

## Oscillatory granular segregation in a long drum mixer

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PACS. 45.70.Mg – Granular flow: mixing, segregation and stratification.

PACS. 45.70.Qj – Pattern formation.

**Abstract.** – Heterogeneous granular mixtures tend to segregate when tumbled in a partially filled, horizontal rotating drum. The dynamical evolution of segregation can, under certain conditions, be oscillatory. Continuum, order parameter-style models of this process posit two coupled fields which oscillate out of phase with one another. Here we examine three candidate fields, the surface concentration, the local streaming angle and the projected concentration of the subsurface core. We find that all these quantities are in phase with one another, in contradiction to a recent order parameter model.

One of the fascinating idiosyncrasies of dry granular materials is their tendency to segregate by size under various flow conditions [1, 2]. Segregation is ubiquitous in natural and industrial processes, and can produce a startling degree of spontaneous order [3, 4]. Probably the best controlled example is segregation along the axis of a partially filled, horizontal “drum mixer” [5–14]. After many rotations, an initially mixed binary distribution sorts itself into almost periodic bands arranged along the axis of the cylinder. The process can display complex, oscillatory wave dynamics during the transient before segregation saturates [9, 10, 13]. Similar oscillatory segregation has been observed in dense suspensions [12] and in multidisperse dry mixtures [14]. Accounting for this rich behaviour has been the objective of cellular automata models [15], continuum theories [16–21], and direct molecular dynamics simulations [22], yet the process remains poorly understood. Here we report experiments in the oscillatory regime and present measurements of three axial quantities; the surface concentration, the surface slope shape and the projected concentration. The latter is sensitive to the subsurface concentration distribution. We find that all three quantities remain in phase during the course of an oscillation; a fact which is very difficult to reconcile with general features of continuum models of this process [20, 21].

The central difficulty in explaining any segregation process is to establish which are the relevant continuum quantities – the correct order parameters, in the language of condensed matter physics – and how they are dynamically related to one another. In the case of axial segregation, we expect that the radially averaged surface concentration, the most obvious measurable order parameter, must be coupled to other axial fields to produce the effects observed. In order for such a theory to be satisfactory, some physical understanding of the fields in terms of microscopic grain motions should be possible. There have been several attempts along these lines, but, as we show experimentally in this paper, even the most sophisticated

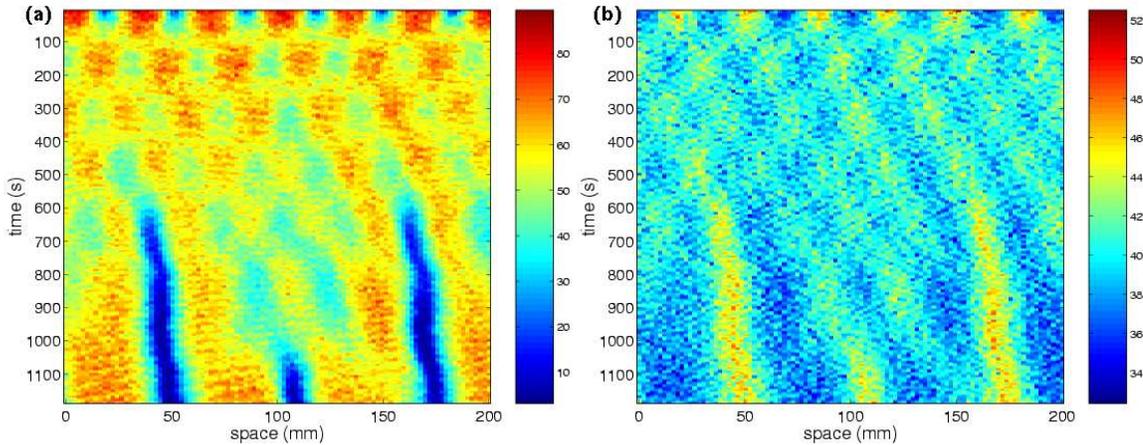


Fig. 1 – (a) Space-time plot of relative concentration for a mixture with a 30 mm presegregated wavelength initial condition. The space coordinate is shown on the horizontal axis, and time increases downward from the top. (b) The corresponding space-time plot of dynamic angle of repose. The intensity of each pixel represents concentration (dynamic angle) values at a particular position and time in the drum. The maximum amplitude of the standing wave in concentration corresponds to the minimum amplitude of the standing wave in the dynamic angle, and vice versa, thus the two fields are  $\pi$  out of phase throughout the oscillatory transient. It is also apparent that the saturated axial bands in both fields maintain this phase relationship.

