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## Coarsening in granular systems

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## ABSTRACT

We review a few representative examples of granular experiments or models where phase separation, accompanied by domain coarsening, is a relevant phenomenon. We first elucidate the intrinsic non-equilibrium, or athermal, nature of granular media. Thereafter, dilute systems, the so-called "granular gases", are discussed: idealized kinetic models, such as the gas of inelastic hard spheres in the cooling regime, are the optimal playground to study the slow growth of correlated structures, e.g., shear patterns, vortices, and clusters. In fluidized experiments, liquid–gas or solid–gas separations have been observed. In the case of monolayers of particles, phase coexistence and coarsening appear in several different setups, with mechanical or electrostatic energy input. Phenomenological models describe, even quantitatively, several experimental measures, both for the coarsening dynamics and for the dynamic transition between different granular phases. The origin of the underlying bistability is in general related to negative compressibility from granular hydrodynamics computations, even if the understanding of the mechanism is far from complete. A relevant problem, with important industrial applications, is related to the demixing or segregation of mixtures, for instance in rotating tumblers or on horizontally vibrated plates. Finally, the problem of compaction of highly dense granular materials, which is relevant in many practical situations, is usually described in terms of coarsening dynamics: there, bubbles of misaligned grains evaporate, allowing the coalescence of optimally arranged islands and a progressive reduction of the total occupied volume.

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## R É S U M É

On décrit quelques exemples représentatifs d'expériences et de modèles concernant des matériaux granulaires, dans lesquels apparaît une séparation de phases avec formation de domaines de plus en plus grands (*coarsening*). On précise d'abord la nature intrinsèque des matériaux granulaires hors de l'équilibre, dans lesquels la température n'intervient pas. On discute ensuite les systèmes dilués appelés «gaz granulaires» : des modèles cinétiques idéalisés, tels que des gaz de sphères dures inélastiques en cours de refroidissement, sont l'outil idéal pour étudier la croissance lente de structures corrélées, par exemple des figures de cisaillement, des tourbillons et des amas. Dans des expériences en milieu liquide, on a observé une séparation liquide–gaz ou solide–gaz. Dans le cas de monocouches de

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particules, la coexistence de phase et la croissance des domaines apparaît dans diverses situations, quand on fournit de l'énergie mécanique ou électrostatique. Des modèles phénoménologiques décrivent, au moins qualitativement, divers résultats expérimentaux, concernant la dynamique de *coarsening* aussi bien que la transition dynamique entre les différentes phases granulaires. L'origine de la bistabilité semble liée à une compressibilité négative, révélée par des calculs d'hydrodynamique granulaire, mais le phénomène est loin d'être bien compris. Un problème important, notamment pour ses applications industrielles, est celui de la décomposition des mélanges ou de leur ségrégation, par exemple dans des tambours tournants ou sur des plaques vibrantes horizontales. Enfin, le problème du compactage des matériaux granulaires très denses, qui se pose dans bien des situations pratiques, est ordinairement décrit comme un *coarsening* : des bulles de grains mal alignés s'évaporent, ce qui permet la coalescence des îlots bien disposés et une réduction progressive du volume total occupé.

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## 1. Introduction

Granular systems are substances made of many *grains*, i.e. particles of average diameter roughly larger than  $10^{-2}$  mm [1]. The size of grains is such that interactions are fairly modeled by dissipative hard core repulsion. Correspondingly, granular systems are athermal, that is they do not possess neither a spontaneous long-term dynamics nor a thermodynamic equilibrium state, except the trivial case of an inert immobile stack (or pile) [2,3].

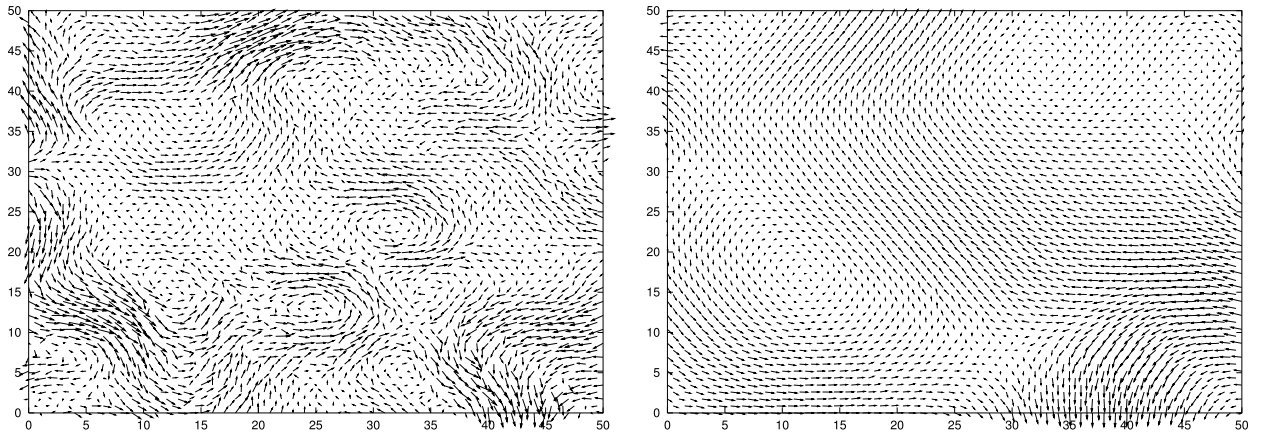
However, injecting energy, usually by means of vibration, shaking, tumbling or falling, leads the granular system to show a variety of dynamical regimes, i.e. different “phases”, roughly analogous to solid, liquid or gas states of molecular matter [4,5]. It is quite hard to push the analogy much forward, since dissipation, in the form of tangential friction and inelastic collisions, makes granular media intrinsically out of equilibrium: in many cases it is evident that not only a Hamiltonian, but even a well-defined thermostat's “temperature” is lacking and therefore no Gibbs distribution can be postulated.

Notwithstanding the inherent non-equilibrium nature of granular phases, many phenomena analogous to equilibrium phase transitions show up in granular experiments and simulations. In most of them, a variation of the input energy flux sensibly changes the internal ordering of the material. Sometimes, the transition from disordered to ordered phase is associated with a growth in time of the size of ordered domains, similarly to what happens in more standard coarsening phenomena. In this case, an abrupt change of the energy input rate plays the role of the usual quench in coarsening dynamics. In this short review, we collect some noticeable examples where the concept of coarsening is empirically meaningful for interpreting and understanding results in the framework of granular systems. For a more extensive review of patterns and collective behavior in granular media, please refer to [6].

The presentation follows a decreasing energy line. In Section 2, we address the more dilute models, i.e. the so-called granular gases, which in the cooling regime display instabilities toward non-homogeneous states with growing domains in the density and velocity fields. Section 3 concerns experiments with dilute or moderately dense shaken granular materials, where several kinds of phase separation appear, with ordered domains that slowly grow in time. The instructive case of electrostatically driven granular fluids is described, with a few noteworthy examples. In Section 4, we discuss the case of demixing or segregation, which usually applies to dense granular materials, slowly agitated or rotated in drums. In Section 5, the compaction dynamics, which is often interpreted as an evaporation of alignment defects or a coarsening of aligned domains, is briefly reviewed. Finally, the last section draws conclusions and perspectives.

