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# Frustrated packing of spheres in a flat container under symmetry-breaking bias

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We study statistical properties of packings of monodisperse spheres in a flat box. After "gravitational" filling and appropriate agitation, a nearly regular (in plane) but frustrated (normal to the plane) triangular lattice forms, where beads at individual sites touch either the front or back wall. It has striking analogies to order in antiferromagnetic Ising spin models. When tilting the container, Earth's gravitational field mimics external forces similar to magnetic fields in the spin systems. While packings in vertical containers adopt a frustrated state with statistical correlations between neighboring sites, the configurations continuously approach the predictions of a random Ising model when the cell tilt is increased. Our experiments offer insights into both the influence of geometrical constraints on random granular packing and a descriptive example of frustrated ordering.

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Optimal regular packing of monodisperse spheres in three dimensions (3D) has been a long-standing problem in fundamental and applied sciences, with many practical implications. Already Kepler in 1611 [1] made a conjecture that equally sized spheres cannot be packed with greater average density than that of the hexagonal close packing or face centered cubic close packing. This was confirmed for regular lattices by Gauss [2], but a general proof was not given until recently [3]. Packing of spheres in 3D necessarily involves geometrical frustration: The locally optimal tetrahedral packing is not space-filling. In contrast, the close-packed equilateral triangular lattice optimizes the packing of disks in a two-dimensional (2D) plane both globally and locally. Confinement adds numerous complications [4–8]. Optimal regular solutions are known only for few special cases, e.g., for cylindrical containers [9–11].

The crossover from 2D to 3D structures is of particular interest: In a rectangular container of little more than a particle diameter's height, beads have the freedom to leave the plane triangular lattice in the out-of-plane direction. In thin colloidal layers, a series of alternating morphological arrangements with hexagonal and square symmetries has been described [12–15]. Buckling and prismlike structures smoothen these transitions [11,16–18]. Numerical simulations and (Ising) lattice models achieved progress in their understanding [19–27], a general mathematical proof of the optimal packing in such geometries lacks. The formation of crystallized domains was also obtained in vibrated granular layers: Parameters are the filling fraction, the gap width, and appropriate excitation [28,29].

However, in practical situations, granular ensembles do not usually reach the regular ("crystalline") ground states. One often deals with stable random packings [30,31] not exceeding volume fractions of $\approx 64\%$. Many aspects of such ubiquitously encountered states are still insufficiently understood [32–34].

The geometry investigated here is a thin cuboid container (gap width $D$ larger than the sphere diameter $d$, but $< 1.5d$). When filled by pouring in monodisperse spheres ("gravitational" filling), regions with nearly regular triangular lattices form, separated by grain boundaries (Fig. 1). Each bead touches either the front or rear wall of the container. Packing of neighboring spheres is *locally* optimized when they are attached to opposite container walls, but this cannot be fulfilled by each sphere in a triangle: While two spheres may occupy opposite sides, the third sphere must share the same side with one of the first two. This generates geometric frustration and disorder in the regular lattice, rendering our system similar to other frustrated systems [35–44]. Structural analogies to the antiferromagnetic Ising model on a triangular lattice [45] are obvious. Its ground state is highly degenerate, i.e., there are numerous constellations characterized by the same fraction of frustrated contacts.

This analogy was already suggested for colloidal systems in horizontal layers [15,35]: Optimal packing is achieved for (kinked) rows of beads alternatingly at the front and back walls, so that each site has exactly $F = 2$ frustrated bonds. The mean number of frustrations per site for uncorrelated particle positions is $\langle F \rangle = 3$. Han *et al.* [35] found $2.1 < \langle F \rangle < 2.5$ experimentally. Corresponding results were obtained for an Ising model on a deformable lattice [27].

In these suspensions, gravity can in general be neglected, while it is essential in our granular experiment. Colloids undergo thermal motion, whereas in the granular system, there is no particle dynamics unless the system is agitated.

We employ gravity as an external bias, comparable to a magnetic field in spin systems. This alters the respective occupation numbers of rear and front positions, and consequently the statistics of the packing.

We perform the experiments in a 2.9-mm-thick cell made out of two 17 cm × 26 cm acrylic glass plates. A Canon EOS 550D camera is mounted 40 cm from the cell, and pointed at the cell center. The whole setup can be tilted by defined angles $\theta$. Monodisperse spherical glass beads of $2.126 \pm 0.072$ mm diameter are filled into the cell from the top. The beads are immersed in drilling emulsion. Due to scattering of the emulsion, beads can be distinguished unambiguously: Those touching the front plate appear darker than those at the rear plate [Fig. 1(b)]. From the images, we detect all beads and their respective positions [Fig. 1(c)].

