Long-time limit of rotational segregation of granular media

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We performed simulations of a model for the segregation of a powder mixture in a horizontal half-filled tube under slow rotation. Fluctuations of the particles' flux play an essential role in our model. We found that an initially homogeneous mixture decomposes on a fast time scale into a sequence of domains of the pure components; this stage is followed by a long coarsening period, with decreasing number of domains. The coarsening rate increases strongly with the level of the fluctuations. After this stage, at very long times, the system approaches an equilibrium state, in which the number of bands fluctuates about its mean value. The obtained results are compared with recent experiments, and a good agreement is found.
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Mixtures of granular materials [1], unlike those of usual liquids, segregate easily under various experimental conditions, in response to macroscopic motion of the mixture [2]. This effect is important for technology [3] (in particular, for pharmaceutics), presenting an interesting physical problem at the same time. Because of the complexity of the granular media dynamics, we cannot write a general equation that would play the role of, say, the Navier-Stokes equations in hydrodynamics of Newtonian liquids. Nevertheless, such common phenomenon as segregation is expected to have a universal *phenomenological* description in different experimental situations [4].

One of the situations in which segregation is simple to observe is the rotation of a long horizontal glass tube, half-filled with a homogeneous mixture of two granular species (say sand and glass beads). This system has been studied experimentally in a number of works [5–7,9,8]. According to these experiments an initially homogeneous glass-sand mixture is decomposed, in about 100 revolutions (at low rate of rotation), into alternating bands, filled predominantly by particles of sand or glass [7]. Since at low revolution rates the bulk of the material behaves like a solid body, the dynamics of a thin layer of particles that flow down upon the inclined free surface of the material should play a dominant role in the segregation.

This system was considered theoretically in Ref. [7] for the first time. The segregation in the rotating tube was explained there as a diffusive instability, described by the diffusion equation with a negative effective diffusion constant

$$\frac{\partial c}{\partial t} = -D \frac{\partial^2 c}{\partial z^2}, \tag{1}$$

where $c(z)$ is the concentration of one of the components of the mixture along the tube's axis z. In this paper we continue the theoretical analysis of this system, and consider its long-time behavior. In order to obtain insight into the underlying physical mechanism, we first present the main physical assumptions leading to Eq. (1), similar to those of Ref. [7].

(1) The particles on the free surface undergo avalanches, whenever the surface is inclined with the angle of repose α_r. Let $\alpha_r^{(g)}$ and $\alpha_r^{(s)}$ be the angles of repose for the glass beads

and the sand particles, respectively (in the experiment of Ref. [7], $\alpha_r^{(g)} \approx 30°$ and $\alpha_r^{(s)} \approx 36°$). Then, for the glass-sand mixture, α_r should vary from $\alpha_r^{(g)}$ to $\alpha_r^{(s)}$, depending on the relative fractions of the ingredients [10].

(2) Consider two neighboring cross sections (sites) "1" and "2." Let the fraction of glass in one of these sites be greater than in the other, $c_g^{(1)} > c_g^{(2)}$, so its repose angle $\alpha_r^{(1)} < \alpha_r^{(2)}$. Naively speaking, the free surface is slightly inclined from site 2 to site 1. This gives rise to on-surface flux of particles from sites 2 to 1. More rigorous analysis of this point can be found in Ref. [7], where the necessary conditions for the validity of this naive argument are discussed.

(3) The *mobility* of the glass particles is greater than that of the sand (that is because they suffer a smaller friction), while the driving (gravity) force is the same. Therefore the portion of the glass particles in the on-surface flux is *higher* than it would be from their bulk concentration.

(4) In order to retain material conservation, this on-surface flux must be precisely compensated for by back flux in the bulk of the material. The sand and the glass particles in this back flux are represented according to their bulk concentrations. This is unlike their populations in the on-surface flux, as described in point (3).

Thus there is an uncompensated glass flux from site 2 to site 1. This leads to the further increase of the difference between $\alpha_r^{(1)}$ and $\alpha_r^{(2)}$, providing a positive feedback. Formalizing these ideas, one can obtain a diffusion equation with negative diffusion constant Eq. (1) for the variation of the fraction of one of the species, that governs the initial stage of the evolution. Experimentally [7], it takes about 3–5 min for the system to approach the segregated state, that consists of alternating domains with majority of glass or sand. This sequence of domains is rather steady on a time scale of hours.

