Granular Segregation Driven by Particle Interactions

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We report the results of an experimental study of particle-particle interactions in a horizontally shaken granular layer that undergoes a second order phase transition from a binary gas to a segregation liquid as the packing fraction C is increased. By focusing on the behavior of individual particles, the effect of C is studied on (1) the process of cluster formation, (2) cluster dynamics, and (3) cluster destruction. The outcomes indicate that the segregation is driven by two mechanisms: attraction between particles with the same properties and random motion with a characteristic length that is inversely proportional to C. All clusters investigated are found to be transient and the probability distribution functions of the separation times display a power law tail, indicating that the splitting probability decreases with time.

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When a mixture of grains is agitated by an input of energy, constituent particles with similar properties may separate to form clusters or stripes [1–3]. This is called segregation and it may be driven by differences in particle size, density, shape, surface properties, restitution coefficient, rolling friction, and so on. Many mechanisms may cause segregation but two qualitatively distinct scenarios can be identified. In the first, particles gather together because each tends to favor a particular location. Examples of this are the Brazil nut effect in vertical shaking [4] and stripe formation during avalanches in heaps [5–7]. Another scenario occurs when there is a certain affinity or "attraction" between similar particles, which cluster without any preferred spatial location. Examples of this are the band segregation in a long drum [8] or stripe formation in a horizontally shaken granular mixture [9], where the bands or stripes can develop at different locations.

In the latter case, considerable debate has arisen over the nature of the forces that lead to the aggregation of similar particles. One possibility is an excluded volume effect [10,11] where the interaction between particles is short range on the order of one particle diameter. Another possibility is long range interactions driven by differences of either pressure [12], density [13], or velocity fluctuations [14] in the region between clustering particles. These experimental results motivated Cattuto *et al.* who found long range repulsive forces in numerical models, which they attributed to a "Casimir" effect [15]. Recently, Shaebani *et al.* [16] demonstrated in a numerical model that these fluctuation induced forces can be attractive or repulsive depending on specific values of control parameters.

Granular segregation has become of significant theoretical interest over the past two decades. So far, studies of segregation have been primarily focused on the dynamics of the developed patterns and less attention has been paid to the specific particle-particle interactions that govern the collective behavior. This is the goal of our work in which we tackle the simplest problem of two particles drifting towards each other forming a cluster that eventually splits. To this end, we have chosen to study a well-known system of a horizontally shaken shallow layer of a binary mixture of dry particles [17–21]. Stripe formation has been found in the direction perpendicular to the forcing, a behavior also observed in colloidal dispersions [22] and granular suspensions [23].

The experimental setup is the one used by Reis *et al.* [18–20]. It consists of a horizontal rectangular tray with dimensions $(x, y) = (180, 90)$ mm connected to an electromechanical shaker driven via a function generator that provides a sinusoidal vibration along the x direction. An amplitude $A = \pm 1.74$ mm and a frequency $f = 12$ Hz were set and monitored using a linear variable displacement transformer and a piezoelectric accelerometer. Most of the experiments were performed with two types of particles: phosphor-bronze spheres (1.50 mm diameter, 3.0% polydispersity, and 8.8 g cm^{-3} density) and poppy seeds which were "kidney" shaped with an average diameter of 1.06 mm, 17% polydispersity, and a 0.2 g cm^{-3} density. Three different phases were identified in this system depending on the filling fraction of the granular layer, C. At low values of C a well mixed state develops analogous to a binary gas phase. Above a critical point ($C_c = 0.65$) phosphor-bronze spheres start aggregating and mobile liquidlike clusters appear. Finally, a segregated crystal phase emerges if $C \gtrsim 0.85$. The transition from a binary gas to a segregation liquid has been systematically

studied using the stripe width as an order parameter revealing characteristics of a second order phase transition, including critical slowing down of the segregation time t_S when C is above the threshold: $t_S \sim (C - C_c)^{-1}$ [18].

