

## Attractive Forces in a Granular Cocktail

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Starting from elementary considerations about the potential energy of bidimensional packings, we postulate the possible existence of an attractive force between two large spherical particles separated by distances of the order of their radius and immersed in a sea of smaller particles. An *a priori* computer simulation of a shaken 3D container of small spheres that embed two large ones does show that the large particles, initially apart, tend to flocculate during the segregation process. [S0031-9007(98)05875-X]

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Granular size and shape segregation, i.e., the natural property of multicomponent granular mixtures to separate their components differing in size, shape, or micromechanical properties, is of tremendous practical importance [1]. Quite generally the industrial handling of food, mineral, and chemical or pharmaceutical granular products, come upon the troublesome problem of granular segregation which prevents complex mixtures of proteinaceous foods, solid chemical products, solid explosive components, etc. In practice, granular segregation results in an expensive use of sophisticated technical procedures which greatly hinders the extensive using of granular mixtures.

Among others (e.g., [2,3]), several computer or theoretical models dealt with this problem assuming that a sandpile configuration at equilibrium results from potential energy minimization [4]. In particular, the Monte Carlo procedure requires the introduction of a sort of artificial temperature into the system in order to allow checking for the various possible positions of the subcomponents during the deposition process. An alternative approach to the simulated building of a 3D and 2D granular pilings using the so-called deterministic “steepest descent algorithm,” have been proposed [5,6]. Recent experimental observations provided substantial support to these computer simulations [7,8] using this sequential and deterministic algorithm which was subsequently implemented with noise and could render most of the observed features. All things considered, it is now generally admitted that sequential building of pilings can simulate correctly the behavior of granulates which undergo a succession of excitation(e.g., upwards launching)—relaxation(e.g., deposit) cycles.

The major question which we address in the following can be roughly stated as follows: We start from a binary mixture of a large number of small monodisperse spherical particles and a couple of larger ones lying initially on the same horizontal line and separated by a given distance. As it is well known, shaking or vibrating this mixture will result in a progressive ascent of both intruders which may, or may not, be reconnected down in the bulk [9]. Elementary tabletop experiments do show

that the larger components of a binary mixture end up at the surface of the piling after a few shakes. The question is the following: Do a couple of large particles tend to approach each other during the ascent or do they climb up independently?

The present Letter starts from elementary considerations in 2D (two dimensions) about the cloud of defects which unavoidably accompanies the introduction of a large particle in a sea of monodisperse smaller particles. When two large particles are sitting apart on a same horizontal line, their clouds of defects intersect giving rise to a sort of depleted zone. This would result in an indirect attractive force favoring flocculation under shaking or vibrating.

As the extrapolation to 3D disordered (frustrated) real situations is certainly not straightforward, we tackle this problem using an *a priori* computer simulation relying upon the “steepest descent algorithm” [5]. We observe that the two large particles tend to approach each other during the ascent induced by a succession of taps onto the bottom of the container. Thus, the basic mechanism seems to work in disordered 3D as well as in 2D ordered configurations. This is a reminiscence of our previous finding that the major results of computer simulations [10] concerning 3D size segregation of a single particle in a sea of smaller particles did not separate from both 2D computer simulations and 2D experimental results [7,8], even though the results obtained in bidimensional configurations made an extensive use of the peculiar symmetry properties of the triangular lattice.

First, we put forward a few preliminary consideration about the gravitational ordering of a bidimensional mixture consisting in a single large particle (a cylinder of radius  $R$ , hereafter called the intruder) imbedded in a sea of smaller cylindrical particles (radius  $r$ ). Be  $\Phi$  the adimensional diameter ratio defined as  $\Phi = R/r$ . We tackled this problem using both experimental observations (described in Refs. [7,8]) and computer generated pilings (Fig. 1). The algorithm we used for computer building such a bidimensional configuration is efficient although rudimentary. We first lay a regular row of small disks

