

Stripe Formation in Differentially Forced Binary Systems

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We consider pattern formation in periodically forced binary systems. In particular, we focus on systems in which the two species are differentially forced, one being accelerated with respect to the other. Using a continuum model consisting of two isothermal ideal gases which interact via a frictional force we demonstrate analytically that stripes form spontaneously above a critical forcing amplitude. The wavelength of the stripes is found to be close to the wavelength of sound in the limit of small viscosity. The results are confirmed numerically. We suggest that the same mechanism may contribute to the formation of stripes in experiments on horizontally oscillated granular mixtures.

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Binary systems subject to an oscillatory driving force are often found to phase segregate and form patterned structures [1,2]. Often this segregation can have important practical applications. For example, it may provide a useful way of separating two mixed species, or conversely, it may be undesirable in an industrial process [3]. The phenomenon is well documented experimentally but a full theoretical understanding is still missing with different mechanisms for the pattern formation being proposed in the literature.

Sanchez *et al.* [4] have carried out experiments and parallel simulations on granular mixtures immersed in water. For sufficiently large amplitudes of vibration a mixture of equal sized glass and bronze particles were found to separate into a striped pattern. Sanchez *et al.* proposed that the differential fluid drag on the two components of the mixture is the mechanism responsible for the segregation.

Mullin *et al.* [5–7] have performed a series of experiments in which they horizontally oscillate a quasi-two-dimensional layer of bronze spheres and poppy seeds. The species are again observed to segregate into a striped pattern. However, they propose a different explanation for the stripes: the effective excluded volume interactions which occur because of the different size of the shaken particles [8].

Therefore our aim in this Letter is to investigate possible mechanisms for binary phase segregation by providing an approximate analytic solution to a one-dimensional isothermal continuum model describing the physics of a binary mixture of two ideal fluids. We find that if the components are differentially forced stripes are formed in the concentration above a critical forcing amplitude. The wavelength of the stripes is controlled by the velocity of sound and standing wave oscillations in the total density play a crucial role in the stripe formation.

We then relate our results more closely to experiments on granular mixtures through a simple particle model. We find that differential forcing of the two types of particle is

enough to cause stripe formation even if the particles have the same mass and volume.

We consider two ideal fluids, labeled *A* and *B*, which are coupled by a frictional force proportional to the difference in their velocities. The physical origin of this force is from collisions between the *A* and *B* particles which tend to equalize the velocities. The *A* fluid is accelerated by a periodic force $a \cos(\omega t)$. The Navier-Stokes and continuity equations for this system may be written [9]

$$\rho^A D_t^A u^A = -\theta \partial_x \rho^A + \xi(u^B - u^A) + \partial_x(\nu \rho^A \partial_x u^A) + a \rho^A \cos(\omega t), \quad (1)$$

$$\rho^B D_t^B u^B = -\theta \partial_x \rho^B + \xi(u^A - u^B) + \partial_x(\nu \rho^B \partial_x u^B), \quad (2)$$

$$D_t^A \rho^A = -\rho^A \partial_x u^A, \quad (3)$$

$$D_t^B \rho^B = -\rho^B \partial_x u^B, \quad (4)$$

where $\theta = k_B T/m$ is the reduced temperature, ν is the kinematic viscosity, and ρ^A , ρ^B and u^A , u^B are the densities and velocities of the *A* and *B* fluids. $D_t^A = \partial_t + u^A \partial_x$ and $D_t^B = \partial_t + u^B \partial_x$ are the material derivatives in the *A* and *B* frames, and the friction coefficient can be written $\xi = \xi^0 \rho^A \rho^B / \rho$, where ξ^0 is constant and $\rho = \rho^A + \rho^B$ is the total density of the mixture.

Figure 1 shows the results of numerically solving Eqs. (1)–(4) using a lattice Boltzmann algorithm. Initially the densities of the two components have the same constant value ρ_0 plus a small random perturbation. Provided the amplitude of the forcing a is sufficiently high, strongly segregated *A* and *B* stripes form as shown in Fig. 1(a). These correspond to an approximately sinusoidal modulation of the concentration $\phi = \rho^A / \rho$. We define the average velocity of the fluid $u = (\rho^A u^A + \rho^B u^B) / \rho$ and the rest frame of the fluid to be that moving at velocity $\langle u \rangle$, where $\langle \dots \rangle$ denotes a spatial average. In this frame the total density performs standing wave oscillations at the forcing frequency but shifted in phase.