Initially, the beads deposit in a disordered packing where only about 65% of them have six nearest neighbors. Then,

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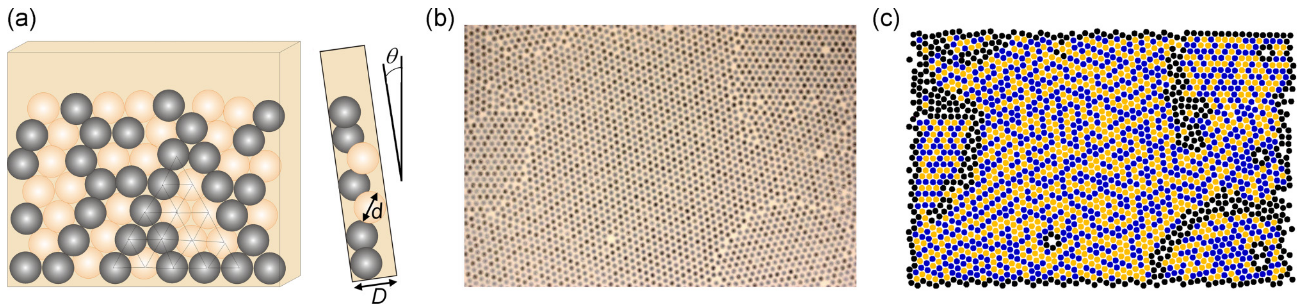


FIG. 1. (Color online) (a) Sketch of the experimental geometry seen from the front and side, arrangement of the spheres in the container. The whole setup can be tilted by angles $0 \leqslant \theta \leqslant 60^{\circ}$. (b) Typical photograph of the evaluation region after annealing: The front beads appear darker than those on the rear side. Some grain boundaries and lattice defects are still present. (c) Processed image: Beads with less than six neighbors at grain boundaries, defects, or cell boundaries (black), front [blue (dark-gray)], and rear [orange (light-gray)] beads with six neighbors.

we agitate the cell by tapping the container bottom to achieve compaction [46]. We find that the statistical properties analyzed in the following remain unchanged after $\approx$50 taps, except for the fraction of regular lattice sites [beads with six neighbors; Fig. 2(a)], which still increases slightly. The average volume fraction reaches $\phi \approx 0.45$. In order to ensure that an asymptotic statistics is established, we perform at least 100 taps in each preparation. Then, a picture of the cell is taken and evaluated by image processing: First, we identify all beads with six neighbors (regular lattice points), and discard all others, i.e., beads at defects and grain boundaries (about 15%) or at the image boundaries (10%). More than 1600 regular sites remain in each image. In order to obtain reasonable statistics, we average ten preparations under identical conditions in each experiment.

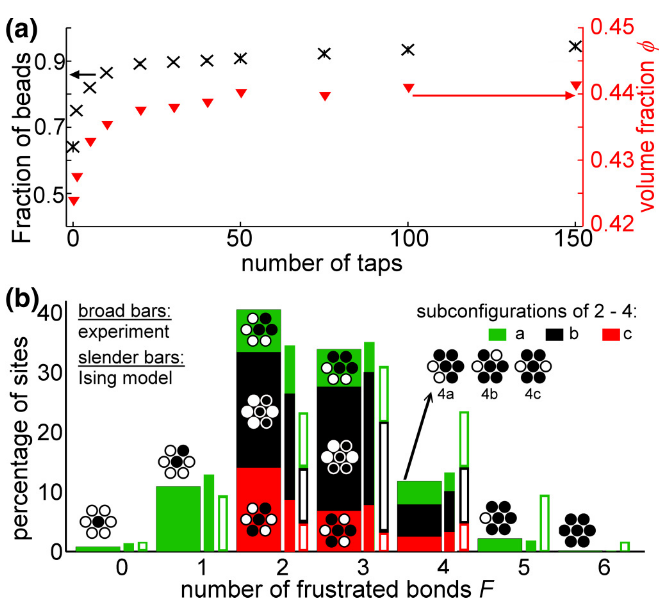


FIG. 2. (Color online) (a) Evolution of the statistics with the number of taps: volume fraction $\phi$ (triangles), fraction of beads with six nearest neighbors (crosses). (b) Distribution of the local geometrically frustrated (sub-)configurations at sites with six neighbors in an experiment for zero cell tilt ($\theta = 0$) after annealing, and comparison to the predictions of noninteracting (empty bars) and nearest-neighbor (filled bars) Ising models.