## 2. Cooling granular gases

Fluidization of granular media is achieved by injecting mechanical energy into the system, typically by shaking the whole container or vibrating one of its sides [2]. When the packing fraction  $\phi$  is low enough (typically lower than 50%), a gas-like or liquid-like stationary state is rapidly achieved, characterized by a “granular temperature”, which is defined as  $T_g = \frac{1}{d} m \langle v^2 \rangle$  where  $d$  is the dimensionality,  $m$  the mass of a grain and  $v^2$  the squared modulus of its vectorial velocity. The granular temperature is determined by a balance between the energy injected and the dissipation in collisions, which is usually parameterized by a restitution coefficient  $\alpha \leq 1$  ( $\alpha = 1$  for elastic collisions). Several examples of “phase transitions” have been recognized in fluidized granular systems. In the absence of an interaction energy scale, due to the hard-core nature of the grain–grain collisions, the transition is usually controlled by the packing fraction, or by the restitution coefficient, rather than the granular temperature. A relevant exception is constituted by the sudden quench protocol, where the fluidizing mechanism is abruptly interrupted and a “cooling” regime intervenes. In this cooling regime, typically studied in simulations and within kinetic theory [3], the growth of ordered structures in the velocity field (vortices or shear bands) and in the density field (clustering) is observed [7]. In kinetic theory, the idealized starting point is the so-called Homogeneous Cooling State (HCS), which is a spatially homogeneous solution of the inelastic Boltzmann equation where the temperature follows the Haff's law, i.e. asymptotically  $T_g(t) \sim t^{-2}$ . Granular Hydrodynamics (GH) [8], which is expected



**Fig. 1.** Coarsening of vortices in the 2d version of the lattice model for the incompressible cooling state, with the size of the system  $512 \times 512$  and the restitution coefficient  $\alpha = 0.7$  [12]. The plots represent two snapshots of the same portion of the system at two different times, measured in number of collisions per particle: left:  $\tau = 52$ , right:  $\tau = 535$ .

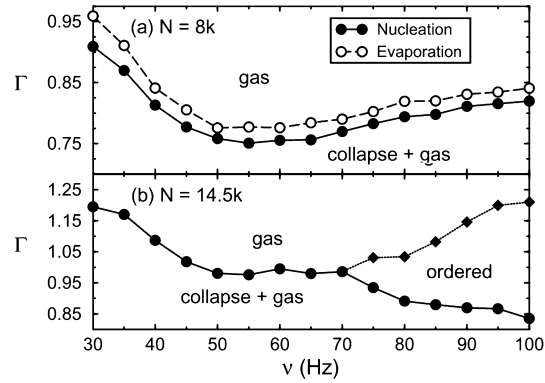
to describe the evolution of “slow fields” (density, macroscopic velocity, and granular temperature), is the simplest theory to predict the instability of the HCS: near the HCS one may linearize the system of GH equations and, in space-Fourier transform, find a linear algebraic system for each wave vector  $k$ . Eigenvalues of the system are guaranteed to be negative only for large enough  $k$ : for  $k < k_{\perp}$  the shear mode (rescaled by  $\sqrt{T_g}$ ) becomes unstable and structures such as vortices display a correlation length growing as  $\sim \sqrt{\tau}$  (where  $\tau$  is the time measured in cumulated number of collisions), while for  $k < k_{\parallel}$  a mode involving density becomes also unstable, growing with a similar law (cluster formation) [7]. Since  $k_{\perp} \sim \sqrt{1 - \alpha^2}$  while  $k_{\parallel} \sim 1 - \alpha^2$ , one has  $k_{\perp} \gg k_{\parallel}$  for  $\alpha \rightarrow 1$ . It is possible, for instance, to choose a linear size  $L$  of the system such that  $k_{\perp} > k_{\min} = \frac{2\pi}{L} > k_{\parallel}$ , which implies that the shear mode is unstable, but the density mode is not: in the linear (initial) stage, there is only the appearance of shear structures in the velocity field, while no clustering is observed.

After some time from the onset of instabilities, the system enters a non-linear regime which is no more described by linear GH. The study of full GH equations is difficult and gives place to many possible regimes depending on geometry, dimensionality, boundary conditions, degree of inelasticity, etc. [9]. Some observations with Molecular Dynamics (MD) in 1d showed that the cooling system asymptotically reaches a regime where energy decays as  $t^{-2/3}$ , which is independent of  $\alpha$  and is therefore equivalent to the dynamics of the sticky gas ( $\alpha = 0$ ) [10]. The sticky gas, characterized by a velocity field with traveling shocks and a density field with coalescing clusters, is well described by the inviscid Burgers equation and is consistent with the observed energy decay. It appears, however, that this scenario breaks down at  $d > 1$ , since there the  $\alpha = 0$  case is no more equivalent to the sticky gas. A series of idealized models on the lattice have been proposed to study the “incompressible” dynamics of the velocity field, i.e. assuming that the density does not change appreciably from the initial homogeneous configuration [11,12]. On each site of the lattice (1d or 2d squared lattice), there is a particle that can collide with its neighbors, dissipating energy and conserving the total momentum. For simplicity, the dependence of the collision rate upon the relative velocity has been neglected, while the so-called kinematic constraint, which forbids collisions between particles going in opposite directions in their center-of-mass reference frame, has been retained. Comparison with MD results in 1d is interestingly good [11], even for the clustering regime, by *unrolling* the coordinates, i.e. replacing the real position with the particle’s index, which is analogous to a sort of Lagrangian coordinate. The study of velocity correlations, through the structure factors, reveals a correlation length  $\xi$  that grows as  $\sim 1/T_g(t)$ . If time is measured by the cumulated number of collisions  $\tau$ , one discovers that  $T_g \sim \tau^{-1/2}$  and  $\xi \sim \tau^{1/2}$ , as in a simple diffusive process. In 2d the lattice model remains consistent with the previous diffusive scenario [12], revealing an energy decaying as  $\tau^{-d/2} = \tau^{-1}$  and a correlation length growing again as  $\tau^{1/2}$ . This correlation length in 2d is clearly associated with the growth of vortices (see Fig. 1).

### 3. Phase coexistence in driven monolayers

The study of granular cooling can be considered as a theoretical framework useful to set up new tools and concepts for non-equilibrium physics. Experimentally, it is much more relevant to study the stationary dynamics resulting from a moderate agitation of the grains. In such a case, correlations usually increase with the density of the system. The dynamics of fluidized granular beds has shown a rich variety of interesting ordering phenomena, mainly the appearance of surface patterns, localized waves and excitations (e.g., “oscillons”). For a small number of particles, the system reduces to an effective 2D geometry, and there is no more distinction between surface and bulk: this is the optimal setup where a phase-separation scenario clearly emerges.

