Coarsening of Self-Organized Clusters in Binary Mixtures of Particles

T. Mullin
Department of Physics and Astronomy, University of Manchester, Manchester, United Kingdom
(Received 2 September 1999)

Experiments on patterned particle segregation in binary granular mixtures which are subjected to horizontal shaking have been performed. A novel mechanism for separation is found, where random forcing of one species by the other causes clustering. A pattern is formed which contains a series of stripes aligned orthogonal to the direction of the periodic forcing. The pattern coarsens with time and shows a power law behavior which is consistent with simple models of the geological process of "stone striping."

PACS numbers: 45.70.Qj, 47.54.+r, 64.75.+g

Segregation of mixtures of granular materials is a topic which is of interest to a broad range of scientists from geologists [1] to physicists [2] and engineers [3]. The process has been reported in avalanching [4,5] and vertical vibration [6] of binary mixtures. Here we report a novel pattern-forming segregation in a binary mixture subjected to periodic horizontal forcing. We uncover a surprising self-organization process which, like some other studies of pattern formation in granular media [7], continually evolves in time. However, we find that the average width of the stripes which make up the pattern follows a power law which is in remarkable agreement with a model problem developed for the geological phenomenon of "stone striping" [8]. In this, larger stones are observed to form stripes aligned downhill on mountainsides where there is soft soil which cyclically freezes and melts. The nightly periodic freezing and melting is believed to induce short term random forcing of the stones by the soil. In our experiment we apply a periodic forcing and observe the random motion directly so that we are able to develop an understanding of this important phenomenon.

Our experimental system was a horizontal tray which contained a shallow layer of particles and was vibrated from side-to-side at a fixed frequency and amplitude. The phenomena described below are robust and found for a range of particles but here we will report on just two mixtures which represent the limits of the density differences between particle types. In both cases there were approximately twice as many small particles as large ones. The first mixture comprised accurate 0.5 mm polystyrene spheres ($\rho = 1050\,\mathrm{kg\,m^{-3}}$) together with approximately 800 "100's and 1000's" cake decorations ("sprinkles") ($\rho = 1588\,\mathrm{kg\,m^{-3}}$) which were roughly spherical with an average size of 1.5 mm. The other mixture was 1000 1.5 mm copper balls ($\rho = 8960\,\mathrm{kg\,m^{-3}}$) and poppy seeds ($\rho = 200\,\mathrm{kg\,m^{-3}}$) which were disklike with an approximate diameter of 1.25 mm and thickness of 0.5 mm.

An example of a pattern of stripes in the polystyrene sphere/"100's and 1000's" mixture is given in Fig. 1. It was taken 15 min after the oscillation ($f = 12.8$ Hz, $a = \pm 1$ mm) is initiated and the dark particles are sitting on top of the layer of polystyrene spheres. The "granular temperature" [9] $\Gamma = A\omega^2/g$ is approximately 0.66, i.e., it is less than 1, which is required for convection and segregation in vertically shaken systems.

We will first make some general remarks concerning the processes involved in the segregation. The poppy seed/copper ball mixture will be used as our example since events were more obvious to the naked eye with this combination. Copper balls alone rolled around the tray in an apparently random fashion with ballistic collisions between balls and the walls. The addition of a small number of poppy seeds slowed this movement down but there was no obvious correlation to the motion. Clumping of the copper balls first occurred when there was approximately a single layer of poppy seeds and thus there appeared to be a critical amount of the small particles required to form structures. The addition of further amounts of poppy seeds produced more definite patterns, and then a series of quantitative measurements were made. The small particles

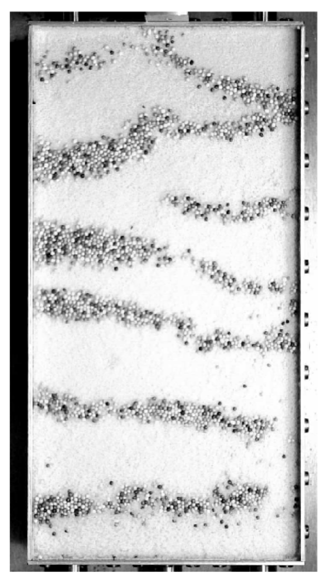

FIG. 1. Stripe pattern formed by oscillating a mixture of "100's and 1000's" and polystyrene beads at ± 1 mm and 12.8 Hz for 15 min. The colored sugar particles are on top of the double layer of smaller particles and the stripes are orthogonal to the direction of the vibration.

moved randomly and a colored trace in a layer of small particles alone was observed to diffuse preferentially in the direction of the forcing with no evidence of the pattern formation found at large amplitudes of excitation [10]. In the mixtures, the small particles retained this random and rapid motion, whereas single large particles moved more slowly with a preference for movement in the direction of the applied forcing.

