Experimental Study of Ordering of Hard Cubes by Shearing

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We experimentally analyze the compaction dynamics of an ensemble of cubic particles submitted to a novel type of excitation. Instead of the standard tapping procedure used in granular materials we apply alternative twists to the cylindrical container. Under this agitation, the development of shear forces among the different layers of cubes leads to particle alignment. As a result, the packing fraction grows monotonically with the number of twists. If the intensity of the excitations is sufficiently large, an ordered final state is reached where the volume fraction is the densest possible compatible with the boundary condition. This ordered final state resembles the tetratic or cubatic phases observed in colloids.

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Granular compaction is considered a canonical example of the inherent complexity related to the equilibrium states displayed by an ensemble of macroscopic grains. Important efforts have been performed to describe these systems inspired on the ideas of classical statistical mechanics [1], but adapted to account for the fact that granular materials are basically "athermal" [2]. Accordingly, ensembles of different mechanically stable arrangements allow calculating stationary averages—like the volume fraction—that can be used as state variables in a hypothetical phase space. Nevertheless, due to the dissipative interactions among grains, these states are closely linked with the excitation mechanisms used to explore their energy landscape [3]. Recently, it has been demonstrated that the shape of the particles is crucial on determining the macroscopic mechanical properties of the ensemble [4]. Moreover, the intrinsic arrangements associated with particle shapes has permitted us to test ideas related to entropy and order [5]. Then, concepts like optimal packing or ordering have been analyzed to unveil the role of the particles' morphology on the geometrical configurations that can be achieved [6–10]. Of note, most of these works are focused in thermal systems such as colloids, and only a few explore the granular limit. For that case, it is known that faceted particles in 2D systems tend to align leading to highly ordered structures [11–14]. Furthermore, 3D tapping experiments of tetrahedral dice were implemented and the different jammed configurations reached were analyzed in terms of the type and number of contacts between particles [15,16]. In this work, we study the packing evolution of an ensemble of cubic particles when submitted to an external perturbation. Different from the standard procedure used to compact grains (where tapping plays against gravity [17]), we apply alternating rotations, or "twists," on the container. This type of excitation leads to particle reorganization by shear, a method highly effective to align both elongated and platy particles [18–21]. After a series of twists, the system reaches the densest limit where concentric rings of horizontally aligned cubes are superimposed in the vertical direction (Fig. 1). The experiment has been performed with cubic particles, which are commercially available as plastic dice. These have regular sides of length $l = 0.50 \pm 0.025$ cm with rounded corners of radius $r \cong 0.068$ cm [22] and are rather monodisperse in mass (0.116 ± 0.001 g). The dice are poured in a cylinder of radius $R = 8.7 \pm 0.01$ cm using a standard protocol that allows obtaining reproducible initial volume fractions. After this, the whole cell is rotated in alternating angular directions (Fig. 1), a kind of forcing that we will name "twist" from now on. Of note, the system is only perturbed when the angular velocity is inverted and the cubes remain at rest during the rest of the rotation cycle. Therefore, we use as the control parameter the cell tangential acceleration $\alpha = 2\mathrm{v}/\Delta t$, where v is the tangential velocity of the container and Δt is the time necessary to invert the turning sense. Just for comparison with the tapping experiments we normalize this magnitude with gravity, $\Gamma = \alpha/g$. Importantly, the range of Γ explored in these twist experiments ($0.3 < \Gamma < 1.5$) is signally lower

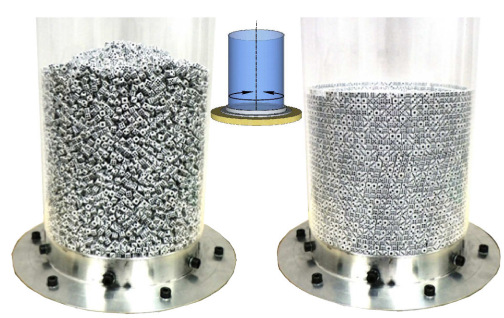

FIG. 1. Experimental cell and sketch of the twist excitation. The pictures show the initial (left) and final (right) states for a ensemble of 25 000 cubes submitted to $N = 3 \times 10^5$ twists of intensity $\Gamma = 1.01$.