The further evolution of this state was recently investigated experimentally by Frette and Stavans [9], who studied the dynamics of the system to extremely long times (one month). It was found that the segregated state evolves very slowly, such that the number of domains decreases. This *coarsening* process continues until only few domains remain. A bit more surprising is the fact that starting from the fully

segregated state (the sand on the left and the glass on the right), one observes the nucleation of new domains.

These long-time coarsening and nucleation processes have not yet been analyzed theoretically. While Eq. (1) explains well the initial stage of segregation, it says nothing about the consequent evolution of the segregated state. In this paper we derive and investigate a dynamical equation for the fraction of one of the species on the basis of the above described physical mechanism. Our approach is different from that of Ref. [7] only in that the fluctuations of flux play an essential role in our model. It turns out that these fluctuations constitute the driving mechanism of the late stage coarsening; if the fluctuations are neglected, the initially segregated system of domains becomes stable. The subject of interest of Ref. [7] was only the initial segregation, and the fluctuations were neglected there.

The nature of these fluctuation is the following. A particle running down on the surface, driven by the gravity force, scatters on the roughness of the surface and collides with the other particles in the avalanche. Consequently, it can diffuse in the transversal direction (along the tube's axis z). This would lead to the self-mixing of the powder (instead of segregation). On the other hand, a spontaneous fluctuation of the glass particles' concentration leads to correlated inclinations of the surface in the z direction, that enhance further this fluctuation (providing the mechanism for segregation), as described above. The competition between the noise-induced self-mixing with the segregation of the two components leads to the long term coarsening of the bands of each of the species.

We turn now to a derivation of the dynamic equation, based on the physical mechanism sketched above. Since most of our assumptions are the same as in Ref. [7], our equation is similar as well. Nevertheless, in order to be consistent, we present a brief derivation. Let n_g and n_s be the number of glass and sand particles at a given site ($n_g + n_s = n$); and let $c_{g,s}$ be the local fractions of the species, $c_{g,s} = n_{g,s}/n$. The total number of particles of both types that go from one site to another on the free surface under the impulse $f \delta t$ is given by

$$\delta n = \delta n_g^{(s)} + \delta n_s^{(s)}. \tag{2}$$

The on-surface motion of the particles of the both species is caused by the same force f, while they have different mobilities $\mu_g \neq \mu_s$. Therefore,

$$\delta n_g^{(s)} = \mu_g c_g f \delta t, \quad \delta n_s^{(s)} = \mu_s c_s f \delta t. \tag{3}$$

This ''on-surface'' flux is compensated for by the ''underground'' back flux of the same number $\delta n = \delta n_g^{(b)} + \delta n_s^{(b)}$ particles, where

$$\delta n_g^{(b)} = c_g \delta n, \quad \delta n_s^{(b)} = c_s \delta n. \tag{4}$$

Using Eqs. (3), (4), and (2), one can calculate the resulting change of the number of the glass particles at a certain site:

$$\delta n_g = \delta n_g^{(b)} - \delta n_g^{(s)} = -\mu_g c_g f \delta t + c_g \delta n$$

$$= [-\mu_g c_g + c_g(\mu_g c_g + \mu_s c_s)] f \delta t.$$

Since $c_s = 1 - c_g$, we obtain $\delta n_g = (\mu_s - \mu_g)(c_g - c_g^2)f$. The dynamics of the system is governed by the continuity equation

$$\frac{\delta c_g}{\delta t} + \frac{\partial J}{\partial z} = 0,$$

with the flux defined by

$$J = (\mu_s - \mu_g)(c_g - c_g^2)f.$$

Note that, by rescaling the time, one can eliminate the factor $(\mu_s - \mu_g)$ from the equation. The force f can be represented as a sum of its regular and random components, $f = F + \eta$. As we noted above, the regular part F is proportional to the gradient of α_r. Since $\alpha_r = \alpha(c)$, we can write, approximately,

$$F = k \frac{\partial c_g}{\partial z}.$$

Since the on-surface motion consists of separate avalanches, we assume that there are no time and space correlations in $\eta(t)$; the material conservation requires the following form of this component:

$$\eta(t) = q \frac{\partial \zeta}{\partial z},$$

where ζ is the normalized white noise; the noise constant q remains a free parameter of our model. Thus we arrive at the following dynamical equation for a deviation of the fraction from its ''unsegregated'' value $\phi = 2c_g - 1$:

$$\frac{\partial \phi}{\partial t} = -\frac{\partial J}{\partial z}, \quad J = k(1 - \phi^2)\frac{\partial \phi}{\partial z} + q(1 - \phi^2)\frac{\partial \zeta}{\partial z}. \tag{5}$$