In a seminal paper, Olafsen and Urbach [13] performed experiments with inelastic particles (stainless steel balls) on a horizontal aluminum plane. When the number of particles is insufficient to complete a single layer, the system is named a *submonolayer* of particles. Such a system can be gently shaken, imposing a sinusoidal vertical displacement of the plate



**Fig. 2.** The phase diagrams for the vibrated monolayer experiment in [13]. Two sets of experiments with different numbers of particles are considered: (a)  $N = 8000$  particles and (b)  $N = 14,500$  particles. The filled circles denote the acceleration where the collapse nucleates. The open circles in (a) indicate the point where the collapse disappears upon increasing the acceleration. The diamonds in (b) show the transition to the ordered (hexagonal) state as the acceleration is reduced.

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( $z(t) = A \sin(\omega t)$ ). The relevant parameter is the dimensionless acceleration  $\Gamma = A\omega^2/g$ , where  $g$  is the gravitational acceleration. The regime of interest is when  $\Gamma$  is not too large, so that particles cannot hop over one another and the motion is effectively on a two-dimensional layer. Within this regime, the system behavior depends on  $\Gamma$ , as well as on the vibration angular frequency  $\omega$  (or the vibration frequency  $\nu = \omega/2\pi$ ) and on the total number of particles  $N$  in the system (see Fig. 2).

For very low  $\Gamma$ , irrespective of the frequency  $\nu$ , a condensate of particles at rest on the plate, while in contact with each other, appears. For a small number of particles, such structure (the so called “collapse”) nucleates as an island surrounded by rapidly moving particles (see Fig. 2, top panel). In systems with a large number of particles (but still submonolayer), ordered clusters of moving particles may also appear, when shaken at high frequency (see Fig. 2, bottom panel). Particles in such clusters move around an average position, disposed in a regular (hexagonal) lattice. The study of the quite interesting phase diagram manifests several analogies with liquid–solid transition, with phase coexistence and hysteretic features. Several other experiments [14–16] confirmed the phase coexistence scenario, suggesting the possibility of phase-ordering kinetics and coarsening of clusters in vibrofluidized granular submonolayers. Recently, pattern formation in submonolayers *horizontally shaken* has also been observed and investigated [17]. The formation of strike-like patterns in monodisperse submonolayers, reproduced in molecular dynamics simulation, is quite fast (about 10 s) and no real coarsening dynamics can be appreciated, at odds with the case of binary mixtures (see below).

In order to better investigate similar phenomena, a different set up has been proposed [18] for much tiny (about 40  $\mu\text{m}$  diameter) metallic particles contained between two horizontal metallic plates. The energy was electrostatically injected, applying an oscillating voltage between the plates. Quite independently from the oscillating frequency, two threshold values for the amplitude of the resulting electric field appear. Above a first, lower value,  $E_1$ , isolated particles detach from the bottom plate and start to bounce. Above an higher value,  $E_2$ , all the particles move and the granular medium forms a uniform gas-like phase.

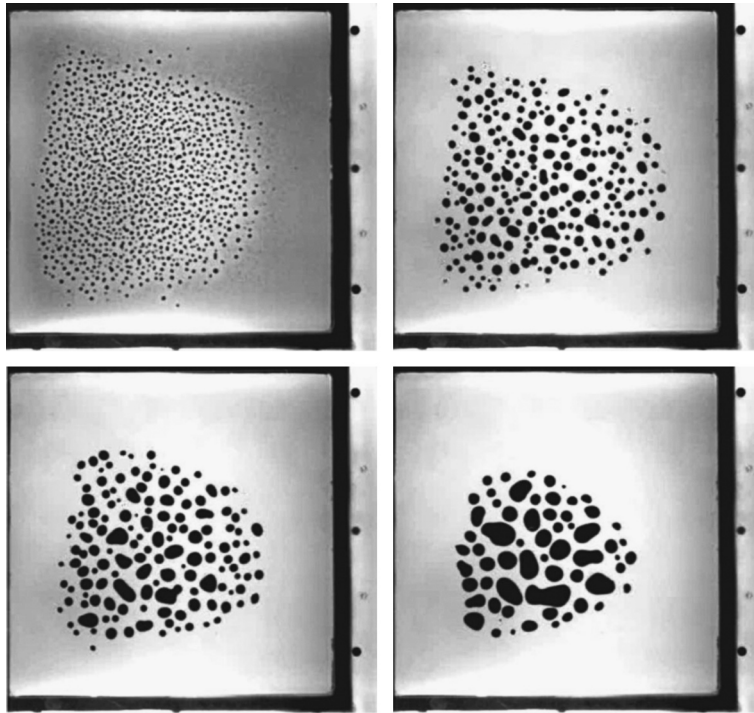
Interestingly, decreasing the field from such a highly mobilized phase to a value comprised between the two thresholds  $E_1 < E < E_2$ , a phenomenon analogous to coalescence dynamics is observed (see Fig. 3). The phenomenon is analogous to the one observed in the vertically vibrating steel spheres described above. In fact, a set-up where the same granular medium could be excited either mechanically or electrostatically has been investigated [19]. The appearance of the same ordered “solid-like” phase (clusters of immobile particles) is reported when the energy injection rate is decreased, both for the vibrated and for the electrostatically driven case.

The extremely small size of the grains allows one to study the dynamics of a very large number of particles. Just after the quench, many “solid” clusters start to form. Their number decreases in time as  $N(t) \propto 1/t$ . On the other hand, their average surface grows as  $\langle \Sigma(t) \rangle \propto t$  (which corresponds to  $\langle R(t) \rangle \propto t^{1/2}$ ), as long as the dynamics keeps quasi 2D. (For low-frequency applied fields, a different large-time growth  $\langle \Sigma(t) \rangle \propto t^{2/3}$  is observed. In this regime the grains can easily hop on each other, the clusters contain more than a monolayer of the particles, and the motion is effectively 3D.)

A phenomenological model has been proposed in [18,21] to describe these experiments. The local density  $n$  of the precipitate phase (the density of immobile particles) is supposed to evolve according to a phenomenological equation

$$\partial_t n = \nabla^2 n + \phi(n, n_g) \quad (1)$$

where  $n_g$  is the number density of bouncing particles (i.e. particles of the gas phase). The function  $\phi$ , at fixed  $n_g$ , has two stable zeros as a function of  $n$ , corresponding to  $n = 0$  (gas) and  $n = 1$  (solid), separated by an unstable zero in between. The function  $\phi$  in Eq. (1) characterizes the solid–gas conversion rate. The effectiveness of the solid–gas transitions



**Fig. 3.** Snapshots of coarsening of granular clusters for experiments with tiny particles (diameter: about 40  $\mu\text{m}$ ) in an electro-static cell at times  $t = 0$  s (top left),  $t = 10^4$  s (top right),  $t = 2 \cdot 10^4$  s (bottom left), and  $t = 5 \cdot 10^4$  s (bottom right). The applied dc electric field  $E = 2.33$  kV/cm.

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is controlled by the local gas concentration  $n_g$ . In other words, at fixed  $n_g$ , Eq. (1) is similar to a time-dependent Ginzburg Landau Equation (GLE), whose solution, from an initial random configuration, displays a coarsening dynamics of the solid domains, where  $n \simeq 1$ . The driving mechanism of the coarsening is the progressive reduction of the curvature of the domain interfaces, as described by the Allen–Cahn equation [22]. This scenario is named “model A” in the classification of Hohenberg and Halperin [23]. In this case the average domain size grows as  $\langle R(t) \rangle \propto \sqrt{t}$  and their number decreases as  $N(t) \propto 1/t$ .

