

Possible Stratification Mechanism in Granular Mixtures

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We propose a mechanism to explain what occurs when a mixture of grains of different sizes and different shapes (i.e., different repose angles) is poured into a quasi-two-dimensional cell. Specifically, we develop a model that displays spontaneous stratification of the large and small grains in alternating layers. We find that the key requirement for stratification is a difference in the repose angles of the two pure species, a prediction confirmed by experimental findings. We also identify a kink mechanism that appears to describe essential aspects of the dynamics of stratification. [S0031-9007(97)03031-7]

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Granular materials exhibit many unusual properties [1], such as size segregation, when exposed to external vibrations or rotations [2]. Size segregation is also observed when a mixture of grains of different size is poured onto a pile [3]; the large grains are found preferentially near the bottom of the pile, while the small grains are found near the top. Recently, it was found [4] that when a mixture of grains of different sizes and shapes is poured between two vertical slabs separated by ≈ 5 mm there appears a spontaneous stratification, with alternating layers of small and large grains parallel to the surface of the sandpile. Additionally, there is an overall tendency for the large and small grains to segregate spontaneously in different regions of the cell [3,4].

Very recently, Bouteux and de Gennes (BdG) [5] treated theoretically the case of granular flow made of two species. They based their work on a set of coupled convection equations to govern the flow of rolling grains and their interaction with the sandpile, introduced earlier by Bouchaud *et al.* in the case of a single-species sandpile [6]. BdG reproduced the phenomenon of segregation, but an understanding of stratification is lacking.

Here we seek to understand segregation and stratification in the conditions of [4], where the two species have different sizes and different shapes. We first introduce a discrete model to give a clear picture of the phenomenology and then develop a continuum approach. In agreement with the experimental findings [4], we find that segregation is related to the difference of size of the grains, and stratification to the difference in repose angles of the two pure species.

In the discrete model, the sandpile is built on a lattice, where the grains have the same horizontal size as the lattice spacing and two different heights, H_1 and $H_2 > H_1$. Each grain belongs to one of two phases: a *static phase* (if the grain is part of the solid sandpile) and a *rolling phase* (if the grain is not part of the sandpile but rolls downward with a constant drift velocity) [5–7]. The local slope $s_i \equiv h_i - h_{i+1}$ of the static grains is the variable controlling the dynamics of the rolling grains, where h_i is the height of the sandpile at column i .

At each time step, we deposit at the top of column 1 of the pile N_1 small grains plus N_2 large grains; these grains belong to the rolling phase. One rolling grain per column of each species interacts with the surface at each time step and can be converted from the rolling phase to the static phase. The remaining rolling grains are convected downward with unit drift velocity—i.e., they all move to the next column at each time step.

The dynamics of each rolling grain interacting with the sandpile surface is governed by its repose angle (the maximum angle below which a rolling grain is converted into a static grain) [6,8]. We note that the repose angle depends on the local composition on the surface, so we define $\theta_{\alpha\beta}$ as the repose angle of a rolling grain of type α on a surface with local grains of type β . We choose $\theta_{21} < \theta_{12}$ to take into account that large grains roll more easily on top of small grains than small grains roll on top of large grains [since the surface “looks” smoother for large grains rolling on top of small grains, see Fig. 1(a)]. The repose angles of pure species $\theta_{\alpha\alpha}$ lie between θ_{21} and θ_{12} .

The stratification experiments [4] use a mixture of grains of different shapes (smaller “less faceted” grains and larger “more faceted” grains). The repose angle of the smaller pure species is then smaller than the repose angle of the large pure species—i.e., $\theta_{11} < \theta_{22}$. To mimic the experimental conditions for stratification [4], we set $\theta_{21} < \theta_{11} < \theta_{22} < \theta_{12}$. We notice that the condition $\theta_{21} < \theta_{12}$ is a consequence of the different size of the species, while the condition $\theta_{11} \neq \theta_{22}$ is achieved, e.g., for mixtures of grains with different shapes.

