Granular Segregation as a Critical Phenomenon

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We present the results of an experimental study of patterned segregation in a horizontally shaken shallow layer of a binary mixture of dry particles. An order parameter for the segregated domains is defined and the effect of the variation of the combined filling fraction, C, of the mixture on the observed pattern formation is systematically studied. We find that there is a critical event associated with the onset of segregation, at $C_c = 0.647 \pm 0.049$, which has the characteristics of a continuous phase transition, including critical slowing down.

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Segregation is a counterintuitive phenomenon where an initially mixed state of dry granular particles separates into its constituent components under excitation [1]. Intriguingly, it does not always happen and the conditions for its occurrence are difficult to predict. Segregation is not only of fundamental interest but it is also of practical importance with applications in areas ranging from industry [2] to geology [3]. There has been an upsurge of interest in small scale laboratory studies have all provided interesting examples of pattern formation in granular segregation [4].

Our focus is on quasi-2-dimensional horizontally driven layers of binary mixtures of particles [5,6] as this gives the practical advantage that any collective behavior is readily visualized and the influence of gravity is effectively minimized. Moreover, the material is in contact with the drive throughout the motion. For this class of binary granular systems a qualitative segregation mechanism has been suggested [7] which uses the idea of *excluded volume depletion* as in colloidal systems and binary alloys [8]. Consider a driven 2D binary system of particles of different sizes, where the number of small particles greatly exceeds the number of large ones. The packing fraction of the system is decreased when self-organized clustering of the larger particles occurs, thereby increasing the number of configurational states of the system. Hence ordered arrangements of the large spheres can increase the total entropy of the system by increasing the free space available to the small particles. This process is called *entropic ordering* [1]. We believe that analogous attractive depletion forces are responsible for segregation in our system.

A schematic diagram of the top view of the apparatus is presented in Fig. 1. It consisted of a horizontal smooth rectangular tray, of dimensions $(x, y) = 180 \times 90$ mm with a flatness of less than ± 5 μm, on which a binary mixture of particles was vibrated longitudinally. The tray, made out of aluminum tool plate, was mounted on a system of four high precision linear bearings and connected to a Ling electromechanical shaker with a servo feedback control. The dynamical displacement and acceleration were monitored by a linearly variable differential transducer (LVDT) and two orthogonal piezoelectric accelerometers. The motion was checked to be unidirectional and sinusoidal to better than 0.1%. The forcing parameters, amplitude and frequency of vibration, were kept constant at $A = \pm(1.74 \pm 0.01)$ mm and $\omega = 12$ Hz, respectively. Static charging effects were eliminated by coating the container's surface with a layer of colloidal graphite.

The binary mixture consisted of poppy seeds ($\rho_s = 0.20$ g cm^{-3}), which were approximately ellipsoidal with major and minor axes $d_s^+ = 1.07$ mm and $d_s^{1-} \approx d_s^{2-} \approx 0.50$ mm, respectively, and larger precision phosphor-bronze spheres ($\rho_l = 2.18$ g cm^{-3}) with diameter $d_l = 1.50$ mm. The amounts used were always such that a shallow layer regime was maintained, with layer height $h < d_l$.

Forcing of individual particles arose principally from the frictional interaction with the base of the container. Although the motion of the container was sinusoidal, the stick-and-slip nature of tray/particle contact induced stochastic thermalization; quasi-2-dimensional trajectories

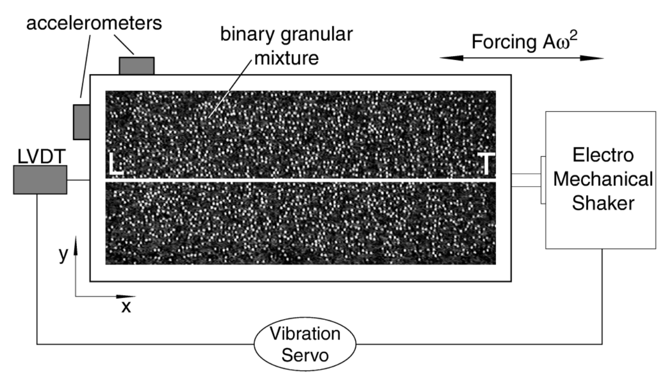

FIG. 1. Top view of the apparatus. The experiment is set horizontally with gravity pointing into the page. The inset photograph shows an example of the homogeneously mixed initial conditions of the granular layer with phosphor-bronze spheres (white regions = 1) and poppy seeds (black regions = 0).