However, here  $n_g$  is not necessarily constant, neither in space, nor in time, and we should provide an equation for its time evolution to couple with Eq. (1). Nevertheless, exploiting the observation that density relaxation in the gas phase is fast compared to the cluster–gas exchange dynamics, one can assume that  $n_g$  is approximately constant in space and depends only on time. On the other hand, the global density of the system is constant in time, hence

$$\int (n + n_g) dx dy \simeq S n_g(t) + \int n dx dy = M \quad (2)$$

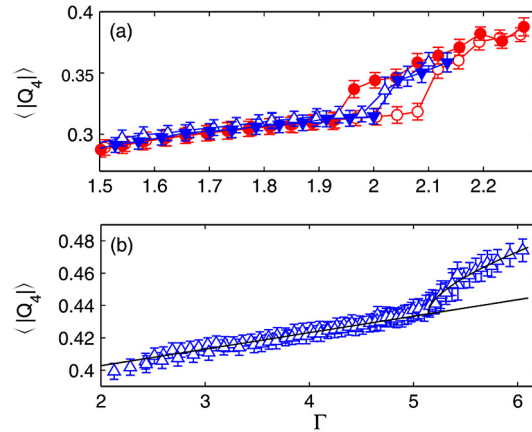
where  $S$  is the surface of the plate and  $M$  the total number of particles. It turns out that  $n_g(t)$  plays the role of a “mean field” interaction between gas and solid, and, on long time scales, it becomes close to a special “equilibrium” value  $n_g^{\text{eq}}$ . Hence, due to the presence of  $n_g$ , the phenomenological model [21] does not properly correspond to model A, since, at odds with model A, here the coarsening field has to satisfy the conservation constraint given by Eq. (2).

In general, conservation properties of the coarsening field affects the dynamics. When the field is conserved, larger domains can grow only at the expenses of smaller ones. For instance, if the coarsening field is locally conserved (which is the case of a dynamics described by a Cahn–Hilliard equation), this can happen through transfer of solid between interfaces via bulk diffusion [24]. The scaling exponents of the average domain size in this case are different (model B in the classification of Hohenberg and Halperin [23]):  $\langle R(t) \rangle \propto t^{1/3}$ .

Nevertheless, the conservation given by Eq. (2) does not have such a dramatic effect. For long time, when  $n_g$  attains a constant equilibrium value, this condition resembles that of a globally conserved order parameter:  $\int n(x, y) dx dy = \text{constant}$ . In this case, the scaling behavior predicted by model A is recovered:  $\langle R(t) \rangle \propto \sqrt{t}$ . According to the mean field theory of Lifshitz–Slyozof–Wagner (LSW) [24,25], the reason for the recovering of Allen–Cahn scaling is that the normal velocity of interfaces is given by the excess curvature with respect to the interface curvature averaged over the whole interface in the system (this mechanism is named Ostwald ripening for interface-controlled dynamics [25]).

In order to corroborate the phenomenological model [21] and its connection with the coarsening with global conservation, the size distribution of clusters has been considered. Experimental investigation in the coarsening dynamics of submonolayer granular media [20], allowed the experimental measure of the (scaling) size distribution of clusters.





**Fig. 4.** (Color online.) Average global fourfold bond-orientational order parameter  $Q_4$  versus relative acceleration  $\Gamma$ . The upper panel (a) shows the case of first-order phase transition (with hysteresis effects). The lower panel (b) shows a continuous transition. The continuous lines in (b) correspond to fits evidencing a linear trend for the sub-critical region  $2.5 < \Gamma < 5$ , on which has been over-imposed a power law in the supercritical region  $\Gamma > 5$  (with a resulting exponent of  $1/2$ ).

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Apparently, the observed distributions, however, do not agree with the theoretical prediction of the mean field LSW theory [25]. In fact, Conti et al. [26] showed that the coalescence of neighboring clusters should not be neglected, as the mean field treatment did. Taking into account such a correction, a good agreement is obtained both with the numerical simulation of the global conserved GLE model, and with the observation in the granular experiment [20].

Recent interesting developments are reported in two papers by Castillo et al. [27,28]. The experiments are performed in vertically vibrated monolayers of 1-mm size steel particles, close to the “solid–liquid” transition. The measure of the system structure factor shows that density fluctuations increase in size and intensity as the transition is approached, but they do not change significantly at the transition itself. On the other hand, the dense, metastable clusters increase their local order in the vicinity of the transition. Exploiting the square symmetry appearing in the dense phase for the specific set-up in study, a bond-orientational order parameter ( $Q_4$ ) has been defined. This quantity evidences a critical behavior at the transition, behaving as a first- or second-order phase transition, depending on filling density and vertical height (see Fig. 4). In the case of a continuous transition, a power law has been observed for the correlation length, the relaxation time and the static susceptibility of the order parameter  $Q_4$ . The corresponding critical exponents are consistent with model C in the Hohenberg and Halperin classification [23] of dynamical critical phenomena, valid for a non-conserved critical order parameter (bond-orientation order) coupled with a conserved field (density).

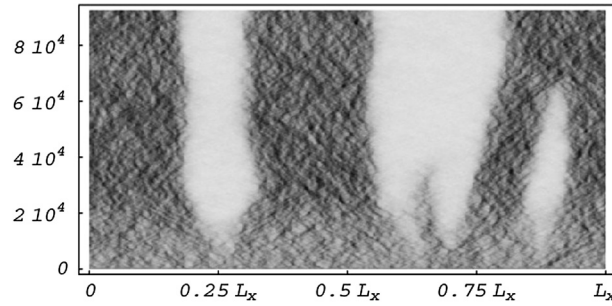
Should these results be confirmed, the observed dynamical phase transition would determine the corresponding coarsening dynamics after a quench. Interestingly, model C displays a quite complex coarsening scenario, with different growing laws for different quenching procedures [29].

The origin of bistability and phase separation in vibrated granular monolayers, in terms of a hydrodynamic description, is not yet fully understood. Several numerical simulations and experiments have been designed in order to advance in this direction.

A transition between a gas-like and liquid-like state with bubble nucleation and subsequent coarsening has been observed in simulations in a two dimensional vibrated box (where the two coordinates represent a horizontal “x” axis and a vertical “y” axis: see Fig. 5 for the time evolution of the system density, averaged on the vertical “y” direction). This transition has been explained by means of a van der Waals-like macroscopic theory: in such a theory, the phase coexistence is usually guaranteed by a nonmonotonous behavior of the pressure as a function of the density, which implies the presence of a bistable region separated by a coexistence and a spinodal curve. The inflection point of pressure versus density (negative compressibility), for granular particles, is due to the energy balance between energy injection (shaking) and energy dissipation in inter-particle collisions, which leads to a stationary  $T_g$ , decreasing with density.

A partial confirmation of this scenario has emerged in a quasi 1d vibrated experiment [31]. In this experiment, phase separation of clusters and coalescence through coarsening has been obtained. The clusters are in a crystal-like (square or triangular lattice) phase and coexist with a fluid phase. Even if the nature of the two phases is different, a similar van der Waals-like theory has been used to explain the experimental result. Measurements of pressure have indicated the existence of a pressure plateau corresponding to the coexistence region. Slow coarsening and coalescence of nucleated crystal islands is observed in time.