With the aim of shedding light on the origin of this transition, we investigate the nature of particle-particle interactions by analyzing the behavior of just two phosphor-bronze spheres immersed in a sea of poppy seeds excited under the above conditions. The measure we use is the distance between the phosphor-bronze spheres ($\Delta - d$ as depicted in Fig. 1). The filling fraction C is defined as the number of poppy seeds N_{ps} times their two dimensional projected area A_{ps} divided by the area of the tray $C = N_{ps}A_{ps}/xy$ (note that C may reach 1 as poppy seeds can overlap). The experiments started with two phosphor-bronze spheres set at a distance $\Delta_0 - d = 3$, 8, or 15 mm apart in the x direction (and the same y position). Depending on the experimental conditions the two particles drifted towards each other to form a cluster, a process that was registered at least 220 times for each C. From this, we obtained the cluster formation probability as the ratio of clusters created with respect to the number of trials. Once formed, we measured the time that clusters stayed in this configuration. For the case of spheres placed at an initial distance of $\Delta_0 - d = 8$ mm, between 20 and 45 videos of 25 s duration were recorded at 240 frames/s for each value of C. A particle tracking program was used to extract the coordinates of the particles during each cycle of shaking (1/12 s.). From this, the dynamics of approach were obtained as well as the distance between the phosphor-bronze spheres when in the cluster configuration.

In addition, we measured the movement of a single phosphor-bronze sphere within the bulk of poppy seeds in order to characterize the effect of C on the individual

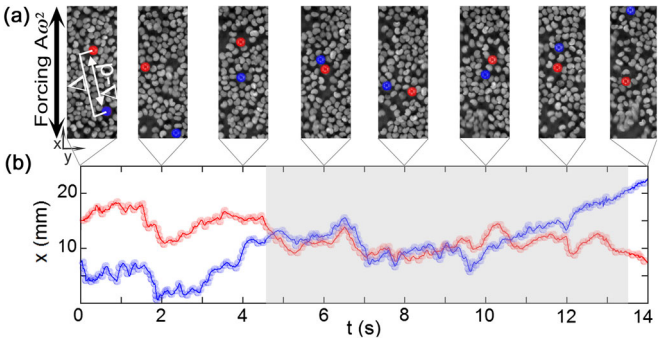

FIG. 1 (color online). (a) Time series of snapshots illustrating the interplay of two phosphor-bronze spheres for $C = 0.91$. The red or blue spots represent each sphere. In the first picture, we indicate the distance between particles ($\Delta - d$) where Δ is the distance between centers and d the phosphor-bronze sphere diameter. (b) Visualization of the trajectory of both spheres in the x direction. The shadowed region indicates the time lapse during which the particles are in a cluster conformation as defined in the main text.

dynamics. Details of these results are provided in the Supplemental Material [24] and are summarized here. The movement of the particle in the x direction was sinusoidal with the frequency of the forcing (12 Hz). In addition, there was a random diffusivelike motion resulting from friction between the tray and the particles. In the y direction only random motion was observed. As a result of the directional forcing, the characteristic displacement of the random motion per cycle of vibration is considerably larger in the x direction than in the y direction. In addition, no correlation was observed in the displacements of consecutive cycles. The effect of increasing C was to reduce the characteristic displacement per cycle in both directions. This can be seen either as a reduction of the motion of the phosphor-bronze spheres or as an increase in the damping by the poppy seeds. A detailed understanding of the complexities of this movement is challenging [25].

Concentrating on the interactions between intruders mediated by poppy seeds, in Fig. 1 we illustrate a typical approach sequence for two phosphor-bronze spheres. In this set of experiments particles were placed at an initial separation of $\Delta_0 - d = 8$ mm. In the majority of experiments, the particles approached one another and a cluster was deemed to be formed when the particles touched for the first time. Typically, the particles subsequently separated and retouched several times. Remarkably, in all the cases studied, the clusters eventually split permanently. As explained below, breakup of the cluster was considered to occur once the particles reached a separation of $\Delta - d >$ 12 mm and this criterion was used to define the cluster splitting time t_s. When particles reached this separation, their motion became uncorrelated and they quickly drifted apart, so the results were not affected by selecting separations greater than 12 mm. Here, we study the effect of packing fraction on three situations: (a) cluster formation (approach dynamics), (b) cluster dynamics, and (c) cluster splitting.

