Mechanisms of granular spontaneous stratification and segregation in two-dimensional silos

Pierre Cizeau, Hernán A. Makse,* and H. Eugene Stanley

Center for Polymer Studies and Department of Physics, Boston University, Boston, Massachusetts 02215
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Spontaneous stratification of granular mixtures has been reported by Makse *et al.* [Nature (London) **386**, 379 (1997)] when a mixture of grains differing in size and shape is poured in a quasi-two-dimensional heap. We study this phenomenon using two different approaches. First, we introduce a cellular automaton model that illustrates clearly the physical mechanism; the model displays stratification whenever the large grains are rougher than the small grains, in agreement with the experiments. Moreover, the dynamics are close to those of the experiments, where the layers are built through a ''kink'' at which the rolling grains are stopped. Second, we develop a continuum approach, based on a recently introduced set of coupled equations for surface flows of granular mixtures that allows us to make quantitative predictions for relevant quantities. This approach includes *amplification* (i.e., static grains entering the flow of rolling grains), a phenomenon neglected in the cellular automaton model. We study the continuum model in two limit regimes: the large flux or thick flow regime, where the percolation effect (i.e., segregation of the rolling grains in the flow) is important, and the small flux or thin flow regime, where all the rolling grains are in contact with the surface of the sandpile. (1) In the thick-flow regime, where most experiments are carried out, the flowing grains are segregated in the rolling phase; as they are flowing down, the large rolling grains are convected to the top of the rolling layer, and only the small rolling grains interact with the sandpile. We include this effect in the continuum model and find results very close to the experiments. (2) In the thin-flow regime, we find interesting results that are close to the thick-flow limit. However, due to the presence of cross-amplification processes, we find a small regime that shows stratification when the small grains are slightly rougher than the large grains, but stratification is much more pronounced if the large grains are rougher. We study in detail the dynamical process for stratification, where the layers are built through a ''kink'' mechanism, and find the dependence of the size of the layers on the parameters of the system. We find that the wavelength of the layers behaves linearly with the flux of grains. We also find a crossover behavior of the wavelength at the transition from the thin-flow to the thick-flow regime. We obtain analytical predictions for the shape of the kink giving rise to stratification as well as the profile of the rolling and static species when segregation of the species is observed. [S1063-651X(99)07104-4]

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I. INTRODUCTION

Segregation is a prominent example of the peculiar properties of granular matter [1–9]. For example, shaking a container filled with two types of grains of different sizes leads not to mixing—as in liquids—but to segregation, with the large grains at the top of the container and the small grains at the bottom. This striking behavior and the importance of mixing problems from a technological point of view have led to a broad interest in granular materials in the physics community.

Several types of segregation have been investigated, namely, segregation by vibrating a container (the ''Brazil nut effect'') [10–14], and segregation in rotating cylinders, where the segregation occurs through surface flow [15–19].

Segregation can also be obtained in the absence of any periodic oscillation by simply pouring a mixture of grains of different sizes onto a pile. One experimental setup, which has attracted much recent attention, consists of a quasi-two-dimensional cell or vertical Hele-Shaw cell where a mixture of grains is constantly poured next to one end of the cell

[Fig. 1(a)]. When a mixture of small and large grains is poured into the cell, a pile builds up and the small grains are observed to segregate near the top of the pile and the large ones near the bottom of the pile [20–26]. This segregation is due to the different grain sizes, because large grains roll down more easily on top of small grains than small grains on top of large grains.

It has been recently observed [27–31] that if the shapes of the species are sufficiently different, another type of segregation can take place: when such a mixture of small rounded grains and large rough grains is poured between two vertical slabs, a spontaneous stratification of the mixture in alternating layers of small and large grains parallel to the pile surface is observed. Additionally, there is an overall tendency for the large and small grains to segregate into different regions of the cell. Spontaneous stratification is also found for more than two species [27]. The layers are then ordered forming a sequence parallel to the surface pile (from bottom to top, small-medium-large, small-medium-large for three species).

This phenomenon could possibly be relevant in different fields, such as geomorphology [32,33]. For example, stones coming from sand dune solidification (sandstone) present successive alternation of layers of different types of grains. This regularity cannot be explained by periodic sedimentation. But one could imagine that the sand dune was built with sand brought by the wind and flowing down the slipface of

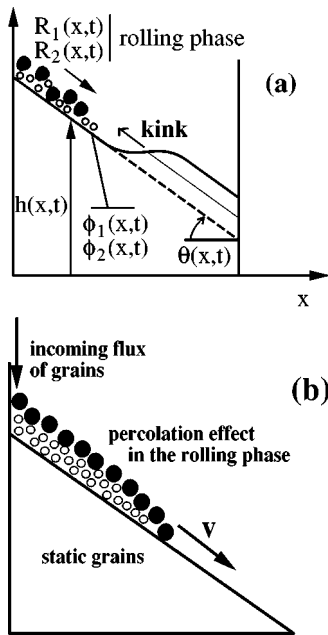*Present address: Schlumberger-Doll Research, Old Quarry Road, Ridgefield, CT 06877.

FIG. 1. (a) Typical experimental setup to study spontaneous stratification and segregation in quasi-two-dimensional cells, along with the different quantities defined in the text. (b) Schematic representation of the percolation effect in the rolling phase. We pour continuously a mixture of large and small grains in a two-dimensional cell and observe a steady thick flow of grains. The percolation effect consists of the size segregation of grains in the rolling phase: large grains are observed to rise to the top of the rolling phase, and small grains drift to the bottom of the rolling phase. Due to the percolation effect, the small grains are the first to be captured at the pile surface, resulting in the segregation of the mixture in the bulk.

the dune [1], where stratification could appear as in the experiments of [27]. This phenomenon may also have important consequences in industry where processing and transport of initially mixed grains could lead to stratification.

Spontaneous stratification has been observed in [27] after a transient regime and only when the large grains are *rougher* than the small grains. When the large grains are the smoothest stratification does not occur but only the segregation of the mixture in different regions of the cell occurs, with the large and rounded being at the bottom of the cell, and the small and rough at the top. These phenomena can be qualitatively understood as the growth of an instability due to the competition of two opposite effects. (i) On the one hand, if the grains have the same shape (spherical, for example) or, more precisely, the same repose angle, then the large grains will roll more easily on the top of the small ones than the reverse. This will lead to segregation, with the small grains at the top of the pile and the large at the bottom. (ii) On the other hand, if the grains have the same size but different shape, the rougher grains will have a larger friction coefficient than the other grains, and we expect segregation with the rougher grains at the top of the sandpile. In the case of large grains rougher than small grains, those two effects will compete, giving rise to an instability, and then to spontaneous stratification. However, in the case where the smaller grains are rougher than the large grains, the two effects will contribute in the same way, and the segregation pattern will

be stable, without the occurrence of stratification [34].

In this paper, we study granular flow in the spirit of [35–38], where one assumes a sharp distinction between the rolling or fluid phase composed of the layer of grains flowing down and the static phase or bulk composed of grains forming the sandpile. The essential feature to understand sandpile formation is then to describe how the rolling grains are flowing down and how they interact with the static grains. In this context, it is important to distinguish between two different regimes, the thin-flow regime and the thick-flow regime, where the interaction between rolling grains and static grains are very different. Moreover, experiments have been done in both regimes, so that both regimes are important to study.

Here we propose a mechanism to understand spontaneous stratification and segregation in two-dimensional silos. We adopt two different approaches. First we introduce a cellular automaton model to give insight into the proposed mechanism. This model displays stratification as soon as the large grains are rougher than the small ones, in agreement with experiments [27]. The dynamics are also very close to the experiments [27], where the layers are built through a ''kink'' going uphill where the rolling grains are stopped.