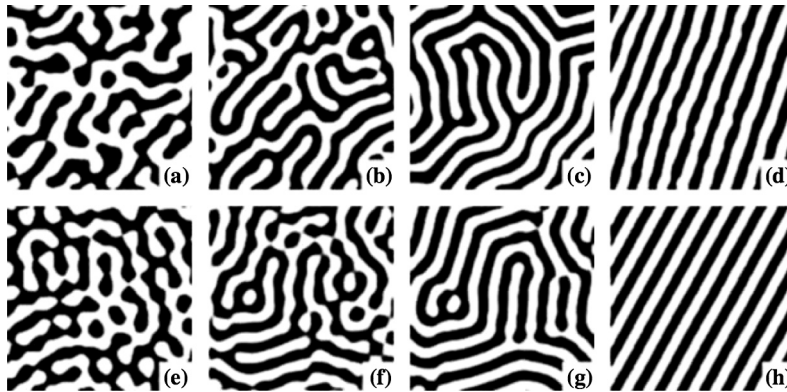
Starting from the Navier–Stokes granular hydrodynamic equation, Khain et al. [32] proposed a different scenario, where bistability results from the nontrivial energy injection mechanism from a vibrating plate to the granular gas. This is different from the mechanism advocated in [30], where the van der Waals instability is rather attributed to energy loss due to inelastic



**Fig. 5.** Spatiotemporal evolution of the two-dimensional system simulated in Ref. [30]. The gray level corresponds to the density averaged on the (vertical)  $y$ -direction of the system (darker regions representing denser regions). In the plot the abscissa represents the time, increasing upward.

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**Fig. 6.** Spatiotemporal evolution of the patterns during experiments and simulations described in [33]. Images (a)–(d): the real images of the free surface of granular layers after a sudden quench from  $\Gamma = 2.4$  to  $\Gamma = 2.8$ : times (in period of vibration) are  $t = 2$  (a),  $t = 10$  (b),  $t = 200$  (c), and  $t = 1000$  (d). The bright parts correspond to the crests of the free surface, and the dark parts correspond to the troughs of the free surface. (e)–(h): The images of the numerical results of the 2D Swift–Hohenberg equation in time (see [33] for the parameters of the simulation).

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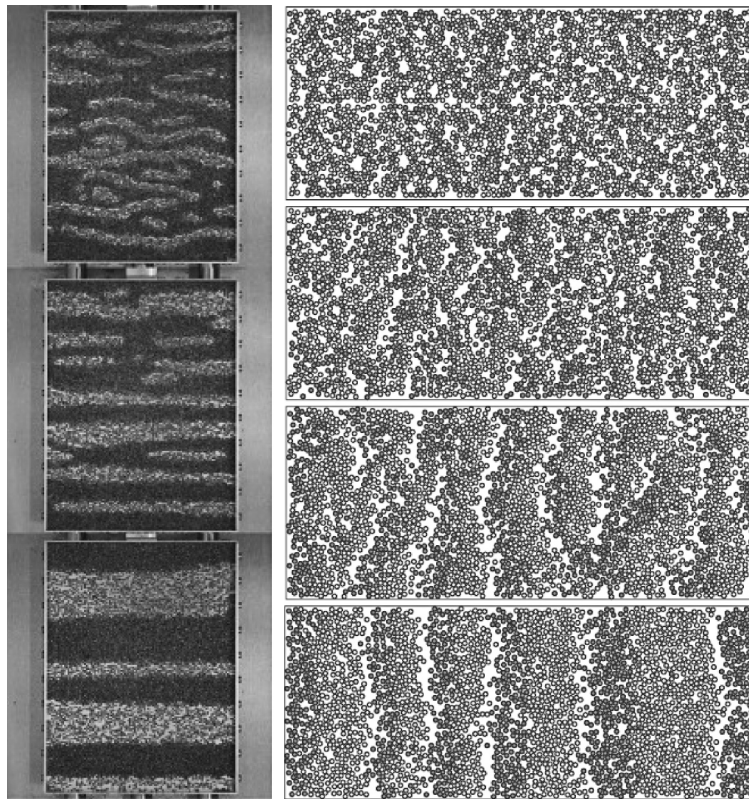
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collision between particles. In both cases, the phase separation is associated with a negative compressibility of the granular gas.

Another example of coarsening in vibrated granular media, where a mono-disperse granular material is studied, is shown in the experiment described in [33]. Here the formation and evolution of regular patterns in a vertically vibrated thin granular layer of phosphor-bronze spherical beads is analyzed. This system shows a rich phenomenology: an initially flat layer can form complex structures, such as stripes, squares, hexagons, etc., see for instance Fig. 6. The analysis focuses on the coarsening dynamics following a sudden change of a control parameter (the vibration amplitude), which leads to stripe structure. The first stage of this evolution is characterized by the formation of rolls of well-defined wavelength with random orientation. Later, these stripes are observed to align and an ordered structure is eventually attained. This behavior can be analyzed within the Swift–Hohenberg theory [34], which describes the dynamics of the order parameter in nonlinear dissipative systems, with an instability of the first kind (namely, the instability grows only at a finite wave vector.) This model predicts a power-law growth for the characteristic length  $L(t) \sim t^z$  with  $z = 1/4$ . In the granular experiment, the order parameter is defined as a function of the local stripe orientations  $\theta(\mathbf{r}, t)$ , which is defined as the local angles of the stripes ( $\mathbf{r}$  denoting the position). This allows one to define the local order parameter  $\psi(\mathbf{r}, t) = \exp[2i\theta(\mathbf{r}, t)]$  and to measure the angle averaged correlation function  $C(r, t) = \langle \text{Re}[\psi(0)^* \psi(r)] \rangle$ . This satisfies a scaling relation with the dynamic correlation length in the system  $C(r, t) = g[r/L(t)]$ , where  $L(t) \sim t^z$  is the characteristic length scale of the ordered domain at time  $t$ . The time behavior of this length turns out to be in agreement with the Swift–Hohenberg model.

#### 4. Segregation of mixtures via coarsening

A relevant problem for dense granular systems, with important industrial applications, is the control of mixtures of different grains, which can differ by their sizes, densities, shapes, friction coefficients, etc. A large amount of experimental and numerical work has been devoted to the study of demixing or segregation. This is a huge field and we refer the reader to the review article [5] for experiments and theories on spontaneous segregation in granular mixtures. Here we focus



**Fig. 7.** Segregation of a monolayer mixture in a horizontally oscillated plate. Left: Pictures of the system studied in the experiment [35]. Time runs from the top to the bottom and the oscillation direction of the plate is perpendicular to the stripe orientation. Right: Molecular dynamics simulations of the same system [38] (stripes are perpendicular to the oscillation direction).

