Structure formation in electromagnetically driven granular media

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We report structure formation in submonolayers of magnetic microparticles subjected to periodic electrostatic and magnetic excitations. Depending on the excitation parameters, we observe the formation of a rich variety of structures: clusters, rings, chains, and networks. The growth dynamics and shapes of the structures are strongly dependent on the amplitude and frequency of the external magnetic field. We find that for pure ac magnetic driving at low densities of particles, the low-frequency magnetic excitation favors clusters while high frequency excitation favors chains and net-like structures. An abrupt phase transition from finite length chains of particles to infinite networks as the driving parameters are varied.

The experimental setup is shown in Fig. 1. The design of the electrostatically driven cell is similar to that reported earlier. Conductive particles are placed between two horizontal transparent conducting glass plates (12×12 cm with the spacing of 1.5 mm). An external magnetic field is provided by a set of large magnetic coils (30 cm in diameter) placed around the cell. To excite the granular media contained in the cell, a voltage of 0 - 2 kV with a frequency of 0 - 150 Hz is applied to the plates. The magnetic interaction is controlled by supplying a dc electric current to one of the coils to create a constant magnetic field in the range of 0-80 Oe. In addition, pure "magnetic" driving of the particles is carried out by applying an ac current to a second coil with small or no electric field applied to the cell. The amplitude of the ac magnetic field can be varied from 0-15 Oe with the frequency from 0 to 300 Hz. The granular media used in our experiment consisted of spherical Nickel microparticles with average size of about 90 µm (Alfa Aesar Company). Magnetic moment per particle at the 80 Oe field is 1·10⁻⁵ emu; saturated magnetic moment is 2·10⁻⁴ emu per particle, saturation field is about 4kG. The number of particles was varied in the range 120,000 – 300,000. Experiments were also performed with 40 µ sized particles, but no qualitative difference was found. The experiments were carried out in air and in non-polar low-viscosity liquid (toluene). Real time images were acquired with a high-speed digital camera.

Electrostatic driving works as follows. Conducting particles acquire an electric charge when they are in contact with the bottom conducting plate of the cell. When the magnitude of the electric field in the cell exceeds some critical value, the upward electric force acting on the charged particle overcomes the gravitational force and...
the particles are driven towards the upper plate. Upon contact with the upper plate, the particles recharge and fall back to the bottom plate. This process repeats in a cyclical fashion. The elevation of the particles off the bottom plate can be adjusted by the frequency of the applied ac electric field. Pure “magnetic” driving works in the following way. Magnetic particles with moment $\mathbf{M}$ subjected to an external magnetic field $\mathbf{H}$ experience a torque $-\mathbf{M} \times \mathbf{H}$ forcing the magnetic moment of particles to be aligned with the applied magnetic field. There are, however, two ways in which the magnetic moment of the particle can adjust its direction: (i) the magnetic moment can rotate inside the particle against the internal magnetic anisotropy field and (ii) the whole particle can rotate in order to keep the moment aligned with the applied magnetic field. In the latter case, the magnetic particle needs to overcome the resistance from the friction and adhesion forces between the particle and surface of the plate. Thus, as the magnitude of the external magnetic field exceeds some critical value, each particle begins to rotate by keeping its moment aligned with the field and moves being driven by magnetic drag force $\mathbf{F}_m = M \nabla H_{\text{local}}$ where $H_{\text{local}}$ designates the local magnetic field coming from dipolar fields of the neighboring particles and the external magnetic field. The effective time the particle can move may be controlled by adjusting the frequency of an ac magnetic field.

As reported earlier, [5, 6] the particles remain immobile on the bottom plate of the cell if the applied electric field is less than some critical value $E_1$. When the field exceeds a second threshold value, $E_2 > E_1$, the system undergoes a transformation to a gas-like state. When decreasing the electric field $E$ below $E_2$ ($E_1 < E < E_2$), small densely packed clusters begin to nucleate. Clustering is promoted through electrostatic screening (two particles in contact acquire a smaller charge than two well-separated particles). In the case of magnetic particles, an additional complication arises from the dipole-dipole magnetic interactions and the dynamics of cluster formation and critical electric fields $E_1, E_2$ are expected to change. The phase diagram delineating the primary experimental regimes as a function of applied voltage and external magnetic field for 200,000 nickel particles with average size of 90 µm (about 9% monolayer coverage) is shown in Fig. 2. The frequency $f$ and amplitude $V$ of the ac electric field applied to the cell were chosen to provide quasi-two dimensional motion of the particles ($V<1000V$, $f = 100Hz$). The cell was subjected to an external dc magnetic field from 0 to 80 Oe.