First, we show the probability that two phosphor-bronze spheres placed at 3, 8, and 15 mm approach each other to form a cluster as a function of C [Fig. 2(a)]. The probability of cluster formation increases monotonically with C and depends on the initial separation of the spheres. Curiously, for $\Delta_0 - d = 3$ and 8 mm the number of clusters created is high even for C values below the critical segregation point ($C_c = 0.65$) [18]. This indicates the existence of aggregation forces, which compete with the random movement of phosphor-bronze spheres described in the Supplemental Material [24], leading to the rich phenomenology observed in the segregation process. At the same time, the strong dependence of the cluster formation probability on $\Delta_0 - d$ hints to a decay in the aggregation forces with distance, implying the existence of an effective length above which such forces become negligible. The graph presented in Fig. 2(b) indicates that this length is around 12–15 mm.

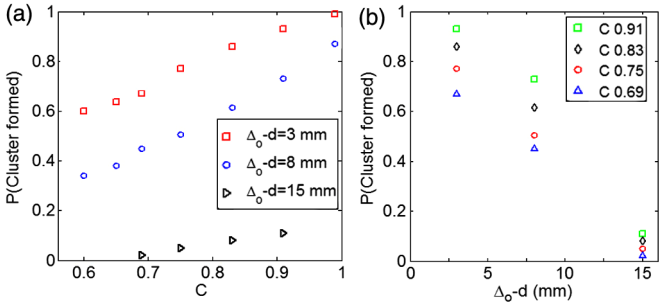

FIG. 2 (color online). (a) Probability that two phosphor-bronze spheres placed at different initial separations $\Delta_0 - d$, as indicated in the legend, approach each other forming a cluster as a function of C. In (b) the same probability is presented as a function of $\Delta_0 - d$, evidencing that for $\Delta_0 - d = 15$ mm the probability of forming a cluster becomes very low.

An indication of the competing mechanisms can be seen using the average distance between the two phosphor-bronze spheres during their approach (Fig. 3). This graph was constructed using data from runs where clusters were formed with particles initially separated by 8 mm. Interestingly, runs with the largest C have the smallest approach velocities, although, in accord with the results in Fig. 2, there is a higher probability for cluster formation. Therefore, although cluster aggregation increased with C, the mobility of bronze spheres was reduced as more poppy seeds had to be displaced, a result which is compatible with the reduction of the typical displacement per cycle with C shown in Fig. 2(b) in the Supplemental Material [24]. In summary, it can be concluded that as C increases, cluster formation is favored but the process slows.

The behavior displayed in Fig. 3 for $t > 0$ reveals that, once a cluster of a pair of particles was formed, they did not always touch each other. The distance between the particles fluctuated until they separated. We present in Fig. 4 the distribution of interparticle distances for several values

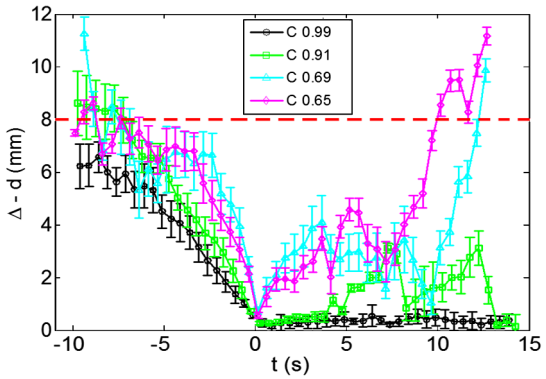

FIG. 3 (color online). Average interparticle distance $(\Delta - d)$ versus time for various values of C. The error bars denote 95% confidence intervals. For each individual trajectory, $t = 0$ indicates when the phosphor-bronze spheres first touched each other (hence, $\Delta - d = 0$). The red dashed line denotes $\Delta_0 - d = 8$ mm, the initial separation of the particles.