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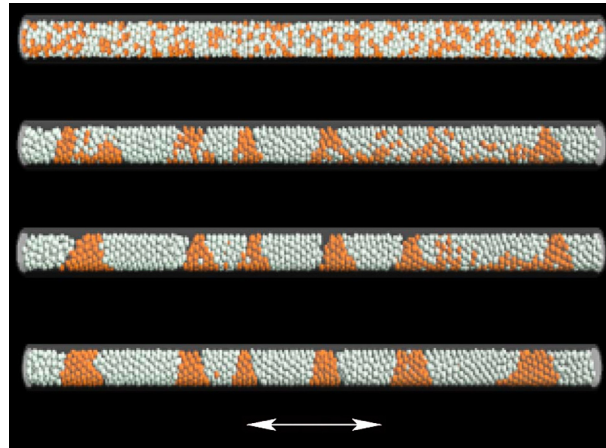
on the cases where such a phenomenon occurs via domain coarsening of the components and we present results for two classes of experiments: vibrated two-dimensional granular systems and rotating tumblers.

*Quasi two-dimensional layers* The simplest setup to study the phenomenon of spontaneous segregation is a quasi two-dimensional configuration, where the granular mixture is spread above a horizontally vibrated plate. In this case, several studies have shown that mixture segregation occurs through stripe coarsening, which develops orthogonally to the vibration direction, and is due to the differential frictional drag acting on the different components of the mixture.

We first consider a mixture of two kinds of particles: poppy seeds and copper spheres [35]. The initially homogeneous mixture is a mono-layer placed on a plate that is horizontally oscillated, with amplitude  $A$  and frequency  $\omega$ . For packing fractions greater than a threshold value, the two components are observed to segregate in stripes of the two components, perpendicular to the oscillation direction of the plate, see left panel of Fig. 7. The evolution in time of these patterns presents a coarsening dynamics: the total number of stripes decreases while their average amplitude,  $L(t)$ , increases with time  $t$ . The behavior turns out to be well described by a power law,  $L(t) \sim t^z$ , with the dynamical exponent  $z = 1/4$ . A stochastic model based on a random walk, proposed in the geological context of stone striping [36], can explain the observed phenomenon. Let us consider the stripe distribution  $f(x, t)$ , that represents the number of stripes of width  $x$  at time  $t$ . The evolution of the stripe width occurs by particles diffusing between stripes and it is described by a diffusion equation:  $\partial f(x, t)/\partial t \propto \nu(t) \partial^2 f(x, t)/\partial x^2$ . Here the frequency  $\nu(t)$  varies in time because the particles have to diffuse in the region between the stripes to produce a fluctuation. This region is expected to scale as the average width of stripes  $L(t) \equiv \langle x(t) \rangle$ , and therefore  $\nu(t) \propto L(t)^{-2}$ . Introducing the variable  $\tau = \int \nu(t) dt$ , the diffusion equation is rewritten as  $\partial f(x, \tau)/\partial \tau \propto \partial^2 f(x, \tau)/\partial x^2$ , which gives  $\langle x(\tau)^2 \rangle \sim \tau$ , and thus, using the definition of the variable  $\tau$ , one readily gets the behavior  $L(t) \propto t^{1/4}$ . In a successive experiment [37], the behavior of the same system has been investigated as a function of the packing fraction, leading to the identification of a continuous phase transition for the segregation phenomenon.

Segregation via stripe coarsening is also observed in granular mixtures immersed in fluids, subjected to horizontal vibration, as reported in [39], see Fig. 8. The two components in this case differ for masses and viscous coefficients, and the force exercised by the fluid produces a differential drag, which is responsible for the segregation mechanism.





**Fig. 8.** (Color online.) Numerical simulations reproducing experiments on the segregation dynamics of a granular mixture immersed in a fluid, from [39]. Time runs from the top to the bottom and stripes are perpendicular to the oscillation direction of the cylinder (indicated by the double arrow). Reprinted with permission from [39]. © 2004 American Physical Society

Numerical studies via molecular dynamics simulations [38,40] of a two-dimensional mixture of granular disks, inspired by the first experiment described above, support the interpretation that the different drag forces acting on the components of the mixture induce the segregation process. Such simulations reproduce the same phenomenology as that observed in the experiments, see right panel of Fig. 7. Since the mixture components have different friction coefficients and masses, their dynamics is ruled by different relaxation times. Therefore, different particles tend to oscillate with different amplitudes and phases, producing the segregation in a stripe pattern.

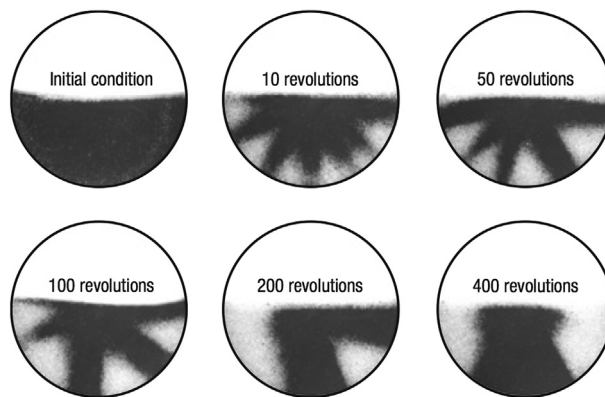
An accurate theoretical description of these systems can be attempted by means of an effective interaction theory [41]. This approach allows one to study an out-of-equilibrium dissipative driven system in terms of an “equilibrium” mono-disperse system. For a granular mixture subject to horizontal oscillations, it has been shown that the effective interaction force is anisotropic and presents a repulsive shoulder at long distances which is prominent in the direction of oscillation. This description leads to a phenomenological Cahn–Hilliard equation that reproduces the observed phenomenology and clarifies the origin of the differential drag mechanism for spontaneous segregation and coarsening in these systems.

*Rotating tumblers* A different experimental setup where the dynamics of granular mixtures is often studied is represented by rotating tumblers (of circular or square section). Several possible segregation patterns are observed, e.g., axial banding, radial streaks, etc., all displaying slow domain growth. For a recent detailed review on this specific topic, we refer the reader to [42]. The mixture segregation appears to occur in three stages: first radial segregation is observed, then axial segregation takes place, with the formation of bands that eventually coarsen slowly with the number of tumbler rotations. Here we focus on three experimental studies in order to illustrate this general phenomenology: the first two are concerned with the phenomenon of axial segregation in long tumblers, while the last one with the observation of coarsening of radial streaks in a quasi-two-dimensional setup.

The first experiment analyzes the axial segregation of dry and wet granular media, in circular or square tumblers [43]. If a cylinder filled with a binary granular mixture is rotated around its longitudinal axis, for specific rotational speeds, which play here the role of control parameter, one observes the formation of alternating axial bands of the two components: initially, the particles separate radially in the plane perpendicular to the rotation axis, and later, a further segregation process occurs, where coarsening of the bands takes place. More precisely, the number of bands,  $N$ , grows in time at short times and then slowly decreases. At long times, one observes for  $N$  a logarithmic decay, as  $-k \log(n)$ , where  $n$  is the number of rotations. This kind of behavior can be explained by some models for standard coarsening in one dimension [22] and it is also predicted by a continuous theory for granular segregation [44].