Second, we study a continuum model that is built on a phenomenological formalism describing granular flow on sandpiles. This formalism was introduced by Bouchaud *et al.* [35] for the case of a single species, and recently generalized by Boutreux and de Gennes (BdG) [37] to the case of a mixture of two species. The essential feature to understand sandpile formation is to describe the interaction between the rolling or fluid phase and the bulk, as well as any segregation which may occur in the rolling phase itself. In contrast to the cellular automaton model, this formalism includes *amplification* processes, i.e., the conversion of static grains at the surface of the sandpile into rolling grains by the flow (this feature is essential to understand avalanching when one tilts a sandpile above the maximum angle of repose, or to understand avalanche dynamics in rotating drums).

In the large flux regime, the rolling grains form a thick phase and the grains are kinematically segregated in the rolling phase, an effect called kinematic sieving, free surface segregation, or percolation [Fig. 1(b)] [23,25,28]. Due to this phenomenon the large grains in the rolling phase are found to rise to the top of the rolling phase while the small grains sink downward through the gaps left by the motion of larger grains in the rolling phase. Thus, small rolling grains form a sublayer underneath the sublayer of large rolling grains. Then only the lower grains of this layer interact with the sandpile. Thus, a segregation effect that we refer to as percolation takes place inside the rolling phase where the large grains are convected along the flow to the top of the layer. Thus, if small rolling grains are present, they will preferentially be on the bottom of the layer, preventing the large grains from interacting with the sandpile.

We will develop a continuum formalism that includes the percolation effect, and show that stratification arises in a way similar to that of the experiments. When the large grains are rougher, stratification is made of layers spreading all along the sandpile; the layers of the two types of grains being of the same size for an equal volume mixture of grains. When the small grains are rougher than the large grains, we observe the complete segregation of the mixture but not stratification,

with the small and roughest grains being found at the top of the pile in agreement with the experiments. A thick rolling phase is a condition for the percolation effect to take place. However, a large difference in size is also necessary to observe the percolation effect: we expect $d_2/d_1 \gtrsim 1.5$, where d_1 and d_2 are the typical size of the small and large grains, respectively.

In the small-flow regime each rolling grain is always in contact with the sandpile and so interacts with it. In this case a thin rolling phase is expected—of the order $\approx d_2$—and percolation effects are expected to be irrelevant. However, strong segregation effects are expected, as in the case of percolation, since we will consider the case of large size difference between the grains. The large grains are not being captured on top of static small grains (although they interact with them) because of the large difference in size, while the small grains are easily captured on top of the large static grains. Using the theoretical formalism adapted to this case, we will show that stratification is observed when the large grains are rougher than the small grains, in agreement with the experiments.

Cross-amplification processes—the influence of which was greatly reduced in the case of thick flows due to percolation effects—now plays an important role since all grains are interacting with the pile and cross interactions of the type small/large grains are expected to occur at the fluid-bulk interface. Due to cross amplification, the transition from stratification to segregation does not occur sharply when the small grains become the roughest. We find a regime where the small grains are slightly rougher than the large grains, which also gives rise to stratification. When the small grains are more rounded than the large grains, then we find the segregation of the grains in agreement with experiments.

In general, we find that the thick-flow regime and the thin-flow regime show similar results regarding the stratification and segregation of the species. This is valid provided that the grains differ appreciably in size (in term of the size ratio we expect $d_2/d_1 \gtrsim 1.5$ [28,29]). Then we study analytically a mechanism for stratification and segregation valid for both regimes. We study the ''kink'' mechanism, and we obtain predictions for the wavelength of the layers as a function of the different parameters of the system. In particular, we find that the wavelength of the layers is proportional to the flux of adding grains (i.e., proportional to the thickness of the rolling phase), and we also find a crossover behavior at the thin-thick flow transition. Finally we discuss our results in regard to previous approaches to segregation [37] and stratification [34] and to the experiments [27–31]. A short report of some of the results presented here has been published in [38].

II. CELLULAR AUTOMATON MODEL

A. Motivation and Definitions

We first introduce a simplified discrete model to understand the mechanism leading to stratification. Discrete models have been used before for modeling the complex behavior of granular materials. The role played by the angle of repose has been used to understand the complex dynamics of granular flows of mixtures [8,35,39,40]. The concept of angle of repose normally involves a macroscopic

definition—i.e., the angle of repose is the angle of the pile surface after the grains are poured onto a heap. Here, we will define a microscopic angle of repose as the maximum angle at which a rolling grain is trapped at the surface of the pile. This definition must be understood in terms of a coarse grained distance along the surface of the pile—of the order of a few grain sizes—over which all the quantities involved in the model are averaged.

In the case of a single-species sandpile, as sand is added to the sandpile we consider a critical angle of repose θ_r. When the local angle of the pile is smaller than θ_r the rolling grain is stable and there is no flow. When the local angle of the pile is larger than θ_r the rolling grain is unstable and flow occurs.

We develop a simple discrete model, based on the idea that grains flow when the local angle of the sandpile is larger than the angle of repose. We consider that the angle of repose depends on the type of rolling grains and also on the composition of the surface. Thus, in the case of granular mixtures of two different species, we consider the existence of four different *generalized angles of repose*.

The sandpile is built on a lattice, where the grains have the same horizontal and vertical size as the lattice spacing. The two species will be distinguished according to different generalized angles of repose. In general, we will call species type 1 to the small grains, and species type 2 to the large grains. The species can also have rough or smooth surface properties, which, together with the different size, will define the different angles of repose of the species. Following [35–37], we regard each grain as belonging to one of two phases: (i) static phase, if the grain is part of the sandpile; (ii) rolling phase, if the grain is not part of the sandpile and rolls downward with a constant drift velocity.

We consider the local slope

$$s_i \equiv h_i - h_{i+1} \tag{1}$$

of the static grains as the variable controlling the dynamics of the rolling grains. Here, h_i denotes the height of the sandpile at coordinate i, and we assume the pouring point at $i = 1$.

At each time step a set of N_1 small grains and N_2 large grains is deposited at the top of column 1 of the pile [see Fig. 2(a)]. These grains are considered to belong to the rolling phase. In the case of thin flow we assume that the rolling phase is homogeneous and both types of species are mixed in the fluid phase and interact with the surface. Then one rolling grain of each species interacts with the surface at each time step, and can be converted from the rolling phase to the static phase. In the thick-flow regime, the percolation effect is expected to take place, and the small grains screen the interaction of the large rolling grains with the pile surface. Thus in the thick-flow regime, we consider that the large grains interact with the pile surface only when the number of small rolling grains at a given position falls below a given threshold $\epsilon \ll N_2$.

The remaining rolling grains which do not interact with the pile surface are convected ''downward'' with unit ''drift velocity''—i.e., they move to the next column at each time step.
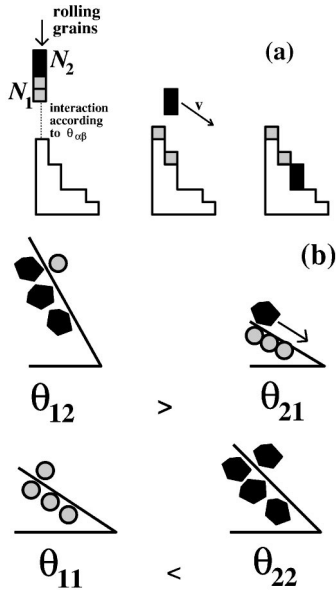
The dynamics of each rolling grain interacting with the

FIG. 2. (a) Description of the discrete model. (b) The four dynamical angles of repose $\theta_{\alpha\beta}$ depend on the composition of grains at the surface of the pile, and are chosen according of the four possible interactions between the two species of grains. As discussed in the text, the angles of repose satisfy $\theta_{21} < \theta_{11} < \theta_{22} < \theta_{12}$, when species 1 are small and rounded and species 2 are large and rough; and $\theta_{21} < \theta_{22} < \theta_{11} < \theta_{12}$, when species 1 are small and rough and species 2 are large and rounded.

sandpile surface is governed by its local generalized angle of repose. We consider that the repose angle depends on the local composition on the surface, so we define $\theta_{\alpha\beta}$ as the generalized repose angle of a rolling grain of type α on a surface with local grains of type β. We propose that

$$\theta_{21} < \theta_{12} \qquad (2)$$

to take into account the fact that large rolling grains roll more easily on top of small static grains than small grains roll on top of large static grains [since the surface ''looks'' smoother for large grains rolling on top of small grains, see Fig. 2(b)]. The repose angles of pure species $\theta_{\alpha\alpha}$ lie between θ_{21} and θ_{12}.