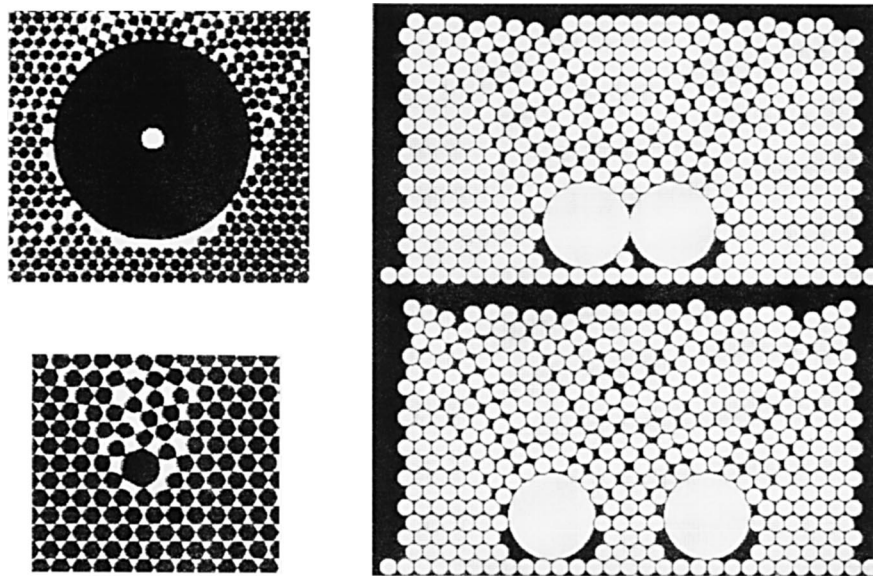


FIG. 1. Bidimensional configurations resulting from the introduction of one (at left) or two (at right) large intruders in a sea of smaller particles. At left, seen through real snapshots of experimentally obtained 2D packings. At right, computer made packings including two intruders. The intersection area of both defect wings is larger when the defects are far apart.

on a horizontal line. We pin a larger intruding disk at a given height above the lowest row. Then we introduce small disks in the piling, one after the other and each one at the lowest possible site. Our previous work [7] reports a detailed analysis of the dependence on  $\Phi$  of the stability of such a piling. Note that our experimental observations deal with relaxed configurations obtained after a gentle shaking of the initially disordered mixture. Doing so, we were able to observe real pilings which quite generally exhibit common features with the computer generated patterns simulating lowest potential energy configurations. The results of experiments and simulations are reported in Fig. 1.

Among others, we observe that the introduction of a larger cylindrical intruder in a sea of smaller particles induces a defect cloud extending above the large particle. As expected, the defect orientation sits at angles  $(\pi/3, 2\pi/3)$  to the horizontal. This certainly arises because of the mismatch between the natural horizontal gravitational ordering of the triangular lattice and the curvature of the external boundary of the intruder. This mere observation is the central point of the following considerations.

We now wonder whether this cloud of defects would possibly allow for some long or medium range interaction between *two* distinct intruders immersed in the piling. From this point of view, the calculation of the potential energy of a computer generated two dimensional piling, including two intruders sitting at a distance  $D$ , is illustrative (hereafter  $D$  is the difference between the center-to-center distance and the intruder diameter). Figure 2 reports an example of typical results obtained at two different size ratio  $\Phi = 5$  and 8, using this procedure.

Starting from a distance  $D = 0$  and progressively increasing the separation between the two intruders lying

on the same horizontal line in the bath, we observe the following interesting features: up to a distance  $D_0$  approximately equal to  $0.7R$ , the piling potential energy  $E$  increases progressively up to a maximum value  $E_m$ , and again increasing progressively the distance we observe a slight and approximately constant very small decrease in the piling potential energy (hardly visible in Fig. 2).