At each time step, the rolling grain interacting with the sandpile surface at coordinate i will either stop—by being converted into a static grain—if the local slope of the surface $h_i - h_{i+1} < s_{\alpha\beta} \equiv \tan \theta_{\alpha\beta}$ or will continue to roll (together with the remaining rolling grains) to column $i + 1$ if $h_i - h_{i+1} \geq s_{\alpha\beta}$. We iterate this algorithm to form a large sandpile of typically 10^5 grains.

Figure 1(b) shows the resulting morphology. The stratification is qualitatively the same as that found experimentally [4], not only in regard to the *statics* of

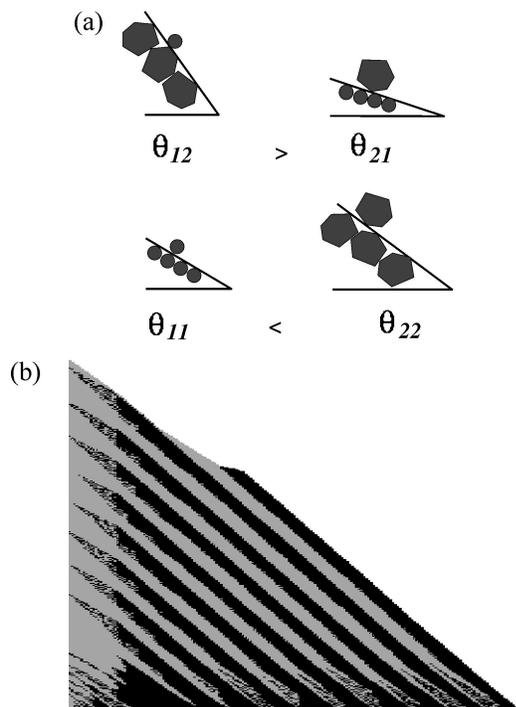


FIG. 1. (a) The slopes of repose $s_{\alpha\beta}$ depend on the composition of grains at the surface of the pile and are chosen according to the four possible interactions between small and large grains. The slopes of repose satisfy $s_{21} < s_{11} < s_{22} < s_{12}$ (see discussion in text). (b) The result obtained with the discrete model (for $H_1 = 1$, $H_2 = 2$, $s_{21} = 2$, $s_{11} = 6$, $s_{22} = 7$, $s_{12} = 10$, $N_1 = 20$, and $N_2 = 10$).

the sandpile [seen in Fig. 1(b)], but also in regard to the *dynamics*. After a pair of static layers is formed with a layer of large grains on top of a layer of small grains, the angle of the sandpile is close to θ_{22} . Since the surface of the sandpile is made of large grains and $\theta_{22} < \theta_{12}$, a thin layer of small grains is trapped on top of the layer of large grains. These small grains smooth the surface without changing significantly the sandpile angle and allow rolling small grains to go further down (since $\theta_{11} < \theta_{22}$). The large grains are rolling down on this thin layer of small grains without being captured ($\theta_{21} < \theta_{22}$) and are the first to reach the bottom of the sandpile, giving rise to segregation. When the flow reaches the base of the pile, the grains develop a profile which displays a “kink” where the grains are stopped: The small grains are stopped first since $\theta_{21} < \theta_{12}$, and a pair of layers begins to form, with the small grains underneath the large grains. The kink moves upward (in the opposite direction to the flow of grains) until it reaches the top of the pile and a complete pair of layers has been formed.

If, on the other hand, we consider $\theta_{22} < \theta_{11}$ in the model (corresponding to a mixture of smaller more faceted grains and larger less faceted grains) we find segregation but no stratification. Thus the control parameter for stratification appears to be the difference in the repose angle of the pure species.

As seen in Fig. 1(b), before the layers appear there is an initial regime where only segregation is found. At the onset of the instability leading to stratification, a few large grains are captured on top of the region of small grains near the center of the pile where the angle of the pile is $\theta \approx \theta_{11}$. The repose angle for large grains is now θ_{22} . Thus, if $\theta \approx \theta_{11} < \theta_{22}$, more large grains can be trapped (since the angle of the surface is smaller than the repose angle), leading to the first sublayer of large grains and then to stratification. On the other hand, if $\theta \approx \theta_{11} > \theta_{22}$, no more large grains are captured, the fluctuation disappears, and the segregation profile remains stable. Thus this picture suggests that, in agreement with [4], the segregation profile observed in the initial regime is “stable,” so long as $\theta_{22} < \theta_{11}$, and unstable (evolving to stratification for large enough systems) when $\theta_{11} < \theta_{22}$.