Since a large grain rolls easier than a small rolling grain on top of a layer of small static grains, we have

$$\theta_{21} < \theta_{11}. \qquad (3)$$

Conversely, a large grain rolls easier than a small grain on top of a layer of large static grains, so that

$$\theta_{22} < \theta_{12}. \qquad (4)$$

The stratification experiments [27] use a mixture of smaller less faceted grains and larger more faceted grains. Thus, the last inequality between the angles of repose is provided by the fact that the grains differ in shape of friction properties. Since the small species are the most rounded, then the repose angle of the smaller pure species is smaller than the repose angle of the large pure species—i.e.,

$$\theta_{11} < \theta_{22}. \qquad (5)$$

Therefore, to mimic the experimental conditions for stratification [27], we propose

$$\theta_{21} < \theta_{11} < \theta_{22} < \theta_{12}. \qquad (6)$$

We notice that conditions (2)–(4) are a consequence of the different size of the species, while the condition $\theta_{11} \neq \theta_{22}$ is achieved for mixtures of grains with different shapes, and does not depend on the size of the grains. Thus the model incorporates the size segregation and shape segregation in the definition of the angles of repose. In general the grains with the larger angle of repose will tend to be captured first. Thus the small grains will tend to be captured at the top of the pile, and the smoothest grains at the bottom of the pile. The percolation effects tend to segregate the small grains at the top of the pile, thus, it acts in the same way as the size segregation due to different angles of repose, Eqs. (2)–(4).

At each time step, the rolling grain interacting with the sandpile surface at coordinate i either will stop (by being converted into a static grain) if the local slope of the surface

$$h_i - h_{i+1} \leq s_{\alpha\beta} \equiv \tan \theta_{\alpha\beta}, \qquad (7)$$

where $s_{\alpha\beta}$ is the local slope, or will continue to roll (together with the remaining rolling grains) to column $i+1$ if

$$h_i - h_{i+1} > s_{\alpha\beta}. \qquad (8)$$

We iterate this algorithm to form a large sandpile of typically 10^5 grains. We assume that the substrate is made of a layer of large grains.

The discrete model in the thick-flow and thin-flow regimes gives rise to similar results. Then in the following we will present results of the discrete model in the thick-flow regime where most of the experiments are done, and where the grains are segregated in the rolling phase. Indeed, we will see when discussing the continuum model that the differences between the thin-flow regime and the thick-flow regime arise when cross-amplification processes are taken into account and for small difference between the pure angles of repose.

B. Kink mechanism

Figure 3(a) shows the resulting morphology predicted by the discrete model when the large grains are rougher than the small grains. This fact is quantified by taking into account that the angle of repose of the pure large species is larger than the angle of repose of the pure small species, $\theta_{22} > \theta_{11}$. The stratification is qualitatively the same as that found experimentally [27], both in regard to the *statics* of the sandpile [seen in Fig. 3(a)], and also in regard to the *dynamics* (seen in Fig. 4).

The mechanism for spontaneous stratification involves the formation of a kink at which the grains are stopped during an avalanche. 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} [Fig. 4(a)]. 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 as more grains are poured onto the cell. These small grains smooth the surface without changing significantly the sandpile angle, and allow rolling small grains to go further
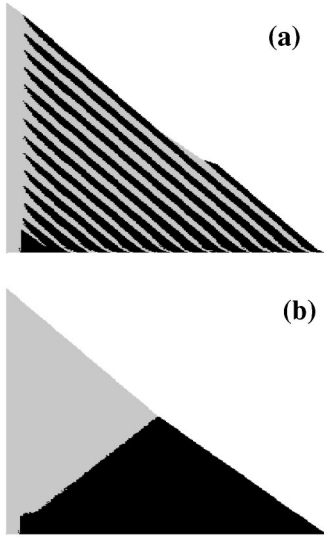
FIG. 3. (a) Spontaneous stratification: results obtained with the discrete model for $N_1 = 10$, $N_2 = 10$, $\epsilon = 1$, $s_{11} = 4$, $s_{12} = 7$, $s_{21} = 2$, $s_{22} = 5$. Note the kink profile at which grains are stopped during an avalanche. The large rough grains are black and the small rounded grains are gray. (b) Segregation: results obtained with the discrete model when the angle of repose of the large rounded grains is smaller than the angle of repose of the small rough grains for $N_1 = 10$, and $N_2 = 10$, $\epsilon = 1$, $s_{11} = 5$, $s_{12} = 7$, $s_{21} = 2$, $s_{22} = 4$.

down (since $\theta_{11} < \theta_{22}$) [Fig. 4(b)]. 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 [Fig. 4(b)]. 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 at the kink since $\theta_{21} < \theta_{11}$, and a pair of layers begins to form, with the small grains underneath the large grains [Fig. 4(c)]. 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 [Fig. 4(d)].

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 complete segregation of the mixture but no stratification [Fig. 3(b)]. In this case, the small and faceted grains are to be found near the top of the pile, while the large and rounded grains are found near the bottom of the pile. Thus, the control parameter for spontaneous stratification appears to be the
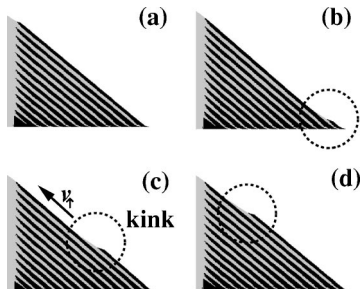

FIG. 4. Dynamics obtained with the discrete model, showing the formation of a pair of layers at the kink. Notice the kink moving uphill at constant velocity $v_\uparrow$.

difference in the repose angle of the pure species, which quantifies the difference in shape of the grains.

As seen in Fig. 3(a), 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 $\theta \simeq \theta_{11}$. The repose angle for large rolling grains is now θ_{22}. Thus, if $\theta \simeq \theta_{11} < \theta_{22}$, 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 $\theta \simeq \theta_{11} > \theta_{22}$, no more large grains can get captured, the fluctuation disappears, and the segregation profile remains stable. Thus this picture suggests that, in agreement with [27], 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 $\theta_{11} < \theta_{22}$. This explains qualitatively why the control parameter for the stratification-segregation transition is the angle of repose of the pure species.

III. CONTINUUM THEORY FOR GRANULAR FLOW

A theoretical approach to segregation in granular flow is not a simple task. On one hand, extensive numerical simulations (such as molecular dynamics [3,5,41–44], or lattice gas models [45–48]) have been able to reproduce several segregation phenomena observed experimentally. On the other hand, an analytical approach of segregation, considering granular medium as continuum, would be instructive, since it would allow to reduce the problem to a small set of parameters that control the system, and then to get a clear phenomenological understanding of the problem. However, an important question is whether the grains can be treated as a continuum medium. A continuum approach means that we will be able to replace the grains belonging to a same region of space by some average quantities, for example, the density, or the averaged speed. But specific grains can play a peculiar role, i.e., in the case of arching effects, where an arrangement of several grains in a special way (an arch) supports the above grains, prevents the flow by gravity, and creates large discontinuities for the local density or the force [49,50].

In this section, we study spontaneous stratification using a continuum formalism. We will show that the results obtained are very close to the experiment, and then we extract the important parameters of the problem. Our results shows that a continuum formalism is able to reproduce a complex phenomenon in granular matter such as spontaneous stratification.