The behavior of the magnetic particles in low external magnetic fields (below 20 Oe) is rather similar to that reported for nonmagnetic conductive particles. The isolated particles are immobile until the electric field exceeds a critical value $E_1$. At electric field values above $E_2$, the granular medium transforms into a uniform gas-like phase. Cluster formation is observed in the interval, $E_1 < E < E_2$, in agreement with earlier studies on nonmagnetic granular systems [5, 6]. However, the clustering dynamics of the magnetic particles is very sensitive to the external magnetic field. Clusters formed in the absence of a magnetic field (a) and at a magnetic field of 10 Oe (b) are shown in Fig. 3. The cluster grown in an external magnetic field looks less compact and sufficiently less smooth in shape. Moreover, cluster formation in an external magnetic field is characterized by arrested coarsening due to depletion of the gas phase.

We analyzed the growth rates to characterize the time evolution of the clusters at different magnetic fields. We find that the change of the cluster size for both zero and nonzero external magnetic fields can be well described by

![FIG. 1: Schematic view of the experimental setup.](image1)

![FIG. 2: Phase diagram in the magnetic field, voltage plane for 90µm Nickel particles in the air filled cell. Surface fraction of particles is 9% (≈200,000 particles)](image2)
a simple equation:

\[ S(t) = S_\infty (1 - \exp \left(-\frac{(t - t_0)}{\tau}\right)) \]  

Here \( S_\infty \) is the cluster size at \( t \to \infty \); \( t_0 \) is the time when the cluster nucleates; \( \tau \) is the growth time. The quantity \( 1 - S(t)/S_\infty \) for the clusters formed at zero field and at 10 Oe is plotted in Fig. 3. Solid lines in Fig. 3 represent the functional dependence of Eq. (1) with the corresponding characteristic growth times. The larger growth time \( \tau \) in the presence of an external magnetic field could be explained as follows: since the particles are large enough to contain multi-domains, the higher external magnetic field induces larger magnetic moments in the particles (due to the growth of magnetic domains within the particle with orientation along the external field) leading to amplification of the dipole-dipole interactions. Consequently, the interaction between particles becomes highly anisotropic and increases the characteristic growth time since (a) aggregation proceeds predominantly through coalescence of chain segments which are less mobile; (b) some particles are repelled from the cluster due to unfavorable magnetic moment orientation. For elevated magnetic fields (20-45 Oe) the formation of immobile vertical chains consisting of two to four particles were observed when the amplitude of the electric fields was reduced below \( E_1 \). The threshold electric field \( E_1 \) decreases with increasing external magnetic field since the formation of immobile chains diminishes the effective spacing between cell plates resulting in higher electric field intensity for particles at the top parts of the chains. Upon exceeding \( E_1 \) chains start to move and form localized clusters of chains. At amplitudes of electric field above \( E_2 \), the system transforms to a gas of short chains. Increase of the magnetic field above 50 Oe creates static long vertical chains touching the upper plate of the cell.

We explored pure ac magnetic driving by placing the magnetic particles into a cell filled with toluene to dampen the kinetic energy in the system. Before each experiment, the system was driven to a gas-like state with an ac electric field in zero magnetic field to create a uniform distribution of particles over the cell. Subsequently, the electric field was turned off and the system was subjected to ac 15 Oe magnetic field with frequency from 0 to 200 Hz. The magnetic dipole-dipole interaction favors completely different particle organization. Fig. 3 (upper part) demonstrates some selected structures.

