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Coarsening of granular clusters: Two types of scaling behaviors

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We report on an experimental study of small cluster dynamics during the coarsening process in driven granular submonolayers of 120-μm bronze particles. The techniques of electrostatic and vertical mechanical vibration were employed to excite the granular gas. We measure the scaling exponent for the evaporation of small clusters during coarsening. It was found that the surface area of small clusters S vs time t behaves as $S\sim(t_0-t)^{2/3}$ for lower frequencies and $S\sim(t_0-t)$ for higher frequencies. We argue that the change in the scaling exponent is related to the transition from three-dimensional (3D) to 2D character of motion in the granular gas.

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One of the most fascinating phenomena observed in freely cooling and driven granular gases is spontaneous separation into dilute and dense regions [1,2]. This phenomenon stimulated great interest among theorists and experimentalists [3–6]. The clustering phenomenon does not appear to be limited to just the traditional granular systems with short-range hard-core collisions. It was shown that some granular systems with competing long-range electromagnetic interactions and short-range collisions exhibit somewhat similar behavior [7]. In the case of mechanically vibrated granular matter where the energy is injected into the system through local collision with the bottom plate, clustering takes place due to the increase of the dissipation rate with the increase of the density of granular gas [4,5]. In the case of electrostatically driven granular matter there is an additional effect of electrostatic screening, which decreases the electrostatic force acting on particles inside dense clusters [7].

The phase diagram for these two seemingly different systems appears to be rather independent on the specifics of driving [5–7]. Particles are immobile for small values of the driving force (amplitude of vibration acceleration in the case of mechanical system and the amplitude of electric field for the electrical system). When the driving force exceeds the first critical value, isolated particles begin to move. If the driving force value is increased above a second threshold value, the granular medium forms a nearly uniform granular gas. Between these values the uniform gas phase is unstable and a fraction of particles can form immobile clusters. The clusters exhibit coarsening dynamics of Ostwald-ripening type: small clusters disappear and large clusters grow with time.

Here we report on the experimental study of small cluster dynamics during the coarsening process in driven granular submonolayers. We systematically compare two techniques of energizing the granular gas: mechanical and electrostatic. We measure the scaling exponent for the evaporation of small clusters during coarsening. We have found that in both systems the surface area of small clusters S vs time t behaves as $S\sim(t_0-t)^{2/3}$ for lower frequencies and $S\sim(t_0-t)$ for higher frequencies. This transition in scaling behavior coincides with the transition from three-dimensional (3D) dynamics of the granular gas to 2D dynamics as the driving frequency is increased for a fixed acceleration.

The experimental setup for mechanical shaking is similar to that in Refs. [5,6,8,9]. We performed experiments in a rigid circular container of 13.5 cm diameter vibrated vertically by an electromagnetic shaker. The spherical bronze particles (120–150 μm) constituted approximately 2/3 of a monolayer coverage on the optically flat horizontal plate (single-crystal silicon wafer). The applied frequency varied in the range of 20–100 Hz. Most of the measurements were performed under atmospheric pressure in air. Some results were verified in vacuum.

To excite a granular medium electrostatically, particles were placed between plates of a large capacitor and energized either by dc or ac electric fields [7]. We used 27 $\times$27 cm transparent capacitor plates (glass coated by indium-doped tin oxide) and the plate spacing was 1.5 mm. The particles used for these series of experiments are the same as for the mechanical shaking measurements and constituted approximately 1/3 of a monolayer coverage on the bottom plate. The applied field was varied in the range 0–7 kV/cm and its frequency was varied from 0 to 120 Hz. The experiments were performed in an atmosphere of dry nitrogen to reduce adhesion of particles on the plate due to humidity in air [10]. The charge-coupled device camera was suspended above both cells. We analyzed the total number of clusters, evolution of individual clusters, mean cluster sizes, etc.

In the electrocell, energy injection works as follows [7]. Particles in the contact with bottom plate acquire an electric charge and the electric force exerted on the particle is $F_0=1.36R^2E^2$, where R is the radius of the particle and E is the external field. When F_0 exceeds the gravitational force, the particle moves upwards, recharges upon colliding with the upper plate and falls down. In the case of DC fields the process repeats in a cyclical manner. By applying AC field one may turn the particle back before it reaches upper plate. Thus, increasing the frequency of the field, one controls the extent of vertical motion of the particles. Clustering of granular gas with consequent ripening occurs if $E_1<E<E_2$ (E_1 and E_2 is the first and second threshold values). The