of these [20,21] fail. Two important qualitative observations are that the segregation is both axial and radial, with the smaller component also differentially moving to form a core near the axis of rotation [6,23], and that the surface shape (or dynamic streaming angle) is concentration dependent [24]. Radial segregation has been extensively studied in thin cylinders and using simulation [2,25,26], but these results appear to be difficult to generalise to the slower axial segregation.

Early theoretical models regarded axial band formation as the result of a diffusion process where a negative diffusion coefficient arises from the coupling of the concentration to the dynamic angles of repose [16,17,19]. These models ignored the radially segregated core. Reverse diffusion models could not, however, account for the rich dynamics found during the transient that precedes complete segregation in some mixtures. In particular, an oscillatory traveling wave state exists [9,10,13] which apparently demands that the basic dynamics be at least second order in time, rather than merely diffusive. A comprehensive new model [20,21] reproduces both axial segregation as well as the oscillatory transient state. The crucial feature is that two essentially diffusive fields, the concentration and the dynamic angle of repose, are coupled such that they oscillate  $\pi/2$  out of phase during the initial transient. They are in phase for later times. Similar phase evolution is likely to be generic for models containing oscillatory behaviour, regardless of how the order parameters are defined. The order parameters have a conjugate relationship in the sense that they act like position and momentum in a mechanical oscillator. We show experimentally below that the surface concentration and dynamic angle do not have the required phase relationship, falsifying this model.

The dynamics of the radially segregated core is often held to be important in the axial segregation process [6,23]. Indeed, the axial bands can be seen to be due to a core instability leading to a periodic thickening of the core. When these thickened regions break the surface,

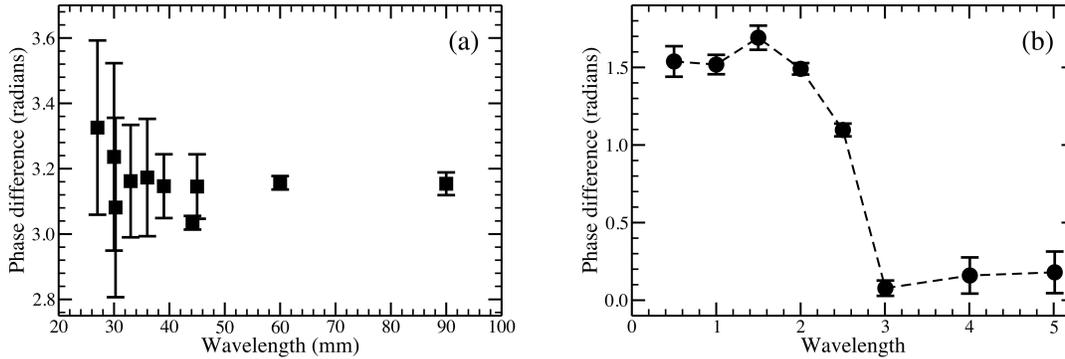


Fig. 2 – (a) The absolute value of the phase difference as a function of wavelength, including both presegregated initial conditions and random initial conditions. The data indicates that the two fields have a constant phase difference of  $\pi$ . (b) The absolute value of the phase difference as a function of wavelength for the theoretical simulation used in Refs. [20,21]. The wavelength is given in dimensionless units. Note the longer vertical scale: the theoretically predicted phase difference changes by  $\pi/2$ .

bands are observed. Thus, a clear candidate for the second order parameter is the thickness of the subsurface core. In this view, the oscillations result from a periodic exchange of material between the surface and the core. Again, in order to capture oscillations, we might expect order parameters based on the surface concentrations and core thickness to have a conjugate relationship. Using a projection technique to visualise the core, we show below that this is not the case. Our results place severe constraints on the mechanism of axial segregation and suggest new directions in which theories of this process might proceed.

