Dynamically Induced Effective Interaction in Periodically Driven Granular Mixtures

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We show that a granular mixture subject to horizontal oscillations can be reduced to a monodisperse system of particles interacting via an effective interaction. This interaction is attractive at short distances and strongly anisotropic, and its features explain the system rich phenomenology, including segregation and stripe pattern formation. Finally, we show that a modified Cahn-Hilliard equation, which takes into account the characteristics of the effective interaction, is capable of describing the dynamics of the mixture.

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Dynamical instabilities and pattern formation have only recently begun to be fully investigated in granular materials $[1,2]$ $[1,2]$ $[1,2]$, which are collections of macroscopic particles interacting via dissipative forces where thermal effects are negligible. Understanding their origin in these systems, which is crucial for scientific reasons and substantial industrial applications, is today a challenge as granular systems cannot be simply described by usual statistical or fluid mechanics $[2-7]$ $[2-7]$ $[2-7]$. In this Letter we make a step further in this direction by showing that a binary granular mixture subject to a periodic drive can be reduced to a monodisperse undriven thermal system of particles interacting via an effective potential. This result can be considered as an extension, to the nonthermal and driven case, of the ''depletion potential'' approach introduced by Asakura and Oosawa [\[8](#page-3-5)] to reduce an undriven thermal binary mixture to a monodisperse system, which has been also investigated in granular systems [[9](#page-3-6)]. However, while the depletion potential has a purely entropic origin as related to the size difference of the two components, the effective interaction we introduce here has a purely dynamical origin, as it results from the different response of the mixture components to the oscillating drive. Our approach could be also of value in the study of driven thermal binary mixtures.

We apply our ideas to a granular mixture subject to horizontal oscillations, a system previously investigated both experimentally and numerically $[2,10-12]$ $[2,10-12]$ $[2,10-12]$ $[2,10-12]$ $[2,10-12]$, whose complex and not well-understood phenomenology encompasses both instabilities and segregation. First we discuss how to measure the effective interaction force between like particles. This is obtained by studying a system in which two particles of a given species are held at a fixed distance in a bath of particles of different species, in presence of the external drive. Then we show that this effective force captures the physical mechanism responsible for the observed segregation and instability. Indeed, simulations of a system of one species of particles interacting via the previously determined effective force, and in absence of the external oscillating drive, show that these particles evolve as if they were a component of the original driven granular mixture. Finally, we introduce a phenomenological CahnHilliard equation which reproduces the observed phenomenology, but also allows for analytical predictions.

*Model —*We have investigated via molecular dynamics simulations a two-dimensional model [\[2\]](#page-3-3) of the experiment of Ref. [\[10](#page-3-7)], where a monolayer of a granular mixture is placed on a horizontally oscillating tray of size $160D \times$ $40D (D = 1 cm)$. Contacting particles interact via a spring dashpot repulsive force with constant coefficient of restitution $e = 0.8$, while the interaction between a grain and the oscillating tray is given by $\mathbf{f}_{\text{tray}} = -\mu(\mathbf{v} - \mathbf{v}_{\text{tray}})$, where $\mathbf{v}_{\text{tray}}(t) = 2\pi A \nu \sin(\nu t) \mathbf{x}$ is the velocity of the tray and **v** the velocity of the disk, plus a white noise force $\xi(t)$ with $\langle \xi(t)\xi(t') \rangle = 2\Gamma \delta(t - t')$ [see [\[2,](#page-3-3)[13\]](#page-3-9) for details]. The two components of our mixture have masses $m_h = 1$ g, $m_l = 0.03$ g, frictional coefficients $\mu_h = 0.28$ g s⁻¹ and $\mu_l = 0.34$ g s⁻¹, while $\Gamma = 0.2$ g² cm² s⁻³, *A* = 1.2 cm, and $\nu = 12$ Hz. The diameters of the two species considered here are equal, $D_h = D_l = 1$ cm, to remark that no ''entropic'' depletion forces are present.