than the one used in tapping. To test the influence of the cell radius on the packing process we have also implemented a half radius cell where, qualitatively, the same dynamics was observed. More details about the experimental setup and measuring protocol can be found in Ref. [23]. We start by presenting results of the temporal evolution of the packing fraction ϕ for different values of Γ [Fig. 2(b)]. ϕ is calculated as the total volume of dice normalized with the volume of a cylinder whose height is the mean value of the free surface position as indicated in Fig. 2(a). The data are averages of five repetitions in the same conditions. Clearly, the packing fraction evolution depends on the excitation intensity. For $\Gamma > 0.5$, ϕ grows logarithmically until it saturates at the densest packing possible compatible with the boundary conditions. Indeed, although the rigorous calculation of such packing is a stiff optimization problem [24], a visual inspection of the setup confirms that the dice arrange in concentric rings as shown in Fig. 3. The emergence of these arrangements was confirmed by registering the free surface along the complete sequence of twists, and also by manually "peeling" the upper layers of the column at its final state to check the order degree inside the bulk (see Ref. [23]). For $\Gamma < 0.5$, the volume fraction growth is much slower than for higher excitation intensities. It is difficult to elucidate whether the densest state will be reached for these low excitation intensities but, in the affirmative case, a linear extrapolation of the observed tendency suggests that ten years of twisting will be necessary to reach such a hypothetical configuration. Moreover, the images of the free surface shown in Fig. 3(a) evidence that the central region remains disordered or with domains of cubes aligned with one of its diagonals (between opposite corners) with gravity. Incidentally, this orientation is analogous to the one obtained when filling a silo [11]. Interestingly, for intermediate Γ [$\Gamma = 0.52$ in Fig. 2(b)] the packing fraction evolution shows slightly distinctive features: it is slow at the initial stages and, then, it suddenly jumps to what it seems is a stationary state whose density is below the highest possible. This feature correlates with the development of very stable dice clusters aligned at different directions (also seen in tapping simulations of spheres [25]) that seem to be behind a reduction in the repeatability of the packing evolution among the different runs. In order to further investigate this effect, we now focus in the compaction dynamics of the external layer of dice (the one in contact with the cylinder wall). We define the lateral volume fraction φ as the number of cubes that have any face parallel to the container wall, normalized by the maximum number that allows the geometrical restriction imposed by the circular boundary conditions (see Ref. [23]). The temporal evolution of φ is reported in Fig. 2(c). Clearly, the larger the twist acceleration the quicker the increase of the lateral volume fraction. Although the evolution of φ with N is similar to the one reported in tapping experiments [26], we have checked that

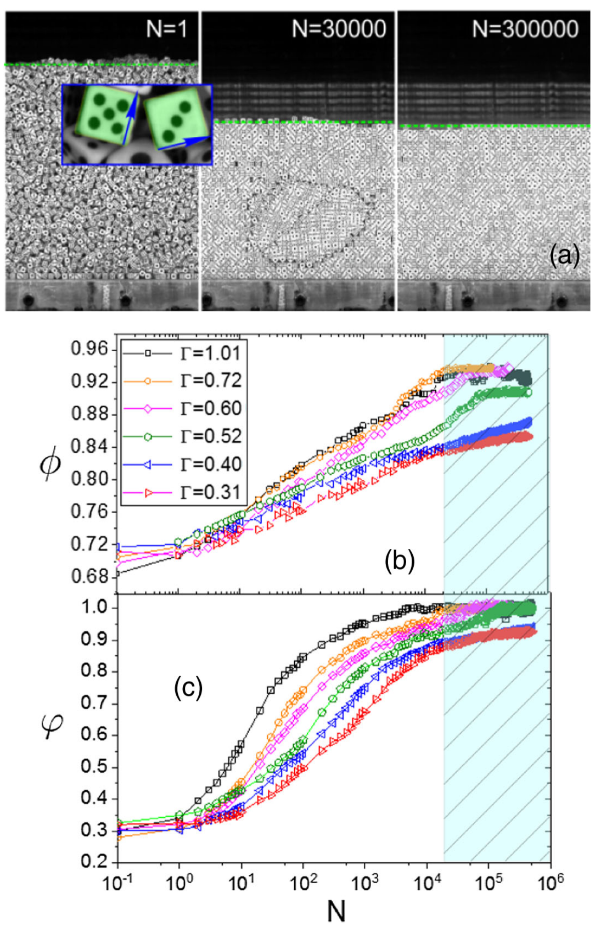

FIG. 2. (a) Lateral view of the cell after different numbers of twists N. The green dotted lines indicate the mean height used to calculate the packing fraction ϕ. In the inset, the green shadows correspond to the area used to calculate the lateral volume fraction φ, and the arrows define the dice orientation with respect to the horizontal $\mathbf{u}_i$. (b) Packing fraction ϕ versus N in logarithmic scale for various Γ as indicated in the legend. (c) Evolution of the lateral volume fraction φ as a function of N for the same values of Γ.

