# Liquid-Gas Phase Separation in Confined Vibrated Dry Granular Matter

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A new phase transition is observed experimentally in a dry granular gas subject to vertical vibration between two horizontal plates. Molecular dynamics simulations of this system allow us to investigate the observed phase separation in detail. We find a high-density, low temperature liquid, coexisting with a low-density, high temperature gas moving coherently. The importance of the coherent motion for phase separation is investigated using frequency modulation.

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Finding the general principles determining the formation of collective steady states in systems far from thermal equilibrium can be regarded as one of the fundamental unsolved problems of statistical physics. In this context, driven granular gases have received growing interest in recent years as model systems for complex collective behavior [1]. Phase-transition-like phenomena have been found in many granular systems subject to vibration, both experimentally and in simulations [2–11]. Vertically vibrated confined monolayers exhibit distinct solid- and fluidlike phases which are analogous to those of an equilibrium hard-sphere gas [3]. Solid-liquid coexistence of granular matter has also been observed experimentally in a similar geometry [4,5]. These experiments show that phase separation arises due to spinodal decomposition, and have been explained using simulations involving thermalized boundary conditions and deeper beds [6,7]. However, recent simulations of confined monolayers suggest that phase separation does not occur if the system is driven by random forcing [8]. A detailed understanding of the role played by vibration and confinement on granular phase separation is still lacking.

In this Letter, we report on a novel liquid-gas phase separation phenomenon in a vertically vibrated, confined, dilute, granular system. We have carried out experiments using a cell containing glass spheres which we observe to phase separate under vertical vibration as the amplitude is increased beyond a threshold. The phase diagram has been determined as a function of volume filling fraction and driving amplitude. To understand this behavior in some detail we have carried out simulations using a simplified model which is able to reproduce the experimental results. The simulations suggest that phase separation is driven by spinodal decomposition; the spinodal arises from the sudden crossover from a thermalized liquid to a coherently moving gas. This behavior is clearly distinct from previously known separation mechanisms, such as clustering induced by inelastic collisions [12] or the presence of cohesive forces between grains [9,10].

Our experimental apparatus consists of a cell containing glass spheres which can be vibrated vertically using an electromagnetic shaker. The cell is constructed out of a lower, anodized aluminum plate on top of which there is attached a square aluminum frame (interior side 13 cm) and an upper, square, glass plate. The gap between the bottom and top plates could be varied by changing the metallic frame: we used gaps, $h$, of 0.5 cm, 0.75 cm and 1 cm. The particles used in the experiments were slightly polydisperse spherical glass spheres (Ballotini) of mean diameter $d = 610\ \mu$m. The cell was rigidly attached to an electromagnetic shaker (Ling-Dynamics V406) and driven by a sinusoidal signal from a function generator. The vertical motion was monitored using a capacitance cantilever accelerometer. Experiments were carried out over the frequency range $f = 23$ to 65 Hz. The dimensionless driving acceleration $\Gamma = A(2\pi f)^2/g$ was varied between 1 and 18, where $A$ is the amplitude of the driving and $g$ is the acceleration due to gravity. Care was taken to ensure that the cell was levelled prior to each experimental run. The construction of the cell and the controlled environment in the laboratory minimized charging effects of the particles. The particles and the cell were cleaned and dried in an oven, and a certain volume of particles was then poured into the cell. We define the volume filling fraction, $\bar{\phi}$, as the volume of particles in the cell divided by the volume of the cell. The cell was then closed and vibrated at a fixed frequency and the amplitude was varied.

Figure 1 shows typical snapshots of the system for different values of $A$. As $A$ increases, the initial homogeneous state separates into a low-density (dark) and high-density (bright) region. For each $A$ the pattern is stationary apart from small fluctuations. For higher $A$ the particles eventually spread out homogeneously. The appearance of two-phase coexistence is detected by visual inspection, and the corresponding amplitude was reproducible within experimental error. No crystalline order in the high density phase was visible. We found that the amplitude at which phase separation first occurred was independent of vibration frequency within the range investigated, 23 Hz $< f <$ 65 Hz (cf. inset of Fig. 2). We also note that a small amount of hysteresis was found for some of the phase boundaries.

