Collapse-driven formation of a tetratic structure of confined quasi-2D granular tubes

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A tetratic phase, characterized by four-fold orientational symmetry, is observed in an experimental quasi-2D system of tubular particles subject to granular collapse. Evidence is presented that the cell's aspect ratio primarily affects the fraction of particles aligned along the length of the cell and not the overall degree of tetratic order as measured by the tetratic order parameter S_4, and that granular collapse enhances tetratic order beyond the effect of increasing the effective local density.

Keywords: Quasi-2D; granular collapse; tetratic order; driven granular media; tubular particles.

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1. Introduction

Systems of elongated particles with partial or no flexibility are common experimentally, and include seeds such as rice [1], molecular systems such as liquid crystals [2], some biological systems (see, for example, Dogic and Fraden [3] or Volfson *et al*. [4]), and minerals such as halloysite [5]. Of these systems, granular particles driven by an external energy source are not as readily simulated as analogous colloidal and molecular systems in equilibrium due to exhibiting inelastic collisions. Driven granular systems can form dense clusters of particles with little kinetic energy [6], which has been described by Tobochnik [7] as granular collapse, and can lead to a percolation transition. In the simulations by Tobochnik [7], granular collapse is driven, in addition to inelastic collisions, by a spatially inhomogeneous energy supply. As particles are driven into "cold" highly-packed regions, they lose nearly all their kinetic energy due to inelastic collisions. Stationary states with spatial kinetic energy variations are more poorly understood than homogeneous systems.

Quasi-2D systems are systems such that 3D constituents are constrained, by walls, external potentials or an interface, to move essentially along a plane and can thus be described in terms of 2D variables. Granular quasi-2D systems have been studied experimentally before (see, for example, Reis *et al.* [8] or Pacheco-Vázquez *et al.* [9]), but to the best of the authors' knowledge tubular quasi-2D systems exhibiting granular collapse have not been reported in the literature. Furthermore, most experimental work on elongated particles has focused on spatially homogeneous systems in equilibrium. Also, most of the relevant literature focuses on either rigid spherocylinders [10-15] or partially flexible systems [16]. However, there is evidence that 2D particles with flat ends, such as hard squares [17] and rectangles [18], exhibit isotropic-tetratic phase transitions, the tetratic phase being defined as having long-range or quasi long-range orientational order along two perpendicular axes while only exhibiting short-range positional order [18]. This is in contrast with the nematic, smectic and isotropic phases seen for particles with rounded ends; for example, for *Escherichia coli* in a long channel, nematic and not tetratic order is observed [4]. This difference in phases alone, given the importance of particles with flat ends, such as tubular systems like carbon and halloysite nanotubes, makes producing a granular analogue of such systems of considerable interest. In addition, the tetratic phase is not unique to particles with flat ends, as Narayan *et al*., [1] have demonstrated using basmati rice in a cell without granular collapse, yet it has not been nearly as thoroughly researched as other analogous phases, particularly in the presence of granular collapse.

A system's orientational order can be characterized using the tetratic order parameter [18]:

$$S_4 = \max_{\theta_0} \langle \cos[4(\theta - \theta_0)] \rangle \ , \tag{1}$$

which is unity for a perfectly tetratic phase and zero for an isotropic phase. θ_0 corresponds to the direction relative to which particles in a tetratic phase tend to be either parallel or perpendicular. Donev *et al*. [18] report, at a packing fraction $\sim$0.75, an isotropic-tetratic phase transition for hard rectangles of aspect ratio 2. As this transition takes place, S_4 goes from below ~ 0.2 to being greater than ~ 0.8 with increasing packing fraction. Donev *et al*., [18] explain their results in terms of the tetratic configuration's entropy, but for granular systems with external agitation, as they are not in thermodynamic equilibrium, the same behavior cannot be assumed *a priori*.

We report a system of quasi-2D near-monodisperse rigid granular tubes with inhomogeneous agitation leading to granular collapse, and observe tetratic phases, while also finding important differences with both previous simulational work on hard rectangles by Donev *et al*. [18] and novel simulations.