Front and back sides are equivalent for the nontilted cell, thus both layers should be equally populated on average. For individual regular sites, 13 possible local configurations can be distinguished [35], characterized by the number of frustrated bonds $F = 0 \ldots 6$ and their relative positions (subconfigurations a–c). We measure a mean number of frustrations per site of $\langle F \rangle = 2.5$ owing to local correlations of the states of neighboring sites. A ground state of minimal frustration would correspond to $F = 2$, e.g., spheres in the bottom layer chained on one side of the container and the subsequent layers alternating at rear and front sides. With respect to the degree of frustration, all zigzag arrangements [35] are equivalent to that packing configuration. In contrast to colloids, our grains are not affected by thermal fluctuations, and reaching one of these ground states is highly unlikely even after extensive agitation (comparable to spontaneous crystallization of spheres in 3D [47]). The average size of striped domains (configurations 2b or 2c) is only six sites.

Assuming that the positions of neighboring beads are uncorrelated, a noninteracting Ising model is a first approximation to predict the frustration statistics. For example, it yields the the probability $p(F) = \binom{6}{F}$ of configurations with $F$ frustrations. Compared to this prediction, sites with two frustrated bonds are clearly overrepresented in the experiment [see Fig. 2(b)]. Those with four to six frustrated bonds are underrepresented. The measured ratios of subconfigurations also deviate. An improved model includes interactions between nearest neighbors. We performed Monte Carlo simulations to obtain the configuration statistics. Each frustrated bond is penalized with a statistical weight $\exp(-\beta)$. Agreement of $\langle F \rangle$ with the experiment is obtained for $\beta = 0.475$ [see Fig. 2(b)]. The model reproduces the experimental ratios of the (sub-)configurations reasonably well. Deviations, e.g., for configurations 2c and 3c, indicate that this model with only nearest-neighbor interactions is somewhat too simple.

We now extend the analysis to the effects of an "external field" bias on the configuration statistics. When the cell plane forms an angle with the vertical, a relocation of beads from the front to the rear position changes their potential energy. In our notation, beads at the back plate of the tilted cell are energetically favored.

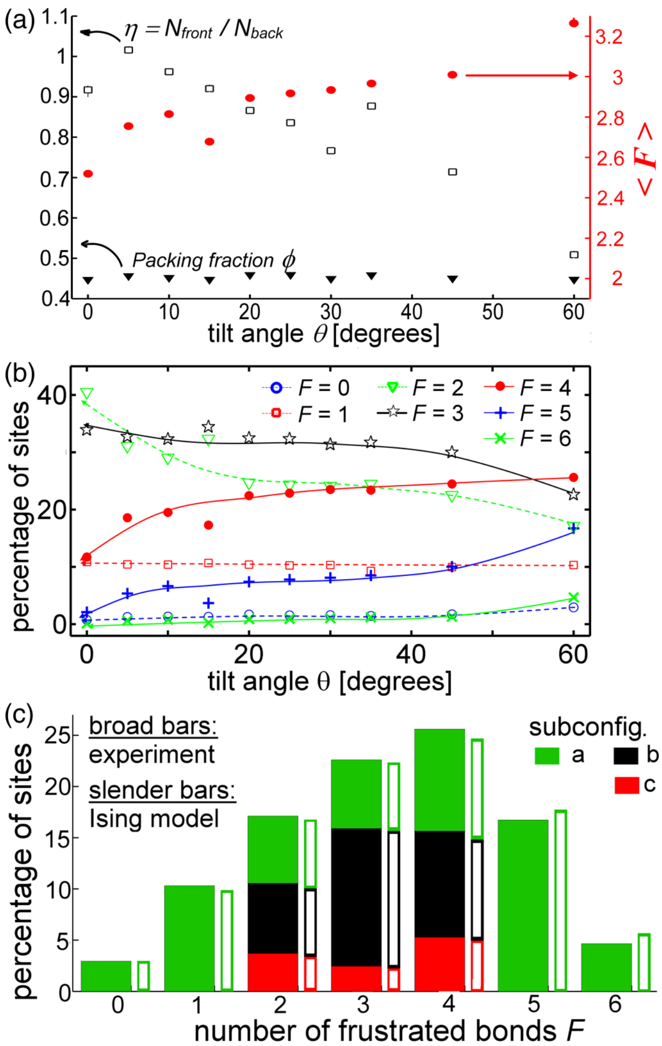


FIG. 3. (Color online) (a) Volume fraction $\phi$ (triangles), ratio of beads at the front (i.e., upper) cell wall $N_{\text{front}}/N_{\text{back}}$ (squares), and average number of frustrations per site with six neighbors (dots) in dependence on the cell tilt $\theta$. (b) Percentage of sites in the different configurations as a function of $\theta$; lines visualize the trends. (c) Distribution of the local frustration states [cf. Fig. 2(b)] for the $\theta = 60^\circ$ tilted cell, and corresponding predictions of the noninteracting Ising model [Eq. (2)]. Error bars in (b) are smaller than the symbol size.