To offer further insight into the physical mechanism of stratification, we now develop a continuum approach in the spirit of Refs. [5–7]. The variables are the two thicknesses of rolling grains $R_\alpha(x, t)$, with $\alpha = 1, 2$, respectively, for small and large grains, the height of the sandpile $h(x, t)$, and the volume fraction of grains of β type in the static phase $\phi_\beta(x, t)$. The equations of motion are [5,9]

$$\frac{\partial R_\alpha}{\partial t} = -v_\alpha \frac{\partial R_\alpha}{\partial x} + \Gamma_\alpha, \quad (1a)$$

$$\frac{\partial h}{\partial t} = -\sum_\alpha \Gamma_\alpha. \quad (1b)$$

Here v_α is the downhill convection velocity of species α , and Γ_α describes the interaction of the rolling grains with the surface—i.e., how rolling grains are stopped and become part of the sandpile (capture), and how grains of the sandpile can enter the rolling phase (amplification). The concentrations are given by $\phi_1 + \phi_2 = 1$ and $\phi_\alpha = -\Gamma_\alpha / (\partial h / \partial t)$.

As in the discrete model, we focus on the dependence of the repose angle on the composition of the surface $\phi_\beta(x, t)$. The repose angle θ_α of each type of rolling grain is now a continuous function of the composition of the surface $\theta_\alpha = \theta_\alpha(\phi_\beta)$ [see Fig. 2(a)]. The repose angle $\theta_{\alpha\beta}$ defined for the discrete model is now $\theta_\alpha(\phi_\beta)$ with $\phi_\beta = 1$. We propose that $\Gamma_\alpha = \Gamma_\alpha(R_\alpha, \phi_\beta, \theta_{\text{loc}})$ obeys the relation

$$\Gamma_\alpha \equiv \begin{cases} \gamma_\alpha [\theta_{\text{loc}} - \theta_\alpha(\phi_\beta)] R_\alpha & \text{if } \theta_{\text{loc}} < \theta_\alpha(\phi_\beta) \\ \gamma_\alpha \phi_\alpha [\theta_{\text{loc}} - \theta_\alpha(\phi_\beta)] R_\alpha & \text{if } \theta_{\text{loc}} > \theta_\alpha(\phi_\beta) \end{cases}, \quad (2)$$

where $\theta_{\text{loc}}(x, t) \equiv -\partial h / \partial x$ is the local surface angle. The parameter γ_α represents the effectiveness of the interaction: $v_\alpha / \gamma_\alpha \sim d_\alpha$ (where d_α is the linear size of the grain) is the length scale on which a rolling grain will interact significantly with the surface when θ_{loc} is slightly different from the repose angle [7].