The continuum theory of granular flow for a single-species sandpile was proposed by Mehta [51] and Bouchaud et al. [35]. Bouchaud et al. recognized the necessity of treating differently the rolling grains (rolling on the surface of the pile), and the static grains that form the sandpile. The rolling grains are in a "liquid" state, flowing down by gravity and interacting with the static grains in the "solid" state. The dynamics of the sandpile is then given by how the rolling grains move on the surface and how they interact with the sandpile. The equations for those two quantities for the case of single-species sandpiles are

$$\frac{d}{dt}R[x(t)]=\Gamma(R,h), \qquad (9a)$$

$$\frac{\partial h(x,t)}{\partial t}=-\Gamma(R,h). \qquad (9b)$$

Here $R[x(t)]$ is the height of the layer of rolling grains in the Lagrange representation (following the rolling grains), and $h(x,t)$ the height of the sandpile [Fig. 1(a)]. The rolling grains interact with the sandpile through $\Gamma(R,h)$. Equations (9) conserve the number of grains.

We need to define how the rolling grains move on the surface, and how they interact with the sandpile. Two forces are acting on the rolling grain: (i) gravity and (ii) friction coming from the collisions of the rolling grain with the grains of the sandpile. The number of collisions at the scale of interest for continuum equations (several times the size of a grain) is large enough so that at this scale, the speed is always at its limit value where the two forces balance exactly, and the motion of a grain is overdamped. Equations (9a) and (9b) can be rewritten in Euler representation

$$\frac{\partial R(x,t)}{\partial t}=-v\frac{\partial R(x,t)}{\partial x}+\Gamma(R,h), \qquad (10a)$$

$$\frac{\partial h(x,t)}{\partial t}=-\Gamma(R,h). \qquad (10b)$$

Here $R(x,t)$ is now a function of the time and the position x, and we assume the pouring point at $x=0$. The rolling grains are moving down at a speed $v(x,t)$ in the positive x direction, where $v(x,t)$ could depend on the local slope of the sandpile. However, in the spirit of capturing only the essential physical mechanisms to recover the observed phenomena, we will consider the speed v to be constant all over the sandpile.

Bouchaud *et al.* [35] proposed a form for $\Gamma(R,h)$ in the general case, that has been simplified by de Gennes [36] for the simpler case of a continuous flow of rolling grains. In this case, there is no discrete avalanching and only the repose angle must be included in the formalism [52]. The interaction term $\Gamma(R,h)$ is

$$\Gamma(R,h)=\gamma[\theta(x,t)-\theta_r]R, \qquad (11)$$

where

$$\theta(x,t)\equiv\frac{-\partial h(x,t)}{\partial x} \qquad (12)$$

is the local angle of the surface (we will make no distinction between the angle of the surface and the tangent of the angle).

Equation (11) states that the rate of the interaction is proportional to the number of grains interacting with the sandpile. Rolling grains will become part of the sandpile if the angle of the surface $\theta(x,t)$ is smaller than the repose angle θ_r (''capture''), while static grains will become rolling grains if $\theta(x,t)$ is larger than θ_r (''amplification''). The constant γ is the inverse of the time on which this interaction is

effective because the distance on which a rolling grain is stopped when $\theta(x,t)$ is smaller than θ_r is v/γ. This distance must scale with the size of the grain d. Thus [36]

$$v/\gamma\sim d. \qquad (13)$$

The linear dependence of $\Gamma(R,h)$ on the quantity $[\theta(x,t)-\theta_r]$ can be understood as a first-order development of a more complicated function of $\theta(x,t)$. As soon as the angle of the sandpile is far from the repose angle, nonlinear terms must be added (avoiding the nonphysical divergences that could be found with the linear development). Finally the proportionality of $\Gamma(R,h)$ to $R(x,t)$ is meaningful in the case where all the grains interact at each time with the surface (thin-flow limit). This imposes the height of the rolling layer to be of the order of the grain size $R(x,t)\simeq d$. However, we will argue that this approximation might be still valid in the case of thick flows as well, since the interaction might be proportional to the pressure exerted by the fluid phase, which in turn is proportional to $R(x,t)$ for a fluid [16]. For very large fluxes (thicker rolling phase) we expect this linear approximation to break down, and we discuss this point at the end of Sec. V C.

To treat the problem of segregation in granular flow composed of binary mixtures, Boutreux and de Gennes (BdG) [37] have extended this formalism to the case of flows made of two types of grains, identifying three variables: the two heights of rolling grains $R_\alpha(x,t)$ (i.e., the total thickness of the rolling phase multiplied by the local volume fraction of the α grains in the rolling phase at position x), and the height of the sandpile $h(x,t)$. BdG generalized Eqs. (10) to

$$\frac{\partial R_\alpha}{\partial t}=-v_\alpha\frac{\partial R_\alpha}{\partial x}+\Gamma_\alpha, \quad \alpha=1,2, \qquad (14a)$$

$$\frac{\partial h}{\partial t}=-\Gamma_1-\Gamma_2, \qquad (14b)$$

where the interaction term Γ_α takes into account the conversion of the rolling grains into static grains, and is defined through the collision matrix $M_{\alpha\beta}$,

$$\Gamma_\alpha\equiv\sum_{\beta=1}^{2}M_{\alpha\beta}R_\beta. \qquad (15)$$

The collision matrix, governing capture and amplification, depends on the local angle of the pile $\theta(x,t)$ and on the local composition of the surface of the sandpile $\phi_\alpha(x,h)$, which is a function of x and h. However, writing an equation of evolution for $\phi_\alpha(x,h)$ is not easy. When both rolling species are captured, the height of the sandpile increases, and $\phi_\alpha(x,h)$ is given by

$$\phi_\alpha(x,h)=-\frac{\Gamma_\alpha}{\partial h/\partial t}, \qquad (16)$$

and

$$\phi_1+\phi_2=1. \qquad (17)$$

As soon as amplification dominates for one or both species, Eq. (16) is no longer valid. When $\Gamma_\alpha > 0$ for both species, the height of the sandpile decreases, and $\phi_\alpha(x,h)$ does not have to be updated. Finally, when grains of type α are captured and grains of type β are amplified, the composition will be $\phi_\alpha(x,h) = 1$.

The general form of the collision matrix is defined by taking into account a set of binary collisions between a rolling and a static grain [37],

$$\hat{M} \equiv \begin{pmatrix} a_1(\theta)\phi_1 - b_1(\theta) & x_2(\theta)\phi_1 \\ x_1(\theta)\phi_2 & a_2(\theta)\phi_2 - b_2(\theta) \end{pmatrix}. \quad (18)$$

This definition involves a set of a priori unknown collision functions contributing to the rate processes: $a_\alpha(\theta)$ is the contribution due to an amplification process (when a static grain of type α is converted into a rolling grain due to a collision by a rolling grain of type α), $b_\alpha(\theta)$ is the contribution due to capture of a rolling grain of type α (when a rolling grain of type α is converted into a static grain), and $x_\alpha(\theta)$ is the contribution due to a cross-amplification process, (the amplification of a static grain of type β due to a collision by a rolling grain of type α).

BdG [37] proposed an analytically tractable form for the collision matrix that includes capture and amplification, in the case where the two species have the same size but differ in respect to their repose angle. BdG found the steady state solution in the geometry of the silo, in the case where the repose angle does not depend on the composition of the surface. The BdG model explains the segregation of the two type of grains in different regions of the sandpile, but not the phenomenon of spontaneous stratification. A generalization of the model to include also the different sizes of the grains [53] shows a smooth segregation of the species with the concentrations of static grains behaving as a power law of the position along the surface pile; results valid only when the species do not differ too much (a fact that allows to perform linear approximations of the collision functions around the angles of repose [53]). In the following we propose a form of the collision functions valid when there is a large difference in size and shape between the species and we treat the thin- and thick-flow regimes, in order to understand stratification and segregation as seen in the experiments.

