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## Phase transitions of granular disks with a magnetic dipole

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We present a simple experiment to study phase transitions of a granular system composed of disks with a magnetic dipole. The particles are contained within a cylindrical container and are excited through alternating magnetic fields. The initial configuration of this system is a low-density isotropic phase. By increasing the density, a critical value is reached and the system starts to organize into columns. At higher densities an ordered state is obtained where structural columnar patterns are formed. The set up of the experimental device is very easy to implement, using standard high-school and undergraduate physics laboratory facilities. The experiment can be used, combined with computer simulations, to introduce the subject of phase transitions in complex materials.

**Keywords:** Granular media; liquid crystals; Monte Carlo simulations.

Presentamos un experimento simple para estudiar transiciones de fase de sistemas granulares compuestos de discos con un dipolo magnético. Las partículas están contenidas en un recipiente cilíndrico y son excitadas por medio de campos magnéticos alternos. La configuración inicial de este sistema es una fase isotrópica de baja densidad. Al incrementar la densidad se alcanza un valor crítico y el sistema comienza a organizarse en columnas. A densidades mayores se observa un estado ordenado con patrones de orden columnar. El arreglo experimental es muy fácil de implementar, utilizando instalaciones típicas de laboratorios de enseñanza media superior y superior. El experimento se puede utilizar, combinado con resultados de simulación computacional, para introducir el tema de transiciones de fase en materiales complejos.

**Descriptores:** Medios granulares; cristales líquidos; simulaciones de Monte Carlo.

PACS: 75.70.Kw, 64.70.mf, 64.70.mj

### 1. Introduction

The study of phase transitions and the understanding of the molecular mechanisms involved in these phenomena is one of the most exciting research areas in statistical physics. This subject has a wide scope of interests, from the understanding of the different physical and chemical processes determining these transitions and the unified approaches developed to explain them, up to the associated technological innovations, such as devices based on magnetic or electric transitions in fluids, crystals, liquid crystals, etc.

In the area of liquid-crystalline phenomena, novel materials have been obtained based on the application of specific phase transitions. Nematic, smectic, chiral and columnar phases of organic molecules are novel phases that can be used to produce different types of electronic devices [1]. The teaching of the subject of phase transitions at an undergraduate level is rather limited, since there are a reduced number of demonstrative experiments and simulations that can be performed in standard elementary laboratories focused on these phenomena. Liquid crystal materials can be used to show this subject [2–4].

On the other hand, granular systems formed by a large number of particles can display complex collective behavior. They are also useful to study phase transitions [5–9]. Distinct patterns have been observed experimentally in vertically vibrated rod-shaped granular materials [10–15], and vertically vibrated monolayers of magnetic spheres [16–18]. Although it could seem inappropriate to use granular media to introduce analogies with molecular systems, since granular

systems are dissipative and the thermal effect is introduced mechanically, some of their features have a strong similarity with their microscopic counterpart.

In this paper we present a simple device to study phase transitions, using granular magnetic disks excited through alternating magnetic fields. We perform a qualitative study of the evolution of the system when the number of disks is varied, and we discuss its analogy with the statistical behavior of a system formed by molecules. The competing interactions between particles determine different structured phases and abrupt changes of the structural organization. The behavior of macroscopic disks resembles phases observed with fluids composed of discotic molecules [19–22]. All the materials required for the device can be easily obtained, and it can be built with standard tools and materials.

This paper is organized as follows: In Sec. II we present the experimental details and procedure to perform the study. Then we describe in Sec. III the observed patterns obtained, and in Sec. IV a full discussion of the associated phenomena, including a comparison with molecular simulations of discotic fluids, is given. Section V contains a summary of the findings and conclusions of this work.