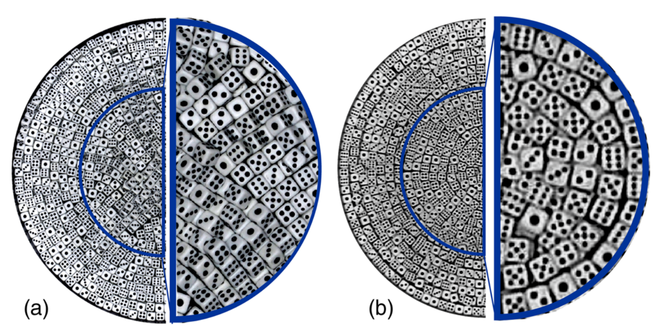

FIG. 3. Free surface after $N = 1 \times 10^5$ twists for (a) $\Gamma = 0.31$ and (b) $\Gamma = 1.01$. The central region has been amplified to remark the difference between both final states. More detailed images can be found in Ref. [23].

an inverse logarithmic correlation does not adequately fit the data. A rapid growth in φ is observed at the beginning that is caused by the movement of cubes in the radial direction and their alignment with the container wall. After that, the lateral packing fraction changes its tendency as shown in the shadowed region of Fig. 2(c). This change is related to the development and competition among ordered domains of cubes. Notably, two dice orientations are dominant: with sides parallel to the horizontal direction or with the diagonal aligned with gravity [central snapshot in Fig. 2(a)]. Similar configurations have been previously reported in bidimensional deposits of square particles [18].

In order to shed light on the cluster formation process we have looked at the evolution of the dice orientation. This is characterized by means of a unitary vector $\mathbf{u}_i$ [see the inset of Fig. 2(a)] whose orientation is measured with respect to the horizontal (note that $\theta = 0$ or $\theta = \pi/2$ for a horizontally aligned dice and $\theta = \pi/4$ for an inclined one). The evolution of the angular orientation distribution $pdf(\theta)$ with the number of taps is presented in Figs. 4(a) and 4(b) for two twist amplitudes. For each case, the analysis was performed in three different regions of the cell (top, middle, and bottom), all of equal height. This is done in order to explore the possible effect of the bottom and free surface in the ordering process. For the lowest intensity [$\Gamma = 0.31$, Fig. 4(a)], the *pdf*'s are rather flat and some dependence on the height is observed. In the bottom region the system evolves to a situation where horizontal and diagonal alignment coexist (see the peaks at $\theta = \pi/4$ and $\theta = \pi/2$), in the middle region only a small peak at $\theta = \pi/4$ can be distinguished, and in the top, after a transitory situation for $N \approx 10^4$ with horizontal and diagonal alignment, the system ends up with most of the dice aligned horizontally. For the highest twist amplitude [$\Gamma = 1.01$, Fig. 4(b)] the behavior is completely different as, independently on the explored region, a quick growth of the number of particles aligned horizontally is observed.

Complementarily, we characterize the global orientational order in the external layer of dice by means of the cubatic order parameter S_4. This was originally introduced to describe colloidal superballs [27,28] and is defined as $S_4 = (1/8N)\sum_i(35|\mathbf{u}_i\cdot\mathbf{n}|^4 - 30|\mathbf{u}_i\cdot\mathbf{n}|^2 + 3)$, where we consider $\mathbf{n}$ as the set of unitary vectors aligned with the experimental setup axis. $S_4 = 0$ means no cubatic order and $S_4 = 1$ corresponds to a perfect ordering. This calculation is done over the whole external layer without separating the different height regions. In Fig. 4(c) the evolution of S_4 is represented for three values of Γ. In all cases, S_4 remains almost zero during a certain number of twists N_c, a behavior that is associated with the increment of randomly aligned particles touching the lateral wall. For high and intermediate intensities, S_4 rises after N_c as the cubes start to get aligned to each other. Both, N_c and the growth rate of S_4 depend on the tapping intensity: N_c is reduced and the growth rate is increased when augmenting Γ. At the final stages of the S_4 evolution (i.e., when S_4 approaches 1) the magnitude becomes very fluctuating. This is attributed to the development of small clusters aligned at different directions that finally disappear when the system becomes completely ordered. Fluctuations are specially visible for $\Gamma = 0.52$ suggesting the competition among clusters of dice aligned horizontally and with the diagonal in the direction of gravity. Finally, let us remark that, for the lowest Γ, there are no signs of global orientational order even after $N = 5\times10^5$, strengthening the idea that this intensity is not high enough to induce particle alignment. Finally, in Fig. 4(d) the S_4 evolution is displayed against φ revealing that a packing fraction sufficiently large ($\varphi > 0.8$) is necessary to observe global orientational order. In this figure we also notice that, for the lowest Γ, lateral volume fractions above $\varphi = 0.9$ are reached without signs of global order.