The nature of the self-assembled structures is quite straightforward. Since the local arrangement is dominated by a highly anisotropic dipole-dipole magnetic interaction, the head-to-tail interaction is the strongest, favoring formation of the quasi-one dimensional chain structures. Each chain has a certain rigidity against bending since in a bent chain the dipoles are not fully aligned. When the bending angle exceeds some threshold value, the energy barrier to bending starts to decrease since closing of the chain now becomes more favorable due to the attraction of the opposite ends of the chain. It can be shown that a ring is more energetically favorable than a chain if the number of particles in the chain exceeds four. However, to form a ring from a chain, one needs to overcome a strong potential barrier associated with chain bending. Consequently, a more plausible mechanism for forming a ring is a "fusion" of several short chain segments of appropriate orientation, polarization and position, see Fig. 4. Chains can also form compact structure, such as a cluster of dipoles. Such dipole clusters can nucleate in the situation when two neighboring parallel chains have opposite polarization and attract each other to form a cluster.

One can vary the frequency of the ac magnetic field to control the time it takes the particles (or chains) to rotate in order to keep its moment aligned with the local field and to move along the local field gradient. If the frequency is too high (200 Hz in our case), nothing hap-
pens since the characteristic reaction time of the system is higher than the period of the ac magnetic field. At some point while decreasing the frequency, the particles start to react to the external ac magnetic field and short chains appear in the system. A further decrease of the frequency leads to the creation of more energetically favorable configurations such as rings and branched chains.

Compact clusters as shown in Fig. 4 can be initiated by decreasing the frequency of the ac magnetic field. The bottom panel of Fig. 4 shows various patterns formed under the influence of an external ac magnetic field at 20, 50, and 100 Hz for a cell with 5.3% surface coverage of Ni particles. Each experiment was started from randomly dispersed configuration of the nickel spheres over the bottom plate of the cell. Clearly, the pattern is determined by the frequency of the external ac magnetic field: low-frequency excitations (0-30 Hz) produce a clustered phase; high-frequencies (80-200 Hz) assemble short chains while intermediate-frequencies favors net-like patterns. The resulting pattern is strongly history dependent. For example, a continuous decrease of the magnetic field frequency from 200 to 10 Hz results in the formation of a net-like structure, in contrast to the formation of clusters obtained when the system "relaxes" at a constant frequency of 10 Hz. Denser configurations ($\phi$=0.115 and 0.133), however, do not exhibit the cluster phase and instead, a phase transition to the network phase (infinitely long multi-branch chains) is observed at low frequencies. To demonstrate the phase transition to the network phase, the average chain length as a function of the ac magnetic field frequency is plotted in Fig. 4 for different densities. Data was taken after the system was left to "relax" in the applied field for about 10 minutes to attain their respective equilibrium state. As the frequency of the magnetic field decreases, the average length of the chains tends to diverge for dense configurations ($\phi$=0.115 and 0.133) and saturates for less dense ones. The solid lines in the figure are fits to the expression $y = A + B/(x - f_0)$, where $f_0$ designates a critical frequency when the system undergoes a phase transition to the net-like phase. The critical frequencies extracted from the fits are 14.10 Hz, and 6.11 Hz for $\phi$=0.133, and $\phi$=0.115, respectively. The critical frequency for surface coverage of $\phi$=0.07 resulted in a negative value, $f_0$=-5.62 Hz, indicating that there is no transition to the network phase. Indeed, for such low density configurations, compact clusters were observed at low frequencies.

Inset to Fig. 5 demonstrates the effect of an ac electric field on the system of 240000 Ni particles ($\phi \sim 0.10$) in ac magnetic field ($H_{max} = 15$ Oe; $f$=25 Hz). The frequency of the electric field was kept at 100 Hz to provide mostly two-dimensional motion. The average length of the chain decreases with increasing ac electric field, suggesting that the latter acts as an analogue to temperature.

We studied the self-assembly of magnetic microparticles in ac electric and magnetic field. Excitation of the system by an ac magnetic field revealed a variety of patterns that can be controlled by adjusting the frequency and the amplitude of the field. We found that at low particle densities the low-frequency magnetic excitation favors cluster phase formation while high frequency excitation favors chains and net-like structures. For denser configurations an abrupt transition to the network phase was observed. We thank Ulrich Welp for help with magnetization measurements. This research was supported by the US DOE, grant W-31-109-ENG-38.

\[\text{See EPAPS Document No. or http://mi.msd.anl.gov/SelfAssembly/paper.htm for movies illustrating formation of structures. A direct link to this document may be found in the online article's} \]
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