*Effective interaction—*The effective interaction is determined by making simulations in which two heavy disks are placed at fixed relative positions $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2 = (x, y)$ in a system of lighter disks covering an area fraction $\phi \approx 0.63$

FIG. 1 (color online). Radial component of the effective force along directions forming an angle θ with the *x* axis. For $r/D < 1$ the force is strongly repulsive.

(we have sampled the range $0 \le x, y \le 6D$). When the tray oscillates horizontally the two heavy disks are moved as a single object of mass $M = 2m_h$ subject to a force $\mathbf{f} = \mathbf{f}_1 + \mathbf{f}_2$ $\mathbf{f}_2 + (2\mathbf{f}_{\text{tray}} + 2\mathbf{f}_{\text{noise}})$, where \mathbf{f}_1 (\mathbf{f}_2) is the force acting on particle 1 (2) due to collisions with lighter disks. The two disks can translate horizontally and vertically, but \mathbf{r}_{12} remains fixed. The effective force that particle 1 exerts on particle 2, averaged over one period of oscillation, *T*, is given by

$$
\mathbf{f}^{\text{eff}} = (f_x^{\text{eff}}, f_y^{\text{eff}}) = \frac{1}{2T} \int_0^T [\mathbf{f}_1(t) - \mathbf{f}_2(t)] dt. \tag{1}
$$

Our numerical results show that this force is attractive at short distances, and strongly asymmetrical in the *xy* plane: it has a repulsive shoulder at long distances, which vanishes along the *y* axis. This is shown in Fig. [1](#page-0-0), where we plot the radial component of the effective interaction force along directions forming an angle θ with the *x* axis. The effective force is weakly dependent on Γ (in the investigated range $0 < \Gamma < 10 \text{ g}^2 \text{ cm}^2 \text{ s}^{-3}$), and its amplitude increases with the area fraction of the smaller species. Variations of the amplitude and frequency of oscillation do not change the qualitative features of the effective force (asymmetry, attraction at short distances, and repulsive shoulder), but they change the range of attraction and the position of the repulsive shoulder. Rigorously, the effective force **f**eff cannot be derived by an effective scalar potential, as the curl of f^{eff} ($\nabla \times f^{\text{eff}} = \partial_y f_x^{\text{eff}} - \partial_x f_y^{\text{eff}}$) varies in space [the presence of a solenoid component in the effective interaction has been also reported in [[14](#page-3-10)] for a system of two disks in a constant fluid flow]. However, as the solenoid component of **f**eff appears to be negligible with respect to its irrotational component $[15]$ $[15]$, an effective interaction scalar potential can be introduced within a good approximation.

The effective force we derive (see Fig. [1](#page-0-0)) appears to be the key ingredient in understanding the phenomenology of our system. It follows from the different response of the two components to the oscillating drive (the tray): due to their differences in mass and friction coefficient, in fact, the two species are forced to oscillate with different amplitudes and phases [[6](#page-3-12)]. A qualitatively understanding of the microscopic origin of the effective force can be obtained by considering the simpler case where two disks of a given species [striped disks in Fig. $2(a)$ and $2(b)$] are immersed in stream of disks of a different species flowing along the *x* axis with velocity $v > 0$. Figure [2\(a\)](#page-1-0) illustrates that, as $\theta_1 < \theta_2$, the majority of the collisions experienced by grain *A* push it closer to grain *B*, explaining the attraction between particles *A* and *B* along *y*. Figure [2\(b\)](#page-1-0) shows that, for small distances, particle *A* screens particle *B*, inducing an effective attraction between them along *x*. For larger distances, and in the presence of the oscillating drive, light disks tend to be caged between the two heavy ones, and their density may become higher than average, as shown in Fig. $2(c)$. In this condition the competition be-