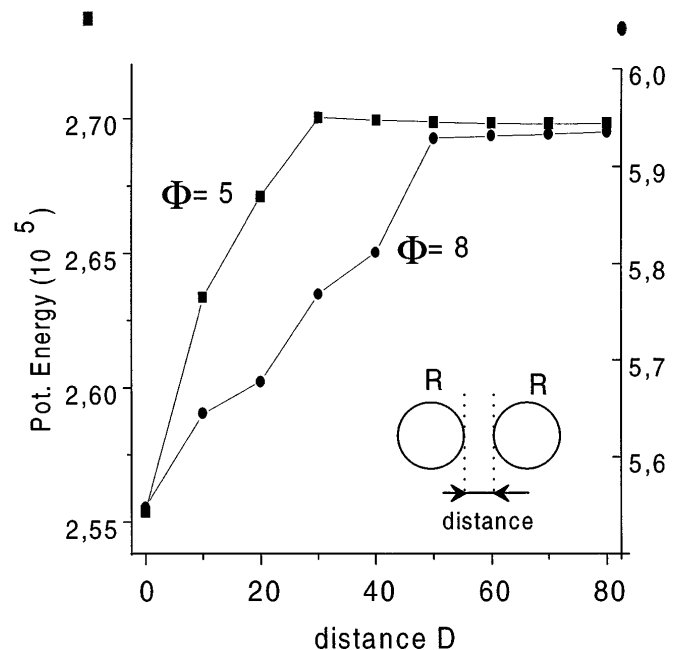


FIG. 2. Potential energy dependence of the piling on the separation between two intruders. Note that the potential energy curve is traced at integer multiples of  $r = 5$  units.

Without attempting to go into the details of a rigorous topological analysis, a careful scrutiny of the computer generated patterns is instructive. The right hand side of Fig. 1 reproduces pictures of the central part of a typical series of patterns obtained at different separations  $D$ . Quite generally, and as can be seen on this figure, both intruders interact via the creation of a central intersection zone of both defect lines generated by each intruder. As can be seen on the picture, this zone has the same vertical symmetry axis as the couple of intruders. We observe that this intersection defect zone, approximately lying at  $\pi/4$  to the horizontal, is largely depleted in comparison with the regular stacking of the triangular lattice. Considering that the existence of a depleted cloud of defects in the bulk of the lattice results in a potential energy increase, one can explain simply the preceding observations related to the potential energy dependence on separation  $D$ . For  $0 \leq D \leq 0.7R$ : the intersecting cloud (the depleted zone) increases in size when the separation  $D$  increases. This results in a fast increase in the potential energy and, correlatively, in an attractive force. For  $0.7R \leq D \leq \infty$ : The depleted zone surface remains constant. It climbs up in the bulk as the distance is increased. This results in a slight decrease of the potential energy and, correlatively, in a weak repulsive force.

The extension of this reasoning to 3D is not a trivial task as the bulk, which is in this case a "frustrated" packing of small spheres, is disordered. Therefore, we have decided to perform numerical simulations to directly test the possible existence of such an interaction. Our calculation is a straightforward extension to two large spheres of the one previously done with a single sphere imbedded in a packing of small ones [5]. A large number  $N$  of spheres are deposited one after the other on a square base of size  $L \times L$ . All the small spheres, of radius  $r$ , are deposited along randomly chosen vertical trajectories, while the two large ones, of radius  $R = \Phi \times r$ , which are the  $n$ th and  $(n + 1)$ th deposited spheres, have their trajectories located at  $L/2 \pm [R + D(0)/2]$ , where  $D(0)$  is the initial desired distance. The number  $n$  is a sufficiently large fraction of  $N$  in order to ensure that both large spheres will be totally immersed in the small spheres packing, without being disturbed by the base plane.

Each further sphere follows the path of steepest descent on the previously deposited packing before the next one is deposited. Periodic boundary conditions are used in both lateral directions during this procedure. Because of the roughness of the packing surface, both large spheres do not necessarily reach at rest a mutual distance exactly equal to  $D(0)$ . If this distance differs from  $D(0)$  by more than  $|0.2 \times R|$ , the packing is discarded and a new packing is built. After the  $N$  spheres have been deposited, the shaking procedure is performed as in [5]: all the particles are placed in a list in order of ascending height (of their centers) and then redeposited in that order retaining their previous horizontal coordinates and keeping to the steepest

descent rules. The process of ordering according to height and redeposition is repeated several times. Here, the time  $t$  is simply the number of shakes, running up to a maximum number  $t_M$ . After each shake, the distance  $D(t)$  between the two large spheres is computed (this distance is defined as above as the difference between the center-to-center distance and the sphere diameter).