IV. CONTINUUM MODEL FOR STRATIFICATION OF GRANULAR MIXTURES

A. Collision matrix and generalized angle of repose

We present the collision matrix $M_{\alpha\beta}$ that includes the effects of capture and amplification, and the dependence of the repose angle on the composition of the surface as discussed in [38]. Considering one type of rolling grain α, we define capture and amplification as follows: if the local angle of the surface $\theta(x,t) = -\partial h(x,t)/\partial x$ is smaller than the generalized angle of repose of the rolling specie α, θ_α, the α rolling grains will be captured. In the reverse case $\theta(x,t) > \theta_\alpha$, the static grains at position x will be amplified. Amplification will cause locally a small hole, i.e., the two types of static grains will be amplified the same way according to their local concentration ϕ_α.
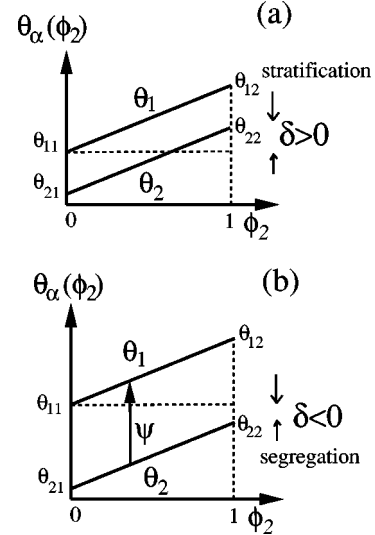

FIG. 5. Dependence of the repose angle for the two types of rolling grains on the concentration of the surface of large grains ϕ_2. (a) An essential ingredient to obtain stratification is that $\theta_{22} > \theta_{11}$, (b) while for segregation we require $\theta_{11} > \theta_{22}$. For the numerical integration, we use the linear interpolation between $\phi_2 = 0$ and $\phi_2 = 1$ as plotted here.

The generalized 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. 5),

$$\theta_1(\phi_2) = m\phi_2 + \theta_{11}, \quad \theta_2(\phi_2) = m\phi_2 + \theta_{21} = -m\phi_1 + \theta_{22}, \quad (19)$$

where $m \equiv \theta_{12} - \theta_{11} = \theta_{22} - \theta_{21}$. We have assumed the difference

$$\psi \equiv \theta_1(\phi_2) - \theta_2(\phi_2) \quad (20)$$

to be independent of the concentration ϕ_2. For simplicity, we choose a linear interpolation between the extreme values $\theta_\alpha(\phi_\beta = 0)$ and $\theta_\alpha(\phi_\beta = 1)$. If we label the small grains with $\alpha = 1$ and the large grains with $\alpha = 2$ and denote the extreme values of the repose angle by $\theta_\alpha(\phi_\beta = 1) = \theta_{\alpha\beta}$ as in Sec. II, the difference in size implies [see Fig. 2(b)]

$$\theta_{12} < \theta_{21}. \quad (21)$$

If species 1 are the smallest then $\theta_1(\phi_2) > \theta_2(\phi_2)$ for any composition of the surface ϕ_2—i.e., the small grains are always the first to be captured. Thus, the angular difference ψ is determined mainly by the difference in size between the species, and the value of ψ determines the degree of segregation; the larger ψ, the stronger the segregation. The repose angles of the pure species θ_{11} and θ_{22} will be intermediate between θ_{12} and θ_{22}, and their relative values will depend on the shape of the two species as discussed in Sec. II A.

For mixtures of grains with different shapes or friction coefficients we have $\theta_{11} \neq \theta_{22}$, and $\theta_{12} = \theta_{21}$ if the species have the same size. If θ_{11} is the repose angle of the pure round species and θ_{22} is the repose angle of the pure rough species, then $\theta_{11} < \theta_{22}$. If the species have the same shape or friction coefficients then $\theta_{11} = \theta_{22}$.

When the size ratio is close to one ($d_2/d_1 \lesssim 1.5$), the angle ψ is expected to be small and then it is plausible to linearize the collision functions around the angles of repose as done in [53]. When there is a larger difference in size ($d_2/d_1 \gtrsim 1.5$), we expect ψ to be large, and we approximate the collision functions and define the following collision matrix as follows:

$$\hat{M} = \begin{pmatrix} \gamma_{11} \ \delta\theta_1 \ \Theta_{\text{self}}[\delta\theta_1, \phi_1] & \gamma_{12} \ \delta\theta_2 \ \Theta_{\text{cross}}[\delta\theta_2, \phi_1] \\ \gamma_{21} \ \delta\theta_1 \ \Theta_{\text{cross}}[\delta\theta_1, \phi_2] & \gamma_{22} \ \delta\theta_2 \ \Theta_{\text{self}}[\delta\theta_2, \phi_2] \end{pmatrix},$$

(22)

where

$$\delta\theta_\alpha \equiv \theta(x,t) - \theta_\alpha.$$

(23)

Here, $\gamma_{\alpha\alpha} > 0$ has dimension of inverse time, and $v/\gamma_{\alpha\alpha}$ represents the length scale at which a rolling grain will interact significantly with a surface at an angle above or below the angle of repose.

The functions $\Theta(\delta\theta_\alpha, \phi_\alpha)$ distinguish capture from amplification. Θ_{self} treats the case of self amplification, i.e., amplification of static grains α by rolling grains α, and Θ_{cross} the case of cross-amplification, i.e., amplification of static grains α by rolling grains β. More specifically, we consider

$$\Theta_{\text{self}}(\theta - \theta_\alpha, \phi_\alpha) = \begin{cases} 1, & \text{if } \theta - \theta_\alpha < 0, \\ \phi_\alpha, & \text{if } \theta - \theta_\alpha > 0. \end{cases}$$

(24)

These equations mean that in the case of capture, the amount of rolling grains of type α converted to static grains is proportional to the number of grains interacting with the surface, i.e., R_α; in the case of amplification, the amount of static grains converted to rolling grains is proportional to R_α again and to the number of static grains α on the surface, i.e., ϕ_α. Accordingly, we obtain

$$\Theta_{\text{cross}}(\theta - \theta_\alpha, \phi_\beta) = \begin{cases} 0, & \text{if } \theta - \theta_\alpha < 0, \\ \phi_\beta, & \text{if } \theta - \theta_\alpha > 0. \end{cases}$$

(25)

In terms of the collision functions defined in Eq. (18) we have

$$\begin{aligned} a_\alpha(\theta) &\equiv \gamma_{\alpha\alpha} \Pi[\theta(x,t) - \theta_\alpha(\phi_\beta)], \\ b_\alpha(\theta) &\equiv \gamma_{\alpha\alpha} \Pi[\theta_\alpha(\phi_\beta) - \theta(x,t)], \\ x_\beta(\theta) &\equiv \gamma_{\beta\alpha} \Pi[\theta(x,t) - \theta_\beta(\phi_\beta)], \end{aligned}$$

(26a)

where

$$\Pi[x] \equiv \begin{cases} 0, & \text{if } x < 0, \\ x, & \text{if } x \geq 0. \end{cases}$$

(26b)

This form for the matrix $M_{\alpha\beta}$ supposes that amplification (capture) occurs only when the local angle is larger (smaller) than the repose angle. Thus, for a given angle only capture or amplification can occur at a given time; an assumption which is expected to be valid when the grains differ appreciable (ψ large). This fact gives rise to strong segregation effects with exponential behavior of the concentrations as we will corroborate when calculating the steady state solution of the problem in Sec. V A. In contrast, the form of the collision

functions proposed in [37,53] assumes that amplification and capture are linear functions of the local angle. For a given angle both amplification and capture act simultaneously, and the repose angle corresponds to the angle where amplification and capture are equal. This linear approximation is valid when the species do not differ much (for small ψ), and gives rise to a weaker segregation of the species—with a power law behavior of the concentrations as a function of the position along the pile surface—than the segregation found in this study which is valid when the species differ appreciable in size and shape.

We focus here on the dependence of the repose angles on the composition of the surface. We will consider that the other parameters—i.e., $\gamma_{\alpha\alpha}$ and v_α—do not depend on the composition of the surface and on the species considered

$$\gamma_{11} = \gamma_{22} = \gamma, \quad v_1 = v_2 = v.$$

(27)

In the conditions of the experiment [27], where an equal volume of the two species is poured at the left side of the cell, the boundary conditions are

$$R_\alpha(0,t) = R_\alpha^0 = \frac{R^0}{2}.$$

(28)

These equations are meaningful if all the rolling grains interact each time with the surface, and do not interact directly with each other. Then, when there is percolation in the rolling phase, the interaction terms have to be modified accordingly. In the next section we will show how to introduce the percolation effect characteristic of the thick-flow regime. However, we will show that the above equations—although strictly valid in the thin-flow limit, i.e., $R_\alpha \simeq d_\alpha$—will be also needed to describe completely the thick-flow regime.

