with a charge-coupled-device (CCD) camera fixed to the moving table.

During the experiments we noticed global changes in the dynamics of the system while crossing a critical threshold of the filling fraction μ . In particular, the spheres became arranged in regular, triangular patches. To study this phenomenon, a monolayer of *monodisperse* glass spheres is placed inside the dish. After an early stage, during which the system loses all traces

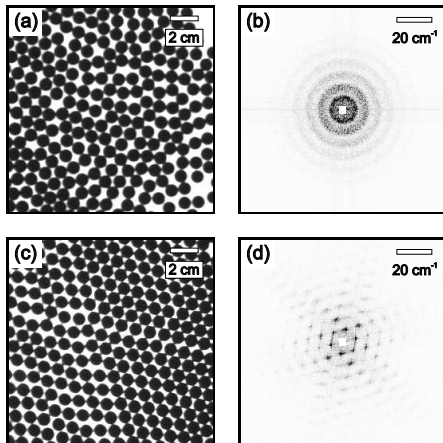


Fig. 2. (a) Image of a monolayer of 410 spheres (filling fraction $\mu = 0.49$) rolling on a table shaken with an amplitude $A = 2.22$ cm and a frequency $f = 1.67$ Hz. (b) Contour plot of the corresponding 2D power spectrum. (c) Image of a monolayer of 580 spheres ($\mu = 0.69$) under the same excitation, and (d) its 2D power spectrum. The snapshots are taken in a square of 15 cm at the center of the cell.

of the arbitrary initial configuration, an image is taken. To underline the developed structured state a 2D Fourier transformation (FFT) is performed. Results of these measurements are shown in Fig. 2, for low and high filling fractions of spheres. At low density, the configuration of grains does not show any structures. Its 2D power spectrum displays a continuous intensity distribution within a circle of radius $k_d = 2\pi/d$. In contrast, at high density, the small spheres arrange in a triangular lattice. In the 2D power spectrum six peaks appear at a wave number $k_0 = 2\pi/\frac{\sqrt{3}}{2}d$, evenly spaced by an angle of $\pi/3$.

The dynamics of the particles is obviously different in these two regimes. At low density each particle is free to follow its own trajectory until it collides with its neighbor, like in a fluid. On the other hand, at high density, the particles are forced into a collective motion inside a 2D crystal. The crossover between these two regimes is reminiscent of a liquid-solid transition.

To specify the transition point, a characteristic order parameter is extracted from the power spectra. The spectral intensity is integrated radially in an annulus $0.98 k_0 < k < 1.02 k_0$ around the expected peaks. The resulting averaged intensity defines a function of the azimuthal angle φ . In the structured state this function is supposed to present six equidistant peaks, see inset of Fig. 3. To obtain a *single* characteristic number, which quantifies the order of the structured state, the angular space $[0, 2\pi]$ is subdivided into six equal parts. The intensities in each interval are summed up yielding a singly peaked function $I(\varphi)$ in the reduced angular range $\varphi \in [0, \pi/3]$. Dividing $I(\varphi)$ by its arithmetic mean yields a normalized intensity

$$I_n(\varphi_i) = \frac{I(\varphi_i)}{\frac{1}{i_{\max}} \sum_{i=1}^{i_{\max}} I(\varphi_i)}, \quad \varphi_i = i \cdot \frac{\pi}{3i_{\max}}, \quad i_{\max} = 12,$$

