Self-organized patterns in collections of chains

T. Sykes and T. Mullin*

Manchester Centre for Nonlinear Dynamics, The University of Manchester, Manchester M13 9PL, United Kingdom

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Results are presented of an experimental investigation into patterned segregation in thin layers of poppy seeds and short lengths of metal chains subjected to vibration. Critical phenomena are uncovered and both continuous and discontinuous transitions are observed. A phase diagram for the behavior is mapped out and a tricritical point that separates hysteretic from continuous segregation is identified. Remarkable similarities are found between the observed behavior in this driven granular system and phase separation phenomena in mixtures where the dynamics of the constituent components are markedly different.

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Spontaneous pattern formation in dissipative systems driven far from equilibrium is of broad interest across the sciences [1,2] primarily because of its widespread importance in the natural world. Examples include convection cells in the solar atmosphere [3] and dried mud [4] to the emergence of complex organisms from a single cell [5]. The specific example we study here is concerned with driven granular segregation [6] in which both the creation of a transient network and the formation of a striped pattern are found. The investigation is concerned with the self-assembly of a striking pattern formed in a thin layer of a mixture of small metal chains and poppy seeds which is subjected to vibration. We find it helpful to draw analogies with ideas from equilibrium phase separation [7,8] in multicomponent mixtures to aid progress in understanding observed phenomena.

Segregation has attracted widespread attention in recent years [6,9–11] both for its practical and fundamental interests. Insights into the mechanisms which drive granular segregation remain aloof since differences in size, shape, roughness, and density of the particles can all play a role. Previous investigations of horizontally driven granular mixtures [12–14] have indicated that a critical concentration ratio of the species is required to initiate segregation. These findings indicate that insights can be gained into the underlying physics using ideas from nonequilibrium statistical mechanics. This new approach to segregation has subsequently stimulated several numerical investigations [15–19], which have all had success in reproducing patterned segregation but have not yet been able to duplicate critical behavior. Here, we show that the introduction of strong shape anisotropy introduces unique aspects into the nature of the critical phenomena indicating that even deeper connections with ideas from phase transitions may be possible. In particular, we find the transition from supercritical to subcritical behavior with associated hysteresis.

Our investigation of patterned segregation was performed by horizontally vibrating a tray holding essentially single layers of particles. The initial conditions for each experiment were mixtures of small anisotropic poppy seeds with interspersed flexible chains of linked metal spheres. Analogies between long vibrated chains of this type and the dynamics of polymers driven far from equilibrium have been discussed previously [20,21]. Here, we extend the analogy by immersing the chains in a bed of poppy seeds which acts as a fluid or a damping medium. The dynamic asymmetry in the time scales of the motion of the constituent components of the mixture [8] suggests that certain features will be present in the segregation process. An example of this can be seen in the sequence of snapshots of the observed pattern formation presented in Fig. 1. The initially mixed state progresses to a striped pattern after ≈300 s by passing through a transient network stage between ≈60 and ≈180 s. This clear evidence for segregation under excitation contrasts with the static phase behavior in the packing of anisotropic mixtures of this type where it has been shown that complete segregation does not occur [22].

The mixtures were held in a horizontal tray with an aspect ratio of 2 and dimensions of 90×180 mm^2 mounted on precision bearings and attached to a large electromechanical shaker with proportional feedback. The tray was vibrated at a frequency of 12 Hz with an amplitude of 1.74 ± 0.01 mm and the phenomena are robust over a range of these parameters. Full details of the experimental arrangement and its control are given elsewhere [12]. The approximate monolayer of particles comprised poppy seeds and small lengths of commercial bath plug chains. The poppy seeds were disklike ellipsoids with approximate dimensions of

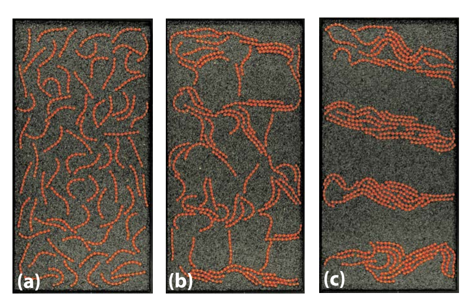

FIG. 1. (Color online) Snapshots for the development of patterned segregation where the images are of the view from vertically above the apparatus. The direction of the applied periodic excitation was along the long direction of the images. The tray was vibrated at a frequency of 12 Hz with an amplitude of 1.74 ± 0.01 mm. There were 74 chains present which occupied ~16% of the area of the tray. (a) Mixed initial state, (b) transient network state formed after 75 s, and (c) pattern formed after 100 s.