The drum mixer consisted of a horizontal Pyrex tube, 750 mm long with an inner diameter of 27 mm, rotated about its long axis at a constant angular frequency of 4.8 rad/s. The grains had a sharp colour contrast to allow sensitive visualisation of the segregation. The larger grains were white table salt, which had a cubic shape and a size distribution peaked between 300 and 710  $\mu\text{m}$ . The smaller component was black “hobby” sand [27], which was irregularly shaped, and had a size distribution peaked between 75 and 212  $\mu\text{m}$ . The mixtures were composed of 2/3 white salt and 1/3 black sand and the filled volume fraction of the tube was 28%. Under these conditions, an oscillatory transient is known to occur before segregation saturates [9,10]. Qualitatively similar oscillatory behaviour has since been observed in certain mixtures of glass beads [13].

Since pouring or shaking the granular mixture causes segregation, filling the tube is a nontrivial task. We loaded the granular mixture in a long U-shaped channel, which was then inserted lengthwise into the drum and rotated to deposit its contents. This procedure ensures an even filling, with reasonably random initial conditions. To obtain reproducible, quantitative dynamical information, however, it is advantageous to use “presegregated” initial conditions, in which the tube is loaded with a known, periodic concentration distribution. Previous experiments [9,10] have established the similarity of the oscillatory dynamics for random and presegregated initial conditions. A presegregated pattern was made by placing thin spacers in the U-shaped channel forming volumes that were alternately filled with the sand and salt

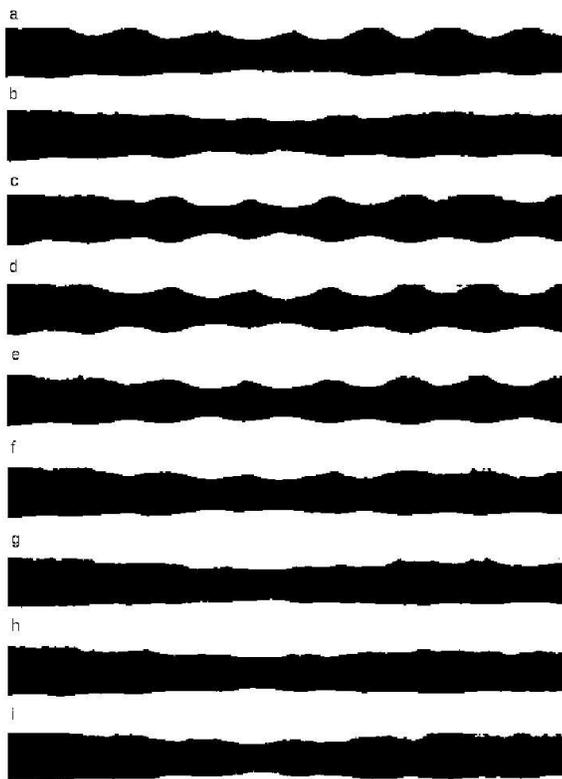


Fig. 3 – Axial projections of the radial core at: (a) 66 seconds, (b) 88 seconds, (c) 111 seconds, (d) 133 seconds, (e) 156 seconds, (f) 178 seconds, (g) 201 seconds, (h) 223 seconds, and (i) 246 seconds. The core was imaged perpendicular to the flowing granular surface. Here we can see that the transient results from an oscillatory radial core instability.

mixtures, keeping their relative proportions constant. Oscillatory transients are observed for wavelengths shorter than a certain cut-off, as discussed below.

The contents of the drum were illuminated by a thin laser sheet which struck the flowing granular surface perpendicular to the axis of the drum. The laser-illuminated surface was imaged by a high speed CCD camera. The average intensity of the illuminated part of the granular surface was used to quantitatively determine the local relative concentration of large (white salt) to small (black sand) grains. The intensity scale was calibrated using samples of known composition ranging from 0 % salt to 100 % salt. The camera viewed the granular surface obliquely. The images were time averaged and thresholded to eliminate noise, and a geometric projection was applied to determine the surface profile as though it were viewed from the end of the drum. Mixtures rich in salt have a smooth surface profile that is close to linear. However, mixtures rich in sand have a pronounced kink in their surface profile. To model the profiles, we fit them to polynomials. The surface slope below the kink was rather insensitive to the concentration. The surface slope above the kink was more sensitive to variations in concentration than below. We therefore calculated the dynamic angle of repose from the average arctangent of the slope of the profile above the kink.