As an example of numerical result, we have reported in Fig. 3 the mean curve  $\langle D(t) \rangle$  resulting from an average over 20 independent runs, for  $\Phi = 5$  and  $D(0) = 0.6 \times R$ . The number of particles  $N$  has been chosen large enough so that the large spheres have not reached the top after  $t_M = 40$  shakes. Even with such a wide sampling, the mean curve  $\langle D(t) \rangle$  is quite noisy. This comes from the fact that the individual trajectories  $D(t)$  differ significantly from each other because they are subject to a strong random noise due to the stochastic character of the shaking procedure. This is illustrated in the inset of Fig. 3, where two typical individual  $D(t)$  curves are shown. As it can be seen in this inset, sometimes, after being in contact [i.e.,  $D(t)$  staying equal to zero for a while], the large spheres can separate again. However, when averaging over a large enough number of independent runs, one obtains a *net decrease of the averaged distance*  $\langle D(t) \rangle$  with time. Therefore, in addition to an unavoidable random force of zero mean, there definitively exists an attractive force between the large particles.

In order to investigate the dependence of this force on distance, we have compared the averaged curves  $\langle D(t) \rangle$

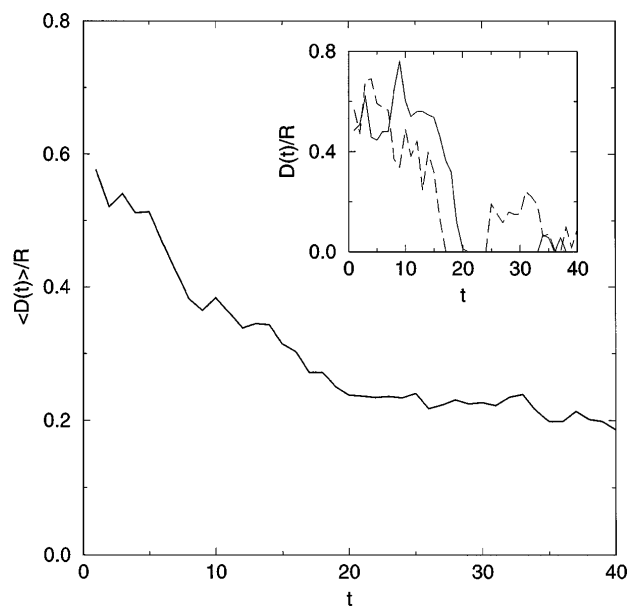


FIG. 3. Time dependence of the separation distance between two large spheres imbedded into a shaken packing of small spheres. The parameters of the simulation are  $\Phi = R/r = 5$ ,  $L = 16 \times R$ ,  $N = 64\,000$ ,  $n = 8000$ ,  $t_M = 40$  and  $D(0)/R = 0.6$ . The main curve  $\langle D(t) \rangle$  corresponds to an average over 20 independent runs. Two particular examples of  $D(t)$  trajectories are shown in the inset.

for different  $D(0)$  values in Fig. 4. As in 2D, the attractive force decays with distance, but vanishes at a larger distance, namely for  $D$  of order  $1.4 \times R$ . A more precise determination of this characteristic distance would necessitate much more computer time to reduce the noise in the  $D(t)$  curves (such computing effort will be performed for a forthcoming publication). Note the existence of a minimum in the  $\langle D(t) \rangle$  curve clearly visible for  $D(0) = 0.4 \times R$ . This minimum is the consequence of possible detachments due to the random noise. We have also studied the influence of varying the size ratio  $\Phi$ . We have found that, despite randomness, the averaged  $\langle D(t) \rangle$  curves, when the distance is counted in unit of  $R$ , are almost  $\Phi$  independent. Note that the two potential energy curves of Fig. 2 becomes also almost superimposed when  $D$  is divided by  $R$ . This means that the linear size of the depletion zone is proportional to  $R$  in both two and three dimensions. Other quantities of interest have been calculated, providing some more details about the ascending and relative motions of the large balls, and they will be reported elsewhere. Let us just mention that, when the two large spheres are very close to each other, they rotate during their upward motion to progressively reach a position in which one sits on the top of the other.

In conclusion, the analysis of the potential energy dependence on the separation between two large intruders in a sea of small granulates in two dimensions allows one to predict and understand the existence of a significant attractive force between neighboring large particles. This sort of steric effect, resulting from the interference between topological perturbations induced by intruders

in a sea of smaller particles may be reminiscent of the well studied problem of polymeric stabilization of (free) polymeric suspensions (see, e.g., [11]). First, the potential energy dependence on distance is similar in both cases. Second, and as in the present case, the attractive force between colloids results from the formation of a depletion zone between neighboring surfaces of colloids and the repulsive interaction results from the restoration of the normal distribution of the free polymers in the solution when the intercolloids distance is increased.