 

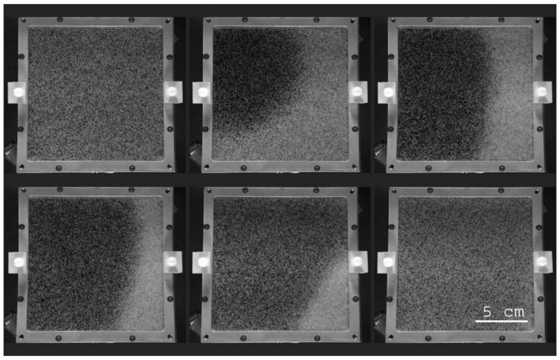


FIG. 1. Snapshots showing the cell containing the glass spheres (bright) viewed from above. The volume filling fraction occupied by the particles was $\bar{\phi} = 0.059$. The cell was vibrated at $f = 45$ Hz and from top left to bottom right the maximum acceleration $\Gamma$ is 5.9, 7.5, 9.0, 10.6, 12.0, and 13.7 respectively; the corresponding amplitudes, $A/d$, are 1.19, 1.51, 1.81, 2.13, 2.41, and 2.76. In the first snapshot (top left) the particles are distributed in the horizontal plane homogeneously throughout the cell. As $A$ is increased a dilute region suddenly appears at the top left corner in the system (top middle). This dilute region expands as $A$ is increased further (top right, bottom left and bottom middle). Eventually at large enough $A$ the system returns to a homogeneous state (bottom right).

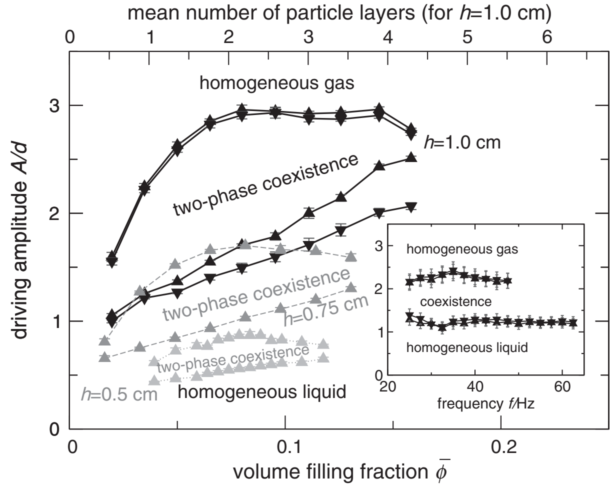


FIG. 2. Experimental phase diagram in the space of the dimensionless amplitude, $A/d$, and volume filling fraction, $\bar{\phi}$, for three gap heights, $h$: 0.5 cm (dotted), 0.75 cm (dashed) and 1.0 cm (solid). The lines show the approximate boundary of the coexistence region obtained by increasing (triangle up) or decreasing (triangle down) the amplitude at fixed filling fraction. We only show the hysteresis for the system with the largest gap. The upper horizontal axis shows the volume filling fraction expressed as the mean number of particle layers, assuming a hexagonal close packing, for $h = 1.0$ cm. The inset shows the appearance and disappearance amplitude for phase separation as a function of frequency at a filling fraction of $\bar{\phi} = 0.035$. For large enough frequencies it can be seen that the appearance and disappearance of phase separation is almost independent of frequency.

When the system phase separates the particles in the dilute region always hit the top plate of the cell during their motion.

Figure 2 shows the experimental phase diagram. For a given gap between the bottom and top plates, the conditions for phase separation depend on both the amplitude and the filling fraction, but are independent of frequency. Current theoretical understanding of phase separation in vibrated granular media is generally based on systems driven by thermalized boundary conditions with zero amplitude [6,13,14]. In order to gain an understanding of our system we have to carry out simulations which include the *plate motion* explicitly.

We used event-driven simulations to model the motion of grains enclosed in a geometry similar to that of the experimental cell. We assume that the particles are hard spheres of mass $m$ and mean diameter $d$, and slightly polydisperse (uniformly distributed with a standard deviation of 6% of $d$, similar to the experiment). They are confined by an upper and lower plate with a gap height of $16d$ and a side of $210d$, and move in three dimensions under the influence of gravity. The ratio $h/d$ is similar to the experiment corresponding to the black curve in Fig. 2. Collisions between particles dissipate energy with a velocity-independent coefficient of restitution which was taken to be 0.9 in all simulations presented here. Collisions with the top and bottom plate of the cell are assumed to be elastic. To drive the system, the top and bottom plates are moving sinusoidally in the vertical direction as in the experiment, while the system is assumed to be periodic in the horizontal directions. We define the granular temperature, $T$, to be the mean kinetic energy per particle determined from the horizontal components of the velocity. Two-phase coexistence is identified by a significant difference in the maximum and minimum of the time-averaged local densities.

Figure 3 shows the phase diagram obtained from simulations. For low driving amplitudes the system remains homogeneous; for intermediate values of $A$ and $\bar{\phi}$ there is a two-phase coexistence (orange shaded region); for large $A$ the system becomes homogeneous again. The simulation predicts a similar crescent-shaped coexistence region to that observed experimentally. Some simulations were carried out for different gap heights; the corresponding variation in the phase diagram was qualitatively similar to that found in experiment. We attribute quantitative differences to other dissipation channels, such as inelastic collisions with the base, sliding friction and rotational degrees of freedom, which have been neglected in the simulation. Simulations also do not include air effects; we have carried out experiments down to an air pressure of 0.03 Torr and still observed the phase separation. We also note that even though the particles are slightly polydisperse no size segregation is observed.