### 2. Experimental details and procedure

The granular anisotropic particles are obtained from a 0.5 mm width flexible magnetic vinyl sheet, that is perforated using a handheld one-hole punch. Each disk has a 6 mm diameter (Fig. 1a). A transverse magnetic dipole can be induced on

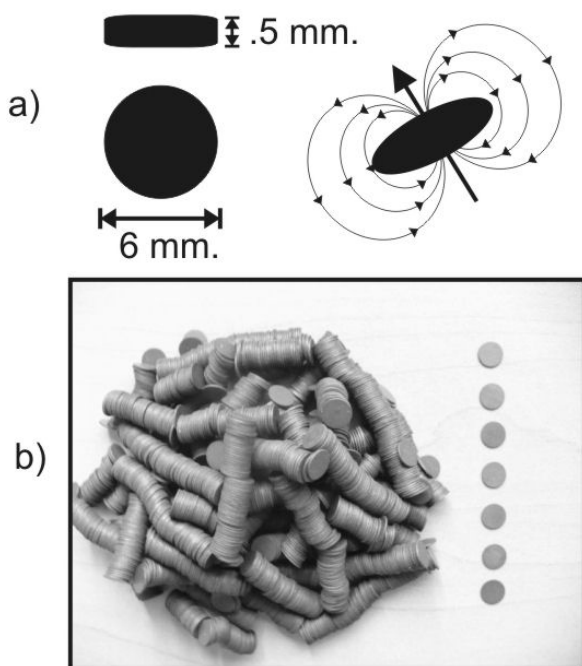


FIGURE 1. a) Anisotropic particle with magnetic dipole; b) Formations of columnar aggregates using magnetic disks.

each disk by placing the disks over the planar surface of a magnet. The magnetic disks can aggregate to form columns, as we can see in Fig. 1-b, and can be excited through a device that produces alternating magnetic fields, as shown in Fig. 2a. This device is made of an electric motor (Fig. 2a, (1)) with a rectangular plate with two 0.35 T square magnets with inverted poles (Fig. 2a, (2)), in order to generate alternating magnetic fields by spinning the motor. The rectangular plate has the dimensions:  $10 \times 1.5 \times 1.5$  cm. The rotation frequency of this motor is tuned through a dimmer (Fig. 2a, (3)), and its maximum value is 27.5 Hz. Disks are contained within a cylindrical vessel with a transparent cap (Fig. 2b), which helps to maintain a constant number of particles. The diameter of this vessel is 15 cm, and its height is 10 cm.

The experiment is described as follows. First, the electric motor is turned on and its spinning frequency is controlled by the dimmer. Second, 100 particles are added to the container and the system evolves by a 5 minute interval. Third, several photographs are taken from the observed configurations. This procedure is repeated adding 100 particles at regular intervals of time, until 2300 particles are introduced in total. In this way, a series of photographs are obtained with an increasing number of particles.

### 3. Observed patterns

The experiments were performed starting with 100 particles (Fig. 3a), and with a constant spinning frequency. The concentration of particles was changed at regular intervals of time. When 300 particles were reached, the disks were ran-