*tom@reynolds.ph.man.ac.uk

$1.07 \times 0.5 \times 0.5\ \text{mm}^3$ and a polydispersity of 17%. The chains were flexible and consisted of nine linked mild steel spheres of diameter 2.2 ± 0.11 mm connected by 0.5 ± 0.02 mm diameter rods. Each chain weighed 0.37 ± 0.005 g, which is approximately 1000 times the mass of a poppy seed. Hence, there were clear differences in the size and weight of the constituent components and the smaller poppy seeds also generally covered a significantly greater proportion of the surface area of the tray (see axis scales in Fig. 3).

The particles were driven by stick-slip interaction with the surface of the tray which randomizes the motion of the poppy seeds while the chains displayed a preference to align in a direction orthogonal to the applied vibration. This nematiclike alignment [22] was also observed in the absence of poppy seeds although no obvious clustering was found in this case. In the mixtures, the motion of the chains mainly comprised sliding and no obvious rolling was observed. The distribution of the step lengths of the motion of the poppy seeds when sampled at a fixed point in the cycle was found to be Gaussian with an approximately 50% greater standard deviation in the direction of the excitation.

The two parameters that were varied were the numbers of poppy seeds (N_{ps}) and chains (N_{ch}). These control parameters were made nondimensional by dividing the respective projected areas of the particles by the total available area of the tray (xy).

$$C(N_{ch}N_{ps}) = \frac{N_{ch}A_{ch} + N_{ps}A_{ps}}{xy} = \varphi_{ch} + \varphi_{ps},$$

where $A_{ps}=0.90 \pm 0.15\ \text{mm}^2$ and $A_{ch}=63.05 \pm 0.25\ \text{mm}^2$. These will be called the packing fractions of the respective species and define the axes of control parameter space. Experiments were usually performed by keeping the number of chains fixed at a prescribed value and varying the number of poppy seeds. In this way one-dimensional sweeps were made across parameter space by changing the number of the smaller particles, which permitted sensitive changes in this parameter to be made. The number of chains was changed, another sweep was made, and so on.

Each rehearsal of the experiment was initiated by establishing a uniform layer of poppy seeds under excitation. The chains were placed on the layer at random positions with no preferred orientation. Video images of the tray were recorded and it was found that between 2 and 12 min was sufficient time for transient behavior to die out with lower times required for higher packing fractions. Hence, a stationary state was established after this period and it was either mixed or segregated depending on the concentration ratio.

The width of clusters was estimated by averaging 300 horizontal video lines of individual images which were also passed through a threshold filter. The average stripe width was measured and used as the order parameter which enabled the construction of response diagrams for the system. Examples of response diagrams are presented in Fig. 2 where the average stripe width is shown plotted against the packing fraction of poppy seeds for 74 chains in Fig. 2(a) and 25 chains in Fig. 2(b). At low packing fractions the horizontal portions of the plots in both cases correspond to the width of

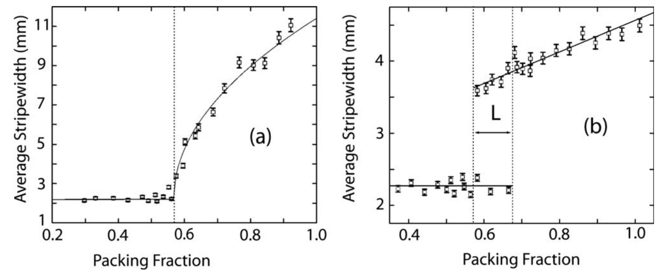

FIG. 2. Response diagrams for mixtures of poppy seeds and (a) $\varphi_{ch}=0.156$ (74 chains) and (b) $\varphi_{ch}=0.052$ (25 chains). The average stripe width is plotted as a function of total filling fraction. The horizontal lines drawn through the data correspond to the width of a single chain. The fitted curve to the right of the vertical line in (a) is a square root and all other fitted lines are linear.