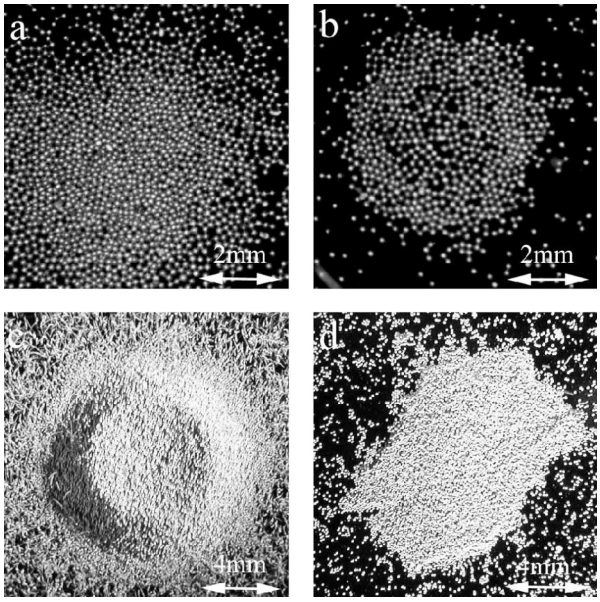

FIG. 1. Shapes of granular clusters. Upper row shows clusters in electrostatically driven system. (a) $f=20$ Hz; (b) $f=120$ Hz. Bottom row shows clusters in mechanically vibrated system. (c) $f=25$ Hz; (d) $f=95$ Hz. Left column: heaplike clusters corresponding to 3D behavior. Right column: flat monolayer clusters corresponding to 2D regime.

clustering comes from two effects: increased dissipation of the kinetic energy in the regions with high granular media density and electrostatic screening (two close particles acquire smaller charge than two well-separated ones). In the mechanical cell, vibration amplitude plays the role of electric field.

The primary objective of our studies was to observe the scaling behavior of individual clusters as function of time. In certain cases one can relate the scaling exponent governing evaporation of small clusters with global scaling properties of coarsening process due to conservation laws [11,12]. For example, if the area of small cluster S in 2D system evolves as $S\sim(t_0-t)^{\beta}$, where t_0 is the instant of the cluster's disappearance and β is scaling exponent, and the total number of clusters (N) evolves as $N\sim 1/t^{\alpha}$, for interface-controlled coarsening regime the exponents α and β are related, $\alpha=\beta$ [11]. In many cases the studies of individual cluster dynamics are more reliable than direct studies of global scaling exponents that are often masked by the finite size effects.

The experiments in the mechanical cell were performed in the following manner. A uniform gaslike phase was prepared by setting the dimensionless acceleration level $\Gamma=4\pi^2 f^2 A_0/g$ [f is vibration frequency, A_0 is the amplitude of driving $A=A_0\sin(2\pi ft)$, and g is gravity acceleration] approximately at $\Gamma=(2.28\text{–}2.72g)$. The acceleration was then suddenly dropped to the value 1.00–1.05, leading to spontaneous nucleation of immobile clusters from the gas phase [Figs. 1(c,d)].

The area of a selected cluster S was measured as a function of time. If the cluster is not the biggest one in the system, eventually it begins to shrink. We find that the area S evolves according to a power law with an exponent that depends on the vibration frequency. For frequencies less than 35 Hz, $S\sim(t_0-t)^{\beta}$ with $\beta=0.63$–0.7, see Fig. 2. For the frequencies higher than 55 Hz, $S\sim(t_0-t)^{\beta}$ with $\beta\approx 1$. A crossover-type behavior is observed for an intermediate range of frequencies. In this case the area evolves as follows: while the cluster is above a certain size, its area decreases linearly with time. However, when the cluster decreases below a certain size, its area decreases quicker, as $S\sim(t_0-t)^{\beta}$ with $\beta\approx 2/3$.

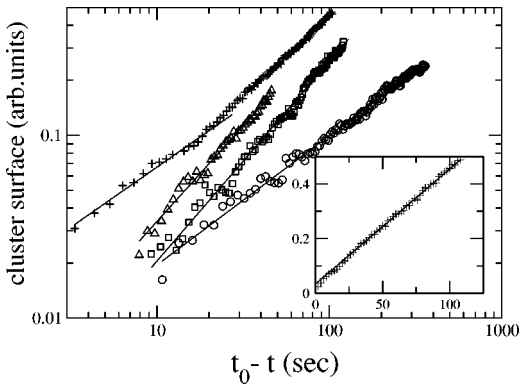

FIG. 2. Cluster area vs time for mechanical system. Circles: $f=25$ Hz, $\beta=0.7$. Crosses: $f=35$ Hz. $\beta=1.03$ at the beginning of evaporation (see inset) and $\beta=0.66$ at the end of collapse. Triangles: $f=55$ Hz, $\beta=1.04$. Squares: $f=85$ Hz, $\beta=0.7$. Inset: S vs t for $f=35$ Hz in linear scale.