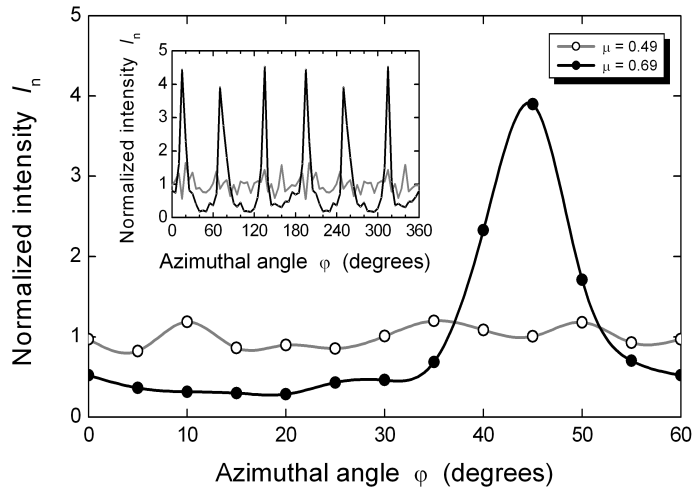


Fig. 3. Normalized intensity I_n of the power spectrum for $\mu = 0.49$ ($N = 410$) and $\mu = 0.69$ ($N = 580$), respectively. The absolute maxima $I_{n,\max}$ are extracted to obtain the order parameter. The inset shows the full range of intensities within rings of same width around the centers of the 2D power spectra.

from which an order parameter

$$\alpha \equiv I_{n,\max} - 1$$

is derived. The open circles in Fig. 3 show that at a low density of spheres, where no coherent structure exists, no peak survives this analytic procedure ($I_{n,\max} = 1.2$), whereas at a high density a pronounced peak remains (solid circles in Fig. 3). Its height $I_{n,\max} = 4.0$ reflects the degree of symmetry established in the triangular lattice of the particles.

The values of α versus the filling fraction μ are shown in Fig. 4 for the driving conditions $A = 2.22$ cm and $f = 1.67$ Hz. A *liquid-solid like* transition can be observed around a critical filling fraction $\mu_{c,ls}$.

For the segregation experiments, a binary mixture of glass beads consisting of 19 large particles of diameter $d_l = 1.0$ cm imbedded in a variable number of small spheres with diameter $d_s = 0.4$ cm are prepared. Initially the large spheres are placed on a regular triangular lattice with 3 cm spacing between the centers of two nearest neighbors. This distance $L_{nn}(t)$, averaged over all the 19 large spheres, is measured in real time [9]. For a clear characterization of the segregation the mean value of the distance between nearest neighbors in the asymptotic regime, L_∞ , is measured as a function of the filling fraction μ . Here, a transition between a non-segregated state at low density and a segregated state at high density can be defined.

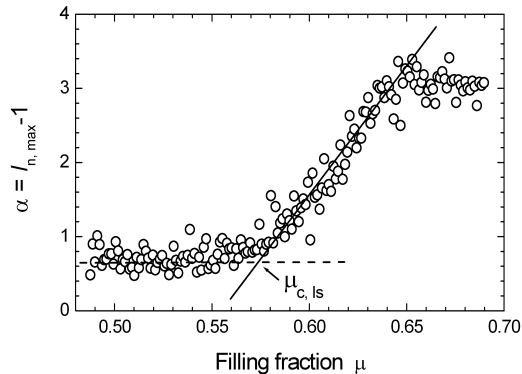


Fig. 4. Evolution of the order parameter α with the filling fraction μ . The critical point $\mu_{c,ls} = 0.57 \pm 0.01$ is found from the intersection of a linear fit of the data with $1.5 < \alpha < 3.0$ with the mean level of the precritical data.

Since both transitions depend on the shaker amplitude we explored this dependence further. Fig. 5 shows the resulting phase diagram of the critical filling fractions $\mu_{c,seg}$ and $\mu_{c,ls}$, respectively, vs. shaker amplitude A . These critical filling fractions seem to be independent on the driving frequency but decrease with increasing shaker amplitude. The regime where segregation occurs is always slightly above the liquid-solid transition line. This suggests that the granular phase transition is a precondition for segregation.

The transition is in the regime from 0.55 to 0.75 and depends on the amplitude of the driving shaker in a counterintuitive way: the liquid-solid transition line can be crossed by increasing the amplitude and thereby the energy input. A similar phenomenon was coined ‘freezing by heating’ in the context of driven mesoscopic systems [10].