As in experiment, the phase diagram is found to be independent of frequency. As a further test, we have carried out simulations in which $g$ was set to zero. No significant change was found in the simulation results. The frequency only sets the time scale of the dynamics and does not influence the steady state configuration. As a result, the phase diagram is independent of frequency.

The snapshot in Fig. 3 shows two-phase coexistence in a simulation. In the phase-separated state the interface between the phases is either circular or forms a stripe (due to the presence of periodic boundary conditions); these configurations suggest that the interface has an effective surface tension, even though there is no attractive force between particles. Some simulations were run with solid reflective sidewalls and no significant change of the phase diagram was found. Simulations also allow us to measure the pair-correlation function. In the dilute phase the pair-correlation function decays monotonically, whereas in the dense phase short-range order is visible. This behavior is reminiscent of a classical gas and liquid; therefore we use the same terminology here. In neither phases is there any evidence of long-range ordering.

In the phase-separated state, we can measure the local density $\phi$ in the two different phases. These are shown in Fig. 3 as filled circles. The locus of the circles defines a "binodal line" surrounding the coexistence region. In order to understand the mechanism responsible for the phase separation, we performed simulations in a system with a small horizontal area. The small box allows for the measurement of the horizontal component of the pressure tensor, $p$, under conditions which would otherwise give rise to phase separation in a large system [6,15]. The inset of Fig. 3 shows $p$ as a function of $\bar{\phi}$ for three different amplitudes in the small system. From such simulations the range of values of $A/d$ and $\bar{\phi}$ where we find a negative compressibility, the "spinodal region", can be determined and is given by the triangles in the main panel of Fig. 3.

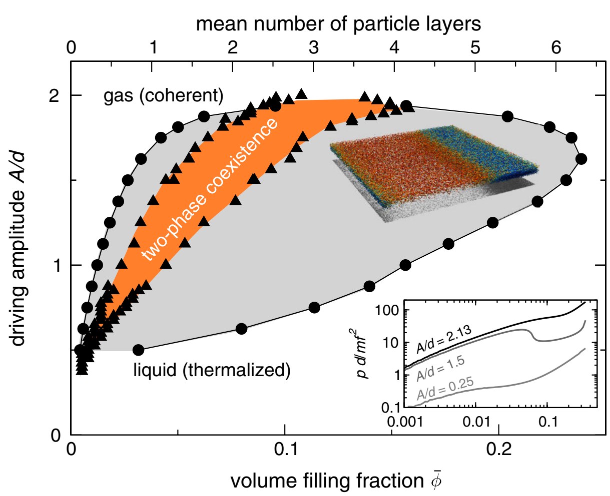


FIG. 3 (color online). Phase diagram in the space of $A/d$ and $\bar{\phi}$ obtained from simulations of the large system. The orange shaded region shows where phase separation occurs, while outside that region the system remains homogeneous within the time scales simulated. For systems which phase separate, the locally coexisting densities $\phi$ are given as black circles. They form a "binodal line". The black triangles, confine the region of negative compressibility (the spinodal region) obtained by the simulations of the small system. The inset shows the pressure, $p$, as a function of $\bar{\phi}$ from simulations of a small system for three different amplitudes. The intermediate amplitude shows a negative compressibility for a range of values of $\bar{\phi}$. The snapshot shows a phase-separated system for $A/d = 1.5$ and $\bar{\phi} = 0.085$, where the color indicates high (red) and low (blue) kinetic energy of each particle and the shadow underneath is a measure of the local volume filling fraction, where the dense phase appears darker. The coexisting temperatures can be found in the inset of Fig. 4 as black circles.

It has been suggested [6] that a spinodal can arise if $T$ decays sufficiently strongly with filling fraction. In the inset to Fig. 4 we show the mean value of $T$ as function of $\bar{\phi}$ for three different amplitudes. For the amplitudes shown, phase separation is only present for the intermediate value $A/d = 1.5$; phase separation occurs for the range of $\bar{\phi}$ for which $T$ decreases rapidly. What is the cause of this rapid decay?