Qualitatively similar behavior is observed in an electrostatically driven system (Fig. 3). In the electrocell, the uniform gas was prepared by applying an external field of 7 kV/cm before "'quenching" the gas below E_2. For the case of a dc electric field applied, as well as for an ac field with the frequency less than 60 Hz, we find that the scaling exponent $\beta\approx 2/3$ (actually the range is 0.63–0.7). Accordingly, the cluster area shrinks linearly in time for frequencies above 90 Hz. In the range from 60 to 90 Hz, there is a crossover behavior observed with these two exponents.

The apparent frequency dependence of the scaling expo-

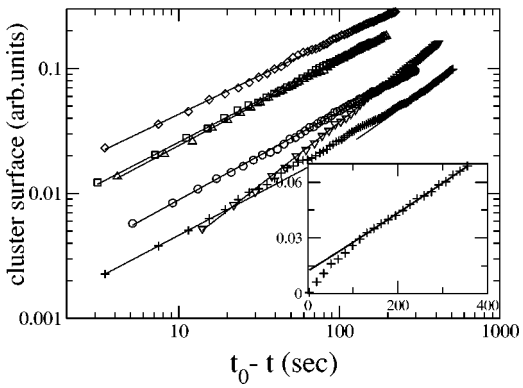

FIG. 3. Cluster area vs time for electrocell. Circles: $f=0$, $\beta=0.7$. Squares: $f=10$ Hz, $\beta=0.67$. Diamonds: $f=20$ Hz, $\beta=0.63$. Up triangles: $f=50$ Hz, $\beta=0.68$. Crosses: $f=75$ Hz, $\beta=1.04$ at the initial stage of evaporation (see inset) and $\beta=0.67$ at the end. Down triangles: $f=100$ Hz, $\beta=1.02$. Inset: S vs t for $f=75$ Hz in linear scale.

nents β for cluster evaporation can be related to the transition from two- to three-dimensional behavior in the granular gas. For the case of dc or low-frequency ac driving, the particles have enough time to reach the upper plate and the granular gas exhibits 3D behavior. Thus, gas particles can fly above the cluster and the exchange of particles between gas and solid phases occurs at entire surface of the cluster. Consequently, the clusters contain more than a monolayer of the particles [Fig. 1(a)]. Measurements show that in the electrostatic cell with a gap equal to 1.5 mm, cluster thicknesses equal to four monolayers are observed in the central part of the cluster. The thickness is limited due to an increase of the electric field above the cluster as the gap between its surface and upper plate shrinks. When the field value at the cluster surface reaches the second threshold value E_2, particles cannot remain immobile, even in the middle of the cluster, and will rejoin the gas. As the extent of vertical particle motion becomes smaller than the particle diameter (for high frequencies), the granular gas becomes two dimensional. In this case, the exchange of particles between gas and solid takes place only at the outer boundary and clusters have a monolayer thickness [Fig. 1(b)].

Similar transition occurs in mechanically vibrated cell. If we keep the value of the dimensionless acceleration Γ fixed, lowering of the frequency implies shaking at higher amplitudes, and consequently, increase of the extent of vertical motion. Thus, at lower frequencies the vertical particle jumps will eventually exceed the particles size d, i.e., the motion is effectively more 3D than it is at higher frequencies. The dimensional transition in the collision dissipation rate has been previously observed in Ref. [13].

The crossover frequency can be determined from the comparison of the height of particle jumps ξ and the particle size d. In mechanical system the height may be roughly estimated to be equal to the vibration amplitude A_0, i.e., $\xi = g/4\pi^2 f^2$ since clusterization occurs when $\Gamma \approx 1$. From the condition $\xi \approx d$, one find the crossover frequency $1/2\pi\sqrt{g/d} = 40$ Hz for $d = 150$ μm, which is in a good agreement with experiment. The value of ξ decreases as the frequency increases and when it becomes smaller than a particle diameter, the granular gas exhibits 2D dynamics. In this case clusters are flat [Fig. 1(d)]. As the frequency decreases, the magnitude of vertical jumps increases and the clusters assume a heaplike form [Fig. 1(c)]. Their evaporation will reflect features of 3D gas dynamics as is the case of electrostatic excitation. The saturation of the cluster occurs when the cluster height achieves the magnitude of the vertical jumps of isolated particles. It is necessary to mention that this heaplike form of the clusters is not necessarily related with the interaction between particles and trapped air flow through the bulk of the medium as it was in Refs. [14]. Clusters keep heaplike form even in the evacuated cell (pressure is 5 mTorr).