FIG. 2 (color online). The effective interaction between two heavy disks is determined via screening effects by their relative position, as discussed in the text an exemplified in panels (a), (b). Panel (c) is a contour level plot of the density field of light disks in an oscillating system. Panel (c) has been obtained by averaging over 100 configurations taken in the frame of reference centered in the midpoint between the two heavy disks (kept at the fixed distance of 3.75*D*) at time $t_n = nT + t_0$, $n =$ 1, ..., 100, where $t_0 = T/4$. Qualitatively similar results are obtained for different values of $t_0 < T$. The regularity of the isodensity lines is due to steric constrains. Filled area represent regions where the density is above (about twice) the average.

tween the collisions experienced by the heavy disks from the caged light disks, which push them apart, and those experience from the surrounding disks, which push them together, is won by the caged disks and results in an effective repulsive force.

*Segregation and instability—*The effective force, **f**eff, was derived for a system of just two heavy grains in a "bath" of lighter ones. For a system with many heavy grains this is, in general, expected to be just an approximation. We checked, however, that **f**eff captures the basic physical mechanism responsible for segregation via stripes formation in the investigated system. To this aim we run simulations of a monodisperse system of heavy disks only, where disks interact pairwise via the effective force previously determined. As in the original mixture the disks are also subject to a frictional force $(f_{\text{tray}} = -\mu_h \mathbf{v})$ and to a white noise force ξ . They are placed on a *fixed* tray of size $160D \times 40D$ and cover an area fraction $\phi \approx 0.5$.

Figure $3(a)$ shows that this system evolves, from an initially disordered configuration, via the formation of interconnected clusters that at long times break into a pattern of stripes parallel to the *y* direction. This is precisely the behavior exhibited by each single component of the original horizontally oscillated binary mixture of disks in simulations [see Fig. 1b of Ref. [\[2](#page-3-3)]] and experiments [\[10\]](#page-3-7). An insight on the system behavior can be gained via an analogy with thermal systems with short range attrac-

FIG. 3 (color online). Panels (a), (b): Evolution of a system of disks interacting via the effective force shown in Fig. [1](#page-0-0). Lengths are expressed in units of particles diameters. Panel (a) (periodic boundary conditions in both direction) shows the evolution from an initially disordered state at $t = 0$, 4, and 100 s, while panel (b) (periodic boundary conditions only alog x) shows the evolution from an initially ordered state at $t = 0$, 8, and 300 s. Each single component of a granular mixtures subject to horizontal oscillations evolves in a similar way [[2](#page-3-3)]. Panels (c), (d): solution of the modified Cahn-Hilliard equation [Eq. ([2\)](#page-3-15)], with $K_y/2 = K_x = 1$ in a system of size 200×32 with periodic boundary conditions along x and no flux boundary conditions along y. Panel (c) shows the evolution from an homogeneous initial condition at times $t = 0$, 100, 2500, while panel (d) shows the evolution from a segregated initial condition at times $t = 0$, 750, 2500. The color scale is a measure of the density difference field.

tion and long range repulsion [\[16\]](#page-3-13). In these systems the short range attraction, which tends to induce a macroscopic phase separation with the formation of a single large cluster in the system, is frustrated by the presence of the long range repulsion adversing large clusters. Depending on the relative strength of attraction and repulsion, the attraction may dominate and the system will eventually phase separate via the standard coarsening mechanism; otherwise, if the repulsion dominates the coarsening process eventually stops when a typical domain width is reached, resulting in the formation of striped patterns [\[16\]](#page-3-13). In our granular system, as the long range repulsive component of the effective force vanishes along the *y* axis (see Fig. [1](#page-0-0), $\theta = \pi/2$), cluster growth along *y* is not frustrated. Along *x* the relative strength of the repulsion is small compared to the attraction, and the coarsening process will therefore proceed slowly as long as the external driving keeps going. The asymmetry of the interaction force therefore explains the formation and the orientation of the striped pattern.