We offer the following thoughts to elucidate the connection between the segregation in the binary mixture and the phase transition in the monodisperse layer of spheres. It seems that the granular material does not stay uniformly distributed but chooses the configuration which minimizes the energy input for a given number of particles and excitation parameters, in order to

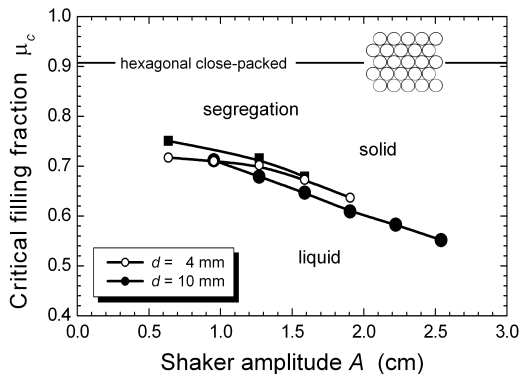


Fig. 5. Phase diagram of the critical filling fraction μ_c vs. shaker amplitude A showing the regime $\mu > \mu_{c,seg}$ (squares), where segregation occurs. The liquid-solid transition line $\mu > \mu_{c,ls}$ (circles) is obtained from the normalized Fourier intensity data for two different particle sizes. Data partially taken from [8].

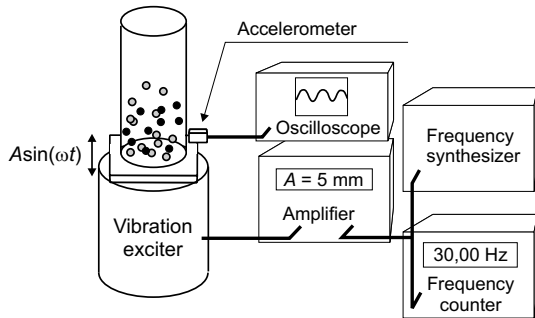


Fig. 6. Sketchy representation of the experimental device for the vertical vibration of a binary mixture of spheres. The diameter of the container is 9.4 cm.

reduce the amount of energy to be dissipated. For $\mu \leq \mu_c$ all particles move randomly with a uniform distribution, i.e., they prefer not to collide with the lateral wall. Indeed at low densities only few particle-wall collisions can be observed. We have measured that, in this case, large and small particles roll inside the dish in almost the same fashion. No segregation occurs. On the other hand, if the filling fraction exceeds μ_c , a boundary-hitting regime begins. This change can be seen and even heard in the experiment. In this regime the granules are continuously hit by the lateral wall. This process tends to decrease the extension of the monolayer which organizes itself in a more and more condensed state, leading to a triangular lattice, when the excitation is increased even further. Following the ideas of Edwards [11], this is also the reason why segregation occurs: beyond the critical filling fraction the collisions with the lateral wall become the dominant driving mechanism. The granular material is continuously compressed by the collisions with the lateral wall and therefore tends to increase its compactness by organizing itself in a triangular lattice. Since the larger particles disturb this ordering process the system pushes these intruders into the central region of the dish thereby reducing the number of holes and defects [12,13].

We conclude that the main mechanism for size segregation in our binary system is the compression force exerted by the lateral boundary and mediated by the developing lattice of the smaller spheres.

3 Vertical vibration

As demonstrated in the previous section, granular media consisting of small and large particles tend to de-mix when shaken vigorously enough. The ‘Brazil-nut effect’ – if a particle mixture is shaken *vertically*, the larger particles will end up on top of the smaller ones, like the nuts in a muesli package – became the *Drosophila melanogaster* of granular media research [4,14–17]. Numerical simulations could validate the rise of larger particles [18,19]. Proposed mechanisms are, for example, convective motion of the smaller particles, which drag the larger ones to the top [15], or the filling of voids by the small particles, thereby lifting the larger ones [4,18,19]. Contrary to these

Table 1. Densities and diameters of the spherical particles used in the experiments. Data as used in [25].

Material	ρ (g cm ⁻³)	d (mm)							
Wood	0.8	8				12			
Synthetic resin	1.1								
Polyurethane	1.2								
Polypropylene	1.5								
Glass	2.5	4	6	8	10				
Aluminum	2.6	2	6						
Steel	7.7	2	4	6	10				
Bronze	8.5								

common observations Shinbrot [20], however, noticed that a large particle, depending on its density, could also sink to the bottom of the container. Recent theoretical investigations [21–23] explained that both effects, the rise or descend of the larger particles, may occur. The latter case has become known as ‘reverse Brazil-nut effect’. The borderline between both effects has been predicted by Hong, Quinn, and Luding [21] in a simple relation between the size d_l/d_s and mass ratios m_l/m_s of the large and small particles.