a single chain indicating that the material remains in a mixed state. Above a poppy seed packing fraction of ≈ 0.58 the results for 74 chains shown in Fig. 2(a) indicate that the averaged stripe width grows continuously and has a square-root dependence on the poppy seed packing fraction. This is consistent with a second-order transition and is in accord with previously reported results for poppy seeds and spheres [12].

On the other hand a qualitatively different growth of the pattern is found for smaller chain numbers as illustrated with the response diagram for 25 chains given in Fig. 2(b). In the range of the packing fraction denoted by "L" in Fig. 2(b), both segregated and mixed states were observed, i.e., the state realized in any given experiment depended on the initial conditions. The mixed state was formed from disordered initial conditions and persisted for all periods of excitation up to packing fractions of ≈ 0.68. Above this value the application of excitation produced a definite pattern within a few minutes of the initiation of the drive. Yet further increase in the initial packing fraction gave rise to an approximately linear growth in the final average stripe width.

In order to obtain estimates of the lower stability limits of the patterned state, a different experimental procedure was adopted. The initial state was generally set to a four-stripe pattern with an appropriate stripe width. (This was changed to three clusters when less than 20 chains were used.) Vibration was applied and the pattern either remained or disintegrated within a few tens of seconds depending on the packing fraction of poppy seeds. When the pattern remained, it was found that the final stripe width after vibration was smaller than the initial one, i.e., some compaction of the artificially created stripe occurred. The experiment was restarted with a reduced packing fraction and the procedure repeated iteratively enabling the section of the upper branch in Fig. 2(b) in the region labeled "L" to be traced out. Iteration proceeded until excitation caused the pattern to disappear within a few tens of seconds of applying the excitation. Subdivision of the change in the packing fraction of the poppy seeds provided an estimate of the limit point. Hence, both mixed and patterned segregated states were stable and observable over a range of packing fractions.

The set of critical points that separate the mixed and segregated states was mapped out by using a sequence of re-

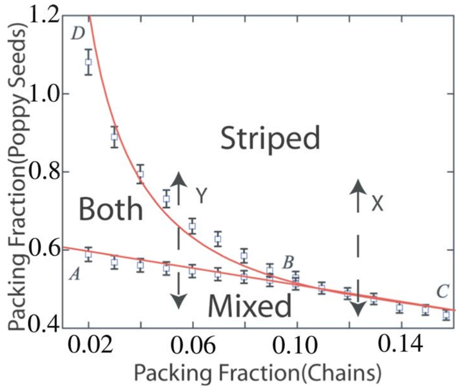

FIG. 3. (Color online) Phase diagram for the mixture of chains and poppy seeds. The critical points were measured by varying the quantities of poppy seeds for set numbers of chains, i.e., vertical slices taken across the parameter space. The fitted lines are *AC*, $y=(1.011\pm0.032)x+0.614\pm0.042$, and *DBC*, $y=(0.0175\pm0.005)/x+0.342\pm0.023$. Mixed states were found below *ABC* and patterned segregation above *CBD*. Both states were observed inside the region denoted by *ABD*. The arrowed paths labeled "X" and "Y" indicate typical slices taken by changing the number of poppy seeds.

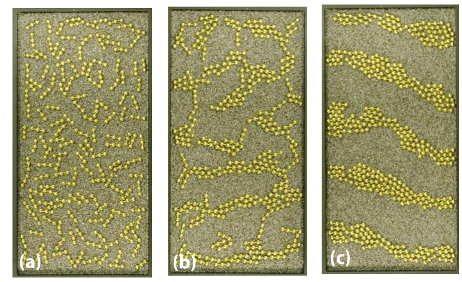

FIG. 4. (Color online) Mixture of poppy seeds and brass chains vibrated at a frequency of 12 Hz with an amplitude of 1.74 ± 0.01 mm. The chains are ≈ 12 mm long and each comprises three 3.25 mm hollow spheres linked by 0.7 mm cylinders. (a) Initial conditions, (b) after 75 s, and (c) after 100 s.

sponse diagrams which were constructed by either adding or removing poppy seeds while keeping the number of chains fixed. These gave one-dimensional cuts through the two-dimensional parameter space and are indicated by the arrowed example paths "X" and "Y" in the "phase" diagram presented in Fig. 3.