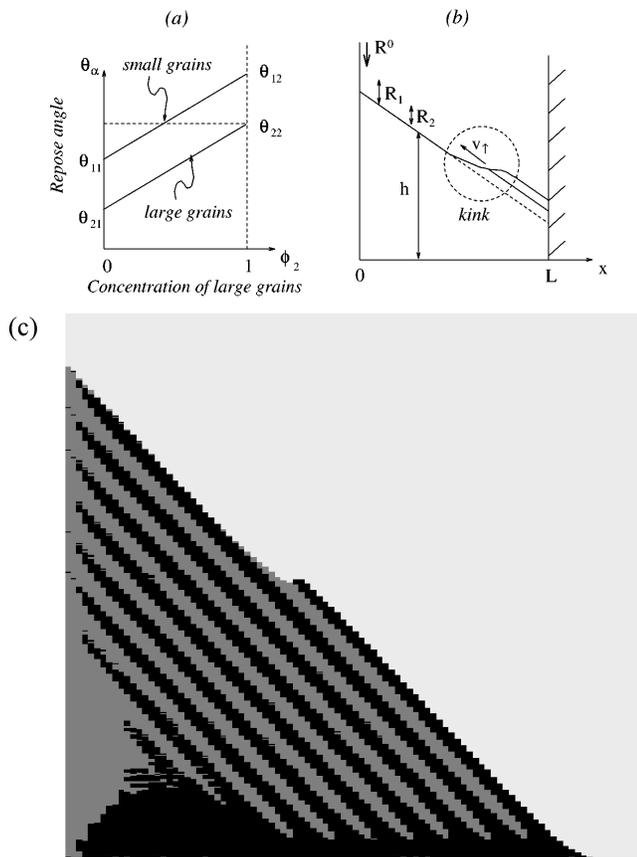


FIG. 2. (a) Dependence of the repose angle for the two types of rolling grains on the concentration of the surface of large grains ϕ_2 . An essential ingredient to obtain stratification is that $\theta_{22} > \theta_{11}$. For the numerical integration, we use the linear interpolation between $\phi_2 = 0$ and $\phi_2 = 1$ as plotted here. (b) Picture with the different quantities appearing in the text. The dash-circled zone is the kink. (c) Resulting morphology of the numerical integration of the continuum equations. The parameters used are $\tan \theta_{11} = 1$, $\tan \theta_{22} = 1.1$, $\tan \theta_{12} = 1.4$, $\tan \theta_{21} = 0.7$, $\gamma_1 = \gamma_2 = 0.8$, and $v_1 = v_2 = 1$.

The interaction term Γ_{α} includes two types of processes [5]. (a) *Capture*: as in the discrete model, rolling grains are captured if $\theta_{\text{loc}} < \theta_{\alpha}(\phi_{\beta})$. The capture is proportional to the number of grains interacting with the surface, and, therefore, is proportional to R_{α} . (b) *Amplification*: if $\theta_{\text{loc}} > \theta_{\alpha}(\phi_{\beta})$, then some static grains of the sandpile are converted into rolling grains. This conversion of α type grains is proportional to their surface concentration ϕ_{α} , and to the number R_{α} of rolling grains acting in amplification.

We next solve Eqs. (1) and (2) numerically. The results, shown in Fig. 2(c), are qualitatively similar to the discrete model. We find stratification whenever the repose angle has the qualitative behavior shown in Fig. 2(a), the key requirement being $\theta_{22} > \theta_{11}$. We also find a kink, corresponding to the growth of the new pair of static layers, with a well-defined steady-state profile and upward velocity [10].

To find the conditions under which stratification occurs, we first calculate the steady-state solution of the full set of Eqs. (1) and (2) and then study its stability under perturbations. To describe the experimental situation, we consider a 2D cell with vertical walls at $x = 0$ and $x = L$ [5]. We assume that the difference $\psi \equiv \theta_1(\phi_2) - \theta_2(\phi_2)$ is independent of the concentration ϕ_2 , and we set $v_1 = v_2 \equiv v$ and $\gamma_1 = \gamma_2 \equiv \gamma$ [Fig. 2(b)]. We seek a steady-state solution, where the profiles of the sandpile and of the rolling grains are conserved in time. Thus stratification cannot be observed for this solution. The conservation of the grains gives $\partial \bar{h} / \partial t = v R^0 / L$, and we impose $\partial \bar{R}_{\alpha} / \partial t = 0$, with boundary conditions $\bar{R}_{\alpha}(0) = R_{\alpha}^0 = R^0 / 2$ and $\bar{R}_{\alpha}(L) = 0$.