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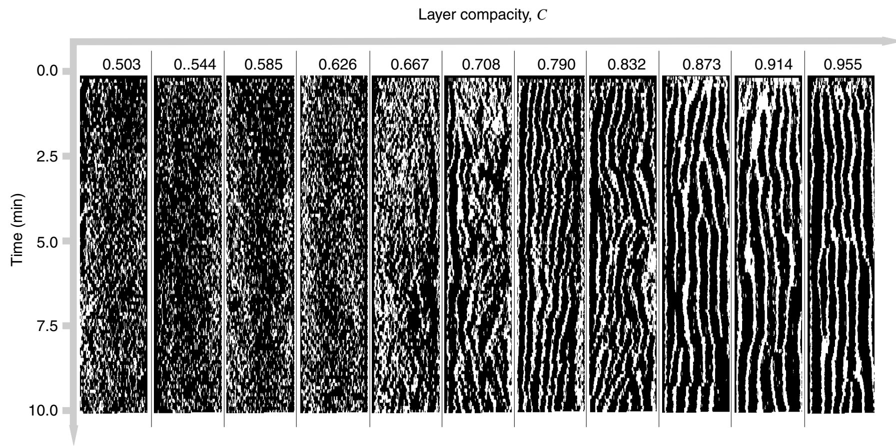

FIG. 2. A series of space-time diagrams for increasing C. Each diagram is a stack, in time, of the central line (LT in Fig. 1). The LH side of the diagram corresponds to a *gaseous* binary state. For larger values of C, on the RH side, segregated structures form within the first minute of forcing. All runs were started from homogeneously mixed initial conditions.

were obtained in x and y, with the characteristics of a 2D random walk [9]. The surface of the tray was accurately leveled with fine-pitch adjustable screws to within $\pm 0.0086°$. This ensured homogeneous forcing across the whole layer.

The granular layer was imaged using a CCD camera mounted directly above the apparatus. Image processing techniques were employed to transform the acquired video frames into binary format, giving a value of 0 (black) to pixels corresponding to regions of poppy seeds and a value of 1 (white) for those of phosphor-bronze spheres. Frames were thereafter analyzed as 600×300 digital matrices, allowing measurements on the dynamics of the granular patterns to be performed.

We define a measure of the combined filling fraction of the layer, or *layer compacity*, to be

$$C = \frac{N_s A_s + N_l A_l}{xy}, \tag{1}$$

where N_s and N_l are the numbers of poppy seeds and phosphor-bronze spheres in the monolayer, A_s and A_l are the 2-dimensional projected areas of the respective individual particles and x and y are the longitudinal and transverse dimensions of the tray. We found the results to be qualitatively robust over a range of the forcing parameters, our choice for A and ω corresponding to maximal thermalization. Hence we regard C as the principle parameter of the system.

An image of a mixture of the two types of particles has been superposed on the schematic diagram of the apparatus in Fig. 1. It is representative of the initial conditions which were consistently set using the following method. First, N_s poppy seeds were vibrated at large amplitudes, $A \sim \pm 5$ mm, creating an homogeneous and isotropic submonolayer. The phosphor-bronze spheres were then suspended above the layer, on a horizontal perforated plate with (57×28), 2 mm diameter, holes arranged in a triangular lattice. A shutter was then opened and the 1596 phosphor-bronze spheres fell onto the layer of poppy seeds, creating a near homogeneous mixture of the two types of particles.

The focus of the investigation was to systematically increase C, consequently decreasing the mobility of individual particles, and explore the conditions under which granular segregation took place. To achieve this, we incrementally increased the number of poppy seeds, N_s, in the layer, in measured steps, with the amount of phosphor-bronze spheres, N_l, held constant; i.e., $C = C(N_s)$. We chose to perform the experiments by changing the numbers of poppy seeds since this provided finer steps in the control parameter.

