Electrostatically Driven Granular Media: Phase Transitions and Coarsening

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We report the experimental and theoretical study of electrostatically driven granular material. We show that the charged granular medium undergoes a hysteretic phase transition from the immobile condensed state (granular solid) to a fluidized dilated state (granular gas) with a changing applied electric field. In addition we observe a spontaneous precipitation of dense clusters from the gas phase and subsequent coarsening—coagulation of these clusters. Molecular dynamics simulations show qualitative agreement with experimental results.

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Despite extensive study over the preceding decade, a fundamental understanding of the dynamics of granular materials still poses a challenge for physicists and engineers [1,2]. Many peculiar properties of granular materials can be attributed to strong contact interactions and inelastic collisions between grains [3,4]. Fascinating collective behavior appears when small particles acquire an electric charge and respond to competing long-range electromagnetic and short-range contact forces.

The electrostatic excitation of granular media offers unique new opportunities compared to traditional vibration techniques which have been developed to explore granular dynamics [3–6]. It enables one to deal with extremely fine powders which are not easily controlled by mechanical methods. Fine particles are more sensitive to electrostatic forces which arise through particle friction or space charges in the particle environment. Their large surface to volume ratio amplifies the effect of water or other surfactants. These effects intervene in the dynamics causing agglomeration, charging, etc., making mechanical experiments uncontrollable. Electrostatic driving makes use of these *bulk* forces, and allows not only for removal of these "side effects," but also for control by long-range electric forces.

In this Letter we report the experimental and theoretical study of electrostatically driven granular material. It is shown that the charged granular medium undergoes a hysteretic phase transition from the immobile condensed state (granular solid) to a fluidized dilated state (granular gas) with a changing applied electric field. A spontaneous precipitation of dense clusters from the gas phase and subsequent coarsening—coagulation of these clusters is observed in a certain region of the electric field values. We find that the rate of coarsening is controlled by the amplitude and frequency of the applied electric field. We have also performed molecular dynamics simulations of electrostatically driven particles. These simulations show qualitative agreement with experiments.

The experimental cell is shown schematically in Fig. 1. Conducting particles are placed between the plates of a large capacitor which is energized by a constant or alternating electric field. To provide optical access to the cell, the upper conducting plate is made transparent. We used 4×6 cm capacitor plates with a spacing of 1.5 mm and spherical copper particles with average size about 35 μm. The particles were sieved to ensure narrow particle size distribution [7]. The field amplitude varied from 0 to 10 kV/cm and the frequencies on the interval of 0 to 250 Hz. The number of particles in the cell was about 10^7. The experiments were performed both in atmospheric pressure and in vacuum (5×10^{-6} Torr).

When the conducting particles are in contact with the capacitor plate they acquire a surface charge. As the magnitude of the electric field in the capacitor exceeds the critical value E_1 the resulting (upward) electric force overcomes the gravitational force mg (m is the mass of the particle, g is the acceleration due to gravity) and pushes the charged particles upward. When the grains hit the upper plate, they deposit their charge and fall back. Applying an alternating electric field $E = E_0 \sin(2\pi f t)$, and adjusting its frequency f, one can control the particle elevation by effectively turning them back before they collide with the upper plate.

The phase diagram is shown in Fig. 2. Isolated particles start to move at $E > E_1$. We have found that at amplitudes of the electric field above a *second* threshold value, $E_2 > E_1$, the granular medium forms a uniform gaslike phase (granular gas). This second field E_2 is (50–70)% larger than E_1 in nearly the whole range of the parameters used. In the field interval $E_1 < E < E_2$, a phenomenon analogous to coalescence dynamics in systems exhibiting first order phase transitions [8,9] was observed. Upon

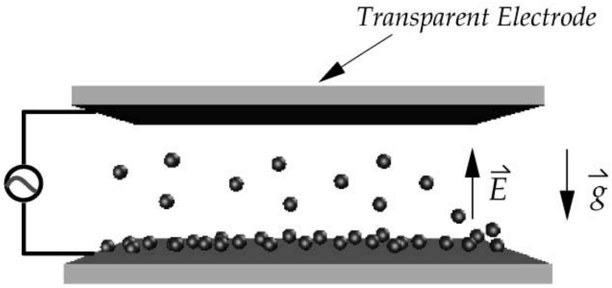

FIG. 1. The electrostatic cell.