Each experiment was initialized by stirring the mixture by hand until it appeared uniform. Of course, there were inevitably small groupings of the copper balls at random locations across the tray and these provided seeds for the formation of clusters. The vibration was initiated and random impulses from the poppy seeds moved the individual copper balls until they encountered others, i.e., forcing of the copper balls was a random process driven by buffeting from the smaller particles. The copper balls moved preferentially along the direction of the forcing and so when they met other balls they stayed together, since they were forced from the outside as the poppy seeds were excluded from the interstitial space. The individual pairs of balls then met others and soon larger clusters formed at random locations. These were usually observed within a few minutes of the start and again the small particles were excluded from the interior of clusters in a manner similar to that found in binary colloids [11]. The clusters thereby joined to form thin stripes which were orientated with their long axes orthogonal to the direction of the vibration. Merging of the stripes took place on the time scale of hours for the copper ball/poppy seed mixture but was quicker and took tens of minutes for the polystyrene sphere/"100's and 1000's" mixture. In all cases the merging slowed progressively as the stripes broadened, presumably as inertia became more important. The final state was nonunique and could be a single stripe at either end or in the middle of the tray, or even multiple stripes distributed along its length. A range of forcing frequencies and amplitudes was explored and more complex behaviors such as periodic motion and chaos have also been observed. The periodic motion had a time scale of hours and was not obviously related to the driving frequency.

The apparatus was a shallow level tray of length 20 cm, width 10 cm, and depth 1 cm with a machined flat surface mounted on a set of linear bearings connected to an electromagnetic linear vibrator. This was driven by an oscillator with a proportional integral derivative feedback control. The motion of the carriage was monitored using a linear displacement transducer and accelerometer which showed the movement to be sinusoidal to better than 1%. The entire apparatus was mounted on a heavy base which could be leveled using adjusting screws and was checked to be horizontal using a machinist's spirit level.

The motion of the particles was monitored using a CCD camera and both real- and elapsed-time processing was performed. Patterns were identified and tracked using edge detection methods and the averaged width of the clusters was estimated from 100 equally spaced lengthwise line scans taken at preset time intervals. This statistical measure was essential since the stripes were not always uniform across the tray. Each experiment took between one day and one week to perform, depending on the mixture of particles and the amplitude of the drive which was typically less than 2 mm peak-to-peak.

We show in Fig. 2 a sequence of images taken when the forcing frequency was 12.8 Hz and the amplitude was ± 2 mm. The images were taken at intervals of 5 min, 10 min, 15 min, 30 min, 1 h, and 6 h after the vibration was started. The ordering in the figure is left-to-right and downwards. The light-colored stripes containing copper balls are orientated orthogonal to the direction of the motion of the carriage. Interesting features such as disclinations, which were an essential part of the merging process, can be seen. The stripes appear to be nearly two dimensional but the detailed motion of individual balls within the stripes was observed to be three dimensional. A feature of these particular stripes is that the copper balls displace the adjacent poppy seeds, causing them to pile up, opening up spaces in regions which are rich in copper balls. The above formation sequence was also observed for the polystyrene beads/"100's and 1000's" mixture. However, in this case, the clusters rolled on top of the polystyrene beads and remained compact, i.e., gaps between the larger particles were not evident.

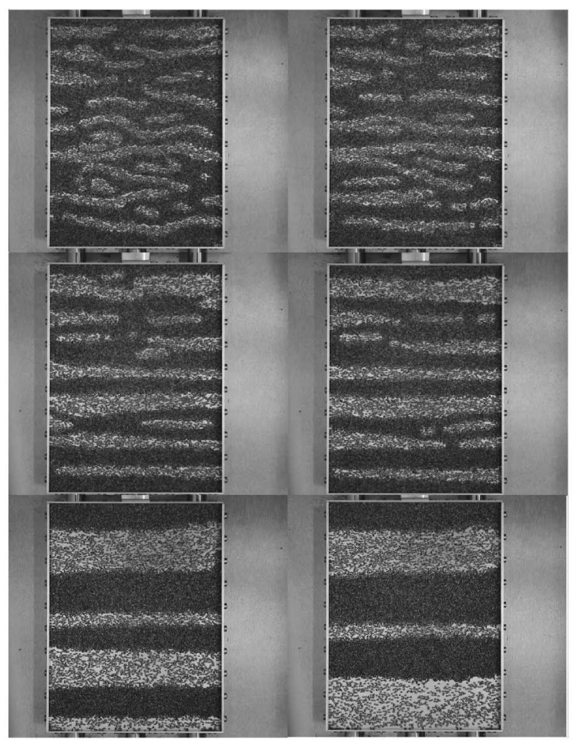

FIG. 2. Sequence of images of the evolution of the pattern for the copper ball/poppy seed mixture vibrated at ± 2 mm and 12.8 Hz. The images were taken at 5 min, 10 min, 15 min, 30 min, 1 h, and 6 h. The stripes are orthogonal to the direction of the vibration.