of C. The data were gathered between the initial touch and the split. The highest concentration ($C = 0.99$) displays a peak at $\Delta - d = 0$ reflecting that particles forming the cluster remained in contact. A slight decrease of the concentration leads to a secondary peak at $\Delta - d \approx 1$ mm, which correlates with a configuration where the two bronze spheres were separated by a single poppy seed. The peak becomes wider with further reduction in C, until it disappears for $C \approx C_c$. For all concentrations we find a negligible probability that the two particles that form the cluster are at a distance $\Delta - d > 9$ mm. This result, and the one reported in Fig. 2(b), prompted us to choose $\Delta - d > 12$ mm as the criterion for destruction of the cluster. Remarkably—in contrast to previous investigations [12–14]—in all the cases studied here the clusters eventually split, a behavior that might be caused by the strong driving force. This was also found in ongoing research with clusters of 3, 5, and 10 particles.

Now, we report results for the cluster splitting time t_s. In Fig. 5(a) the complementary cumulative distribution functions (complementary CDFs) of t_s are shown for several values of C. These distributions (also called survival or reliability functions) indicate the probability that a cluster still remains at a time T equal or larger than t_s, $P(T \geq t_s)$. This is a convenient alternative to traditional methods to obtain information from the distribution tails [26]. All distributions display power law tails ($t_s^{-\alpha}$), which are fitted using the method developed by Clauset *et al.* [26] that provides the exponent of the power law tail and the minimum value $t_{s(\min)}$ from which the fit is valid. We obtain the same exponent for all distributions: $\alpha = 2.5$ (note that the slope of the complementary CDF is $\alpha + 1$). However, the values of $t_{s(\min)}$ increase with C as detailed in the caption of Fig. 5. This enables the collapse of all distributions by rescaling the data by the average splitting time $\langle t_s \rangle$ [inset of Fig. 5(a)]. The dependence of $\langle t_s \rangle$ with C suggests that $\langle t_s \rangle \neq 0$ even below the critical point $C_c = 0.65$ [circles in Fig. 5(d)]. This finding provides further support for the competing aggregation or splitting behavior mentioned above.

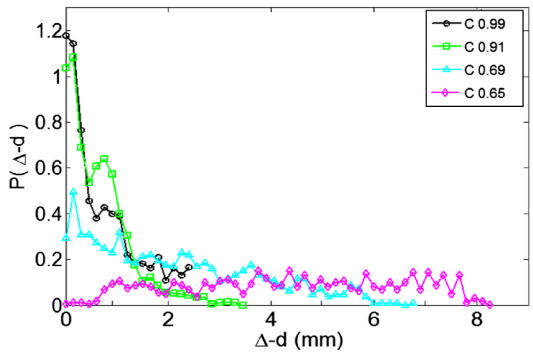

FIG. 4 (color online). Histogram of the interparticle distance $(\Delta - d)$ when the two phosphor-bronze spheres are forming a cluster for various values of C.