A similar equation was obtained in Ref. [7]; Eq. (5) differs from that equation only by the noise term. We will see below that this term is essential for the long-time coarsening behavior. In order to obtain a feeling for Eq. (5), one can represent it in the form

$$\frac{\partial \phi}{\partial t} = k \frac{\partial^2}{\partial z^2}\left(\frac{\partial W}{\partial \phi}\right) + q \frac{\partial}{\partial z}\left(D(\phi)\frac{\partial \zeta}{\partial z}\right), \tag{6}$$

where $D(\phi) = (1 - \phi^2)$, and

$$W(\phi) = -\frac{\phi^2}{2} + \frac{\phi^4}{12}, \quad |\phi < 1|. \tag{7}$$

Note that local minima of the ''potential'' $W(\phi)$, $\phi = \pm \sqrt{3}$, are not physical, because they do not satisfy the condition $|\phi < 1|$. This condition is equivalent to infinitely sharp walls at $\phi = \pm 1$. In our simulations we take care of this effect by adding to the potential $W(\phi)$ the term $\frac{1}{27}\phi^{18}$; this term almost does not contribute for $|\phi < 1|$ but it grows very fast at $|\phi| > 1$, such that $\phi = \pm 1$ become the minima of W.

Let us discuss briefly the simplest properties of Eqs. (6) and (7). The point $\phi(z) = 0$ is an unstable solution of Eq. (6), being a local maximum of $W(\phi)$. Linearizing it about $\phi = 0$, we recover Eq. (1), that governs the initial segregation

of the mixture. This instability is saturated when $|\phi(z)| \approx 1$. The sign of $\phi(z)$ at a given point is determined by the sign of the small initial perturbation, so $\phi(z)$ can become quickly oscillating, flipping between 1 and -1 at each point. Of course, this is an artifact of our model, that operates with a *local* fraction in the pointlike cross sections, while the physical quantities (such as the repose angle) depend on the fraction *averaged* over some length d of the order of, say, the radius of the tube. Therefore, a physically correct model should replace $W(\phi)$ by $W(\bar\phi(z))$ and $D(\phi)$ by $D(\bar\phi(z))$, where

$$\bar\phi(z) = \frac{1}{d} \int_{z-d/2}^{z+d/2} \phi(z')dz' \approx \phi(z) + \frac{d^2}{3}\frac{\partial^2 \phi}{\partial z^2}. \quad (8)$$

The last term of Eq. (8) gives rise to the "energy cost" for domain boundaries in the potential $W = W(\bar\phi(z))$, leading to smooth profiles of $\phi(z)$. Note, however, that the effect of using $\bar\phi(z)$ instead of $\phi(z)$ is analogous to a cutoff at the distance $\sim d$; this effect is achieved automatically when we simulate Eq. (8) on a discrete lattice with a lattice constant of the order of d. Thus, in our *discrete* simulations of the system of equations (6) and (7), we did *not* make use of Eq. (8). However, we did keep $D = D(\bar\phi)$ in the noise term. This is because the discrete scheme gives $|\phi| \approx 1$ at *each* lattice site in the segregated state; hence $D(\phi) \approx 0$. Using $D = D(\bar\phi)$, we permit relatively large noise near the domain boundaries, while the noise almost does not affect the "bulk" of the domains, where $|\bar\phi| \approx 1$.

We simulated Eqs. (6) and (7) numerically on a lattice of 20 000 sites for different levels of noise q, keeping the other parameters fixed: the lattice constant $d = 0.1$ and $k = 0.1$. The random process ζ was represented by random numbers, uniformly distributed in $[-\frac{1}{2}, \frac{1}{2}]$. We used the simplest explicit scheme with periodic boundary conditions. The results of our simulations confirm the qualitative expectations presented above. The homogeneously mixed state is fully unstable, in accordance with the linear stability analysis. A fast process of initial segregation leads to a state where $|\phi| \approx 1$, while the sign of ϕ changes almost at each lattice site.