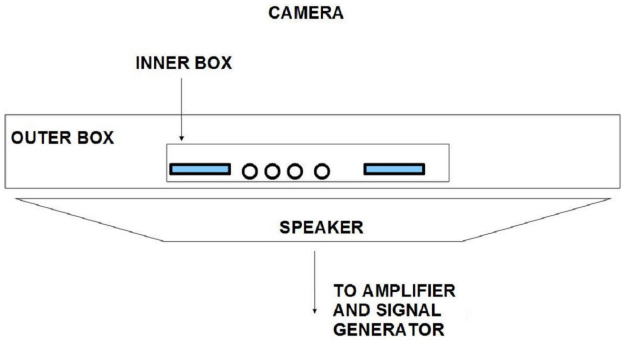

FIGURE 1. The experimental setup used is shown schematically. The filled rectangles depict the particles used (not shown to scale) seen sideways, and the empty circles depict their profiles seen from an end. The largest inner cell used has dimensions 9.1×7.1 cm.

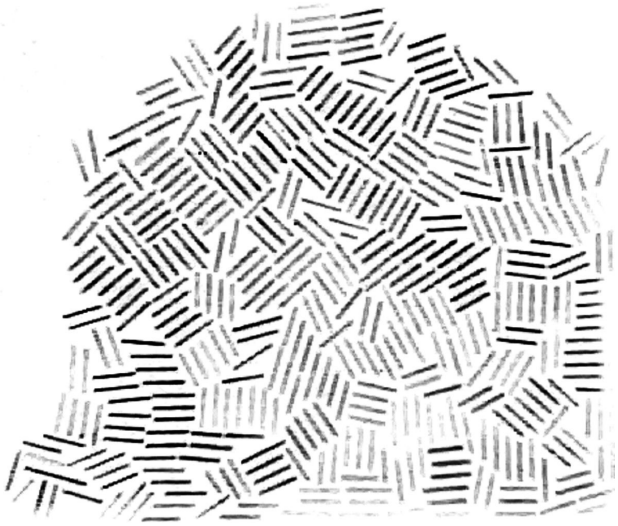

FIGURE 2. Sample image, inverted and with contrast enhanced for clarity, of the particles in a cell of aspect ratio 1.28.

2. Materials and Methods

The cell was made to vibrate by a speaker (Sony, 190 W peak output, 16 cm) driven by a 150 Hz sinusoidal wave from a signal generator (Phillips PM5132) and an amplifier (Steren AMP-010, 35 W rms), as shown schematically in Fig. 1. Images were captured by a Microsoft LifeCam VX-800 webcam at a rate of 5 frames per second. The driving frequency was chosen to be over an order of magnitude faster than the camera frame rate to ensure different frames correspond to different microstates (see Fig. 2), while still using an easily accessible frequency; the choice of 150 Hz is otherwise arbitrary. This system is reminiscent of the one used, for example, by Gradenigo *et al.* [19], but using a much more inexpensive apparatus. The amplitude was chosen so that the system remained quasi-2D, and care was taken to level the setup before each experiment. Inhomogeneous agitation was obtained as a result of using a flat cell. Since the speaker produces curved wavefronts, this results in a maximum amplitude near the cell's center. That is, the plane the particles can move in is not equidistant to the speaker center, resulting in inhomogeneous oscillation amplitudes and thus inhomogeneous agitation. To reduce, but not eliminate, inhomogeneity, the cell containing the particles was itself placed within another plastic cell. Inner cells of various aspect ratios were used (the aspect ratio of the particles themselves is fixed); for the lowest aspect ratio, 400 particles were used. Details of the particles used (smooth, rigid, silver-colored hollow plastic cylinders) are shown in Table I. The packing fraction (area fraction) η was calculated taking the particles as rectangles. Based on the speaker producing spherical wavefronts, the amplitude is, to a first approximation, expected to decrease linearly with r^2, with r being the horizontal distance from the cell center. For the maximum amplitude, estimated as ~ 1 mm, the corresponding dimensionless acceleration $A\omega^2/g$ is ~ 90.

The obtained image sequences were analyzed by fitting ellipsoids using the software ImageJ. In all cases, hundreds of frames per video and multiple videos per system were used to ensure good statistics. The order parameters were calculated with code written in-house. Based on the work of Donev *et al*. [18], we take states with $S_4 \gtrsim 0.8$ as tetratic.

Monte Carlo simulations (MC) were also carried out to better understand the origins of the differences between our experimental results and other systems, such as the simulations of Donev *et al*., [18]. The simulations carried out for the present work were of strictly 2-dimensional monodisperse hard disk composites (200 composites) in equilibrium. These composites consist of a dimer and a third disk whose center is at the point of contact of the disks forming the dimer (see Fig. 3), and each composite is treated as a rigid body. The simulations were carried out using a smectic initial configuration, and were allowed to proceed by expansion (lowering

TABLE I. Dimensions of the tubular plastic particles used.