One approach to describe an externally driven granular medium is to consider the individual particles as hard spheres. The driving could be – as in the case of the Brazil-nut effect – a vertical vibration of the container, which confines the granules. For strong driving, where all particles are in continuous motion, the ideas of the kinetic theory of gases can be applied. Then it is possible to define a granular temperature T in analogy to a gas, using the mean kinetic energy:

$$T \propto \frac{1}{N} \sum_i \frac{m_i}{2} (v_i - \langle v \rangle)^2, \text{ with } \langle v \rangle = \frac{1}{N} \sum_i v_i, \quad (1)$$

where N is the total number of particles, and m_i and v_i are the mass and the velocity of the i -th particle, respectively. Hong [24] calculated, on the basis of this model, a critical granular temperature $T_c = mgd\mu/\mu_0$, below which a system of monodisperse spheres condensates. This critical temperature depends on the diameter d of the particles and the initial filling height μ , measured in units of d . g denotes the gravitational acceleration and μ_0 is a constant, which depends on the spatial dimension and the underlying packing structure [24]. For a binary particle mixture different critical temperatures do exist, as pointed out by Hong *et al.* [21]. If a binary mixture of spheres is agitated by an external shaker, such that the granular temperature is in between the two critical values, one type of particle condensates while the other remains fluidized. It depends on the size and mass of the particles, which particle species will condensate and therefore sink to the bottom of the container. Following Rosato *et al.* [18], Hong *et al.* claimed that the cross-over condition is given when the ratio of the critical temperatures is equal to the

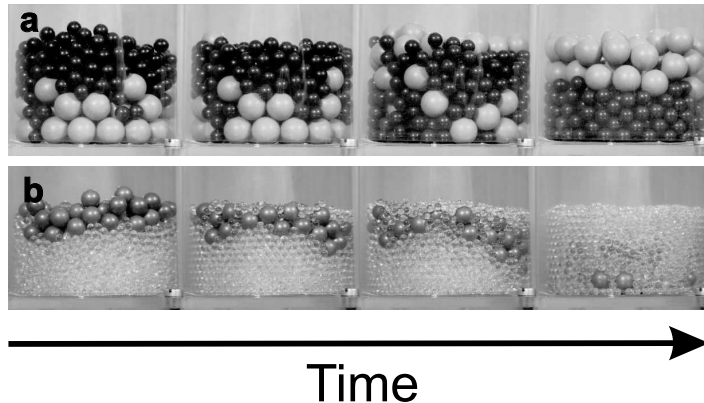


Fig. 7. Temporal evolution of the system. (a) Initially 8 mm glass beads on top of 15 mm polypropylene, which show the classical Brazil-nut effect. (b) 10 mm bronze spheres on 4 mm glass beads showing the reverse Brazil-nut effect. From [25].

volume ratio of the two particle species, which leads, in D dimensions, to the simple relation

$$\frac{d_l}{d_s} = \left(\frac{m_l}{m_s} \right)^{\frac{1}{D-1}}. \quad (2)$$

If, in $3D$, the diameter ratio is larger than the square root of the mass ratio, the particle mixture should show Brazil-nut effect and vice versa.

The prediction of formula (2) was put to the test [25] by preparing an instable layering, followed by a controlled shaking of the container. Our experimental device (see Fig. 6) operates at frequencies f between 0 and 100 Hz and normalized accelerations $\Gamma = A(2\pi f)^2/g$ up to 40, where A is the shaking amplitude and g the gravitational acceleration. The acceleration Γ is measured with an accelerometer attached to the base plate of the Perspex cylinder. The granular materials used were spherical particles with properties listed in Table 1.