B. Thick-flow regime and the percolation effect

An important consideration of the model proposed above is that the two types of rolling grains do not interact significantly each other when the flow of grains is small, i.e., $R_\alpha(x,t) < \epsilon$, where ϵ is a cutoff of the order of the grain size d_α. This hypothesis may not be valid in experiments where the input flux is large, where the layer of rolling grains could be larger than one grain size.

In the case of a thick layer of rolling grains, the phenomenon of percolation occurs; during the flow of grains down an inclined plane, the small rolling grains percolate downward through the gaps left by the motion of larger rolling grains [23,25,28] [Fig. 1(b)]. Therefore, as they are convected down, the rolling grains segregate, the concentration of small grains being large at the bottom of the rolling layer, and the concentration of large grains large at the top of the rolling layer. As only the lowest grains of the rolling layer interact with the sandpile, the above effect imposes that small grains will preferentially interact with the static grains. We include the percolation effect in the continuum model and see how the results related to spontaneous stratification are affected.

To model this effect, we consider the extreme situation where the segregation is present as soon as the grains start to roll, i.e., up to the top of the sandpile, and that only the small grains will interact with the sandpile as soon as $R_1(x,t)$

$>\epsilon$, with $\epsilon \ll R^0$. ϵ is thus the height of small grains below which the percolation stops to be effective, and for which the large rolling grains start to interact with the surface. The interaction terms discussed in Sec. IV A have to be modified accordingly.

This model implies that except for a small region where $R_1 < \epsilon$, the surface of the sandpile is always divided into two regions. The upper region corresponds to the part of the surface where small rolling grains are present. As they are the only one to interact with the sandpile, the sandpile will be made only of small grains. In the lower part, only large grains are present (except for the ϵ part of small grains). The division point between the two regions is able to move with time.

For the upper part of the sandpile, defined by $R_1(x,t) > \epsilon$, only small grains are captured, with $R_2(x,t) = R_2^0 = R^0/2$ and $\phi_1(x,t) = 1$. The equations become

$$\frac{\partial R_1}{\partial t} = -v\frac{\partial R_1}{\partial x} + \gamma(\theta - \theta_{11})(R_1 + R_2), \qquad (29a)$$

$$\frac{\partial h}{\partial t} = -\gamma(\theta - \theta_{11})(R_1 + R_2). \qquad (29b)$$

When $R_1(x,t) < \epsilon$, the percolation effect disappears and the equations of evolution of R_α and h can be considered to be the same as the one defined by the collision matrix (22) valid for the thin-flow limit.

The effectiveness of the interaction (given by γ before) must be multiplied now by the vertical pressure acting on the lowest rolling grain interacting with the surface, i.e., the weight of the column of rolling grains above the interacting grain [16]. This explains the presence of the $R_2(x,t)$ terms in the interaction term Γ_1 in Eq. (29).

We numerically solve Eqs. (14) with the interaction terms given by (22) when $R_1(x,t) < \epsilon$ and by Eq. (29) when $R_1(x,t) > \epsilon$ as discussed above. We find qualitatively the same results as in the experiments [27], with stratification appearing when the large grains are rougher than the small grains [$\theta_{22} > \theta_{11}$, Fig. 6(a)]. The dynamics of stratification show also the formation of the kink as observed in the experiments and in the discrete model shown in Fig. 4. When the small grains are rougher than the large grains ($\theta_{11} > \theta_{22}$), then we obtain the strong segregation of the mixture with the small rough grains found at the top of the heap [Fig. 6(b)] and a small region of mixing in the center of the pile [54].

C. Thin-flow regime

In the absence of the percolation effect, Eq. (22) remains valid along all the pile since in the small flow limit a thin rolling phase is expected with $R_\alpha(x,t) < \epsilon$. However, we assume a large difference in size, so that strong segregation effects are expected anyway, and the collision functions are expected to behave as in Eq. (22). We then solve Eqs. (14)–(22) numerically. Defining the ''control parameter''

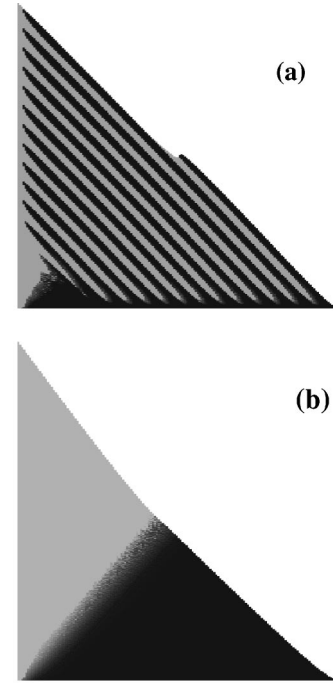$$\delta \equiv \theta_{22} - \theta_{11}, \qquad (30)$$

FIG. 6. Thick-flow regime: morphology resulting from the numerical integration of the continuum equations when the percolation effect is present. (a) Stratification of a mixture of large rough grains and small smooth grains. The parameters used are $R_1^0 = R_2^0 = 1$, $\epsilon = 0.25$, $\tan(\theta_{11}) = 0.5$, $\tan(\theta_{22}) = 0.6$, $\tan(\theta_{12}) = 0.9$, $\tan(\theta_{21}) = 0.2$, $\gamma_{11} = \gamma_{22} = 1$, $\gamma_{21} = \gamma_{12} = 0.1$, and $v_1 = v_2 = 1$. The dynamics obtained by the numerical integration of the equation of motion proposed in the text show the formation of a pair of layers through the kink mechanism. We also see that the rolling grains are stopped at the kink in similar fashion as was observed in the stratification experiment [27] and in the discrete model in Fig. 4. (b) Complete segregation of a mixture of small rough grains and large smooth grains obtained with the same parameters as in (a) except for the angle of repose of the pure species $\tan(\theta_{11}) = 0.7$, $\tan(\theta_{22}) = 0.5$. Notice the total segregation of the mixture. The region of mixing is concentrated in a small region in the center of the pile.

we obtain stratification when $\delta > 0$, i.e., when the large grains are rougher than the small grains as in the experiments and in the thick-flow regime above. However, we find that the transition to segregation does not occur sharply at $\delta = 0$ but occurs at a small negative value $\delta_c < 0$, which depends on the value of the cross-amplification rates $\gamma_{\alpha\beta}$. When the cross rates are zero, then the transition occurs at $\delta = 0$ as we found for the thick-flow regime above. For smaller values of $\delta < \delta_c < 0$ we find the complete segregation pattern found in the experiments. Thus, the presence of cross-amplification processes—which appear only in the low-flux limit—shifts the transition from stratification to segregation to a value δ_c different from zero [55]. However, the stratification we find for $\delta_c < \delta < 0$ is less pronounced than in the case $\delta > 0$: the layers do not go to the top of the sandpile, and the layer of small grains is very thin. Similar results at the neighborhood of the stratification-segregation transition have been found with a microscopic model of grains dynamics [56]. 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 for $\delta > 0$.

This model, valid for thin flows, is qualitatively close the preceding one valid for thick flows, where we also found a regime of complete segregation separating the surface of the sandpile in two regions. This can be understood noting that the dependence of the repose angle on the composition of the surface in the thin-flow model is comparable to the effect of percolation in the rolling layer in the thick-flow regime. Since for a given composition ϕ_β, the repose angle of the small grains is always larger than the repose angle of the large grains (Fig. 5), the small grains are also the first to be trapped before the large ones. The composition of small grains on the surface increases, amplifying the effect, up to the point where the surface is made of only small grains.

A difference between the models for the thick rolling phase and the thin rolling phase is in the role played by amplification processes. When neglecting amplification we find identical results in both regimes, as we found with the discrete model. Moreover, the influence of amplification processes seems to be confined to the case when the angles of repose of the pure species are very close, and only to the thin-flow regime. The fact that we have obtained similar results in the thin-flow regime and the thick-flow regime with the discrete model is due to the fact that the discrete model neglects amplification processes.