As expected, the preparation by gravitational filling and tapping of the tilted cell does not produce a regular ground state but frustrated triangular lattices again. We find no effects of the cell tilt on the density of lattice defects. However, we observe a systematic shift of the occupation ratio of front to back positions, $\eta = N_{\text{front}}/N_{\text{back}}$ [see Fig. 3(a)]. At our maximum tilt angle of 60°, the ratio is $\eta \approx 0.5$. With the gradual decrease of $\eta$, the average number of frustrations of sites at the less populated front wall decreases systematically, while it increases at the rear. The average number of frustrations reaches $\langle F \rangle = 3.26$ for $\theta = 60^\circ$ [Fig. 3(a)]. This is connected with a significant change of the distribution $p(F)$ of configurations [Fig. 3(b)]: The main trend towards higher frustration is accomplished by a strong decrease of the fractions of sites in configurations 2 (and partly 3), while the fractions of sites in configurations 4 and 5 increase to more than twice their values at zero tilt. For our maximum tilt, $\theta = 60^\circ$, the distribution $p(F)$ is shown exemplarily in Fig. 3(c). The considerable increase of bond frustration, compared to Fig. 2(b) for $\theta = 0$, is evident. Assuming that particle positions are uncorrelated, we can model the effect of the tilt with the noninteracting Ising model, allowing for a variable occupation ratio $\eta$: The probability for a particle in front to have $F$ frustrations is then given by

$$p_{\text{front}}(F) = \binom{6}{F} \left(\frac{\eta}{1+\eta}\right)^F \left(\frac{1}{1+\eta}\right)^{(6-F)} \tag{1}$$

and correspondingly obtained for the rear layer. Then, the average number of frustrated bonds is

$$\langle F \rangle = \sum_{F=0}^{6} F \left[\frac{\eta}{1+\eta} p_{\text{front}}(F) + \frac{1}{1+\eta} p_{\text{back}}(F)\right]. \tag{2}$$

A comparison of this model's predictions with the experimental data at $\theta = 60^\circ$, including the sub-configurations, yields astonishing agreement [see Fig. 3(c)]. Not only does the bond frustration increase with increasing tilt, the packing also becomes more random. This coincides with an observation of details in individual runs of the experiment: regions with chain patterns disappear

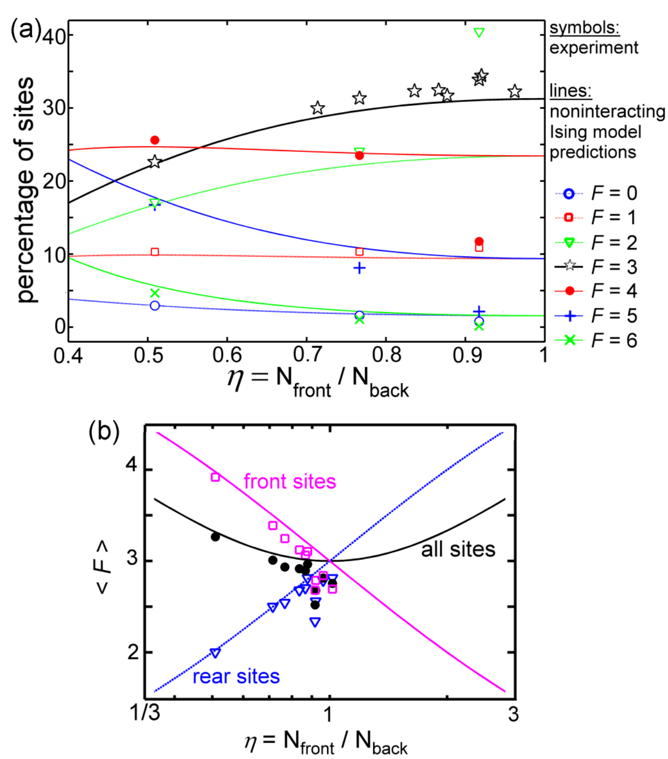


FIG. 4. (Color online) (a) Predictions of the noninteracting Ising model for the probabilities of configurations (lines) and corresponding experimental data in dependence on the fraction of front beads [cf. Fig. 3(a)]. In order to keep the plot simple, the complete configurations are shown only exemplarily for $\theta = \{0^\circ, 30^\circ, 60^\circ\}$. For configuration 3, all tilt angles are included. (b) Average numbers of frustrations predicted by the noninteracting Ising model for the front, rear, and all sites compared to experimental data (symbols).