Our 3D simulation effectively demonstrates the existence of an efficient attractive force. We find that it decays with distance as in 2D. Even if the small sphere packing is here intrinsically disordered, due to geometrical frustration, one may, however, invoke the existence of a depletion zone, which might be due to boundary effects in the region where the small sphere packing is within the close neighborhood of the large sphere surfaces. We were not able to detect any repulsive force at large distances in our 3D simulations. The existence of this very weak repulsive force might be peculiar of 2D geometry where the depletion zone may extend up to the surface, due to the regular character of triangular geometry of the small spheres packing.

It might be worth checking these results by performing real shaking experiments. They are underway in our laboratory. We note that much care must be taken in order to ensure that spurious internal convection due to side walls effects would not perturb the experimental results.

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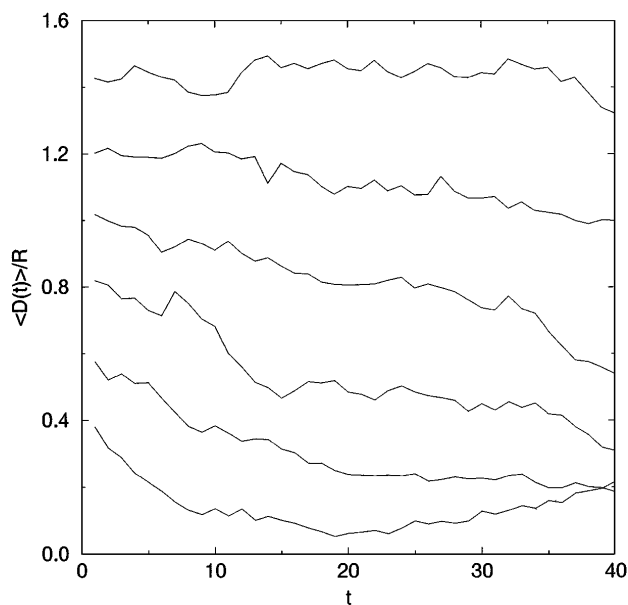


FIG. 4. Time dependence of the mean separation distance for different initial distances. The parameters of the simulation are the same as in the preceding figure except for  $D(0)/R$  which is equal to 0.4, 0.6, 0.8, 1.0, 1.2, and 1.4, from bottom to top.

- [1] A. D. Rosato, T. Vreeland, Jr., and F. B. Prinz, *Int. Mater. Rev.* **36**, 45 (1991).
- [2] J. A. C. Gallas, H. J. Herrmann, T. Pöschel, and S. Sokolowski, *J. Stat. Phys.* **82**, 443 (1996).
- [3] G. C. Barker, A. Mehta, and M. J. Grimson, *Phys. Rev. Lett.* **70**, 2195 (1993).
- [4] A. D. Rosato, K. J. Strandburg, F. Prinz, and R. H. Swendsen, *Phys. Rev. Lett.* **58**, 1038 (1987).
- [5] R. Jullien, P. Meakin, and A. Pavlovitch, *Phys. Rev. Lett.* **69**, 640 (1992).
- [6] R. Jullien, P. Meakin, and A. Pavlovitch, *Europhys. Lett.* **22**, 523 (1993).
- [7] J. Duran, J. Rajchenbach, and E. Clément, *Phys. Rev. Lett.* **70**, 2431 (1993).
- [8] J. Duran, T. Mazozi, E. Clément, and J. Rajchenbach, *Phys. Rev. E* **50**, 5138 (1994).
- [9] J. B. Knight, H. M. Jaeger, and S. R. Nagel, *Phys. Rev. Lett.* **70**, 3728 (1993).
- [10] R. Jullien, P. Meakin, and A. Pavlovitch, *Phys. Rev. Lett.* **70**, 2195 (1993).
- [11] D. H. Napper, *Polymeric Stabilization of Colloidal Dispersions* (Academic, London, 1983).