In order to check whether a binary mixture would show Brazil-nut behavior or its reverse, we prepared a presumed instable configuration, i.e., we put several layers of the type, which were predicted by (2) to rise during shaking on the bottom of the Perspex vessel. On top of these we placed one or more layers of the second type. If the prediction was correct, we would observe that the particles on top started to move through the layers of the other particle type, ending up at the bottom of the vessel (see Fig. 7). On the other hand, if the prediction turned out to be wrong the initial layering would be stable.

Most experiments have shown clearly either the Brazil-nut effect (Fig. 7(a)) or the reverse form (Fig. 7(b)). For some particle combinations a mixed state was stable. An overview of the results of the experimental tests as well as

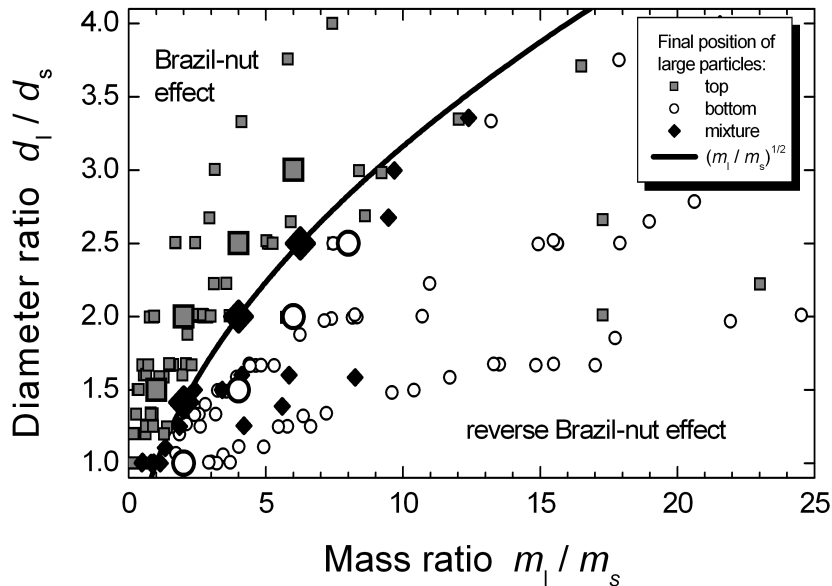


Fig. 8. Phase space for particle properties. The plot shows the regimes where reversed and classical Brazil-nut effects occur depending on the particle properties. Each small symbol represents one of 178 experiments (data taken from [25]). The solid line separating both areas is given by expression (2). The large symbols indicate the prediction of 3D molecular-dynamics simulations performed by Hong *et al.* [21] with up to 3600 particles.

numerical simulations by Hong, Quinn, and Luding [21] together with their theoretical borderline are shown in Fig. 8. For 81% of the tested combinations (145 out of 178) the prediction of (2) was correct. The prediction failed when one particle type was made of aluminum or polyurethane. We assume that for these materials the condition of hard spheres, which is one of the main propositions in the theory was not met. We also noticed that, in the case of the reverse Brazil-nut effect, it is crucial to choose an appropriate filling height. It turned out that the effect is completely destroyed, if the initially lower layer is too large.

We currently focus on experimental verifications of the premise of this segregation mechanism via condensation. If, at a fixed external driving, a monodisperse set of glass beads exceeds a critical number of particles a phase transition can be observed from a fluidized ‘gas-like’ state to a condensed ‘crystalline’ state (see Fig. 9). Systematic studies of the dependence of the condensation temperature T_c on the internal parameters (number, size, and material density of the spheres) as well as the external conditions (amplitude and frequency of the shaker) are promising [26].

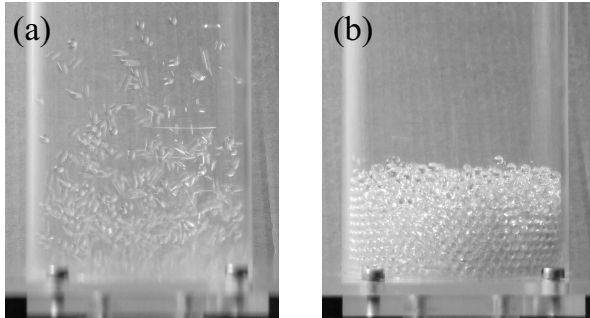


Fig. 9. Behavior of an ensemble of monodisperse glass beads (diameter = 4 mm) at fixed acceleration: (a) granular gas for subcritical filling, (b) crystalline block for supercritical filling.