The coarsening of the pattern may be seen in Fig. 3 which was constructed from a single video line taken along the center of the tray and plotted approximately every 30 s for a total period of 3 h. The light-colored stripes containing copper balls can be seen to merge as time progresses from left-to-right. The initial state at the left-hand edge of the figure is random but this is covered by the first two video lines in the time scale used here and hence is not resolved. The pattern emerges quickly from the randomized initial state at the left-hand edge and then coarsens on a much longer time scale as individual stripes meet.

We next show in Figs. 4(a) and 4(b) the average width of the clusters measured as a function of time plotted on a log-log scale. The unit of time we have used is the period of the applied oscillation cycle. The results shown in Fig. 4(a) are for the copper ball/poppy seed mixture and are averaged over three experimental runs, each of which took approximately 34 h. The steps in the graph correspond to periods when the merging of larger stripes took place and the process slowed down noticeably. The slope of the solid line is 0.25, which is power law expected from Mulheran's model of stone striping. The results shown in Fig. 4(b) are for the polystyrene beads/"100's and 1000's" mixture and the complete data sets from seven runs of the experiment, each of which took approximately 2 h. The slope of the solid line is again 0.25.

The experimental results are remarkably consistent with the power laws found in a numerical simulation of a lattice model of stone striping [8]. The robustness of the scaling law with very different density ratios is particularly noteworthy. The model contains a random walk, where the diffusion term is allowed to vary with time as in the model of normal grain growth by Louat [12]. The forcing in the model is random and diffusionlike and has a preference for motion along the direction of the stripes and thus is orthogonal to the present case. Merging of the stripes in the model takes place by stones diffusing between stripes and so the natural length scale for the diffusion coefficient is the average distance between stripes. A power law of $t^{0.25}$ is then found so that the stripes initially merge quickly and then the increase in width slows down as the inertia of the stripes increases. Coarsening of the pattern in the experiment takes place when neighboring stripes merge and so it is not clear if the averaged width is the correct length scale. However, the fact that the same scaling law is found suggests that the appropriate length scales are the same. One possibility is that, since the inertia of a stripe is

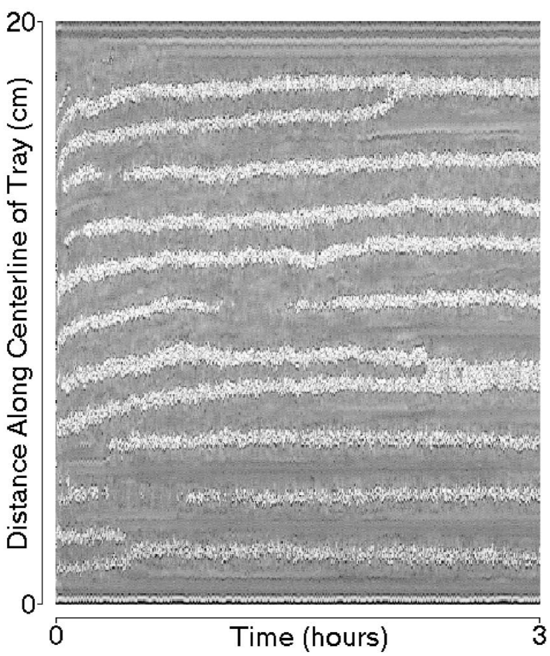

FIG. 3. Time-lapse image of the evolution of the pattern for the copper ball/poppy seed mixture vibrated at ±2 mm and 12.8 Hz. A single video line taken along the center of the tray is plotted as a function of time. It may be seen that a definite pattern was established rapidly from an initially random state, whereas coarsening by merging takes place on longer time scales.