The consequent evolution depends on the level of noise. For $q = 0$, we did not observe any dynamics; for $q > 0$ the initially segregated state evolves slowly (the rate strongly increases with q), decreasing the number of bands. The time dependence (to about four decades of time) of the number of bands, N, vs $\log_{10}t$ is presented in Fig. 1 for two relatively high values of q. Saturation at long times is clearly seen. Saturation is due to the fluctuations which cause the appearance of new domains, as discussed above. We have plotted also the experimental results of Frette and Stavans [9] for comparison. Figure 2 presents the data for smaller values of the noise parameter, $q = 10$ and 12. For $q = 10.0$, $N(t)$ decreases without saturation until very late times, although with a smaller rate, whereas, for $q = 12$, $N(t)$ still approaches a constant at long times. Note that well before saturation both curves have the same slope. The same plot presents also the data of the experiments by Frette and Stavans [9], demonstrating good agreement.
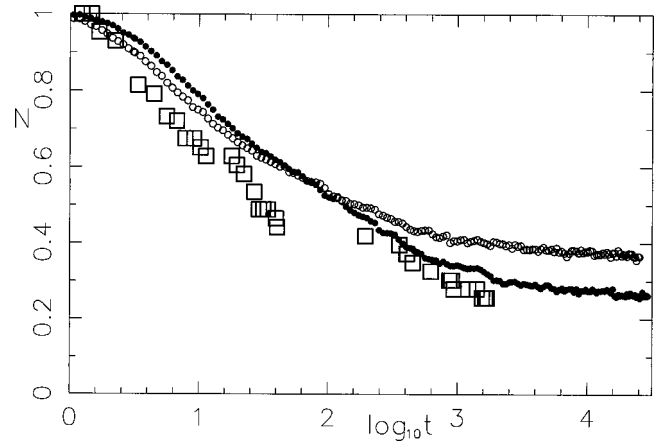

FIG. 1. Number of domains, N (in the units of its maximal value) vs $\log_{10}t$ (t in arbitrary units) obtained by our simulation for two values of the noise, $q = 15$ (full circles) and $q = 13$ (empty circles), together with the experimental data [9] (squares). The maximal number of domains at the simulation presented at this plot is $\sim 14\,000$; in the experiment, ~ 45.

In order to obtain a visual impression of the coarsening process, let us consider Fig. 3, which presents the space-time diagram of the evolution at an intermediate value of the noise, $q = 12.0$ (we used a lattice of 200 sites for this run). Black lines correspond to the domains with $\phi(z,t) > 0$, while the regions where $\phi(z,t) < 0$ are white. The analogous diagram obtained in the experiment (see Fig. 4) looks surprisingly similar to the result of our simulation. Thus our model provides a good description of the experiment. The next step should be a theoretical analysis of Eq. (6), and particularly the logarithmic decay of $N(t)$.

The behavior of the sand mixture in the tube resembles that of binary liquids quenched into a two-phase coexistence region, both by the fact of segregation and by the consequent coarsening process. In this context it is interesting to note that our equation resembles the Cahn-Hilliard equation, that describes the segregation of binary liquids. There is an important difference between the two systems, however. Coarsening of binary liquids is governed by energy reduction and
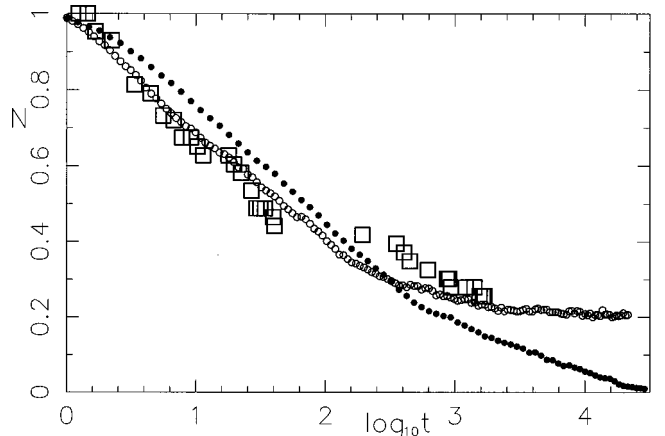