According to the model presented in Ref. [15], when the cluster size becomes small (i.e., near the moment of evaporation), the normal velocity component of the interface between solid and gas phase for 2D dynamics is governed by the equation $\dot{R} \approx -K/2R$. Here R is the local radius of the interface, K is surface tension coefficient that can be expressed through the diffusivity of particles. Thus, for the cluster area $S = \pi R^2$, one derives $S = \pi K(t_0 - t)$, i.e., S decreases linearly during cluster evaporation.

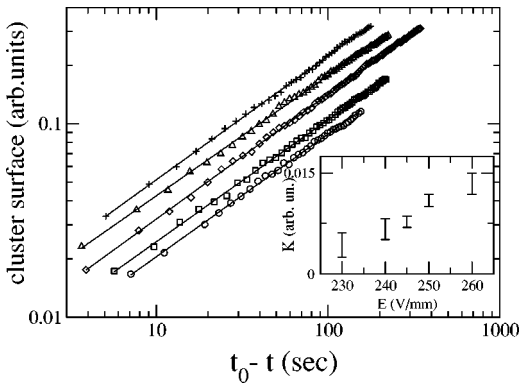

FIG. 4. Cluster area vs time for electrocell. $f = 20$ Hz. Circles: electric field $E = 230$ V/mm. Squares: $E = 240$ V/mm. Diamonds: $E = 245$ V/mm. Triangles: $E = 250$ V/mm. Crosses: $E = 260$ V/mm. Inset: the dependence of cluster surface tension on the applied electric field.

To match the low-frequency scaling, one assumes that the evolution of small clusters is governed by the equation $\dot{R} \approx -\tilde{K}/R^2$. It yields $R^3 \sim (t_0 - t)$ and the area occupied by the clusters scales as $S \sim (t_0 - t)^{2/3}$. This result can be interpreted as a manifestation of the decrease of the surface tension coefficient K in multilayered clusters. One expects that the coefficient K is inversely proportional to the cluster's thickness h (it requires more time to evaporate a multilayer than a monolayer), i.e., $K \sim h^{-1}$. Since for heaplike clusters the thickness h is proportional to the radius R, one obtains $\dot{R} \sim R^{-2}$. This argument applies both to mechanical and electrocells.

We would like to emphasize that the low-frequency exponent $\beta = 2/3$ does not appear to be related to the global scaling exponent α for the number of clusters (N). This is due to saturation of the height of large clusters: when a cluster is large enough ($R \gg h$), the dynamics of its evaporation has a 2D character, as the gas/solid exchange occurs mostly at the

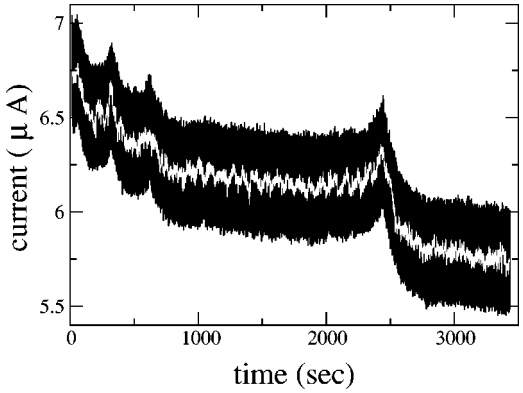

FIG. 5. Current vs time during coarsening in dc electric field. White line shows the signal averaged over 100 points. Current peaks correspond to evaporation of clusters. The signal/noise ratio is greater than 3.

edge of the cluster. However, when the cluster size decreases to $R \sim h$, evaporation occurs on entire surface of the cluster.

We find that the amplitude of driving mostly affects the preexponential factor in the scaling law, i.e., the surface tension. Some selected results are shown in Fig. 4. As one sees from the figure, the value of the surface tension (and, therefore, diffusivity coefficient for the particles) increases with the increase of the applied electric field. This behavior can be attributed to the increase of the granular gas temperature.

One of the interesting predictions of the phenomenological model [15] is a fine structure in the dependence of granular gas density vs time during the coarsening. A small cluster shrinks so quickly at its evaporation point that the gas concentration temporarily increases [15–17], resulting in a peak feature. The electric current through the cell is carried by the particles belonging to the granular gas phase and is, therefore, proportional to the amount of the gas. This leads to a transient increase of the current through the system at the moment of small cluster evaporation. These peaks of current were indeed observed for the case of electrostatic cell with applied dc voltage (Fig. 5). Over time, the current approaches a value corresponding to the coexistence of one cluster and the granular gas. The observation of the peaks reinforces the validity of the continuum model described in Ref. [15].

In conclusion, we studied experimentally the cluster dynamics of evaporation in two driven granular systems, mechanical and electrical. The transition between 3D and 2D behaviors was found. These results are qualitatively similar for both systems. For the cluster coarsening process in a granular gas excited by a dc electric field, the transient increase of current through the system at the moment of a small cluster evaporation was observed.

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