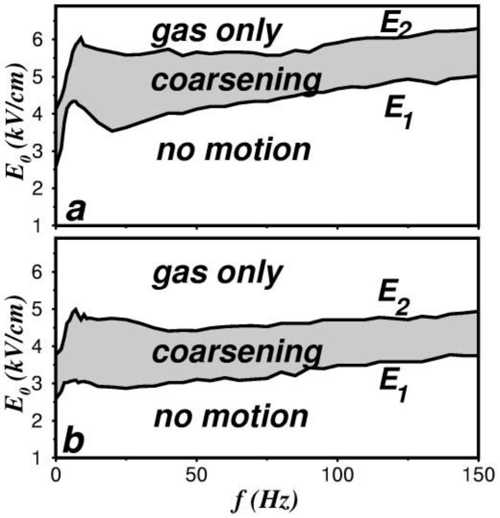

FIG. 2. The phase diagram in the f, E_0 plane for open cell (a) and evacuated cell (b). In *gas only* part clusters do not form; in *coarsening* part clusters and gas coexist.

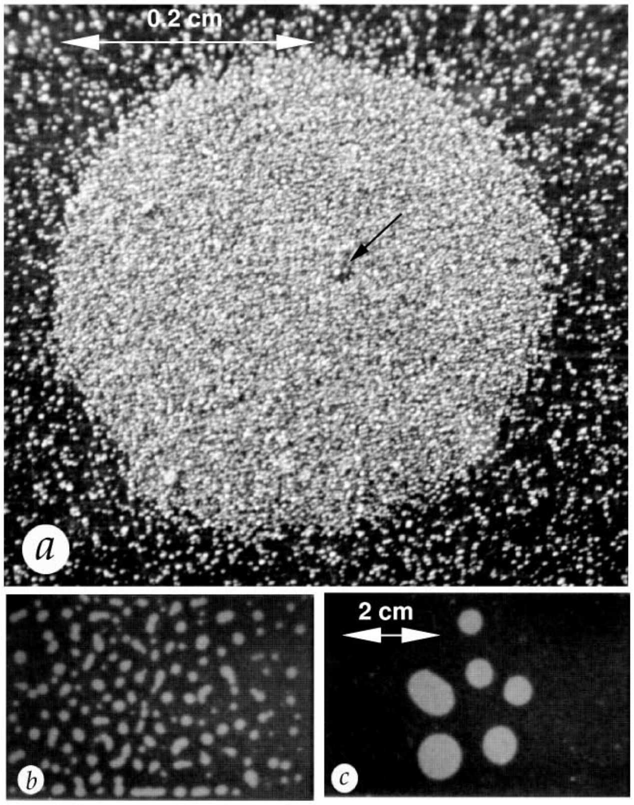

FIG. 3. (a) Close-up image of a single cluster. The larger particle near the center (*arrow*) of the cluster served as the nucleation point. Top view of the cell for (b) $t = 10$ sec, and (c) $t = 109\,10$ sec for $f = 40$ Hz and $E = 5.25$ kV/cm.

decreasing the field below E_2, the gas phase loses its stability and small clusters of immobile particles surrounded by the granular gas form. These clusters then evolve via coarsening dynamics: small clusters disappear and large clusters grow. The number of clusters follows the power law behavior. Eventually the clusters assume almost perfect circular form (Figs. 3a–3c). In the process of coarsening the electric current also changes in time due to the decrease of particles in the gas phase. Approaching the line E_2 from below we observed subsequent decrease in the total area of the solid phase. Approaching E_1 from above the limiting density of the gas phase vanishes and the motion terminates at $E = E_1$. We did not observe hysteresis at the transition lines E_1 and E_2.

A close-up image of one of the clusters is shown in Fig. 3a. Surprisingly, we can even see a larger particle near the center of the cluster which served as the nucleation point at the early stages of the aggregation. Because of its increased mass the large particle is the *first* to become immobile in the lower field. After a very large time (for some parameter values it is as large as $t \approx 300\,00$ sec) all clusters coalesce and a single cluster in the center of the cell survives. The number of particles in the cluster depends upon the value of applied field E_0 and the frequency ω. At the final stage a dynamic equilibrium between the granular solid and the surrounding gas persists; not all the particles join the last cluster.

For the cell under atmospheric pressure (open cell) we found that both fields E_1 and E_2 grow as a function of frequency for large f and show nonmonotonic behavior for $f \approx 12$ Hz. This indicates a characteristic time of the order 100 msec. We suggest that cohesion may be responsible for this relatively large time. Indeed, due to the humidity of the air a surface coating should exist, requiring a characteristic time τ for the grain to detach from the capacitor plate. In order to reduce the cohesion we evacuated the cell to 5×10^{-6} Torr. As demonstrated from Fig. 2b, the frequency dependence is indeed substantially reduced and becomes almost flat, although a rather small peak at low frequency remains. The oscillations in the dependence for low frequency are probably due to a residual coating on the particles which does not completely evaporate in vacuum [10].