In order to delve deeper into this curious effect, we calculate the spatial orientational correlation function [27] $G_4(r) = (1/8N)\langle[35|\cos(\Delta\theta_{i,j})|^4 - 30|\cos(\Delta\theta_{i,j})|^2 + 3]\rangle$, where $\Delta\theta_{i,j} = \theta_i(0) - \theta_j(r)$ quantifies the alignment between two particles whose centers are separated by a distance equal to r. The results obtained for two different values of Γ after applying different numbers of twists are reported in Fig. 5(a). For high twist intensities the dice orientation becomes rapidly correlated (see the curve for $N = 5\times10^4$ when $\Gamma = 1.01$), and finally reaches a globally aligned state (see the curve for $N = 5\times10^5$).

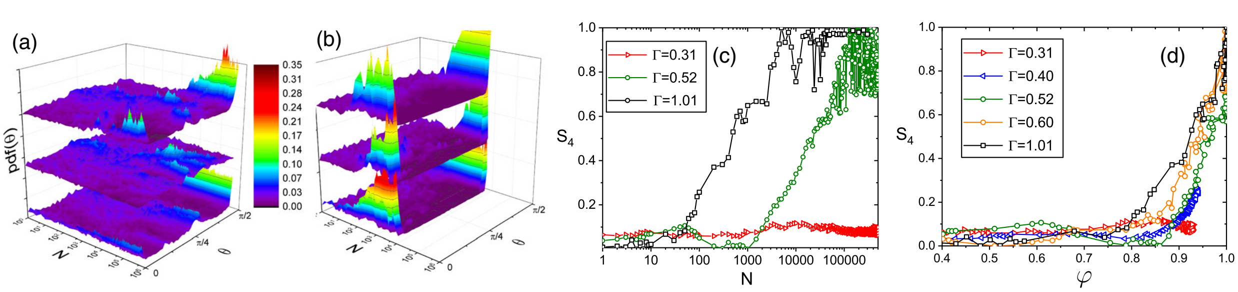

FIG. 4. Dice orientations distribution $pdf(\theta)$ vs N for (a) $\Gamma = 0.31$, and (b) $\Gamma = 1.01$. The bottom, middle, and top curves correspond, respectively, to the distributions obtained for the bottom, middle, and top regions of the cell. (c) Cubatic index S_4 vs N for three values of Γ as indicated in the legend. (d) Cubatic index S_4 versus lateral volume fraction φ for several values of Γ as indicated in the legend.

