Radial distribution function at the particle-bottom interface in granular self-assembly

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Abstract. Dynamic self-assembly of confined non-Brownian spheres under gravity is investigated in terms of the statistical behavior of the like-gaseous particles closest to oscillating bottom. For strong vibrations, these particles resemble a quasi-two-dimensional gas (qtd-gas) that supports upper particles. By vibrational annealing, wet particles inside a commensurate rectangular container self-assembling into face-centered-cubic (fcc). From high temporal resolution images we study, by means of the associated Radial Distribution Function (RDF), the evolution in time of spatial correlations in the transition: qtd-gas to dense liquid of the vibrating particles closest to the oscillating bottom during self-assembly. As the vibrational annealing advances, the particles closest to the bottom reach this final position and the associated RDF shows a clear increase of the spatial correlation, which is congruent with the fcc crystallization phase.

1. Introduction
Due to friction forces, gravity and confinement, static granular materials are into a metastable static states. In this sense, a sudden agitation of the container is enough for the particles composing the granulate system to attain a new spatial configuration (a new metastable state). On the other hand, continuous vibrations (usually sinusoidal) thermalize the system into a stationary state that resembles, for particular vibrating conditions, like granular gases at low density (feature that exclude many experimental conditions of current interest). For this particular case the Boltzmann equations correctly describe the system [1], but for denser gases the Maxwell-Boltzmann distribution, of the horizontal velocities, fails [2]. Despite granulates are inherently dissipative and have no reaction to thermal fluctuations, some sort of stochastic mobility is provided by continuous external excitations, for this the system resemble and have been used as model system for atomic, molecular, and colloidal ensembles in thermal equilibrium [3, 4]. Into a stationary state some thermodynamic-like variables (density, temperature or pressure) are statistically well defined [5-15]. Because of dissipative collisions, such variables exhibit a non-monotonic behavior [13, 14, 16].

In many situations of the life, including the pharmaceutical, food, mineral or construction industries, the granular materials are prominent. Notwithstanding their importance, distinctive phenomena, such crystallization [17-20], convection [21-25], or compaction [26, 27] where nature guides toward one or the other depending, at first instance, on the oscillating conditions, has been extensively studied without considering the dynamics of particles closest to the oscillating bottom.
In this work we study the dynamics of cohesive particles closest to oscillating flat bottom during a granular self-assembly process. It is well known that granulates change their mechanical properties when a small amount of liquid is added to them [28]. Liquid bridges between particles give place to attractive capillary forces and, therefore, granulates behave in a different way. At the beginning of self-assembly process, particles under scrutiny (cohesive ball bearings) behave as a confined (by the vertical walls of rectangular container) quasi-two-dimensional dense gas (qtd-gas), which transfers their kinetic energy by collisions and supports the upper particles (also cohesive). We chose wet beads because an analogous behavior with thermal systems has been established in [29]. As the self-assembly advances, the particles of the qtd-gas slowly evolves to a crystalline phase. Throughout the Radial Distribution Function (RDF), calculated from the analysis of high temporal resolution images of the evolving qtd-gas (a fast camera was mounted underneath of the translucent oscillating bottom, Fig.1), we study the structural evolution of the granular self-assembly. Our results show that spatial correlation of the qtd-gas grows in agreement with the crystalline structure. Hence, the behavior of the whole vibrating granular system can be considered as consequence of the dynamics in the particle-bottom interface.

Figure 1. (Color online) Experimental setup. A rectangular box mounted on the oscillating flat bottom. Two fixed electromechanical vibrators (EV) drives the system to vertical oscillations. The fast speed camera (ccd) was mounted below the square box to capture the movement of the particles closest to oscillating flat bottom.

2. Experimental methodology
The granular self-assembly, by vibrational annealing method, consists in shaking the granular system with a sinusoidal oscillation; where the frequency \( \nu \) and the amplitude \( A \) is increased and decreased by discrete steps, respectively, in such a way that the dimensionless acceleration \( \Gamma = A(2\pi\nu)^2 / g \), \( g \) is the acceleration of gravity) remains constant. The total self-assembly process is completed in about
200 seconds. The frequency and amplitude were changed in intervals of 5 seconds before. After changing the frequency and amplitude the system reaches a stationary state in 0.5 seconds. At the beginning of the process, when the amplitude is large and the frequency small, most of the assembly is a dense gas. Slowly, the assembly becomes a fluid-like system where the beads reduce their velocity but still move all around the container. Finally, at high frequencies and small amplitudes, the system is a solid where beads only vibrate around their equilibrium positions [30].

The initial conditions of the annealing (amplitude and frequency), and the rate of the cooling program, are given to the computer at the beginning of the process. As long as the annealing rate is not too fast to quench the system in a disordered structure, this rate is not crucial. The experimental setup consists of a vertical shaking system driven by a function generator (HP-33120A), an amplifier for the electromechanical vibrators (EV), and a controlling computer. The particles of the qtd-gas were filmed with a high-speed charge-coupled device (CCD) camera (Photron SA3 60K M1 512 × 512 pixels), mounted underneath of a translucent flat bottom. In order to capture the whole dynamic, the particles were filmed at 1000 frames per second. ImageJ free distribution software was used for the video analysis, this give us the particles position for each one frame. In Fig. 2 we show an instantaneous frame (left) and their corresponding reconstruction from the particles positions given by the ImageJ software (right). Unfocused images are the best technique to study only the particles of the qtd-gas. In this way, the sphere's images were recorded just from the bright spot shining at their center, making it easier for the particle center localization algorithm to process the frames. We use cohesive steel beads (a thin film of oil cover the beads surface), of diameter $d=3.18 \text{ mm.}$, inside a commensurate rectangular container made of plexiglas, having dimensions of $L_x = L_y = 61.6 \text{ mm.}$ and $L_z = 150 \text{ mm.}$ The cohesive force between two particles consists of two parts: the surface tension, acting at the wet perimeter, and a term arising from the capillary pressure in the liquid [31].