At low values of Γ the stripes appear to be formed by disks in an ordered state [see Fig. $3(a)$]. At a higher value of Γ the stripes appears fluidlike, while at still higher values the system does not segregate. The same behavior is observed when Γ is varied in simulations [[2\]](#page-3-3), or when the area fraction of one of the two components is decreased [[2,](#page-3-3)[10\]](#page-3-7).

The effective interaction explains as well the occurrence of the dynamical instability generating the above striped pattern [[2\]](#page-3-3). The instability is best visualized when the initial state of the system is not disordered, but disks interacting with the effective force are placed on the tray in a stripe parallel to the *x* direction [see Fig. [3\(b\)\]](#page-2-0). In this condition the initially flat free surface develops a sinelike modulation which grows until it breaks giving rise to a pattern of alternating stripes perpendicular to *x* direction. The same phenomenology is observed when the two components of the granular mixture are placed on the oscillating tray in two stripes parallel to the *x* direction [see, for instance, Fig. 1a of Ref. [[2\]](#page-3-3)].

*Cahn-Hilliard approach—*As the Cahn-Hilliard equation captures the general features of spinodal decomposition of thermal binary mixtures [see [\[17](#page-3-14)] for a review], regardless of the details of the interaction potential between the two components, we expect that a phenomenological Cahn-Hilliard equation may capture the properties of our granular system when coarsening is observed. Therefore, we have investigated a Cahn-Hilliard equation for the density difference $c(\vec{r}) = \rho_1(\vec{r}) - \rho_2(\vec{r})$ of the two components, which takes into account the anisotropy found in the effective force field

$$
\frac{\partial c(\vec{r})}{\partial t} = M \nabla^2 \left[\frac{\partial f}{\partial c} - (K_x \partial_x^2 + K_y \partial_y^2) c \right] + \eta. \tag{2}
$$

Here *M* is a mobility and η a Gaussian random noise [[18\]](#page-3-16), $f(c)$ the usual double-well potential leading to phase separation, whereas the term $K_x \partial_x^2 + K_y \partial_y^2$ accounts for the ''free energy'' cost associated to concentration gradients. In the present case, as the effective interaction is not spherically invariant, the cost of an interface depends on its orientation. Schematically we take this into account by assuming $K_y > K_x$ (in case of radial isotropy $K_x = K_y$) since interfaces (i.e., concentration gradients) along *y* "cost more" than interfaces along *x*. We show in Fig. $3(c)$, which considers the case in which the initial state is homogeneous, and in Fig. $3(d)$, which reproduces the instability of an initially flat interface between the two components, that this phenomenological equation gives a good description of the dynamics. A quantitative agreement is also found as the Cahn-Hilliard equation predicts a coarsening exponent $\nu = 1/4$ during the first coarsening regime [[19](#page-3-17)], which is numerically equal to the one ob-served in both experiments and simulations [[7](#page-3-4)[,10\]](#page-3-7). According to the Cahn-Hilliard equation we expect, how-ever, a crossover [[19](#page-3-17)] to $\nu = 1/3$ at much longer times, not yet observed in experiments and simulations.

*Conclusions—*In this Letter we have generalized the effective interaction approach, widely used in the study of colloidal systems, to out-of equilibrium periodically driven mixtures, and specifically to the case of a granular mixture subject to horizontal oscillations. In this context the effective interaction approach is particularly useful, as it reduces the study of an out-of equilibrium nonthermal driven system (dissipative in the case we have explicitly investigated), to that of an ''equilibrium'' monodisperse system. For a granular mixture subject to horizontal oscillations, the effective interaction force, whose fundamental characteristic is its directional anisotropy, and particularly the presence of a repulsive shoulder at long distances which is prominent in the direction of oscillation, allows for a clear understanding of the observed instabilities and segregation processes. Its features can be cast in a phenomenological Cahn-Hilliard equation which reproduces the observed phenomenology and allows for analytical predictions. Our findings clarify, thus, the origin of the ''differential drag'' mechanism [\[12\]](#page-3-8) proposed to describe the observed phenomena and show how to interpret phenomenological hydrodynamics models [\[11\]](#page-3-18) used to depict the early stages of the stripe dynamics.