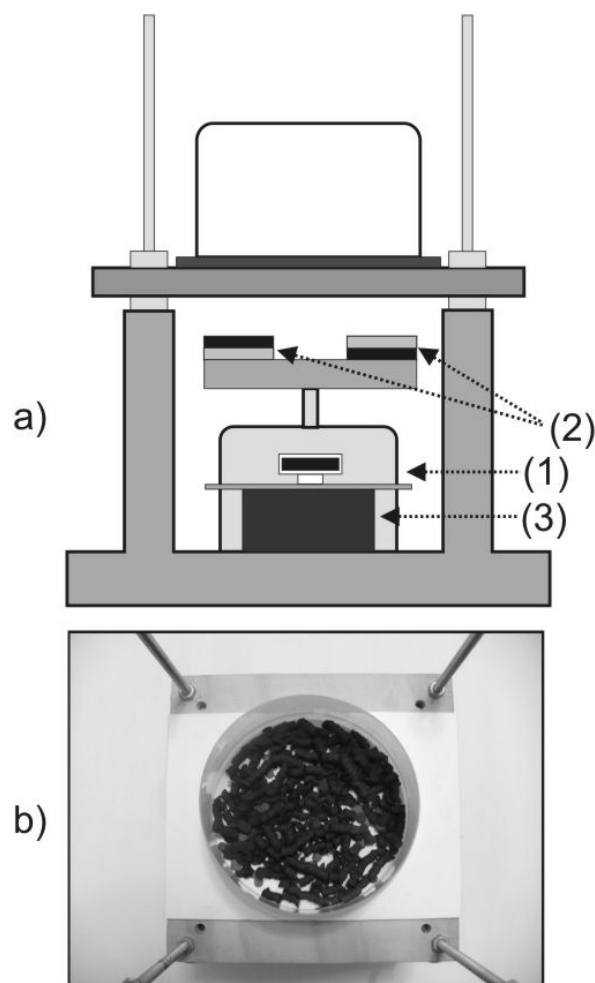


FIGURE 2. a) Diagram of the device that produces alternating magnetic fields; this is made of an electric motor (1) with a rectangular plate with two 0.35 T square magnets with inverted poles (2), in order to generate alternating magnetic fields by the motor's spinning. The rotation frequency of this motor is tuned through a dimmer (3); b) Image of the disks within the container.

domly orientated and some small clusters began to form, (Fig. 3b). This state would correspond to a gas phase in a molecular system, where there is not a strong correlation between particles, and the kinetic energy of the molecules (disks in this case) dominates over the clustering caused by the intermolecular potential (magnetic and hard-body interactions between disks). Small columns were observed when the size of the system was 400 particles; each column was formed by 10 disks on average. This aggregation helped us to conclude that the dipolar moment for each disk was transversally oriented to the disk plane. For 700 particles, the size of the columns increased up to 20 disks, and the density of columns was high enough (Fig. 3c). This state resembled a pre-transitional phase, with the presence of columnar swarms [1].

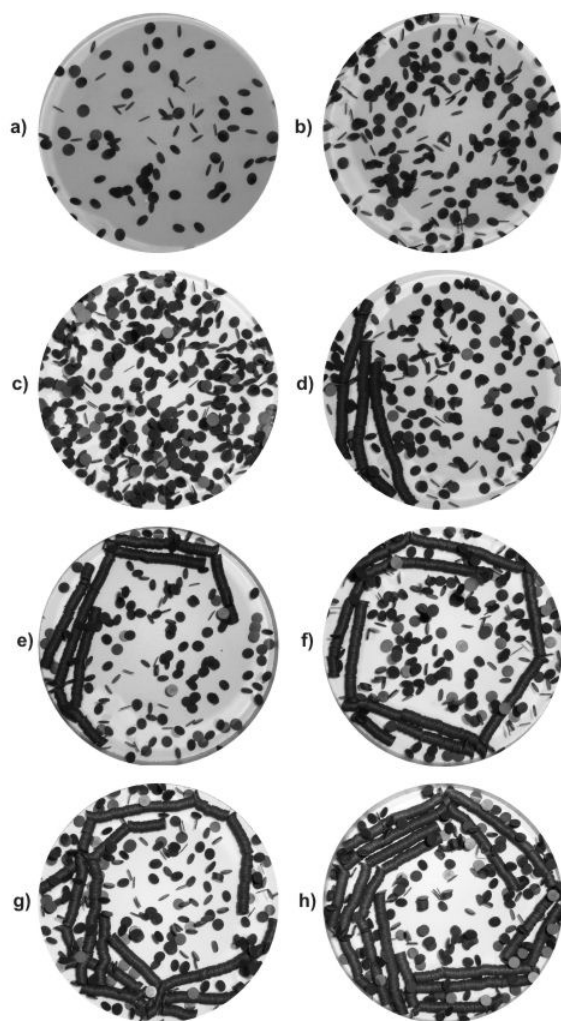


FIGURE 3. Snapshots with different number of granular magnetic disks: a) 100, b) 300, c) 700, d) 800, e) 1200, f) 1700, g) 1900 and h) 2300. States a)- c) would correspond, by analogy with a molecular system, to a low-density fluid, whereas d)- f) would be equivalent to a discotic liquid with columnar structures, and g) and h) to columnar-like phases.