The steady-state solution of (1) and (2) shows almost total segregation. At the *upper* part of the pile, for $0 < x < x_m$, with $x_m \equiv L/2 - v/(\gamma\psi)$, only small grains are present [$\bar{\phi}_1(x) = 1$, and $\bar{\phi}_2(x) = 0$], and the profiles are

$$\bar{R}_1(x) = R^0 \left(\frac{1}{2} - \frac{x}{L} \right), \quad \bar{R}_2(x) = \frac{R^0}{2}, \quad (3a)$$

$$\bar{\theta}_{\text{loc}}(x) - \theta_{11} = \frac{-v/\gamma}{L/2 - x}. \quad (3b)$$

At the *lower* part of the pile ($x_m < x < L$), we find that, after a small region of size of the order of $v/(\gamma\psi)$, mainly large grains are present, and the profiles are

$$\bar{\phi}_1(x) = \exp \left[-\frac{\gamma\psi}{v}(x - x_m) \right], \quad (4a)$$

$$\bar{R}_1(x) = \frac{2v}{\gamma\psi L} \bar{\phi}_1(x) \bar{R}(x), \quad (4b)$$

$$\bar{\theta}_{\text{loc}}(x) - \theta_{22} = -m \bar{\phi}_1(x) - \frac{v}{\gamma(L - x)}. \quad (4c)$$

Here $\bar{R} \equiv \bar{R}_1 + \bar{R}_2 = R^0(1 - x/L)$ and $m \equiv \theta_{22} - \theta_{21} = \theta_{12} - \theta_{11}$.

To analyze the stability of the steady-state solution (3) and (4) for the different phenomenologic parameters, we impose the steady-state solution as the initial condition and then we look numerically for the stability of the profile under perturbations. For $\theta_{11} > \theta_{22}$, the steady-state solution is *stable*: in this case, only segregation is observed and the sandpile conserves in time the profiles (3) and (4). For $\theta_{11} < \theta_{22}$, the steady-state solution is *unstable* (evolving to stratification), just as in [4].

To gain insight about the kink mechanism, we look for a possible steady-state solution for the shape of the kink assuming that (i) far below and above the kink the sandpile has a constant angle θ_0 ; (ii) the lowest part of the kink is made only of small grains, so that large rolling grains are not captured, and the top part is made only of large grains.

To suppose the existence of a stationary solution for the kink implies that $R_1(x, t)$ and $f(x, t) \equiv h(x, t) + \theta_0 x$ are functions only of $u \equiv x + v_{\uparrow} t$, where v_{\uparrow} is the uphill

speed of the kink. For the lowest part of the kink, as only small grains are captured [$\phi_1(u) = 1$, $R_2(u) = R^0/2$], Eqs. (1) reduce to equations for $R_1(u)$ and $f(u)$. We obtain the shape of the low part of the kink: for $u \leq 0$, $f(u) = 0$ and for $u > 0$, $f(u)$ obeys

$$-\frac{1}{w} \log\left(1 - \frac{2wf}{R^0}\right) = \frac{\gamma}{v_\uparrow}(f - \delta_1 u), \quad (5)$$

where $\delta_1 \equiv \theta_0 - \theta_{11}$ and $w \equiv v_\uparrow/(v + v_\uparrow)$. Then the lower layer of the kink is characterized by a linear dependence $f(u) \propto u\delta_1$ for $u \ll R^0/(2w\delta_1)$, plus logarithmic corrections near the boundary with the upper layer of large grains. This solution is no longer valid when the angle of the surface reaches θ_{21} and the large grains start to be captured. We note that this stationary solution exists only when $\delta_1 > 0$.

The solution of the equations for the highest part of the kink where only large grains are present can be obtained in the same way and is

$$f(u) = \left(\frac{R^0}{w}\right) \left[1 - \exp\left(-\frac{w\gamma\delta_2 u}{v_\uparrow}\right)\right], \quad (6)$$

where $\delta_2 \equiv \theta_0 - \theta_{22}$. We then find that the shape of the upper part of the kink is exponential and exists only for $\delta_2 < 0$.

Thus we see that the existence of the stationary solution for the kink implies that $\theta_{11} < \theta_0 < \theta_{22}$: the sandpile is built on an angle intermediate between the two repose angles of the pure species, and the repose angle of the small grains must be smaller than the repose angle of the large grains—in agreement with experiments [4] and the stability analysis performed above.