4 Vertical circular vibration

The combined action of a horizontal *and* vertical vibration of the support has been utilized already for a long time for the controlled transport of bulk cargoes in a whole variety of industrial processes [27–29]. Since the transport phenomena on these so-called ‘vibratory conveyors’ involve the nonlinear interaction of many-particle systems with complex behavior leading to self-organized spatiotemporal patterns, the investigation of their dynamical properties has become a challenging subject to physicists, too.

In this final section we report on a conveyor system designed as a prototype apparatus specially developed to investigate the transport under principal oscillation modes, i.e. linear, elliptical, and circular for a long running time, without disturbing boundary conditions [31,30]. In our experiments, the transporting trough has the form of a horizontally oriented ring with radius $R = 22.5$ cm. The motion of the ring can be described by a trajectory performed on a cylindrical surface, consisting of a vertical oscillation $z(t) = A \cos(\omega t)$ superposed with a torsional vibration $\phi(t) = A/R \cos(\omega t + \varphi)$ around the symmetry axis of the apparatus, where φ is the fixed phase shift between the two oscillations. If, for example, the phase shift φ is chosen to be $\pi/2$, then each point on the trough traces a circular path in a vertical plane tangent to the trough at that point. In short, the support agitates the granules via a *vertical circular vibration*.

The dimensionless acceleration Γ of the conveyor can be varied by changing the rotation frequency f of the driving units in the range $0 < \Gamma < 7$. The granulate used consists of $\approx 520\,000$ glass beads with diameter $d = 1.1$ mm, yielding a layer height of $\approx 10 d$.

Below a critical value $\Gamma_c \approx 0.45$ the grains stay at rest, i.e., they follow the agitation of the tray without being transported. The onset of particle movements is restrained by frictional forces between grains and the substrate. For accelerations above this threshold the granular material sets in motion. Individual particles are unblocked and begin to move freely on top of each other. A net granular flow with constant velocity is observed.

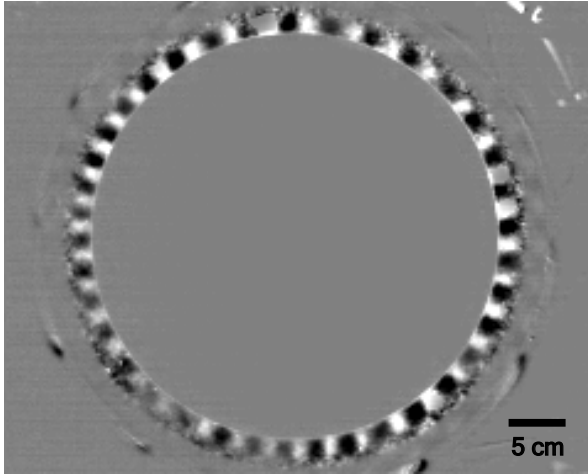


Fig. 10. Granular surface waves ($\lambda = 3.8$ cm) inside a vibratory conveyor with vertical circular motion of the annular trough oscillating with $\Gamma = 3.0$ at a frequency $f = 22.4$ Hz.

If Γ exceeds 1, the vertical component of the circular acceleration will cause the grains to detach from the bulk followed by a flight on a ballistic parabola. This results in a much less dense-packed, ‘fluidized’, state with highly mobile constituents. In a certain driving range at around $\Gamma = \pi$ a locking of the time-of-flight between successive bounces and the period of the circular vibration occurs. The initially flat bed becomes destabilized, and undulations of the granular surface occur [31]. In contrast to the surface waves observed earlier in purely vertically vibrating systems [32–35], here, the wave crests are not stationary but propagate along the trough. The relationship between the wave speed and the transport velocity of the particles is currently under investigation.

5 Conclusion

These three experiments indicate that the four major concepts describing the complex behavior of a vibrated granular system, namely phase transition, segregation, pattern formation, and transport are closely related and yield a rewarding field for future research.

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