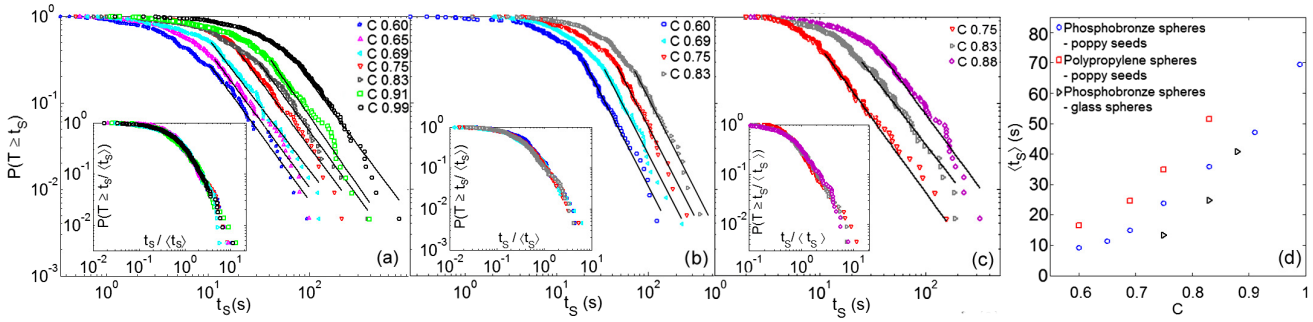

FIG. 5 (color online). (a)–(c) Complementary CDFs of the time that a cluster takes to split (t_s) for various values of C. The solid lines show the fittings of the power law tails $t_s^{-\alpha}$, which are valid for t_s larger than a given value $t_{s(\min)}$ that increases with C. In (a) phosphor-bronze spheres within a sea of poppy seeds; $\alpha = 2.5$ with $t_{s(\min)} = 10.65, 11.74, 10.87, 17.06, 32.23, 41.59, 68.93$ s for $C = 0.60, 0.65, 0.69, 0.75, 0.83, 0.91, 0.99$, respectively. In (b) polypropylene spheres within a sea of poppy seeds; $\alpha = 3.1$ with $t_{s(\min)} = 19.13, 31.79, 40.19, 67.62$ s for $C = 0.60, 0.69, 0.75, 0.83$, respectively. In (c) phosphor-bronze spheres within a sea of glass beads; $\alpha = 2.7$ with $t_{s(\min)} = 11.1, 25.0, 37.5$ s for $C = 0.75, 0.83, 0.88$, respectively. In each panel, the inset shows the collapse of the results after dividing the splitting time by the average for each value of C. (d) Average splitting time versus C for the three cases shown in (a)–(c) as indicated in the legend.

More importantly, the fact that the distribution function of t_s does not decay exponentially implies that the splitting probability is time dependent: it decreases with time. The origin of this behavior is unclear and usually relates to the existence of complex processes (for example, aging or anomalous transport). Also, a combination of exponentials with different characteristic times can lead to a power law [27]. In our case, a splitting probability depending on the spatial direction could cause a power law tail. The robustness of this counterintuitive result is demonstrated using different combinations of aggregating particles and bulk ones. In Fig. 5(b), results are displayed for polypropylene spheres within a sea of poppy seeds. Lighter polypropylene spheres with the same diameter as the phosphor-bronze ones gave an increase of the exponent ($\alpha = 3.1$) and a greater $t_{s(\min)}$ (and hence $\langle t_s \rangle$) for a given value of C [Fig. 5(d)]. This increase of $\langle t_s \rangle$ could be attributed to a reduction of inertial effects on the particles, which might also suppose a decrease of their capability of pushing apart the confining poppy seeds.

Finally, we show that changing the bulk particles also leads to the same behavior. We have measured the splitting time of two phosphor-bronze spheres within a sea of glass beads of the same diameter. Hence, the particles only differ in density. Again, we obtain power law tails emphasizing the generality of this phenomenon. Interestingly, for small values of C the average splitting time is smaller than with poppy seeds suggesting that, in this latter case, differences in sphericity prevent splitting. Nevertheless, as C increases the differences in $\langle t_s \rangle$ become negligible, which could be attributed to crystallization of the glass beads. Finally, we have investigated the effects of rolling using flat-bottomed spheres in a sea of poppy seeds. The same exponent was obtained but smaller values of $t_{s(\min)}$ and $\langle t_s \rangle$ were found (see the Supplemental Material [24]). Although stripe segregation has not been reported with the particular mixtures investigated in Figs. 5(b)–5(c) and the Supplemental Material [24], the generic behavior observed in this work and the robustness of the segregation process shown in Ref. [28] suggest its development under an appropriate parameter range.

We have reported experimental results on the particle-particle interaction of pairs of spheres within a bulk of primarily poppy seeds and other spheres when shaken horizontally. The results suggest a competition between attraction forces and random agitation. As the packing fraction C increases, the random motion reduces and the interaction between particles becomes stronger. Specifically, as C increases (1) the probability of forming a cluster is greater, (2) the peak in the interparticle distance of the clustered configuration is enhanced, and (3) the average splitting time grows. As C increases the approach velocity of the particles is also reduced. This is in accord with intuition since the number of poppy seeds required to be displaced during the approach grows with C. We expect that these detailed quantitative experimental results will stimulate numerical simulations and the development of theories to aid the further understanding of complex processes involved in segregation.

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