Description	Mean	Polydispersity
Length	0.655 cm	4.1 %
Width	0.185 cm	6.5 %
Aspect ratio	3.56	5.8 %

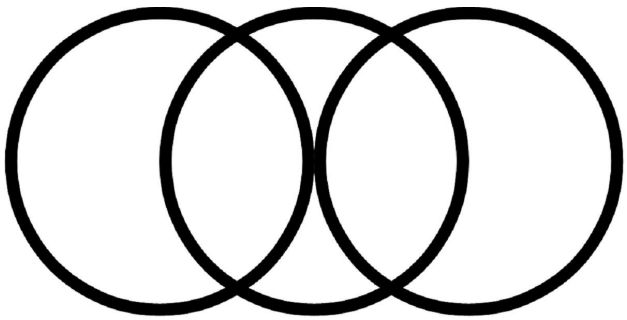

FIGURE 3. Schematic representation of the particles used in our Monte Carlo simulations. Their aspect ratio is 2, as in the simulations by Donev *et al*.

βP). Periodic boundary conditions were used. In order to better identify the effects of inhomogeneous agitation in the experiments, no such inhomogeneity was introduced in the simulations. Finally, different aspect ratios were used in the experiments and simulations in order to test how general the observed behaviors are; in other words, to help identify whether the observed phenomena are particular features of these specific experiments or whether they are of a more general character.

3. Results and Discussion

In contrast with the results reported by Donev *et al*. [18], we obtain values of S_4 close to or above 0.9 for three packing fractions below 0.6, the lowest packing fraction reported by Donev *et al*. and at which they report S_4 to be below 0.1. It is clear that granular collapse in our system leads to a tetratic phase at overall packing fractions far lower than would be the case in its absence. More surprisingly, the highest packing fraction also yielded the lowest S_4, and the behavior of S_4 as a function of η, as well as the comparable behavior of S_2, suggests that order peaks and then decays as the packing fraction increases. An analogous phenomenon has been observed for order characterized using the pair correlation function's first maximum for the jamming transition in quasi-2D experimental systems of colloidal spheres with some overlap between them [20], which was explained as resulting from a wider first peak as the jammed states have a higher degree of particle overlap, and in quasi-2D granular spheres [21]. It is not obvious that a measure of orientational order in granular would follow a trend reminiscent of that of a measure of radial order in a system of spheres, particularly in the case of thermal particles. This result suggests that jamming, regardless of its physical origin and in both thermal and granular systems, hinders order of all kinds.

Our estimates suggest that granular collapse clears about 13 % of the available area for the system at η = 0.749, and thus the effective local packing fraction at the cluster is $\sim$ 0.858, a value well above the transition reported by Donev *et al*., [18]. However, for the other systems studied, the effective local packing fractions were all below 0.65, which is well below the isotropic-tetratic transition reported by Donev *et al*. [18], and so the tetratic ordering we observe cannot be explained solely by the local density. We attribute this phenomenon to the boundary conditions, since the work of Donev *et al*. [18] uses periodic boundary conditions to emulate a bulk system, whereas we have rigid walls. Any granular collapse that is not towards the center of the cell will drive the particles towards the walls. Thus the resulting cluster is constrained by the walls, and as they are straight and at right angles, they provide natural directors for a tetratic phase, which is consistent with the behavior reported by Narayan *et al*. [1] for square and circular cells. Indeed, θ_0 was consistently found to be nearly zero relative to the lengthwise direction of the cell.

A further issue our results can shed light on is the effect of the cell's aspect ratio, which can be expected to be significant if the appearance of a tetratic phase is indeed driven by the boundary conditions. If the cell aspect ratio is unity, the expected orientational order would be purely tetratic, as the particles would have no reason to align themselves along any given wall or the two walls perpendicular to it. At the other extreme, if the cell width is less than a particle length, tetratic order is not sterically possible and nematic order would be strongly favored. The data in Table II correspond to various cell aspect ratios, including one close to unity, and at the other extreme, the cell with an aspect ratio of 8.57 has a width of only 1.7 particle lengths. Although the system exhibits a high S_4 for all cell aspect ratios, this does not imply that there are similar numbers of particles aligned along the length and width of the cell, but only that they tend not to lie along other directions. Thus we proceeded to calculate the parameter χ, which we define as the ratio of particles aligned within 45° of the width to the rest. Thus χ would be unity for a tetratic phase with equal numbers of particles aligned along the length and along the width of the cell, and would be zero for a system with no particles aligned along the width of the cell. As θ_0 is always found to be almost exactly along the length of the cell, these are the two extreme cases pertinent to our system.