A detailed analysis of the specific mechanisms driving the coarsening process in similar geometries for wet granular materials is reported in [45]. In this experimental study, the authors investigate, via optical methods and nuclear magnetic resonance imaging, the dynamics of a binary mixture of glass beads of different radii immersed in water, in a long cylindrical rotating drum. The axial segregation phenomenon is found to exhibit a slow dynamics, where the number of stripes decays logarithmically with the number of rotations. A significant difference with respect to the dry case is that the coarsening phenomenon is observed at lower rotation speeds in the presence of water. The physical mechanism responsible for coarsening is related to the redistribution of the small particles. However, a clear understanding of the late stage of the coarsening dynamics in these systems is still lacking, and the problem is the focus of very recent studies [46].

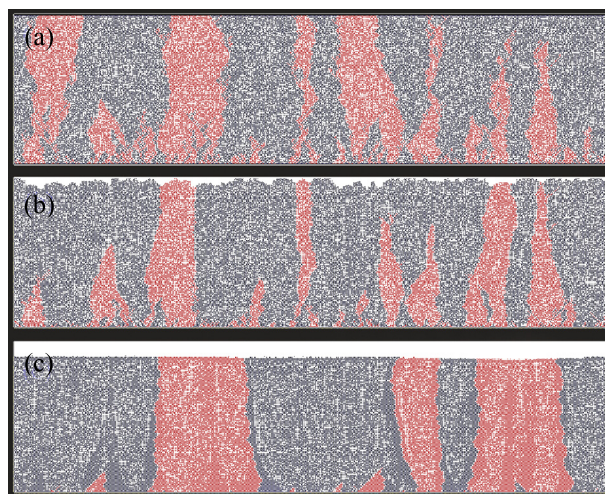
A different geometry used to study mixture segregation consists of a quasi-two-dimensional slowly rotating tumbler, usually filled up to  $\sim 50\%$  with particles. In the experiments reported in [47], the very long time coarsening regime, leading to a complete final segregation, was studied (see Fig. 9). The experiment investigates the dynamics of a binary granular



**Fig. 9.** Pictures of the experiment [47], where a quasi-2D circular tumbler, filled up to 55% with a binary mixture of granular particles, is slowly rotated (two revolutions per minute). The radial streaks are observed to coarsen into one, producing a final segregation of the components (notice that in an angular phase space coarsening cannot last forever and a complete segregation is expected).

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**Fig. 10.** (Color online.) Pictures of simulations from Tetris-like model for granular compaction [50]. The three panels reproduce the compaction dynamics at three different times  $t_a < t_b < t_c$ . Colored pixels are granular particles, while white pixels are voids. Note how the upper free surface of the granular media decreases in height, because of compaction. The colors of the pixels (red and blue) indicates the two different possible arrangements of the grains. An evident coarsening of domains appears. The average width of the domains grows at first as  $t^{1/4}$ , until all the domains span vertically the whole system. Then, a growth like  $\sqrt{t}$  follows, due to the diffusion and annihilation of domain interfaces.

mixture of glass particles, differing in size or in size and density. Even in this geometry, starting from a homogeneous state, when a slow rotation is applied to the tumbler, the mixture initially segregates into radial streaks, which eventually coarsen into one, provoking a final separation of the components.

## 5. Granular compaction

Finally, in the pure solid phase, an interesting ordering phenomenon is compaction [48,49]. This industrial relevant procedure consists in a weak, sometimes sporadic, energy injection aimed at increasing the packing fraction of the system. There is no general optimal protocol to compact a granular medium, and the most accepted theoretical scenario considers the dynamics of bubbles of misaligned grains that evaporate, allowing the coalescence of optimally arranged islands and a progressive reduction of the total occupied volume. In this context, many lattice models have been proposed [50], where the slow coarsening of domains of higher compactivity is observed (see Fig. 10). In order to discriminate different solid, mechanically stable states of a granular, a general analogy with equilibrium statistical physics has been proposed by Edwards [51], where energy is replaced by the occupied volume, and the entropy is a measure of the number of configurations in the position of grains that result in the same occupied volume.

After an external perturbation, due to shaking or vibration, the system organizes itself in a new metastable configuration. In this way, it explores a phase space where each accessible point corresponds to a mechanically stable configuration. This observation suggested the idea to introduce a formalism analogous to the standard statistical mechanics [51]. The central

hypothesis is that the system can be described by an ensemble average over the mechanically stable states (called “inherent states”). Then, assuming that each state has an equal probability a priori to occur, the probability  $P_r$  to find the system in a inherent state  $r$  is obtained maximizing the entropy  $S = -\sum_r P_r \log P_r$ , with the macroscopic constraint that the total energy of the system  $E = \sum_r P_r E_r$  is fixed. This gives the Gibbs result  $P_r \propto \exp(-\beta_{\text{conf}} E_r)$ , where  $\beta_{\text{conf}}$  is a Lagrange multiplier called inverse configurational temperature. A generalized partition function can be then introduced. For example, in the explicit case of a monodisperse system of hard spheres with mass  $m$  in the gravitational field  $g$ , where the centers of mass of grains are constrained to move on the sites  $i$  of a cubic lattice, one has the Hamiltonian:  $\mathcal{H} = \mathcal{H}_{hc} + mg \sum_i z_i n_i$ , where  $\mathcal{H}_{hc}$  represents the hard-core interaction between grains, preventing the overlap of nearest neighbor grains,  $n_i$  is the occupation variable on site  $i$  and  $z_i$  its height [52]. In order to validate the Edwards hypothesis, one has i) to introduce a dynamics allowing the system to explore the phase space; ii) to verify that, at stationarity, the system properties do not depend on the specific parameters of the dynamics; and iii) to check that the temporal averages obtained from such a dynamics are consistent with those obtained from the Gibbs distribution. This approach turned out to be effective in several idealized models for granular compaction [52–55].

## 6. Conclusions

In this short review, we have described some relevant examples where coarsening phenomena are observed in the context of granular media. Remarkably, the study of these non-equilibrium athermal systems reveals that, in many cases, a dynamics analogous to the phase ordering kinetics of standard critical phenomena takes place. Concepts such as “order parameter” and “phase transition” turn out to be effective to describe instabilities, clustering, mixture segregation, compaction, and other observed behaviors. This suggests that the tools of statistical mechanics can be extended and generalized to a wider context, where fluctuations are induced by external forcing, rather than by thermal agitation, and dissipation drives the systems out of equilibrium. In that respect, the physics of granular matter shares many features with other non-equilibrium systems, such as glasses, suspensions, foams, traffic flow, active matter. In particular, in the context of self-propelled particles, the phenomenon of motility-induced phase separation can be explained by the same models of coarsening as those used for describing vibrated granular media, see for instance [56]. These considerations strengthen the idea that a general theory can be able to describe a wide gamut of different physical systems, at least in those cases where the emergent behavior can be characterized in terms of few macroscopic parameters. Here, as in many aforementioned systems, a derivation of the coarse grained models from the microscopic dynamics is, in our opinion, an important step, deserving further investigation.