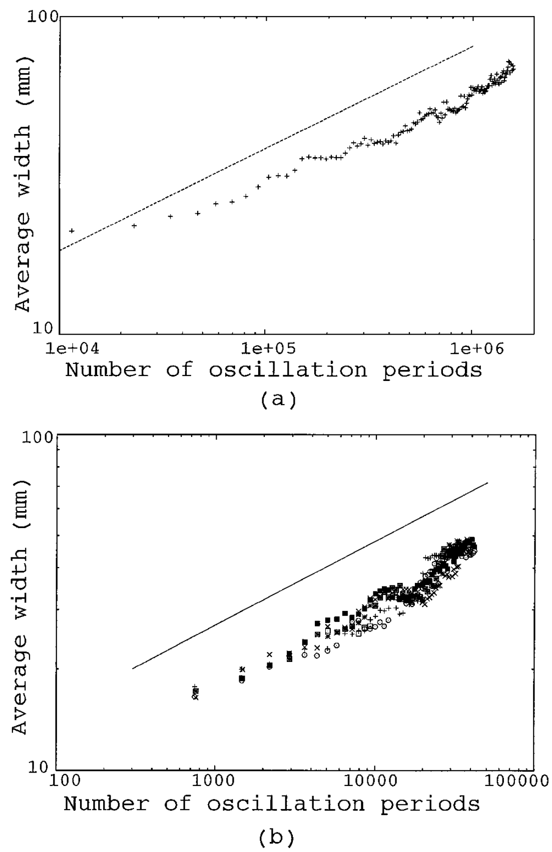

FIG. 4. The averaged width of the stripes plotted as a function of the number of periods of the oscillation. The spatial averages were taken over 100 slices in each case. Both sets of experiments were carried out with an amplitude of ±1 mm and a frequency of 12.8 Hz. (a) The average of three experimental runs with the copper ball/poppy seed mixture. (b) The data from seven experimental runs with the "100's and 1000's"/polystyrene bead mixture. The solid line in each figure has a slope of 0.25 and the kinks in the experimental data sets occur when large clusters merge and the whole process slows down.

proportional to its width then this will increase the diffusion coefficient accordingly. An alternative view has recently been provided by [13] who shows that if the drag in a Brownian diffusion model is made proportional to the width of the stripe then a $t^{0.25}$ relationship is found.

The agreement between the present observations and models containing scaling laws is interesting. In the stone striping model there are strong similarities between the processes involved since the forcing is essentially random and biased and the mobility of the stripes changes with time as they broaden. In fact, the results of recent simulations [14], where the direction of the bias has been changed to be in accord with the present experiments, also show clustering. Hence we believe that it is indicative of a critical phenomenon which has relevance to a wide range of problems where spontaneous segregation is found in binary mixtures.

The author is grateful to S. May for manufacturing the apparatus, S. Shipton for help with computations, and A. Armstrong, D. Binks, M. Cates, A. Juel, J. Jiminez, P. Mulheran, and N. Stocks for helpful discussions.

[1] B. Werner and B. Hallet, Nature (London) **361**, 142 (1993).
[2] H. M. Jaeger and S. R. Nagel, Science **255**, 1523 (1992).
[3] J. C. Williams, Powder Technol. **15**, 245 (1976).
[4] J. M. N. T. Gray and K. Hutter, Contin. Mech. Thermodyn. **9**, 341 (1997).
[5] H. A. Makse, S. Havlin, P. R. King, and H. E. Stanley, Nature (London) **386**, 379 (1997).
[6] W. Cooke, S. Warr, J. M. Huntley, and R. C. Ball, Phys. Rev. E **53**, 2812 (1996).
[7] V. Frette and J. Stavans, Phys. Rev. E **56**, 6981 (1997); K. Choo, T. C. A. Molteno, and S. W. Morris, Phys. Rev. Lett. **79**, 2975 (1997); K. Choo, M. W. Baker, T. C. A. Molteno, and S. W. Morris, Phys. Rev. E **58**, 6115 (1998); I. S. Aranson and L. S. Tsimiring, Phys. Rev. Lett. **82**, 4643 (1999).
[8] P. A. Mulheran, J. Phys. I (France) **4**, 1 (1994).
[9] H. M. Jaeger, S. R. Nagel, and R. P. Behringer, Rev. Mod. Phys. **68**, 1259 (1996).
[10] G. Strassburger, A. Betat, M. A. Scherer, and I. Rehberg, in *Traffic and Granular flow,* edited by D. E. Wolf, M. Shcreckenberg, and A. Bachem (World Scientific, Singapore, 1996).
[11] A. D. Dinsmore, A. G. Yodh, and D. J. Pine, Phys. Rev. E **52**, 4045 (1995).
[12] N. P. Louat, Acta Metall. **22**, 721 (1974).
[13] N. Stocks (private communication).
[14] P. A. Mulheran (private communication).