We have measured the number of clusters N and the averaged radius of the clusters $\langle R \rangle$ as a function of time t (see Fig. 4) using images taken in time lapse by a digital camera and then processed on a computer. The values of $\langle R \rangle$ for both open and evacuated cells are consistent with a power law $\langle R \rangle \sim \sqrt{t}$. However, there is a substantial difference in the behavior of N. In open cells (no vacuum) we observed slow coarsening $N \sim 1/\sqrt{t}$; whereas the coarsening for the evacuated cell is much faster, $N \sim 1/t$. Remarkably, for the evacuated cell the exponents for N and $\langle R \rangle$ coincide with the exponents for two-dimensional bistable systems in the interface-controlled regime of Ostwald Ripening [11,12]. We speculate that the slowdown in coarsening for open cells can be a result of cohesion between the particles and with the capacitor plate. This

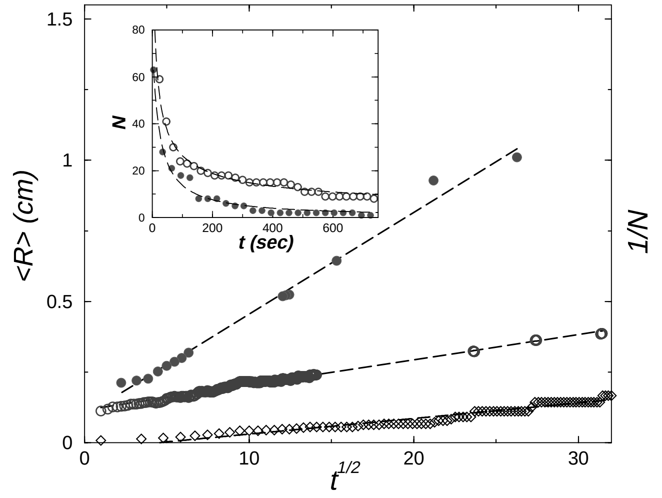

FIG. 4. $\langle R\rangle$ vs $t^{1/2}$ for open cell (○), evacuated cell (●), and $1/N$ for open cell (◇). The conditions were $E = 5.25$ kV/cm and $f = 40$ Hz for open cell and $E = 4$ kV/cm and $f = 8$ Hz for evacuated cell. Dashed lines show linear fit to these curves. Inset: N vs t for open cell (○) and evacuated cell (●). Dashed lines show power law fits $N \sim t^{-1/2}$ and $N \sim 1/t$ correspondingly.

cohesion reduces the mobility of particles at the edges of the clusters, resulting in "pinning" of the front between the granular solid and gas.

Let us compare the forces exerted on an isolated particle and on a particle which is held within a cluster at the same field. The force between an isolated sphere and the capacitor plate in contact can be found as a limit of the problem of two spheres when the radius of one sphere goes to infinity. This problem had been contemplated by several outstanding physicists including Kelvin, Poisson, Liouville, and Kirchhoff [13]. Building on the technique of Ref. [13], we arrive at the force F_e in question:

$$F_e = ca^2E^2, \tag{1}$$

where a is the radius of the sphere and E is the field in the capacitor. The constant $c = \zeta(3) + 1/6 \approx 1.36$ comes from summing of the infinite series derived in [13]. The electric force F_e has to counterbalance the gravitational force $G = 4/3\pi\rho g a^3$, where ρ is the density of the material. Comparing F_e and G we find the first critical field E_1:

$$E_1 = \sqrt{\frac{4\pi\rho g a}{3 \times 1.36}}\,. \tag{2}$$

Our theory indicates no frequency dependence of E_1. For the parameters of our experiments we obtain $E_1 = 2.05$ kV/cm. The experimental critical field is $E_1 \approx 2.4$ kV/cm. This discrepancy seems reasonable since we did not take into account additional molecular and/or contact forces which will increase the critical field.

In order to evaluate the second field E_2 we examine the surface charge distribution in clusters. If the spheres are close to each other, the surface charge will redistribute: each sphere acquires a smaller charge than that of an individual sphere due to a screening of the field by its neighbors. The exact derivation of the force acting on the particle within the cluster is not available at present. The upper bound can be obtained by replacing the square lattice of spheres with radius a by a lattice of squares with area $4a^2$. Since the charge density of the corresponding flat layer is $\sigma = E/4\pi$, the total electric force on the square is $F_2 = 4a^2\sigma E/2$. The ratio of the fields to lift individual particle E_1 and the particle in the square lattice E_2 is $E_2/E_1 = \sqrt{2\pi \times 1.36} = 2.92$. For close-packed hexagonal lattice one obtains a slightly higher value $E_2/E_1 \approx 3.14$. As one sees, the ratio of the critical fields is independent of particle size and the density and the frequency f. This is consistent with the experimental findings (see Fig. 2), although it exceeds the experimental value by a factor of 2. A more accurate account for the surface shape will improve the ratio.