Three distinct regions of behavior were found. The mixture did not segregate when the packing fractions were set to values below those corresponding to the line labeled *ABC* in Fig. 3. Conversely, when particle numbers were such that the mixture concentration was above the curve labeled *DBC* a striped segregated pattern was always produced. Crossing the locus marked *BC* by adding poppy seeds gave rise to the continuous evolution of a striped pattern with square-root growth in the width of the stripes as typified by the results in Fig. 2(a). When the numbers of chains were reduced such that point *B* was approached the growth curve became steeper and was approximately quartic close to *B*. Hence, *B* is analogous to a tricritical point in equilibrium phase transitions [23]. This feature can also be considered as the transition from a supercritical to a subcritical bifurcation as found, for example, in Taylor-Couette flows [24]. While this is an equally appealing description, it raises the vexed question of identifying all the branches of the bifurcation and consequently we prefer the analogy with phase transitions.

Yet further reduction in numbers of chains led to the abrupt appearance of the segregated pattern when path *BD* was crossed. Further, the mixed state was only regained when *AB* was crossed by reducing the number of poppy seeds to a lower value. Clearly, removing poppy seeds could only be carried out in an approximate manner in each rehearsal of the experiment. However, the systematic behavior uncovered suggests that the observed behavior was robust. The divergence between *BD* and *AB* indicates the increasing amount of hysteresis in the transition from mixed to segregated states as the number of chains was reduced.

As stated in the caption of Fig. 3 the least-squares fitted line *DBC* shows that the critical packing fraction of poppy seeds required for the onset of segregation is inversely proportional to the number of chains. Interestingly, we have also found that this proportionality holds when the large particles are 1.5 mm diameter metal spheres [14]. Moreover a steepening of the associated response diagrams was also found but there was no evidence for hysteresis when spheres are used as the larger particles.

The physical process of clustering was observed to proceed in the following way. The chains moved slowly in a random sea of smaller particles and when two happened to meet they moved along each other in a zipping action, i.e., displacement of intervening poppy seeds took place by two chains first forming a wedge. This is an intuitively reasonable process since side-on merging of chains would otherwise require the displacement of all intervening poppy seeds. Individual clusters formed through repetitions of this procedure and subsequent merging of clusters proceeded in a similar manner. On the other hand, if there were insufficient numbers of small particles surrounding any given pair, they glanced one another and then parted. Equally, an artificially created pair would not survive if there were insufficient numbers of poppy seeds surrounding it. While the local interaction between chains was important the $1/N$ dependence suggests that cooperative behavior is of equal importance so that the interaction between all of the particles contributes to the critical conditions.

The lower threshold labeled *ABC* in Fig. 2 displays weak dependence of the packing fraction of poppy seeds required to maintain a segregated pattern on the chain number. This effect is qualitatively different to that found for spheres where there is no evidence for hysteresis. This result is consistent with what one would expect from an entropic force argument [25] where long chains will have a significantly increased external pressure over individual collections of spheres. Hence, a considerable reduction in the numbers of smaller particles was required before the pattern disintegrated and hysteresis was found.

In summary, our experimental results have enabled us to uncover a segregation phase diagram over a range of parameter space where we have found convincing evidence for both first- and second-order phase transitions with an intervening tricritical point. This rich structure has arisen from the introduction of strong shape anisotropy into one of the components in the mixture. Critical phenomena in the segregation process have been shown to be robust and this aspect remains an outstanding challenge to theory and simulation. Moreover, the robustness of the segregation phenomena is demonstrated in the sequence images in Fig. 4 where qualitatively similar features can be seen in mixtures of poppy seeds and brass chains.

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