The main panel of Fig. 4 shows the dependence of granular temperature on amplitude for three different volume filling fractions in a small system which does not phase separate. The amplitude dependence appears to exhibit two distinct regimes with a rapid rise (kink) in granular temperature between them. As $\bar{\phi}$ is increased the shift of the kink to higher amplitudes results in the rapid decay of $T$ with $\bar{\phi}$ shown in the inset. For example, for an amplitude for which phase separation is observed in the large system, $A/d = 1.5$ (dotted line), by increasing $\bar{\phi}$ we move from one regime to the other with a sudden decrease in temperature. The lower and upper volume filling

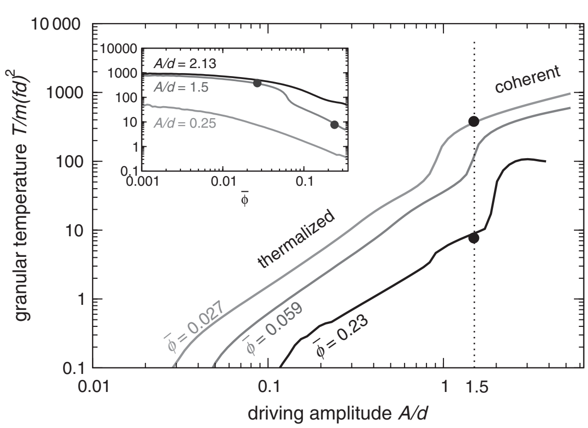


FIG. 4. Granular temperature $T$ as a function of amplitude $A/d$ for three different volume filling fractions $\bar{\phi}$ for a small system which does not phase separate. The curves exhibit two distinct regimes and a rapid rise in $T$ between them. The black circles show the temperature of the coexisting phases obtained in the large system. They lie on different branches of the $T(A)$ curves which correspond to different regimes. The inset shows $T$ as a function of $\bar{\phi}$ for three different amplitudes. Only for the intermediate value, $A/d = 1.5$, shown does phase separation occur due to the presence of the rapid drop in $T$ with $\bar{\phi}$. The circles show the coexisting values of the local temperature $T$ and the local density $\phi$. The amplitude $A/d = 1.5$ is marked in the main panel as a dotted vertical line.

fractions shown were chosen to be the coexisting values for this amplitude in the large system. The figure also shows the temperatures of the two coexisting phases extracted from simulations of the large system (circles). It can be seen that these circles lie on branches of the curves corresponding to the two different regimes.

In simulations we can study in detail the dynamics of the grains, for a given amplitude, in the two different regimes at high and low volume filling fractions. In both regimes energy is injected through collisions with the plates and dissipated by collisions in the bulk. The dense phase looks like a thermalized liquid for which the granular temperature scales as $A^2$ which is approximately recovered in Fig. 4 for small amplitudes. The injected energy is quickly dissipated within the bulk due to the large volume filling fraction. During each cycle shock waves propagate away from the lower and upper plates into the bulk increasing the number of dissipative collisions [16]. In contrast, in the dilute phase a statistically significant number of particles can propagate from one plate to the other without undergoing collisions. The motion of these particles becomes synchronized with the driving and energy is picked up more efficiently due to this resonance [17]. The dissipation is reduced by two mechanisms: there are fewer particles than in the dense phase and the coherent motion reduces the relative velocity of particles undergoing collisions. As a result the temperature of this phase is significantly higher than that in the dense phase under the same driving conditions. It is these two types of dynamical behavior which give rise to the different regimes shown in Fig. 4.

Finally, as a test of the importance of coherent motion for this form of phase separation we have carried out experiments in which the sinusoidal waveform was frequency modulated. As the phase diagram is independent of the driving frequency, one might expect that such a frequency variation should not strongly influence the separation because the amplitude can be kept fixed. We have used a driving waveform given by $A\sin[2\pi ft + (\Delta_f/f_m)\times \sin(2\pi f_m t)]$, where $\Delta_f$ is the peak deviation and $f_m$ is the modulation frequency. By varying $f_m$ and $\Delta_f$ we can find conditions under which the phase separation is suppressed. For example, if we choose $f_m = f/2$ the effect of varying $\Delta_f$ is to shorten and lengthen every alternate driving cycle by a small fixed amount. For $f = 40$ Hz, $f_m = 20$ Hz, $A/d = 2.34$, $\bar{\phi} = 0.099$ and $h = 1$ cm phase separation is switched off for $\Delta_f >= 12.3$ Hz. We have also checked that for sufficiently large frequency modulation we do not observe phase separation for the range of driving amplitudes and volume filling fractions shown in Fig. 2. These findings indicate that if the resonant motion of a single particle is successfully suppressed the collective coherent motion is destroyed and phase separation disappears.

We have investigated a granular system which exhibits a novel form of phase separation. The great virtue of our system is that one can sweep through the phase diagram simply by changing the driving amplitude. This simplicity opens up the possibility of investigating critical point phenomena, the corresponding critical exponents and the dynamics of spinodal decomposition, all in a nonequilibrium setting. Further study of this system may lead to new insights into the fundamentals of nonequilibrium statistical physics.

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