These data yielded the dynamic angle of repose and the relative concentration at the axial position of the laser sheet. The laser and the camera were mounted on the same computer controlled translating stage. The axial time evolution of the concentration and dynamic angle fields were measured simultaneously by moving the translating stage rapidly along a 200 mm long section of the drum located near its centre. With one measurement made every 2 mm, each scan took approximately 5 seconds, the equivalent of about 4 drum rotations. Space-time images of the full spatio-temporal evolution of both fields were reconstructed by smoothly interpolating the time-stamped measurements onto equal times for each axial position. Presegregated initial conditions were used in order to study uniform regions of standing waves. Figure 1 shows the results for an initial wavelength of 30 mm. Decaying left and right travelling waves pass through each other to form a standing wave until they are absorbed into nearby axial bands. Figure 1a shows the relative concentration measurements and figure 1b shows the corresponding dynamic angles. Viewing each spatial position vertically shows the time evolution of the field. Travelling waves do not occur for wavelengths above a cut-off, which was previously reported to be  $54 \pm 1$  mm [9, 10]. We have studied the relationship between the concentration and dynamic angle in numerous runs using presegregated wavelengths both above and below the cut-off as well as with random initial conditions.

Examining the figures 1a and b, it is clear that the phase difference between the concentration and dynamic angle of repose fields must be close to a multiple of  $\pi$ . This is most apparent from the temporal nodes, which occur at nearly the same time for both fields. In order to determine the phase difference between the two fields, we analysed the two-dimensional Fourier spectra of their space-time evolution. Similar Fourier techniques were used previously to determine the dispersion relation for waves in this system [10]. The phase difference was taken to be the average of the absolute value of the angles located within the full-width half-maximum of the peaks in the power spectrum. This phase difference is shown in figure 2a, as a function of the presegregated wavelength. In all cases, the standing waves are  $\pi$  out of phase, within the uncertainties, including those above the cut-off wavelength. Thus, high concentration of the larger, white salt component is directly correlated with small dynamic angles of repose, while black sand concentration is correlated with large dynamic angles. This is the case both during the oscillatory transient and for the fully segregated bands at late times. The same conclusion is reached if the portion of the surface profile below the kink is included.

These results contradict the predictions of theory [20, 21]. In order to make a direct comparison, we ran the full nonlinear simulations described in [20, 21, 28] for presegregated initial conditions and analysed the results using the same procedure as for the actual data. Figure 2b shows the absolute value of the phase difference for the simulated runs; below the cut-off the phase difference is  $\pi/2$ , and the two fields are out of phase as one expects for conjugate order parameters. For initial wavelengths above the cut-off, zero phase difference is found. This clear contradiction with experiment demonstrates that the dynamic angle of repose is not the order parameter which is conjugate to the surface concentration. Since no other superficial feature appears likely, we must turn our attention to subsurface phenomena.

The radially segregated core, which is ignored in the theory, has long been suspected of being involved with the mechanism of axial segregation. Observations using MRI imaging [6, 8] suggest that axial bands begin as subsurface bumps on the core which only become visible when they break the surface. Thus, a natural candidate for the second order parameter would be the thickness of the core. In this view, the waves are caused by the periodic exchange of material between the surface and the core. We know that such a core is present in our sand and salt mixtures from excavating the segregated mixture after the experiment, and from observations through a window forming the end of the drum.

In order to quantitatively investigate the role of the radial core, we developed a bulk