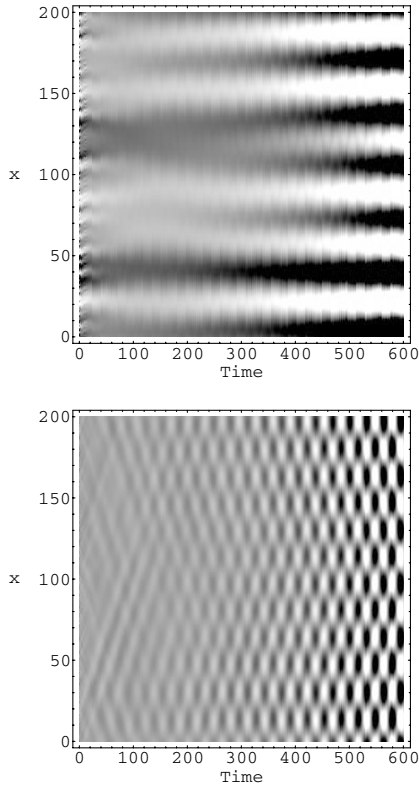


FIG. 1. Evolution in time of (a) the concentration and (b) the total density of a differentially forced binary fluid as a function of the distance x along a one-dimensional system, observed in the rest frame of the fluid. The concentration forms stripes and the total density performs standing wave oscillations, which increase in amplitude, at the frequency of the applied force. The parameters used were $\xi^0 = 6$, $\nu = 0.5$, $\theta = 1$, $a = 0.358$, and $\omega = 0.2$. The wavelength of the resulting stripes $\lambda = 33$ is close to the wavelength of sound $\sqrt{\theta}/\omega = 31$.

We assume that the coupling between the two components is sufficiently strong that it is the fastest relaxation process in the system. Therefore u^A and u^B are always very close in value; hence their time derivatives are essentially the same. This allows us to approximate $D_t^A u^A = D_t^B u^B = D_t u$, where $D_t = \partial_t + u \partial_x$. Adding Eqs. (1) and (2) to remove the coupling term then gives

$$D_t u = \frac{-\theta}{\rho} \partial_x \rho + a \phi \cos(\omega t) + \nu \partial_x^2 u. \quad (5)$$

Similarly adding Eqs. (3) and (4)

$$D_t \rho = -\rho \partial_x u. \quad (6)$$

Motivated by the numerical results we consider the trial solutions

$$\phi = \frac{1}{2} + \Delta \phi \sin(kx), \quad (7)$$

$$\rho = 2\rho_0 + \cos(kx)[\Delta \rho_1 \cos(\omega t) + \Delta \rho_2 \sin(\omega t)], \quad (8)$$

$$u = \frac{a \sin(\omega t)}{2\omega} + \sin(kx)[\Delta u_1 \cos(\omega t) + \Delta u_2 \sin(\omega t)], \quad (9)$$

where $k = 2\pi/\lambda$ is the wave number of the stripes.

Substitution of the trial solutions (7)–(9) into Eqs. (5) and (6), and comparing cosine and sine terms gives

$$\Delta \rho_1 = \left[\frac{2\rho_0 k a (\omega^2 - \theta k^2)}{(\omega^2 - \theta k^2)^2 + \nu^2 k^4 \omega^2} \right] \Delta \phi, \quad (10)$$

$$\Delta \rho_2 = \left[\frac{-2\rho_0 k^3 a \nu \omega}{(\omega^2 - \theta k^2)^2 + \nu^2 k^4 \omega^2} \right] \Delta \phi, \quad (11)$$

$$\Delta u_1 = \left[\frac{k^2 a \nu \omega^2}{(\omega^2 - \theta k^2)^2 + \nu^2 k^4 \omega^2} \right] \Delta \phi, \quad (12)$$

$$\Delta u_2 = \left[\frac{a \omega (\omega^2 - \theta k^2)}{(\omega^2 - \theta k^2)^2 + \nu^2 k^4 \omega^2} \right] \Delta \phi. \quad (13)$$