## References

- [1] B. Andreotti, Y. Forterre, O. Pouliquen, *Granular Media. Between Fluid and Solid*, Cambridge University Press, 2013.
- [2] A. Puglisi, *Transport and Fluctuations in Granular Fluids*, Springer, Berlin, 2014.
- [3] N.V. Brilliantov, T. Pöschel, *Kinetic Theory of Granular Gases*, Oxford University Press, 2004.
- [4] H.M. Jaeger, S.R. Nagel, R.P. Behringer, *Rev. Mod. Phys.* 68 (1996) 1259.
- [5] A. Kudrolli, *Rep. Prog. Phys.* 67 (2004) 209.
- [6] I. Aranson, L. Tsimring, *Rev. Mod. Phys.* 78 (2004) 641.
- [7] T.P.C. van Noije, M.H. Ernst, R. Brito, J.A.G. Orza, *Phys. Rev. Lett.* 73 (1997) 411.
- [8] J.J. Brey, J.W. Dufty, C.S. Kim, A. Santos, *Phys. Rev. E* 58 (1998) 4638.
- [9] B. Meerson, A. Puglisi, *Europhys. Lett.* 70 (2005) 478.
- [10] E. Ben-Naim, S.Y. Chen, G.D. Doolen, S. Redner, *Phys. Rev. Lett.* 83 (1999) 4069.
- [11] A. Baldassarri, U. Marini Bettolo Marconi, A. Puglisi, *Europhys. Lett.* 58 (2002) 14.
- [12] A. Baldassarri, U. Marini Bettolo Marconi, A. Puglisi, *Phys. Rev. E* 65 (2002) 051301.
- [13] J.S. Olafsen, J.S. Urbach, *Phys. Rev. Lett.* 81 (1998) 4369.
- [14] W. Losert, D.G. Cooper, J.P. Gollub, *Phys. Rev. E* 59 (1999) 5855.
- [15] A. Prevost, P. Melby, D.A. E Golf, J.S. Urbach, *Phys. Rev. E* 70 (2004) 1.
- [16] J.S. Olafsen, J.S. Urbach, *Phys. Rev. Lett.* 95 (2005) 1.
- [17] D. Krengel, S. Strobl, A. Sack, M. Heckel, T. Pöschel, *Granul. Matter* 15 (2013) 377.
- [18] I. Aranson, D. Blair, V. Kalatsky, G. Crabtree, W.-K. Kwok, V. Vinokur, U. Welp, *Phys. Rev. Lett.* 84 (2000) 3306.
- [19] M. Sapozhnikov, I. Aranson, J. Olafsen, *Phys. Rev. E* 67 (2003) 010302.
- [20] M.V. Sapozhnikov, A. Peleg, B. Meerson, I.S. Aranson, K.L. Kohlstedt, *Phys. Rev. E* 71 (2005) 1.
- [21] I.S. Aranson, B. Meerson, P.V. Sasorov, V.M. Vinokur, *Phys. Rev. Lett.* 88 (2002) 204301.
- [22] A.J. Bray, *Adv. Phys.* 43 (1994) 357.
- [23] P.C. Hohenberg, B.I. Halperin, *Rev. Mod. Phys.* 49 (1977) 435.
- [24] I.M. Lifshitz, V.V. Slyozov, *J. Phys. Chem. Solids* 19 (1961) 35.
- [25] C. Wagner, *Z. Elektrochem.* 65 (1961) 581.
- [26] M. Conti, B. Meerson, A. Peleg, P.V. Sasorov, *Phys. Rev. E* 65 (2002) 1.
- [27] G. Castillo, N. Mujica, R. Soto, *Phys. Rev. Lett.* 109 (2012) 095701.
- [28] G. Castillo, N. Mujica, R. Soto, *Phys. Rev. E* 91 (2015) 012141.
- [29] J. Kockelkoren, H. Chaté, *Physica D* 168–169 (2002) 80.
- [30] M. Argentina, M.G. Clerc, R. Soto, *Phys. Rev. Lett.* 89 (2002) 044301.
- [31] M.G. Clerc, P. Cordero, J. Dunstan, K. Huff, N. Mujica, D. Risso, G. Varas, *Nat. Phys.* 4 (2008) 249.
- [32] E. Khain, I. Aranson, *Phys. Rev. E* 84 (2011) 1.
- [33] K. Kim, H.K. Pak, *Phys. Rev. Lett.* 88 (2002) 204303.
- [34] J. Swift, P.C. Hohenberg, *Phys. Rev. A* 15 (1977) 319.

- [35] T. Mullin, *Phys. Rev. Lett.* 84 (2000) 4741.
- [36] P.A. Mulheran, *J. Phys. I* 4 (1994) 1.
- [37] P.M. Reis, T. Mullin, *Phys. Rev. Lett.* 89 (2002) 244301.
- [38] M. Pica Ciamarra, A. Sarracino, M. Nicodemi, A. Coniglio, Species segregation and dynamical instability of horizontally vibrated granular mixtures, in: *Traffic and Granular Flow'05*, Springer, Berlin, Heidelberg, 2007.
- [39] P. Sánchez, M.R. Swift, P.J. King, *Phys. Rev. Lett.* 93 (2004) 184302.
- [40] M. Pica Ciamarra, A. Coniglio, M. Nicodemi, *Phys. Rev. Lett.* 94 (2005) 188001.
- [41] M. Pica Ciamarra, A. Coniglio, M. Nicodemi, *Phys. Rev. Lett.* 97 (2006) 038001.
- [42] G. Seiden, P.J. Thomas, *Rev. Mod. Phys.* 83 (2011) 1323.
- [43] S.J. Fiedor, J.M. Ottino, *Phys. Rev. Lett.* 91 (2003) 244301.
- [44] I.S. Aranson, L.S. Tsimring, *Phys. Rev. Lett.* 82 (1999) 4643.
- [45] T. Finger, A. Voigt, J. Stadler, H.G. Niessen, L. Naji, R. Stannarius, *Phys. Rev. E* 074 (2006) 031312.
- [46] T. Finger, M. Schröter, R. Stannarius, preprint, arXiv:1412.7459.
- [47] S.W. Meier, D.A. Melani Barreiro, J.M. Ottino, R.M. Lueptow, *Nat. Phys.* 4 (2008) 244.
- [48] Edmund R. Nowak, James B. Knight, Eli Ben-Naim, Heinrich M. Jaeger, Sidney R. Nagel, *Phys. Rev. E* 57 (1998) 1971.
- [49] E. Ben-Naim, J.B. Knight, E.R. Nowak, H.M. Jaeger, S.R. Nagel, *Physica D* 123 (1998) 380.
- [50] A. Baldassarri, S. Krishnamurthy, V. Loreto, S. Roux, *Phys. Rev. Lett.* 87 (2001) 224301.
- [51] S.F. Edwards, R.B.S. Oakeshott, *Physica A* 157 (1989) 1080.
- [52] A. Fierro, M. Nicodemi, A. Coniglio, *Phys. Rev. E* 66 (2002) 061301.
- [53] J.J. Brey, A. Prados, B. Sanchez-Rey, *Physica A* 275 (2000) 310.
- [54] J. Berg, S. Franz, M. Sellitto, *Eur. Phys. J. B* 26 (2002) 349.
- [55] V. Colizza, A. Barrat, V. Loreto, *Phys. Rev. E* 65 (2002) 050301(R).
- [56] R. Wittkowski, A. Tiribocchi, J. Stenhammar, R.J. Allen, D. Marenduzzo, M.E. Cates, *Nat. Commun.* 5 (2014) 4351.