In Fig. 2 we present a series of space-time diagrams, for increasing C, which were constructed by sampling the midframe cut line along the granular layer's x dimension (shown in Fig. 1 as the white longitudinal line LT), and progressively stacking, in time, 200 of these $(600 \times 1$ pixels) lines (acquisition rate of $1/6$ Hz), for runs of 10 min. This period was found to be long enough for steady state conditions to be achieved.

In the left-hand (LH) image of Fig. 2, with $C = 0.503 \pm 0.035$, it can be seen that the binary layer remains mixed for the duration of the experiment. The system acts as a low density granular gas with two components and individual particles describe quasi-2-dimensional Brownianlike trajectories. The right-hand (RH) image in Fig. 2, for $C = 0.955 \pm 0.081$, displays qualitatively different behavior. Within the first minute of the start of the motion, small localized clusters of phosphor-bronze spheres (white regions) form and progressively coalesce with neighboring ones. Coarsening occurs, so that well-defined stripes are eventually formed, aligned perpendicularly to the direction of forcing as shown in Fig. 3(a).

At intermediate values of C, partially segregated states emerge suggesting that there may be a critical dependence of the process on the layer compacity. Thus we postulate there is a critical value, C_c, below which the binary granular layer remains mixed. Above C_c, as the layer is incrementally compacted, segregation structures develop in a nonlinear manner. Specifically, we use ideas from phase transitions to explore the phenomenon.

We first establish an order parameter, ϕ, which we choose to be the averaged longitudinal width of the phosphor-bronze (white) domains. For each acquired frame this was calculated by scanning through each of the 300 horizontal lines; the length, L, of the *white steps*, $\binom{0\to 1}{1\to 0}$, was measured which typically yielded a distribution of ~6000 identified stripe widths. An example of such a single measure, L_i, is shown on a segregation pattern in Fig. 3(a). These were distributed normally, as shown in Fig. 3(b), where a fitted Gaussian function has been superimposed on the experimental histogram of L. Since the distribution is well defined, we have chosen the order parameter to be $\phi = \langle L \rangle$, i.e., the average value for the distribution of longitudinal widths of the segregated stripes.

In Fig. 3(c) we present a time series of ϕ for various C, where each run has been started from the mixed state. At large compacities, e.g., $C = 0.873 \pm 0.073$, there is a fast initial growth which saturates after ~1 min. The superimposed solid lines are fits of

$$\phi^{\mathrm{fit}}(C, t) = \Sigma_\phi(C) - \alpha \exp\left(-\frac{t}{t_s(C)}\right), \qquad (2)$$

to the experimental time series of ϕ where $\Sigma_\phi(C)$, the value at which ϕ saturates, is the *segregation level*, $\Sigma_\phi(C) - \alpha$ is the initial value, $\phi(t = 0)$, and $t_s(C)$, the time scale associated with the saturation of the order parameter, is the *segregation time*.

The C dependence of the segregation level is presented in Fig. 4. We see that for low compacities, up to a critical value the segregation level remains constant at $\Lambda_\phi(C < C_C) = 3.21 \pm 0.08$ mm. This corresponds to approximately two sphere diameters so that any domains are not classified as structures. As C is increased past C_c, segregated stripes of increasing ϕ emerge and the segregation level exhibits a square-root singularity. The solid curve in Fig. 4 is the line of best fit of the form,

$$\Sigma_\phi^{\mathrm{fit}} = A(C - C_c)^\beta + \Lambda_\phi, \qquad (3)$$

where A is the transition scaling factor, $\{C_c, \Lambda_\phi\}$ is the critical point at which the transition occurs, and $\beta = \frac{1}{2}$ is the order parameter exponent. The numerical values for the fitted parameters were $A = 2.81 \pm 0.07$ mm, $C_c = 0.647 \pm 0.049$. The value C_c was found from the intersection between the linear least square fit of the square of the segregation level, Σ_ϕ^2, and the square of the pretransition level, Λ_ϕ^2, as shown in the inset of Fig. 4.