The crucial information needed is whether a wave with wave number k grows or decays, and the rate of this process. To understand this we must look at the behavior of ϕ as a function of time. The material derivative must now be treated exactly because the concentration changes much more slowly than densities and velocities. Using the continuity Eqs. (3) and (4) and $\rho^A \approx \rho^B \approx \rho_0$ gives

$$D_t \phi = \frac{1}{4} \partial_x (u^B - u^A) + \frac{u^B - u^A}{8\rho_0} \partial_x \rho. \quad (14)$$

Rearranging Eq. (2) and substituting the trial solutions

$$u^B - u^A = -\frac{a}{\xi^0} \cos(\omega t) - \frac{a\omega}{\xi^{02}} \sin(\omega t) + \frac{4\theta \Delta \phi k}{\xi^0} \times \cos(kx) + \frac{2\zeta}{\xi^0} \sin(kx), \quad (15)$$

where

$$\zeta = \left(\omega \Delta u_1 - \nu k^2 \Delta u_2 + \frac{\theta k}{2\rho_0} \Delta \rho_2 \right) \sin(\omega t) + \left(-\nu k^2 \Delta u_1 - \omega \Delta u_2 + \frac{\theta k}{2\rho_0} \Delta \rho_1 \right) \cos(\omega t). \quad (16)$$

Comparing the first order terms in $u_B - u_A$ in Eq. (15) and in u in Eq. (9) we see that the condition that A and B are strongly coupled is equivalent to stating $\xi^0 \gg \omega$.

Substituting (8) and (15) into (14) leads to

$$D_t \phi = \frac{1}{\xi^0} \left(-\theta \Delta \phi k^2 \sin(kx) + \frac{\zeta k}{2} \cos(kx) + \frac{ka}{8\rho_0} \times \cos(\omega t) \sin(kx) [\Delta \rho_1 \cos(\omega t) + \Delta \rho_2 \sin(\omega t)] \right).$$

The time scale over which ϕ changes is much longer than that taken for a single oscillation of the force. This allows us to average the right-hand side over time

$$D_I(\Delta\phi) = \frac{1}{\xi^0} \left(-\theta k^2 + \frac{ka\Delta\rho_1}{16\rho_0\Delta\phi} \right) \Delta\phi. \quad (17)$$

Substituting expression (10) for $\Delta\rho_1$ and integrating gives the exponential solution $\Delta\phi = e^{\Gamma t}$, where

$$\Gamma = \frac{1}{\xi^0} \left[\frac{k^2 a^2}{8} \left(\frac{\omega^2 - \theta k^2}{(\omega^2 - \theta k^2)^2 + \nu^2 k^4 \omega^2} \right) - \theta k^2 \right]. \quad (18)$$

A typical curve of Γ against k is shown in Fig. 2(a). The peak represents the fastest growing wave number. The numerical results, presented for comparison, were obtained by solving Eqs. (1)–(4) for the parameters $\Delta t = 0.001$, $\omega = 0.2$, $\xi^0 = 32$, $\nu = 2$, $\theta = 10/3$, $\rho_0 = 4$, $\Delta\phi = 0.001$, $a = 0.8$, and $k = 2\pi/L$ such that one stripe filled the entire system of length L . The initial condition was taken to be the trial solution.

Using (18) we may make two predictions. First, stripe formation occurs only when the amplitude of the force is above a certain critical value a_c such that Γ is positive for some value of k . To test this numerically we used a system with $L = 1500$, $\Delta t = 0.01$, $\xi^0 = 6$, and $\theta = 1$. Results are shown in Fig. 2(b). In each simulation we initially set the density to be $\rho_0 = 4$ plus a small random perturbation of amplitude 0.02. Stripe formation was defined to occur when $\langle (\phi - \phi_0)^2 \rangle > 0.01$ and the system was observed for up to 20 000 time steps. Exact agreement between the analytic and numerical results is not expected because the simulation size is finite and the value of k is limited to integer multiples of $2\pi/1500$. Moreover, the assumption that $\xi^0 \gg \omega$ breaks down for large ω .