FIG. 2. The same as in Fig. 1 but for smaller values of the noise parameter: $q = 10$ (full circles) and $q = 12$ (empty circles); t in arbitary units. Good agreement with the experimental data [9] (squares) is seen.
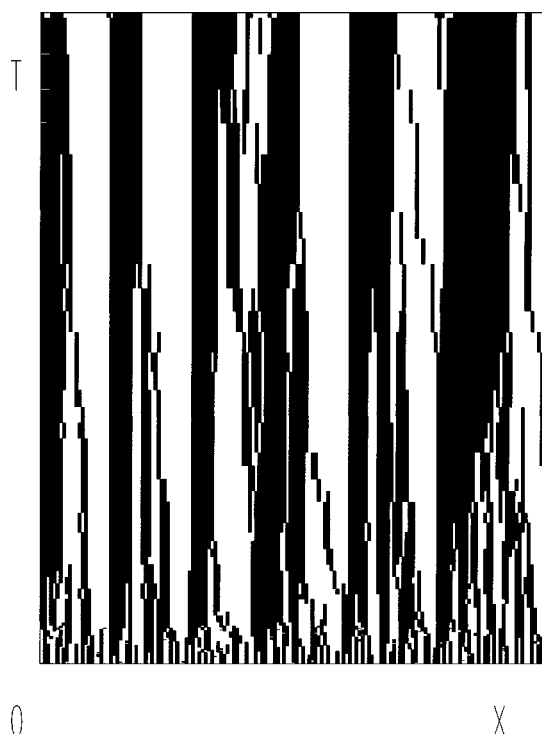
FIG. 3. The space-time diagram of a coarsening obtained by our simulation. The horizontal coordinate represents the tube's axis; the vertical coordinate is time (both in arbitrary units). The initial domain formation takes place at a very short-time scale [of the order of $1/k$; see Eq. (6)], and is not seen here.

is independent of the noise, while in the sand mixtures it is governed by the noise induced motion; if it were not for the noise, there would be no coarsening.

Since there is a nonzero probability of creation of a new domain, the system finally approaches an ''equilibrium'' state, where the number of domains fluctuates about its mean value. The accuracy of the experimental data does not allow one to determine unambiguously whether the saturation is indeed achieved in the experiment (it starts with 45 bands and finishes with only about 12 bands). However, as mentioned above, when the initial state is prepared of only one glass band and one sand band, the appearance of new bands
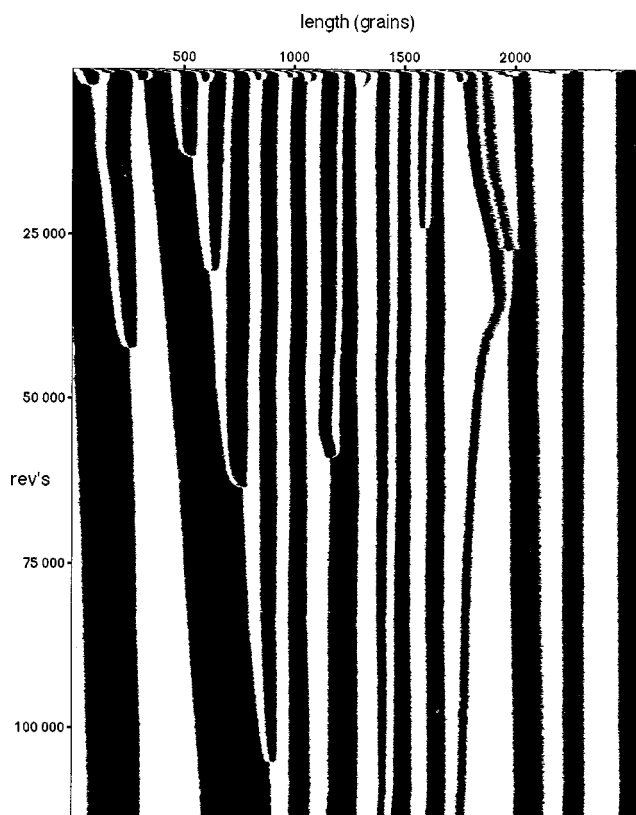

FIG. 4. The space-time diagram of a coarsening experiment by Frette and Stavans [9]. This figure is reproduced from Ref. [9] with the permission of the authors. It is to be compared with our result, presented in Fig. 3.

was found. This means that the equilibrium state could be achieved in an extremely long experiment, similarly to our simulations.

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