In order to elucidate qualitative features of coalescence dynamics in our system we have performed molecular dynamics simulations of conducting spheres in applied electric field. Several simplifications have been implemented: the sphere polarization was neglected; the charge was assumed to be in the center of the sphere; all collisions were assumed to be inelastic. These simplifications are justified by the fact that the particle-particle collisions are rather rare in contrast to deep vibrated granular layers studied in Ref. [14]. Interactions between the spheres are treated as being between point charges if the distance is smaller than the plate spacing. For larger distances we assumed no interaction due to screening of the far charge field by other particles (compare with Debye screening [9]).

In our molecular dynamics simulations we implemented explicitly the fact that the charge acquired by each sphere from the conducting plate decreases gradually with the number of nearest neighbors: it is maximum for an isolated sphere and decreases by a factor of 2 for the sphere inside the cluster. We simulated three thousand 35 μm diameter copper spheres in the domain 7 × 7 mm (200 × 200 particle diameters) with periodic boundary conditions. The capacitor spacing was 1.5 mm, as it was in the experiment. Although the quantitative correspondence between the molecular dynamics simulations and the experiment was not planned (the number of particles and the size of the system are too small), the simulations turn out to be in a qualitative agreement with the experiment. We have obtained a gas phase for high field levels and spontaneous formation of clusters and coarsening for the lower field level (see Fig. 5). However, detailed simulations with a much larger number of particles and a more realistic account for sphere polarization effects are necessary to achieve quantitative agreement with experiment.

We have reported a hysteretic phase transition accompanied by coarsening behavior in the electrostatically driven conducting granular medium. The origin of coarsening

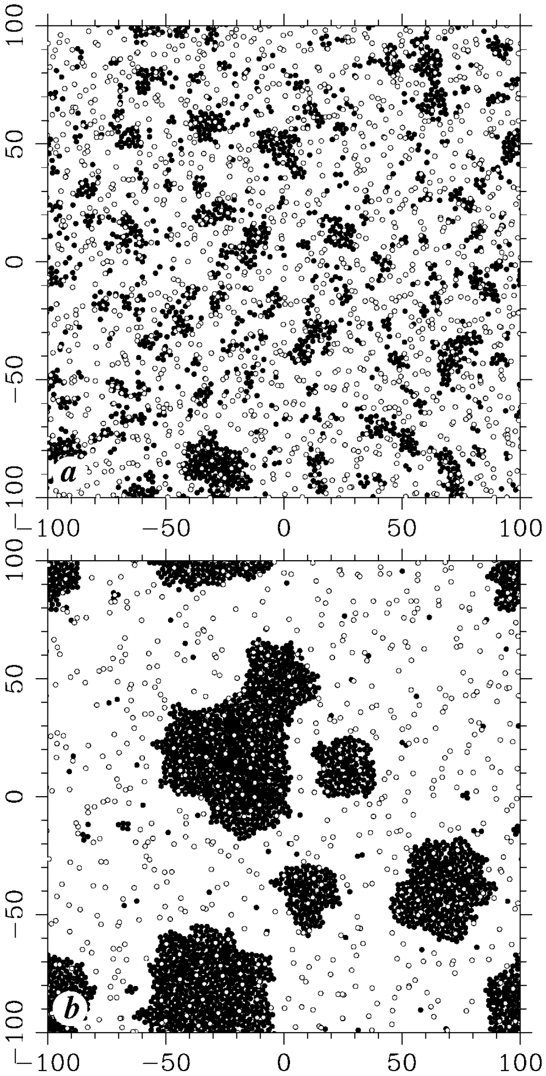

FIG. 5. Top view of simulation domain. Size of the domain is 200 × 200 particle diameters, number of particles is 3000, value of the applied field is $E = 1.1E_1$. The configurations are shown for $t = 1$sec (a) and $t = 10$sec (b). Black bullets correspond to immobile particles in the clusters, open circles correspond to flying particles.

dynamics and hysteresis is due to a screening of the electric field in dense particle clusters. Our results indicate high sensitivity of the phase boundaries to surface coating of the grains due to humidity. Molecular dynamics simulations with conducting particles demonstrate qualitative agreement with the experiment: existence of two critical fields and coarsening.

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