Second, the wavelength of the stripes tends toward the wavelength of sound $\lambda_s = 2\pi c_s/\omega$, where $c_s = \sqrt{\theta}$ for an isothermal gas, in the low viscosity limit. This is clearly illustrated in Fig. 2(c), which shows how stripe wavelength changes as a function of the speed of sound for systems at a_c . The relation to c_s is expected because the evolution of the total density (8) is a standing wave (i.e., a sound wave) which slowly increases in amplitude as the striped concentration profile forms.

Thus we have shown that, within a continuum theory, differential forcing of the two components of a binary fluid can lead to stripe formation in the concentration profile.

We now take the first steps in investigating whether the same mechanism can hold in granular systems by solution of a two-dimensional, particle-based model which includes an excluded volume constraint. We consider two types of particles A and B which have equal diameter d , and equal mass m . The particles undergo free streaming and a periodic force is applied to the A particles.

$$\mathbf{r}_i^{A,B}(t + \Delta t) = \mathbf{r}_i^{A,B}(t) + \mathbf{v}_i^{A,B} \Delta t, \quad (19)$$

$$\mathbf{v}_i^A(t + \Delta t) = \mathbf{v}_i^A(t) + \mathbf{a} \cos(\omega t) \Delta t. \quad (20)$$

Each particle is then identified as belonging to one of a

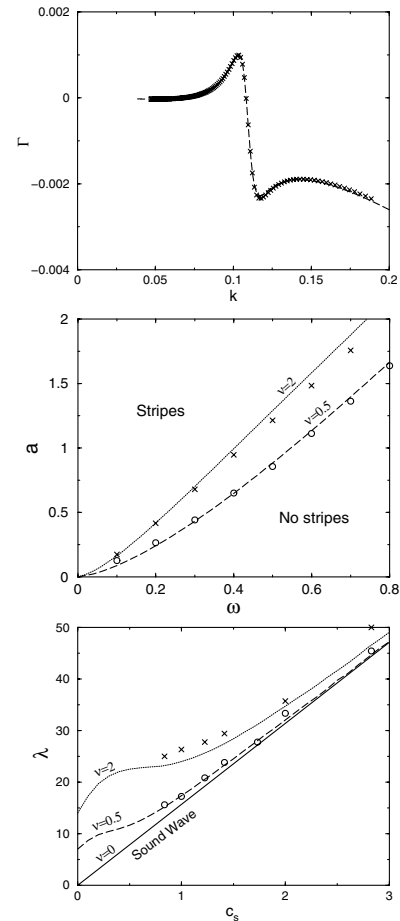


FIG. 2. (a) Growth rate of stripes Γ as a function of wave number k (crosses: simulations; dashed line: theory). (b) Critical forcing amplitude a_c , above which the system forms stripes, as a function of angular frequency ω for two different viscosities (crosses: simulations with $\nu = 2$; open circles: simulations with $\nu = 0.5$, dotted line: theory with $\nu = 2$; dashed line: theory with $\nu = 0.5$). (c) Stripe wavelength λ as a function of speed of sound c_s for an angular frequency of $\omega = 0.4$ and different values of viscosity ν (solid line: wavelength of sound).

grid of square cells with dimension $2d$. Each particle is checked to see if it overlaps with particles in its own cell or the three other closest cells. If two overlapping particles are traveling towards each other then they collide elastically, thus conserving energy and momentum. The smaller the time step Δt the more rigorously the excluded volume condition is enforced. The system is made isothermal by dividing it into slices of width $2d$ perpendicular to the forcing direction and rescaling velocities in the center of mass frame of each slice to accord with equipartition of energy.