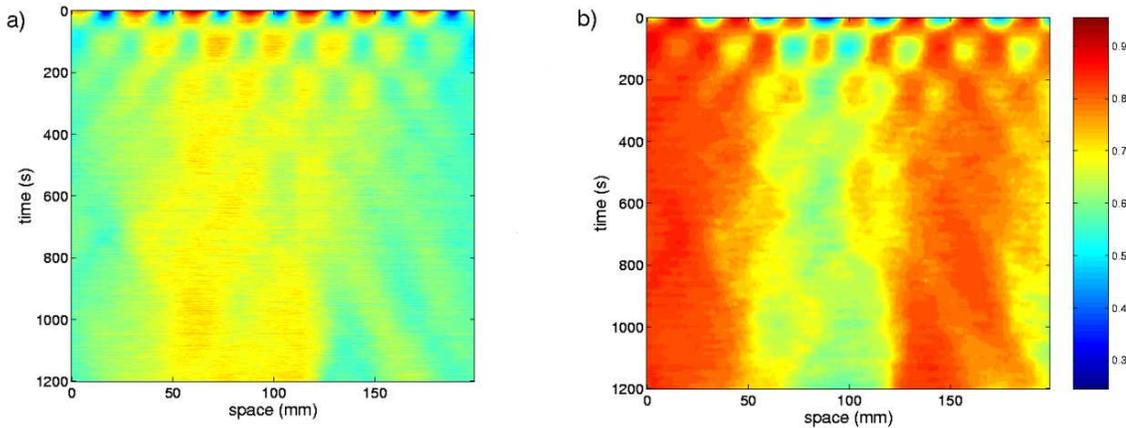


Fig. 4 – (a) Space-time plot of grayscale surface measurements for a mixture with a 30 mm pre-segregated wavelength initial condition. (b) The corresponding space-time plot of the fraction of radial core from axial projections of the radial core. As before, the space coordinate is shown on the horizontal axis, and time increases downward from the top. The maximum amplitude of the surface measurements corresponds to the minimum amplitude of the fraction of radial core, and vice versa, thus the two fields are  $\pi$  out of phase throughout the oscillatory transient.

visualisation technique. The salt grains are translucent and sand grains are opaque. We found that positioning a bright light source behind the rotating drum casts a shadow of the radial core on the front face of the mixture. The shadow is in effect a two dimensional projection of the core. No index-matching interstitial fluid is required [11]. By alternately illuminating the drum from above and behind, we used a stationary camera to collect images of both the surface concentration and the projected core as a function of time. The camera was positioned perpendicularly to the flowing surface.

Figures 3a – i show the actual core projections at different times through the transient. From these images it is apparent that the transient is a result of an oscillatory radial core instability, where the thickness of the core oscillates between the axial bands. Using edge detection, the radial width of the shadow of the core was measured and expressed as a fraction of the full width of the material in the drum. Figure 4 shows the space-time evolution of a mixture with a 30 mm pre-segregated initial wavelength. The oscillatory transient can be clearly seen in both the surface concentration and the core width. We can see from figure 4 that, as in the case of the dynamic angle of repose, the width of the radial core and the surface concentration are not  $\pi/2$  out of phase; they are oscillating in phase with one another. This is confirmed by detailed analysis of the space-time data. Thus, a simple order parameter based on the width of the core also fails to satisfy the requirements of an order parameter conjugate to the surface concentration.

We also performed a number of stopping experiments in which the tube was halted and then restarted after a variable delay time. The delay ranged from a few seconds to over 12 hours and in all cases was sufficiently long that all grain motion ceased. We observed that the decaying oscillation continued after any delay, even when the tube was stopped at a temporal node line. This strongly suggests that the memory of the pattern does not reside in subtle static electric charge effects, which would be expected to decay away with time, or in the momentum of the flow itself.

In conclusion, our results suggest that the surface concentration and surface shape, long thought to be important to the mechanism of the segregation, are in fact not independently dynamically active in the process. Rather, both they and the thickness of the radially segregated core are slaved together throughout the motion. This is consistent with previous observations in the non-oscillatory regime, using MRI techniques [8]. Stopping and restarting the tube after a time delay demonstrates that the segregation does not depend on the momentum of the grains, or on electric charging. Thus, the oscillatory behaviour of the segregation process emerges from the complete three-dimensional configuration of the local concentration alone. Although the streaming surface layer is the only dynamically active region, knowledge of its state alone is insufficient to explain the observed phenomena. The time evolution of the system depends, in addition, upon the state of the grains trapped in the bulk. The goal of our current investigations is to obtain a clear understanding of how this happens.

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We wish to thank Lorraine Courneyea, Alec Campbell, Simon-Philippe Breton, Ed Taylor, Troy Shinbrot and Lev Tsimring. This work was supported by the Natural Science and Engineering Research Council of Canada.

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