V. ANALYTICAL RESULTS

The thick-flow and the thin-flow regimes show similar results as long as there is a large difference in size between the grains. We will now show analytical results valid for both regimes.

A. Steady-state solution

We next study the stationary solution of Eqs. (14) and then we study how a perturbation to this solution can be amplified giving rise to stratification. For simplicity, we consider the geometry of a silo of lateral size L, i.e., all the rolling grains are stopped when they reach a wall at position $x=L$ [Fig. 1(a)]. Moreover, we assume the difference

$$\psi \equiv \theta_2(\phi_2) - \theta_1(\phi_2) \qquad (31)$$

to be independent of the concentration ϕ_2. We seek for a solution where the profiles of the sandpile and of the rolling grains are conserved in time. Thus, stratification that corresponds to periodic variations in time of the different variables of interest cannot be observed for this solution. The conservation of the grains gives [57]

$$\frac{\partial h}{\partial t} = \frac{v R^0}{L}, \qquad (32)$$

and we impose

$$\frac{\partial R_\alpha}{\partial t}(x) = 0. \qquad (33)$$

We assume that, as in the experiment, an equal volume mixture is used, so

$$R_1^0 = R_2^0 = R^0/2, \qquad (34)$$

and the boundary conditions are

$$R_\alpha(L) = 0, \quad R_\alpha(0) = R^0/2. \qquad (35)$$

The steady-state solution of (14)–(22) shows *total* segregation: except in a region of size of v/γ at the center of the sandpile where the grains are mixed, the sandpile is exclusively made of small grains in the upper part of the pile, and of large grains in the lower part of the pile. The details of the calculations can be found in [34]. Here we present a simplified results for the case of $v/\gamma \ll L$, since we expect Eq. (13).

At the *upper* part of the pile, for $0 \le x \le x_m$, with $x_m \equiv L/2 - v/(\gamma\psi)$, only small grains are present ($\phi_1(x) = 1$ and $\phi_2(x) = 0$), and the profiles are

$$R_1(x) = R^0 \left(\frac{1}{2} - \frac{x}{L} \right), \qquad (36a)$$

$$R_2(x) = R^0/2, \qquad (36b)$$

$$\theta(x) - \theta_{11} = -\frac{v/\gamma + \psi\gamma_{21}L/(2\gamma)}{L(1 + \gamma_{21}/\gamma)/2 - x}. \qquad (36c)$$

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

$$\phi_1(x) = \exp\left[-\frac{\gamma\psi}{v}(x - x_m) \right], \qquad (37a)$$

$$R_1(x) = \frac{2v}{\gamma\psi L}\phi_1(x)R(x), \qquad (37b)$$

$$\theta(x) - \theta_{22} = -m\phi_1(x) - \frac{v}{\gamma(L - x)}. \qquad (37c)$$

Here

$$R(x) \equiv R_1(x) + R_2(x) = R^0(1 - x/L), \qquad (38)$$

and $m \equiv \theta_{22} - \theta_{21} = \theta_{12} - \theta_{11}$.

When the system is in this stationary state, the angle of the upper part of the sandpile (where only small grains exist) is almost θ_{11}; without cross amplification ($\gamma_{21} = 0$), the sandpile would be at its repose angle θ_{11}, and because of cross amplification, the angle is significantly reduced. The divergence around $x = L/2$ insures that all the small grains are captured despite cross amplification. Moreover, the angle for the low part of the sandpile is almost θ_{22}. If we now suppose a fluctuation in R_1 such that few small rolling grains enter the low part of the sandpile, capture will be of the order of $\gamma(\theta_{22} - \theta_1(\phi_2))R_1$, and amplification will be of the order of $\gamma(\theta_{22} - \theta_2(\phi_2))R_2$. As $R_1 \ll R_2$, amplification will dominate capture, and the small rolling grains will go downhill up to the point when capture starts to exceed amplification. As soon as this small layer of rolling grains exist, the large grains are convected down without being capture. This excess of large grain creates a kink at the bottom of the pile going uphill where large grains are now stopped; the first layers are formed and the same process starts again. This may explain why when cross-amplification terms are present
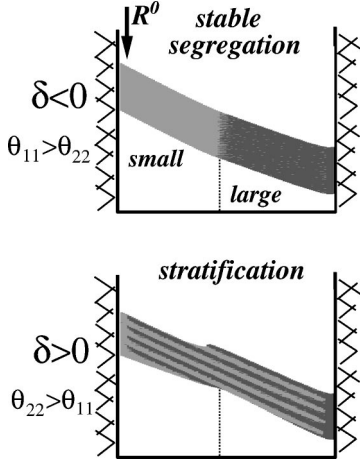
FIG. 7. Stability analysis of the steady state solution of the equations of motion for granular flow of mixtures in the geometry of the silo. When $\delta < 0$ we find that the steady-state solution is stable, so that this solution is the physical solution when $\delta < 0$. On the other hand, when $\delta > 0$ the steady state solution is unstable and it evolves into stratification as in the experiment.

the transition stratification-segregation is shifted from $\delta = 0$ to $\delta = \delta_c < 0$. When percolation is present this shift disappears since cross interactions are greatly reduced by the screening effect of the small rolling grains at the bottom of the rolling phase.

To analyze the stability of the steady-state solution 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 (Fig. 7). For $\theta_{11} > \theta_{22}$, the steady state solution is stable, so that Eqs. (36) and (37) are the physical solution for this case. In this case, only segregation is observed, and the sandpile conserves in time the profiles (36) and (37). For $\theta_{11} < \theta_{22}$, the steady-state solution is *unstable* (evolving to stratification), just as in [27]. The onset of the instability is clearly seen, where the kink first start at the center of the pile where some large grains are captured on top of small grains as seen in Fig. 7 for $\theta_{22} > \theta_{11}$.

Our model predicts the complete segregation of the species when $\theta_{22} < \theta_{11}$. The concentration of small grains drops very fast near the center of the pile in a characteristic region of the order of $v/(\gamma \psi) \sim$ cm according to experiments [28]. As discussed in Sec. IV A this is a consequence of our assumption of large difference in properties of the species.

B. Analytical shape of the kink

We saw in a preceding section that the layers were constructed through a kink where all the grains are stopped. A precise analysis of this kink would allow us to understand the dynamic of the process and the characteristic size of the layers.

From the simulations we observe that the kink has a well-defined uphill speed and that its shape is preserved in time. To obtain an analytically tractable set of equations, we then look for a possible steady-state solution for the shape of the kink, and thus for $R(x,t)$ and $h(x,t)$. We also must make the assumptions that far below and above the kink, the sandpile has a constant angle θ_0 and is only made of large grains.

These assumptions are verified by comparing the obtained result for the shape of the kink with the numerical results.

The existence of a stationary solution for the kink implies that $R_\alpha(x,t)$ and $f(x,t) \equiv h(x,t) + \theta_0 x$ are functions only of

$$u \equiv x + v_\uparrow t, \tag{39}$$

where $v_\uparrow$ is the uphill speed of the kink. For the lowest layer of the kink composed of small grains (the upper part of the pile), as only small grains are captured $[\phi_1(u) = 1, R_2(u) = R^0/2]$, Eqs. (14) reduce to equations for $R_1(u)$ and $f(u)$ (we assume $\gamma_{21} = \gamma$),

$$(v_\uparrow + v) \frac{\partial R_1(u)}{\partial u} = \gamma \left(-\frac{\partial f}{\partial u} - \delta_{11} \right) R_1$$

$$+ \gamma \left(-\frac{\partial f}{\partial u} - \delta_{21} \right) R_2^0$$

$$v_\uparrow \frac{\partial f(u)}{\partial u} = \gamma \left(\frac{\partial f}{\partial u} + \delta_{11} \right) R_1 + \gamma \left(\frac{\partial f}{\partial u} + \delta_{21} \right) R_2^0, \tag{40}$$

with

$$\delta_{11} \equiv \theta_{11} - \theta_0, \quad \delta_{21} \equiv \theta_{21} - \theta_0. \tag{41}$$