The system forms stripes of A and B particles perpendicular to the forcing direction as in the continuum model. Moving to two dimensions and including fluctuations does not qualitatively alter the stripe formation

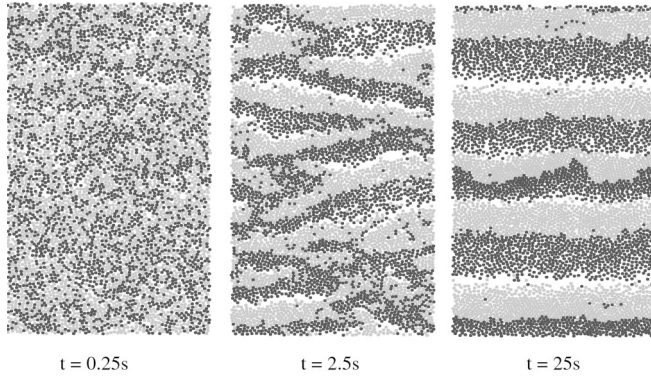


FIG. 3. Time evolution in seconds of a horizontally shaken granular system (this shows the view from above). The different friction coefficients of the particle species cause the stripe formation. The parameters of the simulations are given in the text.

mechanism. However, an interesting difference which we do not yet have an explanation for is that there is no threshold forcing amplitude for the stripe formation. Instead the time taken to form stripes diverges as the forcing tends to zero. The wavelengths of the stripes corresponds to the wavelength of sound to within $\sim 10\%$.

Finally, we discuss how differential forcing of the two components of a granular mixture may be responsible for the experimental findings of Mullin *et al.* [5]. Mullin *et al.* consider a flat tray which is oscillated in the horizontal plane at a frequency of 12 Hz. A mixture of poppy seeds and bronze spheres are placed on the tray. The particles are initially mixed and form an almost complete single particle layer. Forcing of individual particles arises principally from the frictional interaction with the base of the container. Mullin *et al.* observe that in time the two types of particles segregate into a striped pattern (very similar to that in Fig. 3, which shows the results of the computer simulations described below).

We modeled this system using the excluded volume elastic scattering algorithm with the update in the particle velocities given by

$$\mathbf{v}_i^{A,B}(t + \Delta t) = \mathbf{v}_i^{A,B}(t) + (\mathbf{a} \cos(\omega t) - \mu^{A,B} g \hat{\mathbf{v}}_i) \Delta t,$$

where \mathbf{a} is the amplitude of the acceleration applied to both species. The final term represents the frictional interaction with the container base: $g = 9.8 \text{ ms}^{-2}$ is the acceleration due to gravity, $\hat{\mathbf{v}}_i$ is the unit vector in the direction of the particle velocity, and $\mu^{A,B}$ are the coefficients of friction for poppy seeds and bronze spheres, respectively. No thermostat is used. Energy from the forcing is dissipated by friction. Because the friction coefficients of the two types of particles are different they are differentially forced, although this difference at large \mathbf{a} is like a square wave rather than the sinusoidal forcing used in Eq. (20).

We make the approximation that the static and dynamic friction coefficients are equal and ignore questions as to how much the spheres roll or slide. Experimental data for μ^A and μ^B are not available and we use the physically realistic values $\mu^A = 0.6$ and $\mu^B = 0.2$. We find that the qualitative behavior is not dependent on the exact values.

Figure 3 is the result of a simulation showing the evolution in time of a shaken binary system which is initially mixed. The dimensions of the tray are 90 by 180 mm and there are 1500 particles each with radius $r = 1.5 \text{ mm}$. The oscillation frequency was 4.8 Hz and the forcing amplitude $a = g$. Stripes form on a time scale very similar to that found in the experiments, which was $\sim 40 \text{ s}$ [5]. Therefore it seems reasonable that the differential acceleration of the two components can be identified as at least partially contributing to the stripe formation. However, we caution that these simulations are not isothermal, hence direct comparison with the isothermal model presented earlier is difficult. Furthermore the model assumes elastic collisions which may not be a good approximation for the real experiment.

To summarize, we have demonstrated analytically, using a continuum model, that if two isothermal ideal gases interacting via a frictional force are subject to differential forcing, stripes form in the concentration profile above a critical forcing amplitude. The wavelength of the stripes is approximately that of the wavelength of sound. We then used a particle-based numerical model to provide evidence that the same mechanism, differential forcing due to differential friction with the base of the container, is likely to be responsible for stripe formation in horizontally oscillated granular mixtures. Further work will aim to use more realistic models of granular systems to investigate the effect of inelastic collisions and nonuniform temperature distributions on the pattern formation.

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