A clear broken-symmetry phase was obtained when the container had 800 particles (Fig. 3d). The new phase had long columns formed by 150 particles, approximately. The spatial correlation between disks was higher due to an increased dipolar moment of the columns. Once the number of disks was increased to 1200 particles, the number of long columns also increased (Fig. 3e). For 1700 particles, column-column magnetic interactions produced different structural arrangements of the columns, as can be seen in Fig. 3f. Due to the container's wall, there were border effects and the configurations became unstable, surviving just a few seconds. A common feature of these patterns was that columns tended to arrange parallel to each other along the border. Once a configuration was destroyed, a new one appeared in few seconds, and the system was not longer in an isotropic state.

These partially ordered states resemble a liquid crystalline phase with columnar arrangements. More stable patterns were observed when the size of the system was increased to 1900 (Fig. 3g), lasting longer than the previous ones. In this case, excluded volume effects became more significant due to the reduction of free volume, and there was a change in the way that columns arranged themselves, as can be seen when Figs. 3f and 3g are compared. A final state was reached when the number of particles were 2300 (Fig. 3h). The system clearly was highly ordered, and an equilibrated configuration was observed, where columns tended to align along the boundary, in a concentric pattern. This phase was stable, since the pattern was not destroyed, and only local structural changes occurred. In this case the system resembles a solid-like phase. The columns followed a planar arrangement due to the gravitational interaction, although the disks could individually occupy the available volume.

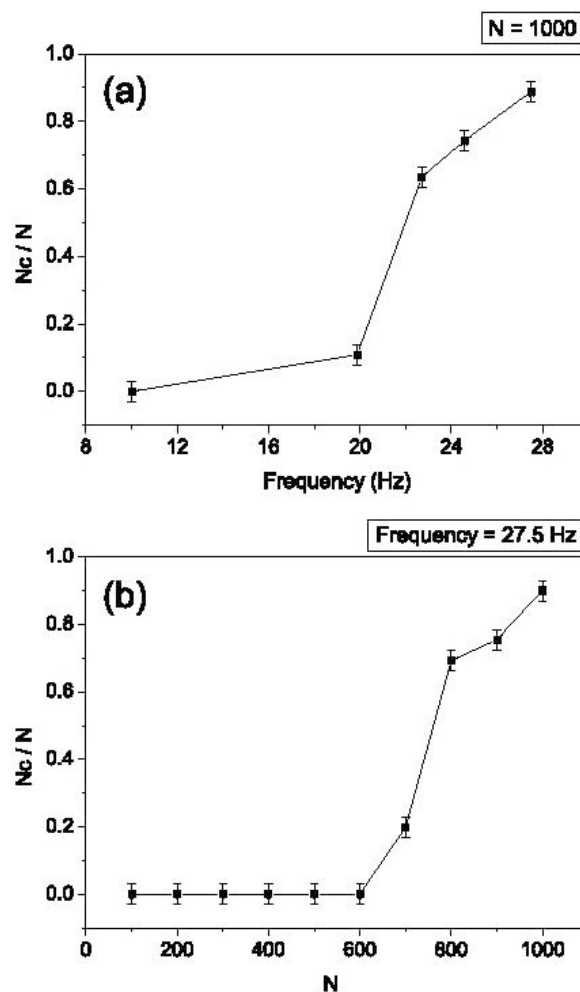


FIGURE 4. Fraction of disks associated in columns,  $N_c/N$ , where  $N_c$  is the number of disks in columns and  $N$  the total number of disks, as a function of the frequency of external magnetic excitation (a) and the total number of particles (b). In (a)  $N = 1000$ , and in (b) frequency = 27.5 Hz.