We obtain the shape of the low part of the kink. For $u \leq 0, f(u) = 0$ and for $u > 0, f(u)$ and $R_1(u)$ obey the following equations:

$$\frac{\partial f}{\partial u} = \frac{(\delta_{11} + \delta_{21}) R^0/2 - w \delta_{11} f(u)}{v_\uparrow / \gamma + w f(u) - R^0},$$

$$R_1(u) = -w f(u) + R^0/2 \tag{42}$$

where $w \equiv v_\uparrow / (v + v_\uparrow)$. Then the lower part of the kink is characterized by a linear dependence

$$f(u) \propto \frac{(\delta_{11} + \delta_{21})/2}{v_\uparrow / (\gamma R^0) - 1} u. \tag{43}$$

Finally, this solution is valid up to the point where the large grains start to be captured, i.e., $\partial f/\partial u = -\delta_{21}$. From Eqs. (42) and (43) we obtain the following inequalities:

$$\theta_0 > \frac{\theta_{11} + \theta_{21}}{2}, \quad v_\uparrow < \gamma R^0. \tag{44}$$

The solution of the equations for the upper layer of the kink (the lowest part of the pile) where only large grains are present can be obtained in the same way and is

$$f(u) = \left(\frac{R^0}{w} \right) \exp \left(\frac{w \gamma \delta_{22} u}{v_\uparrow} \right), \tag{45}$$

where

$$\delta_{22} \equiv \theta_0 - \theta_{22}. \tag{46}$$

We then find that the shape of the upper part of the kink is exponential, and that the stationary solution exists only for $\delta_{22} < 0$.

Thus we see that the existence of the stationary solution for the kink implies that

$$\frac{\theta_{11} + \theta_{21}}{2} < \theta_{22}, \qquad (47)$$

and that the sandpile is built on an intermediate angle θ_0 between those two extreme values.

C. Wavelength of the layers

The layer thickness λ is defined by the width of the kink, $\lambda \equiv \lim_{u \to \infty} f(u)$. From Eq. (45), we obtain $\lambda = R^0/w$, which is a consequence of the conservation law stating that all the rolling grains are stopped at the kink. This relation is obtained assuming that the density of the fluid phase is the same as the density of the bulk. However, in general we have $\mu_{fluid} < \mu_{bulk}$, where μ_{fluid} and μ_{bulk} are the number of rolling grains per unit volume of the fluid phase and bulk, respectively. Then we obtain

$$\lambda = \frac{\mu_{fluid}}{\mu_{bulk}} \frac{(v + v_\uparrow)}{v_\uparrow} R^0. \qquad (48)$$

Furthermore, Eq. (43) implies that

$$v_\uparrow = C_\uparrow \gamma R^0, \qquad (49)$$

where $C_\uparrow$ is a numerical constant that does not depend on v, γ, or R^0. Then we obtain

$$\lambda_{thin} = \frac{\mu_{fluid}}{\mu_{bulk}} \left(\frac{v}{C_\uparrow \gamma} + R^0 \right) \quad \text{(thin-flow regime)}. \qquad (50)$$

This relation is relevant to the thin flow regime where $v/\gamma \sim d$ and $R^0 \sim d$ are both of the order of the size of the grains, and the velocity of the rolling grains is constant since the rolling phase is homogeneous. However, in the thick-flow regime the mean value of the velocity of the grains scale with the thickness of the rolling phase as found recently experimentally [28],

$$v = C \gamma R^0, \qquad (51)$$

where C is a numerical constant that does not depend on γ or R^0, but may depend on the angles of repose and other features of the grains. In this case Eq. (48) becomes

$$\lambda_{thick} = \frac{\mu_{fluid}}{\mu_{bulk}} \left(1 + \frac{C}{C_\uparrow} \right) R^0 \quad \text{(thick-flow regime)}. \qquad (52)$$

Thus we see that we expect a linear dependence of the wavelength of the layers as a function of the thickness of the rolling phase, i.e., as a function of the flux of grains (if the plate separation is maintained constant [28]), but the proportionality constant is different in the thin-flow regime and the thick-flow regime. From Eqs. (50) and (52) we can obtain an approximate estimation of the value of the crossover from the thin-flow regime to the thick-flow regime R_c^0 assuming that $\lambda_{thin}(R_c^0) \approx \lambda_{thick}(R_c^0)$, then $R_c^0 \approx v_{thin}/(C\gamma)$, where v_{thin} is the velocity of the rolling grains in the thin flow regime.

Typical experimental values of the phenomenological constants of the problem for a system of $L = 30$ cm, $d_1 = 0.27$ mm, $d_2 = 0.8$ mm, are [28] $v \approx 10$ cm/sec, $\gamma \approx 20$/sec, $\tan \psi = \tan \theta_{11} - \tan \theta_{21} \approx 0.1$, $\mu_{fluid}/\mu_{bulk} = 0.85$, and $C/C_\uparrow \approx 1$. Therefore, $v/(\gamma \tan \psi) \approx 2.5$–$5$ cm. We also obtain an estimation of $R_c^0 \approx 0.25$ cm as the value of the crossover flux from the thin flow regime to the percolation regime.

Thus we expect a linear behavior of the wavelength of the layers as a function of the thickness of the rolling phase (or as a function of the incoming flux of grains when keeping the separation between the plates of the cell constant) in the thin-flow regime as well as in the thick-flow regime, although the slope of λ versus R^0 differs in both regimes. This prediction regarding the linearity of λ has been confirmed in recent experiments [28,29] for rolling phases in the range below 1 cm thick and fluxes of the order of 1 g/sec and separation between plates of the cell ≈ 0.5 cm [28]. When the rolling phase becomes thicker than 1 cm, we expect deviations from the linear behavior, and our model must be modified. As discussed in [28] nonlinearities may be present in the behavior of the uphill velocity Eq. (49) or in the velocity (51) of the rolling grains. Moreover, the linearity between the interaction term and the total flux of rolling grains $R_1 + R_2$ proposed in Eq. (29) for the thick-flow regime is not expected to be valid for very large flux. In fact, it has been recently proposed in a study of thick avalanches in single-species sandpiles [58] that for a rolling phase thicker than $10d$, the interaction term should saturate to a constant value $v_\uparrow/\gamma$. It would be interesting to explore the consequences of such interaction term for the dynamics of stratification and segregation in mixtures.

VI. DISCUSSION

In summary, we develop a mechanism to explain the spontaneous stratification reported in [27]. This mechanism is related to the dependence of the local repose angle on the local surface composition. When we consider only capture (corresponding to the cellular automaton model), we find that spontaneous 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). This result is in agreement with the experiments. Stratification is also obtained when θ_{22} is slightly smaller than θ_{11} as soon as we include cross-amplification process, as we find in the thin flow regime with the continuum model. When $\theta_{22} < \theta_{11}$, the model predicts almost complete segregation, but not stratification. These results are in agreement with experiments [27].

It is interesting to compare these results with a previous study [27] where a continuum model was used neglecting cross amplification (i.e., amplification of α static grains by β rolling grains). In this case, it was shown that amplification never occurs during the formation of the sandpile, and stratification is obtained in the same conditions as in the experiments.

We discuss the possible importance of percolation effects in the rolling layer, that takes place when the input flux of grains is large. Although the continuum model must be

deeply modified, the results are surprisingly close to the thin-flow model.

The model describes well the static picture of the sandpile of [27] with alternating layers made of small and large grains, and also reproduces the dynamics, where the layers are built through a kink mechanism. The numerical simulations suggest that the motion of the kink is stationary. It allows us to make quantitative predictions for the dependence of the size of the layers on the different parameters of the problem.

From the theoretical point of view, it is the first time that this continuum formalism for granular flow is directly compared to experiments. The quality of the results suggests that this formalism includes the essential features of the physics of granular flow. Using this formalism, it may be possible to have theoretically access to 3D problems, such as periodic segregation in rotating cylinders [16], that are out of reach with present-day computer simulations.

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