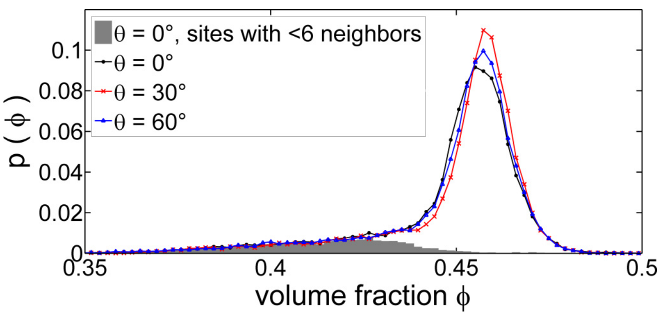


FIG. 5. (Color online) Distribution of the local volume fraction for the untilted, 30° and 60° tilted cells. The contribution of lattice defects is shown in gray shade for $\theta = 0^\circ$. No systematic changes are observable.

completely. Since this analytical approximation already reproduces the experimental distribution satisfactorily, we omitted the MC result, which is practically the same.

Figure 4(a) shows the predictions of the noninteracting Ising model (solid lines) for the configurations with zero to six frustrations as a function of the occupation ratio $\eta$. For clarity, the complete experimental data are shown only for $p(F = 3)$; for the remaining configurations we restrict to tilt angles $\theta = \{0^\circ, 30^\circ, 60^\circ\}$. The experimental trends follow the predictions of the noninteracting Ising model for all configurations. Increasing tilt improves the agreement with the model considerably. This is also reflected in an overview plot of $\langle F \rangle$ vs $\eta$ for both categories of beads [see Fig. 4(b)]. When we use the interaction parameter $\beta$ in the model with interactions to fit the experimental $\langle F \rangle$, we find that $\beta$ systematically decays with decreasing $\eta$ (increasing tilt $\theta$). The model with uncorrelated sites becomes a good approximation at large tilt.

In contrast to the evident consequences of a container tilt on the frustration statistics, the global packing fraction is hardly influenced [Fig. 3(a)]. Even the distributions of local packing fractions, exemplarily shown for $\theta = \{0^\circ, 30^\circ, 60^\circ\}$ in Fig. 5, are practically unaffected. An explanation can be given intuitively: Owing to the jammed character of the packing, the system does not explore the full configuration space and thus does not converge to a densest packed state (which has $\phi \approx 0.488$, few percent above the experimental average value). A perfect hexagonal lattice with all beads on the same side (90° tilt) yields $\phi = 0.443$.

We do not find any trend in frustration numbers or other statistical features along the vertical. This is a direct consequence of the fact that in confined packings, pressure saturates a few layers below the surface and forces are transmitted to the container walls (Janssen's law). The randomness in the local configurations does not significantly affect the volume fraction, neither locally nor globally.

Changing the ratio $d/D$ influences the quantitative characteristics, but not the qualitative results as long as the ground state is a triangular lattice. For smaller $d/D$, where the ground state becomes a square lattice [12–14], frustration is lifted: Any two neighboring beads can occupy positions on opposite sides of the cell.

Summarizing, in vertical cells we find results similar to colloids [35]. The ground state with minimal frustration is not reached, the mean number of frustrations per site levels at 2.5 for the given $d/D$. Tilting the cell respective to the gravitation field breaks the symmetry of back and front planes. In that respect, it can be considered as an external bias acting like a magnetic field on spin states in classical Ising antiferromagnets. Nevertheless, the situation is more complex: While in spin systems, interactions with the field are local and the potential is identical for all spins, this is at first glance not the case for the present system. The reason that we do not find long-range effects of the rearrangement of individual sites and that spheres appear to interact individually with the external bias is the character of force propagation in a granular pile: Force chains redirect most of the weight to the vertical walls.

Even though many statistical features seem to be understood, we list some open issues: First, there is no obvious explanation how the occupancy ratio $\eta$ relates to the tilt angle, only the limits (1 for the upright and 0 for the horizontal cell) are evident. This occupancy is a crucial parameter for all kinds of transport characteristics (heat, electric current, etc.) between the two plates of such a cell. Likewise, the interpretation of $\beta$ is not straightforward. Second, the influence of friction has not been considered. Our beads are frictional; this is crucial for the stability of the packings. Third, the influence of a small polydispersity could have larger effects than in bulk samples due to the dependence of local packing densities on $d/D$. Our descriptive experiment demonstrates how confinement can lead to the emergence of new aspects in packings of granular matter.

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