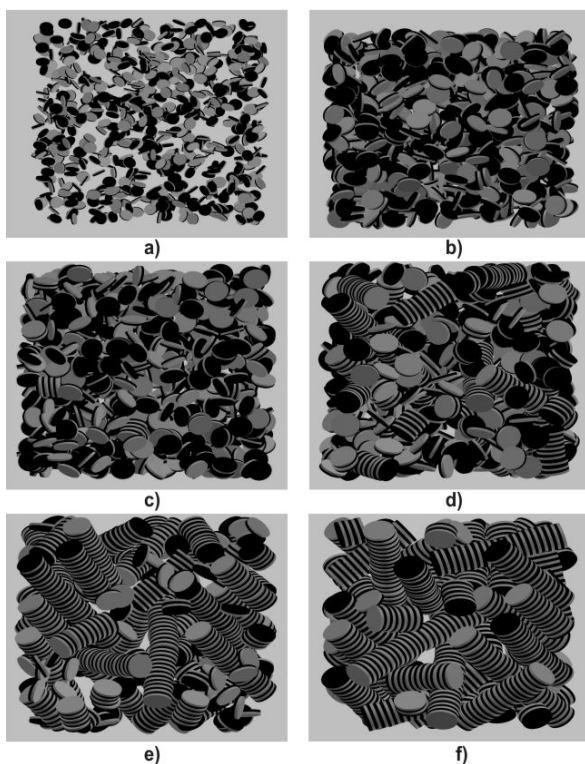


FIGURE 5. Snapshots at different densities and temperatures of disks with a magnetic dipole, obtained from Monte Carlo simulations using the isothermal-isobaric ensemble for 720 particles. Images correspond to phases at the same pressure but different temperatures and densities. Different liquid-crystal phases are obtained, that have a strong similarity with the phases obtained experimentally using granular magnetic disks, as shown in Figs. 1 and 3. Snapshots a) and b) correspond to isotropic phases; c) and d) to isotropic-columnar coexistence phases; e) and f) phases with columnar ordering.

#### 4. Discussion

It is well known that substances formed by anisotropic molecules present mesophases [1]. These phases are due to partial ordering of the translational and orientational degrees of freedom, as a consequence of the intermolecular interactions, as well as molecular symmetries.

In the experiment reported here, we verified that the anisotropic shape of the disk induced the columnar ordering. We performed experiments with different shapes (squares, triangles, two-dimensional spherocylinders) produced with the same magnetic material, and we found that when the system is formed by a pure component shape, columns are clearly formed. However, this long-range ordering is lost when different shapes are mixed. A possible explanation is that the difference of shapes produces similar effects than the presence of defects in an homogeneous phase. The ordering is lost due to this effective-defect behavior.

Keeping constant the frequency of the external magnetic excitation, the system evolved towards an ordered configu-

ration by increasing the number of particles. Some of these phases were metastable with a specific periodic repetition, whereas other configurations were clearly stable, which we considered as proper equilibrated configurations. The external exciting device can be described as an equivalent system of a thermal bath, in such a way that by fixing its frequency, the temperature of this bath is also controlled. As already mentioned before, the transitions between different configurations depended on the number of particles (*i.e.*, density, considering the fixed value of the container volume). The variation of the number of particles induced the evolution of the system from an isotropic to a highly-ordered columnar phase.

The fraction of disks associated in columns ( $N_c/N$ , where  $N_c$  is the number of disks in columns and  $N$  the total number of disks) can be studied as a function of the frequency of the external magnetic excitation, for a fixed total number of particles ( $N = 1000$ , see Fig 4a), and as a function of the total number of particles for a fixed frequency ( $= 27.5$  Hz, see Fig. 4b). In both cases we can observe a clear change in  $N_c/N$  as the frequency and the number of disks are increased, that illustrates how this fraction can be used as an order parameter to characterize the order-disorder transition observed in the system.

This experiment can be used as an educative material to illustrate how a system with a large number of particles behaves. Teachers can stress how competing effects determine the level of ordering in the system: the interaction between a dipolar disk and the external magnetic field, the interaction between disks, and the collision between particles and between them with the container wall. In the low-density state, the driving effect is the first one, and the system behaves as being in a gas phase. As the density increases, the other effects start to play a more important role, and the structuring depends on the balance between interactions. Small columns are induced by the dipole-dipole interaction. As the size of the column increases, there is also a corresponding increment on the total dipolar moment of the column. Columns start to interact strongly and align themselves on the bottom of the container. Due to the interaction with the external field, columns have translational, vibrational and rotational movements. Translation and vibration have a destructive effect on the columns, whereas the collision between single disks and columns does not have a significant effect. Once columns are stable, smaller segments within the columns start to spin around their molecular axis, with different frequencies.