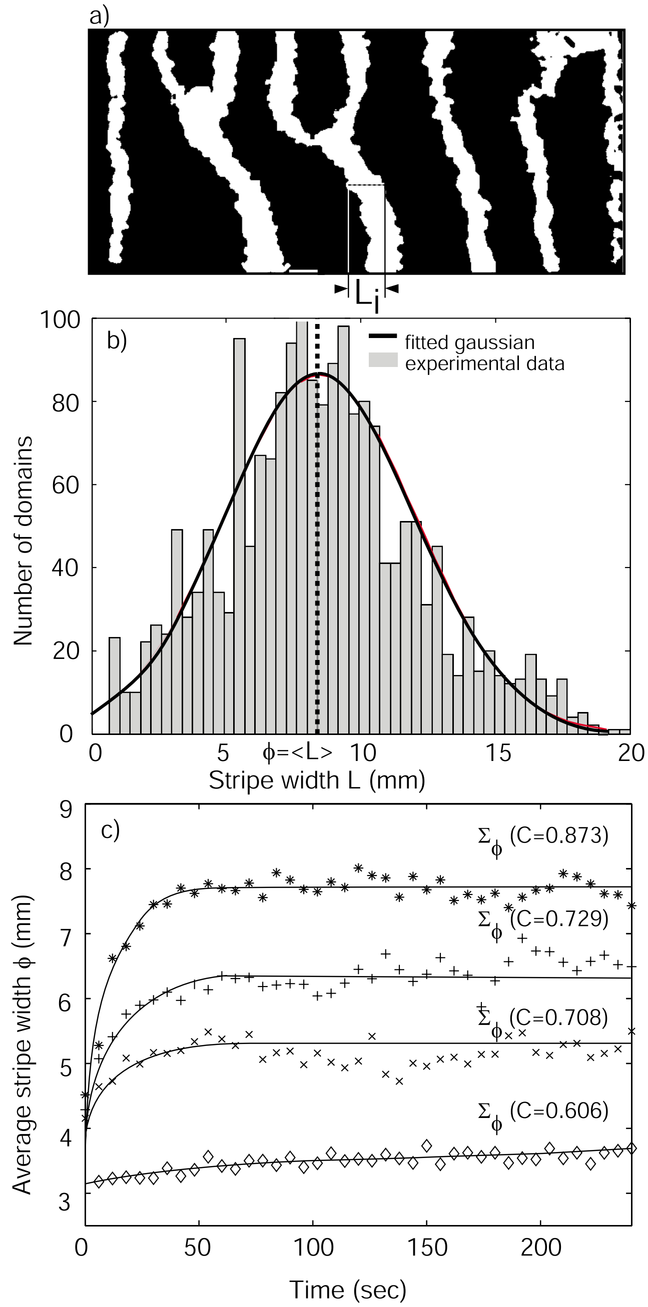

FIG. 3 (color online). (a) Frame of segregation pattern showing a single measure of a longitudinal stripe width L_i ($C = 0.873$, $t = 3$ min). (b) Histogram of the L distribution. Solid curve is Gaussian fit. Dashed line is average value $\phi = \langle L \rangle$ which we define to be the order parameter. (c) Time series for the order parameter ϕ for various values of C. A saturation level is attained after a fast initial segregation period. Solid curves are fits to Eq. (2).

It is interesting to note that the generally accepted value of the maximum packing fraction for a random closed packing for a 2D system of particles [10] is around