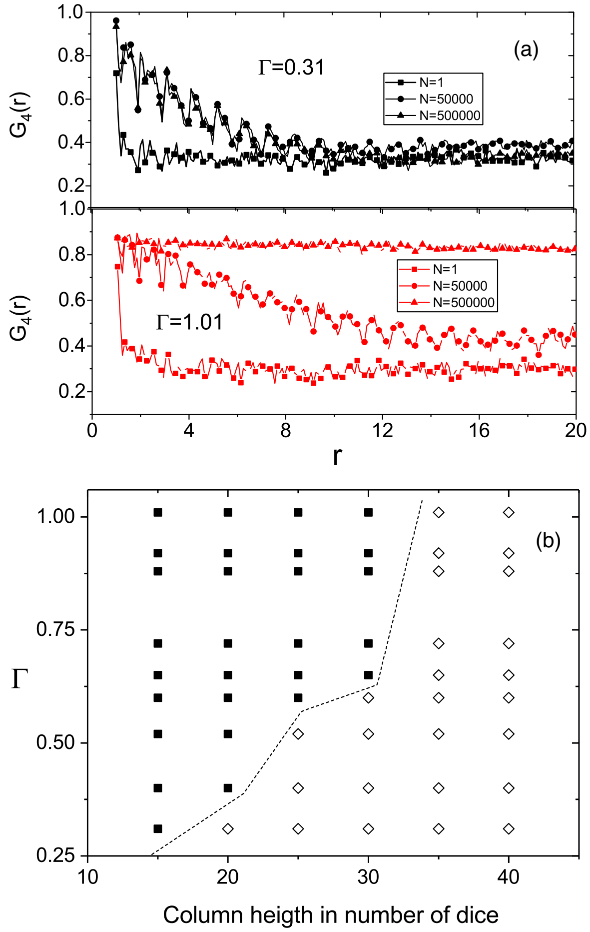

FIG. 5. (a) Orientational spatial correlation function $G_4(r)$ for low and high values of Γ after different numbers of twists N as indicated in the legend. r is the distance normalized by the cube's side length. (b) Sketch of the cube's ordering phase diagram. The black squares correspond to states where, after $N = 3 \times 10^4$ twists, the lateral volume fraction is maximum; the open diamonds indicate states where different cluster orientations coexist. The dotted line is a guide for the eyes.

Otherwise, for low twist amplitudes, even if there is also an initial increase in the correlation (see the curve for $N = 5 \times 10^4$ when $\Gamma = 0.31$) the evolution ceases as is proved by the fact that the $N = 5 \times 10^4$ and $N = 5 \times 10^5$ curves are identical. This feature is perfectly coherent with the idea that, for low twist intensities, the system evolves to a situation in which clusters of particles oriented at $\theta = \pi/4$ and $\theta = 0$ coexist. Therefore, there is some orientational correlation for short distances but the global orientational order remains very low. Up to this point we have evidenced that high twist amplitudes lead to perfectly ordered structures whereas, for low amplitudes, the system evolves to an arrangement that is inhomogeneous in the vertical direction and where clusters of cubes with different orientations coexist. These features suggest that the ordering process is a competition between the shear imposed by the container (which leads to particle alignment in the horizontal direction) and the load carried out by the dice column (which prevents the effectiveness of the shearing forces). If that hypothesis is correct, a variable that should be considered in the ordering process is the number of cubes within the cell (which in our case is quantified by the number of vertical rings that will be hypothetically stacked in a perfectly ordered arrangement). In Fig. 5(b), a phase space is presented separating the states that, after $N = 3 \times 10^4$ twists, are perfectly ordered from those where clusters of different orientations coexist. If the number of cubes inside the cell is small, the shear stress developed for low Γ is sufficient to induce rearrangements and the ordered state is rapidly reached. On the contrary, when the number of dice increases, the load competes with the shear and prevents complete ordering. In the latter case, the final state is characterized by domains of cubes aligned at different directions. Note that the transition between the two states could be slightly displaced if we consider a larger number of twists. In addition, we would like to remark that this phase space seems to be in contradiction with a previous work [29] where a shear stress perpendicular to the tapping direction was proved to promote disorder. However, these results were obtained for packings of spherical beads agitated by means of two mechanisms, tapping and shearing; the former leads to order and the latter to disorder. In our system, the only perturbation is the shearing among layers that promotes the alignment of cubes.

In this work, we have shown that twisting a sample of cubic particles is a highly efficient way to achieve ordered packings. The reason lies in the presence of flat surfaces, which tend to be aligned under shearing conditions. Indeed, contrary to the behavior observed with the standard tapping protocol, the system reaches the same final state regardless of the excitation intensity (provided it is large enough). Besides, the evolution of the packing fraction does not fit the commonly used parking lot model. Instead, the logarithmic growth suggests the necessity of introducing alternative arguments to describe the free volume interchange during the compaction process of sheared cubes. Finally, the asymptotic configuration seems to be the densest one compatible with the boundary conditions. It is a series of concentric rings stacked in horizontal layers. This arrangement implies a notably growth in the number of accessible configurations compatible with the value of the packing fraction. This result suggests that the notion of an ordering transition driven by entropy [5,30] could be adapted to this case. Indeed, the entropic origins of the ordering transition for athermal systems and the corresponding changes in the local environment around particles have been also discussed in Ref. [31]. Based on the established morphologies of concentric rings, similar arguments could be valid also for a systems of cubes under confinement.

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