The layer thickness λ is R^0/w [see Eq. (6)], which is a consequence of the conservation law stating that all the rolling grains are stopped at the kink [4]. Furthermore, Eq. (5) implies $\gamma wf/v_\uparrow \sim 1$. For $f \sim R^0/w$, this gives $v_\uparrow \propto \gamma R^0$, so that we obtain

$$\lambda \propto cv/\gamma + R^0, \quad (7)$$

where c is a numerical constant that does not depend on v , γ , or R^0 . This relation, which we verify numerically, is relevant since v/γ and R^0 are both of the order of the diameter of the grains.

The typical size of the initial regime of segregation, L_x , observed prior to stratification when $\delta \equiv \theta_{22} - \theta_{11} > 0$ [Fig. 2(c)] can be calculated as follows. The condition for the appearance of a first layer of large grains on top of the region of small grains near the center of the pile is that capture of large grains must be larger than capture of small grains, i.e., $|\Gamma_2| > |\Gamma_1|$, where $|\Gamma_1| = \gamma m R_1$ and $|\Gamma_2| = \gamma \delta R_2$. Assuming that the solution (3) is valid for the initial segregation regime, we can evaluate R_1 and R_2 at $x = x_m$. We obtain

$$L_x \approx \frac{v}{\gamma\psi} \frac{m}{\delta} \frac{R^0}{R_2^0} \quad (8)$$

and verify (8) numerically.

In conclusion, we develop a mechanism to explain the observed stratification [4]. This mechanism is related to the dependence of the local repose angle on the local surface composition. We find that stratification occurs only when the repose angle of the large grains is larger than the repose angle of the small grains ($\theta_{22} > \theta_{11}$, corresponding to large grains rougher than small grains). The model describes the static picture of the sandpile of [4] with alternating layers made of small and large grains and also reproduces the dynamics, where the layers are built through a kink mechanism. When $\theta_{22} < \theta_{11}$, the model predicts almost complete segregation, but not stratification. These results are in agreement with experiment [4].

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- [1] H. M. Jaeger and S. R. Nagel, *Science* **255**, 1523 (1992); H. J. Herrmann, in *Disorder and Granular Media*, edited by D. Bideaux and A. Hansen (North-Holland, Amsterdam, 1993).
- [2] J. C. Williams, *Powder Technol.* **15**, 245 (1976); J. A. C. Gallas, H. J. Herrmann, and S. Sokolowski, *Phys. Rev. Lett.* **69**, 1371 (1992); J. B. Knight *et al.*, *ibid.* **70**, 3728 (1993); O. Zik *et al.*, *ibid.* **73**, 644 (1994).
- [3] R. L. Brown, *J. Inst. Fuel* **13**, 15 (1939); R. A. Bagnold, *The Physics of Blown Sand and Desert Dunes* (Chapman and Hall, London 1941).
- [4] H. A. Makse, S. Havlin, P. R. King, and H. E. Stanley, *Nature (London)* **386**, 379 (1997); J. Fineberg, *ibid.* **386**, 323 (1997).
- [5] T. Boutreux and P.-G. de Gennes, *J. Phys. I (France)* **6**, 1295 (1996).
- [6] J.-P. Bouchaud, M. E. Cates, J. R. Prakash, and S. F. Edwards, *Phys. Rev. Lett.* **74**, 1982 (1995); *J. Phys. I (France)* **4**, 1383 (1994).
- [7] P.-G. de Gennes, *C. R. Acad. Sci. (Paris)* **321**, 501 (1995).
- [8] R. A. Bagnold, *Proc. Roy. Soc. London A* **295**, 219 (1966).
- [9] BdG [5] proposed Eqs. (1). They did not consider the dependence of the repose angle on the surface composition and used a different interaction term Γ_α , taking into account another type of amplification process (where static grains of one type are amplified by rolling grains of the other type). We postpone treating this amplification for a subsequent work.
- [10] It is interesting to note that the dependence of the repose angle on the composition of the surface is close to the concept of interparticle percolation, according to which the large grains in the rolling phase are convected down on the top of the small grains. Since, in our model, the repose angle of the small grains is locally always larger than the repose angle of the large grains, the small grains are also trapped before the large ones.