![Figure 2.](image)

**Figure 2.** (Color online) Left, the instantaneous image from the fast camera of the particles closest to the bottom. Right, the reconstruction by means of the particles position obtained from the ImageJ software.

Radial Distribution Function: The radial distribution function $g(r)$ is a standard way of describing the average positional structure of particulate systems [32]. $g(r)$ measures the probability that two particle centers are at distance $r$, regardless of orientation. For hard particles there is zero probability that the particles overlap so $g(r) = 0$ for $r < d$. The construction of the RDF is as follows: consider, for example, a gas of hard particles and chose, in any instant of time, a particle and draw it a series of concentric spheres, set at a small fixed distance $(dr)$ apart. At regular intervals of time the number of particles found in each shell is counted and stored. At the end, the average number of particles in each shell is calculated. Then, this is divided by the volume of each shell and by the average density of particles in the system. The result is the RDF. The mathematical expression for the RDF is given by:
\[ g(r) = \frac{n(r)}{4 \pi r^2 \, dr}, \]

where \( n(r) \) is the mean number of particles in a shell of width \( dr \) at distance \( r \), \( \rho \) is the mean particle density. The method need not be restricted to one particle. All the particles in the system can be treated in this way, leading to an improved determination of the RDF as an average over many particles.

In our experiments, despite the intrinsic effects of the limiting walls of the container and the cohesion forces, we calculate the RDF for particles of the qtd-gas at each one of the stationary state during the self-assembly. For this, we first consider the particle closest to the center (Fig. 3) to calculate the mean number of particles in a ring of radius \( r \) and thickness \( dr \), thereafter we extend the count to the five particles closest to the center and go one for each one of the 5000 frames belonging to some particular stationary state, at the end we averages over time and the five particles to obtain \( g(r) \). We choose \( dr \) enough small compared with the dimensions of the system; hence the particles outside the longest circle do not contribute to the RDF.

![Figure 3. (Color online) Each dot represents the center of a particle.](image)

3. Discussion and Results
The annealing process starts with \( \nu = 22 \text{ Hz} \) and \( \phi = 5.5 \). For these oscillating conditions the system behaves as a dense fluid and the closest particles to the oscillating bottom move erratically throughout the container. Due to periodic oscillations, the number of these particles is not constant, so the packing fraction associated takes values \( \varphi = (0.54, 0.64) \). From one frame to the next, the particles of the qtd-gas could be different; nevertheless the RDF captures correctly the positional correlation of granular gases in a stationary state [33, 34], being these in out of equilibrium but stationary ensembles. In Fig. 4a we show the RDF at the beginning of the self-assembly, this is consistent with lead particles (lesser restitution coefficient, \( \mu \)) in two-dimensional granular gases at low density (\( \varphi = 0.35 \)) [35]. It is interesting to note that particles with bigger restitution coefficient and packing fraction (our system) behave as a gas of particles with minus restitution coefficient and packing fraction. Then, \( \mu \) and \( \varphi \) must be considered for a correct description. The annealing goes one and the positional correlation...
increase. For example, at \( \nu = 28 \) Hz the RDF (Fig. 4b) shows strong correlation, and several peaks indicates that the closest particles to the bottom vibrates as a very dense fluid. At this time the system has been suffered a structural phase transition of first order [36], where dense fluid particles support a more dilute phase in the upper part of the vibrating system. The RDF at \( \nu = 40 \) Hz and \( \nu = 28 \) Hz looks very similar, with a first peak at \( \sqrt{2}r/d \) and the second principal at \( 2r/d \). This indicates that a cubic lattice (face centered cubic) is formed over the oscillating bottom. Before the second principal peak a little peak emerges at \( \nu = 40 \) Hz (Fig. 4c) around the vertical line. We think that this small peak is due to the periodic oscillations. At some particular time the particles collides with the oscillating bottom, and after they leave it, so the system undergoes a small periodic expansion-contraction, this produces an overlap that gives place to the small peak. Despite the dissipative collisions, the effects of the walls, and the increase of the packing fraction, the RDF captures the correlations as the annealing advances and the crystalline phase begin to appear.

![Figure 4](image-url) (Color online) Radial Distribution Function at different stationary states, characterized by the oscillating frequency. With the aim to appreciate, we show only the RDF for three stationary states.
4. Conclusions

We have presented an experimental study of the spatial correlations associated with the closest particles at the particle-bottom interface in a granular self-assembly process. The radial distribution functions of these particles were obtained when the system was in a stationary state, characterized by the frequency of vibration \( \nu \). Despite the effects of the confining walls and the relatively high values considered for \( \rho (0.54 < \rho < 0.75) \), the corresponding RDF's for each stationary state, shows congruency with the conformation of the FCC structure. Our work could be used as the basis for calculating effective potentials (via the Orstein-Zenike equations with the Percus-Yevick closure) at the particle-bottom interface.

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