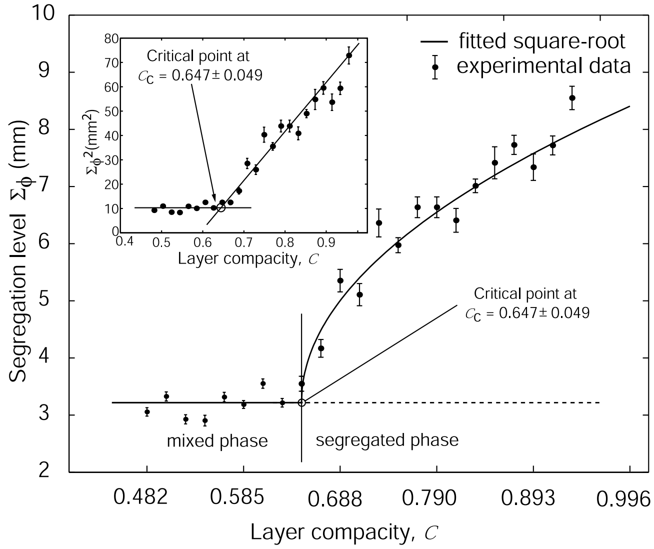

FIG. 4. Phase transition plot for segregation level. Solid line is theoretical curve for square-root singularity. Critical point occurs at $C_c = 0.647 \pm 0.049$. The mixed phase exists below this point and the segregated phase above it. The inset shows Σ_ϕ^2 which was used to determine the numerical value of C_c.

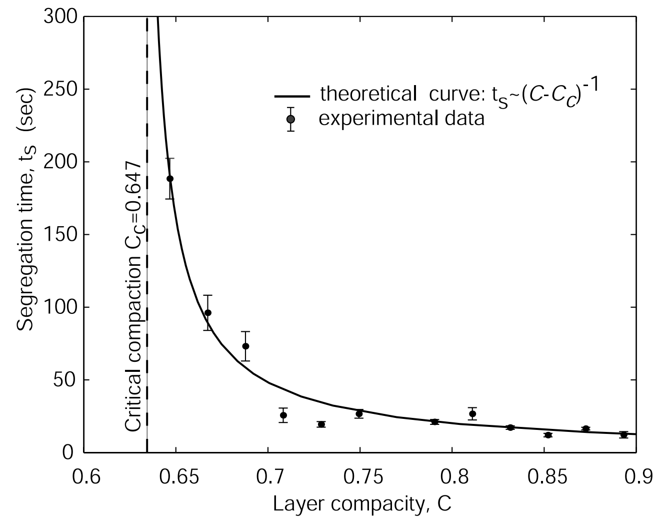

FIG. 5. Plot of segregation time, t_s, which diverges as the critical point C_c is approached from above. Solid line is theoretical curve of the form: $t_s \sim (C - C_c)^{-1}$.

0.82 which is considerably larger than C_c for segregation in our system. This seems to indicate that the segregation transition occurs while the granular mixture is still in the monolayer regime.

A further measure that supports the notion of critical behavior is the C dependence of the segregation time, t_s, extracted from the fittings of $\phi(C, t)$ to Eq. (2). Note that t_s is only defined for $C > C_c$, corresponding to the region where segregation occurs. The measured segregation time is plotted in Fig. 5 as a function C. As expected for a continuous phase transition, t_s diverges, as C is decreased from above, near C_c. This is usually referred to as *critical slowing down* where the divergence has the form of $t_s \sim (C - C_c)^{-\gamma}$ [11]. The solid line in Fig. 5 corresponds to the mean field approximation result, $\gamma = 1$ [12].

We have also performed experiments for various amounts of large spheres, N_l, and found that the location of the critical point C_c remained unchanged over a wide range, indicating that the compacity $C(N_s, N_l)$ is an appropriate parameter for the transition. Furthermore, the scaling parameter of the phase transition, A in Eq. (3), was found to depend linearly on N_l.

In summary, we have shown that granular segregation in a horizontally shaken binary layer displays both qualitative and quantitative features which are consistent with a continuous phase transition. The critical exponents for the order parameter and segregation time agree with those of mean field type behaviors, $\beta = \frac{1}{2}$ and $\gamma = 1$, respectively. Recent theoretical developments on critical behavior in granular hydrodynamics [13] and self-organization in compaction models [14] may be crucial to elucidate the mechanisms underlying the critical nature of mixing and segregation in granular materials. This could be important for practical purposes as, below a critical value of the control parameters, mixing of a binary granular system is guaranteed: a process that, to date, is difficult to predict.

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