The phases experimentally observed can be compared to the liquid-crystal behavior that molecular discotic fluids present. In Fig. 5 we present computer simulation snapshots of a system formed by disk-like molecules with a point dipole at the center, perpendicular to the disk. The aspect ratio of the disks is  $A = 0.2$ , where  $A = L/D$  and  $L$  and  $D$  are the height and diameter of the cylinders, respectively. These simulations were performed using the Monte Carlo (MC) method, in the isothermal-isobaric ensemble for a fixed number of particles (NPT ensemble with 720 particles), using

periodic boundary conditions and the Reaction-Field method in order to model the long-range dipolar interactions, using the standard procedure described in detail in Refs. 23 and 24. The dipolar interaction between disks is given by

$$\phi(r_{12}) = -\frac{\mu^2}{r_{12}^3} \left[ \frac{3(\mathbf{e}_1 \cdot \mathbf{r}_{12})(\mathbf{e}_2 \cdot \mathbf{r}_{12})}{r_{12}^2} - \mathbf{e}_1 \cdot \mathbf{e}_2 \right] \quad (1)$$

where  $\mu$  is the strength of the dipole moment,  $\mathbf{r}_{12}$  is the relative position vector between the disks centers (with magnitude  $r_{12}$ ), and  $\mathbf{e}_1$  and  $\mathbf{e}_2$  denote the dipole moment unit vectors.

The hard-body interactions were handled using the overlap method described in Ref. 25. Snapshots shown in Fig. 5 correspond to states at different values of density and temperature, for the same pressure value,  $P^* = 5$ , where  $P^* = PD^6/\mu^2$ . The final configurations were obtained after  $2.8 \cdot 10^5$  MC initial cycles (*i.e.*, number of movements per particle), in order to reach an equilibrated state; and followed by another  $2 \cdot 10^5$  cycles, in order to obtain proper averaged properties of the system. Using scaled variables for the density and the temperature,  $\rho^* = \rho/\rho_{cp}$  and  $T^* = kTD^3/\mu^2$ , respectively, where  $\rho_{cp}$  is the closed-packing density value, the snapshots in Fig. 5 correspond to different phases. In Figs. 5a ( $\rho^* = 0.025$  and  $T^* = 200$ ) and 5b ( $\rho^* = 0.129$  and  $T^* = 20$ ), the phases are isotropic; whereas, in Figs. 5e ( $\rho^* = 0.234$  and  $T^* = 13$ ) and 5f ( $\rho^* = 0.360$  and  $T^* = 12$ ) the phases have columnar ordering. A region of isotropic-columnar coexistence between the two pairs of states mentioned before seems to be present in Figs. 5c ( $\rho^* = 0.148$  and  $T^* = 16$ ) and 5d ( $\rho^* = 0.225$  and  $T^* = 15$ ).

Although the experimental and simulated systems are not completely identical, the strong similarities observed between the phases shown in Figs. 1b, 3 and 5, suggest two important features of the experimental device and setup illustrated here. In the first place, the use of experiments and simulations exhibiting the behavior of discotic systems would be very beneficial in the teaching of phase transitions. The setup of different kind of imaginative experiments devoted to illustrate the real behavior of molecular fluids and granular material could be used by teachers and students in order to draw

analogies and differences between both types of systems. Our understanding of analogies and differences between molecular and granular liquid crystal systems is an interesting area to be explored through the combined use of experiments with granular systems, like dipolar disks, and computer simulations of models of molecular liquid crystals.

## 5. Conclusions

In this paper we present an experimental study of a system composed of magnetic granular disks, using a simple device. By studying the temporal evolution of this athermal system, it was possible to obtain an isotropic and a columnar stable phases. The appearance of a metastable columnar phase preempts the stable one. The observed phases are consequence of competing factors, according to the dipole-dipole interactions and the excluded volume effects. These phases and the mechanisms behind them are analogous to the liquid-crystal behavior that is observed in molecular discotic fluids.

The experimental device presented in this article is not only of scientific interest but it can be useful from the educational point of view. It is a simple non-expensive experimental model that can be used to analyze the behavior of many-particle systems and complex collective behavior due to competing effects: self assembly, phase transitions, modulated phases, etc. [5]. Although all these phenomena can be studied using standard computational methods in Molecular Simulations, it is useful to have direct experiments showing the interplay of physical principles.

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