TABLE II. Order parameters in various experimental conditions.

Cell Aspect Ratio	Packing Fraction	$\langle S_4 \rangle$
5.43	0.520	0.920
8.57	0.586	0.897
5.43	0.594	0.971
1.28	0.749	0.859

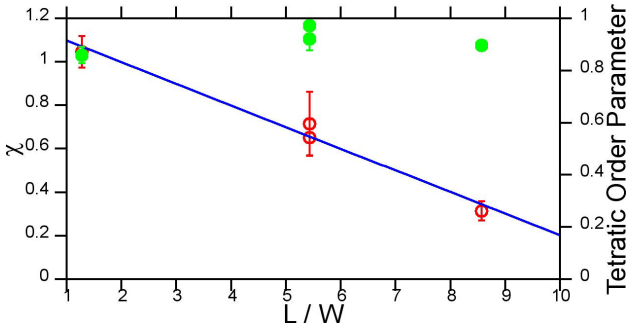

FIGURE 4. The experimentally-obtained ratio of particles aligned along the length of the cell to those aligned along the width (empty circles) is shown as a function of the cell's aspect ratio. The error bars correspond to standard deviations, and a linear fit is shown to guide the eye. Note that although the two data points at an aspect ratio of 5.43 correspond to different packing fractions, they are within a standard deviation of each other. The experimental tetratic order parameter values (filled circles) are also shown; they do not exhibit a strong dependence on the aspect ratio. Error bars again correspond to standard deviations.

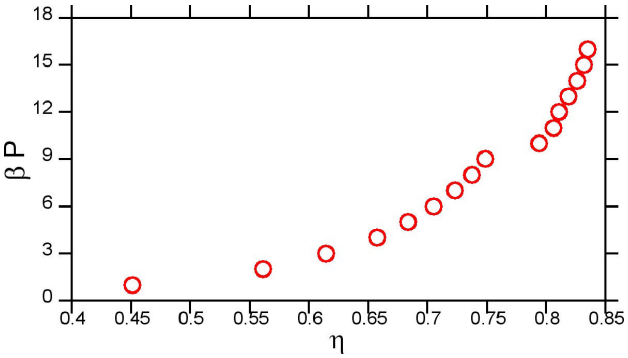

FIGURE 5. Normalized pressure P/k_BT vs. packing fraction as obtained from MC simulations of the composites described in the Materials and Methods in an NPT ensemble. The results suggest a first-order transition at $\eta \sim 0.75$, which is close to the transition reported by Donev *et al*., [18].

Figure 4 shows the behavior of χ as a function of the cell's aspect ratio. For the lowest aspect ratio, χ is close to unity as would be expected, and decreases with increasing aspect ratio. This suggests that, while granular collapse in this system induces a phase whose tetratic order parameter does not depend on the cell aspect ratio, said aspect ratio does govern the fraction of particles that align along the cell's length. The exact role of the aspect ratio and width relative to the particle length in determining the nature of the resulting phase, particularly in the presence of granular collapse, remains a theoretical and experimental challenge.

Figure 5 shows normalized pressure as a function of η (as this is an NPT ensemble, the pressure is fixed *a priori*), and the results strongly suggest a phase transition taking place at a slightly lower packing fraction than the one Donev *et al*. [18] report for hard rectangles. The simulational system we report has the same aspect ratio as said rectangles, but has circular ends. Our results suggest that, at least to a first approximation, and since for any elongated hard particles the low density limit in equilibrium can be expected to be isotropic, the phase boundaries of elongated particles are governed primarily by their aspect ratio rather than by the detailed nature of their ends, which may govern the precise nature of these phases. If so, this would render our experimental system relevant to a wide variety of different systems, even ones with fairly irregular ends such as the rice grains studied by Narayan *et al*., [1]. Finally, the transition seen in the simulations, which do not exhibit granular collapse, is consistent with tetratic order at low packing fractions being driven by granular collapse.

4. Conclusions

The formation of a tetratic phase, a phase with four-fold orientational symmetry, is observed in an experimental quasi-2D system of tubular particles subject to granular collapse. Evidence is presented that the cell's aspect ratio primarily affects the number of particles aligned along the length of the cell and not the overall degree of tetratic order as measured by the tetratic order parameter S_4, and that granular collapse enhances tetratic order beyond what can be attributed to increasing the effective local density. Future work will focus on improving our understanding of the effect of non-periodic boundary conditions, and on extending the simulations carried out to other particles and conditions.

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