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- [1] D. J. Goldfarb, B. J. Glasser, and T. Shinbrot, Nature (London) **415**, 302 (2002).
- [2] M. P. Ciamarra, A. Coniglio, and M. Nicodemi, Phys. Rev. Lett. **94**, 188001 (2005).
- [3] T. Shinbrot and F. J. Muzzio, Nature (London) **410**, 251 (2001).
- [4] P. Richard *et al.*, Nat. Mater. **4**, 121 (2005).
- [5] J. M. Ottino and D. V. Khakhar, Annu. Rev. Fluid Mech. **32**, 55 (2000); A. Kudrolli, Rep. Prog. Phys. **67**, 209 (2004).
- [6] M. P. Ciamarra *et al.*, Phys. Rev. Lett. **96**, 058001 (2006).
- [7] M. Pica Ciamarra, M. Nicodemi, and A. Coniglio, J. Phys. Condens. Matter **17**, S2549 (2005).
- [8] S. Asakura and F. Oosawa, J. Polym. Sci. **33**, 183 (1958).
- [9] J. Duran and R. Jullien, Phys. Rev. Lett. **80**, 3547 (1998); S. Aumaitre, C. A. Kruelle, and I. Rehberg, Phys. Rev. E **64**, 041305 (2001); D. A. Sanders *et al.*, Phys. Rev. Lett. **93**, 208002 (2004); M. Bose, P. R. Nott, and V. Kumaran, Europhys. Lett. **68**, 508 (2004); G. C. M. A. Ehrhardt, A. Stephenson, and P. M. Reis, Phys. Rev. E **71**, 041301 (2005); M. Tarzia *et al.*, Phys. Rev. Lett. **95**, 078001 (2005); P. Melby *et al.*, cond-mat/0507623; D. A. Sanders *et al.*, Europhys. Lett. **73**, 349 (2006).
- [10] T. Mullin, Phys. Rev. Lett. **84**, 4741 (2000); P. M. Reis and T. Mullin, Phys. Rev. Lett. **89**, 244301 (2002); P. M. Reis *et al.*, Europhys. Lett. **66**, 357 (2004).
- [11] C. M. Pooley and J. M. Yeomans, Phys. Rev. Lett. **93**, 118001 (2004).
- [12] P. Sánchez, M. R. Swift, and P. J. King, Phys. Rev. Lett. 93, 184302 (2004).
- [13] L. E. Silbert *et al.*, Phys. Rev. E **64**, 051302 (2001).
- [14] J. Dzubiella, H. Lowen, and C. N. Likos, Phys. Rev. Lett. **91**, 248301 (2003).
- [15] According to Helmholtz's theorem any vector field **V** admits a unique decomposition in a irrotational and solenoidal component, $V = V_I + V_S$. The solenoidal component of **f**eff appears to be negligible with respect to its irrotatioanl component, $\mathbf{f}_S(r) \ll \mathbf{f}_I(r)$.
- [16] S. C. Glotzer and A. Coniglio, Phys. Rev. E **50**, 4241 (1994); M. Seul and D. Andelman, Science **267**, 476 (1995); A. Imperio and L. Reatto, J. Phys. Condens. Matter **16**, S3769 (2004).
- [17] N. Goldenfeld, *Lectures on Phase Transitions and the Renormalization Group* (Addison-Wesley, Reading, MA, 1992); A. Bray, Adv. Phys. **43**, 357 (1994).
- [18] The Gaussian random noise η is obtained as the divergence of a random vector field in order to keep the order parameter conserved.
- [19] F. Corberi, A. Coniglio, and M. Zannetti, Phys. Rev. E **51**, 5469 (1995); C. Castellano and M. Zannetti, Phys. Rev. E **58**, 5410 (1998).