A dynamical phase transition in ferromagnetic granular materials

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We study, using simulations the dynamical properties of complex ferromagnetic granular materials. The system of grains is modeled by a disordered two-dimensional lattice in which the grains are embedded, while the magnitude and direction of the easy axis are random. Using the monte-carlo method we track the dynamics of the magnetic moments of the grains. We observe a transition of the system from a macroscopic blocked (ferromagnetic) phase at low temperature in which the grain’s magnetic moment do not flip to the other direction to an unblocked (superparamagnetic) phase at high temperature in which the magnetic moment is free to rotate. Our results suggest that this transition exhibits the characteristics of a second order phase transition such as the appearance of a giant cluster of unblocked grains which is fractal at the critical temperature, a peak in the size of the second largest cluster at the same temperature and a power law distribution of cluster sizes near the criticality.

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I. INTRODUCTION:

Granular systems composed of small metallic grains deposited on an insulating substrate (see Fig. 1) have generated much recent experimental and theoretical interest [1-9]. These systems show a rich behavior stemming from the properties of the individual grains and from their mutual interaction leading to a complex collective behavior of the granular system. The size of the individual grain has an enormous influence on its magnetic properties [10]. The grains may be classified into three categories according to their magnetic behavior: Multi-domain grains containing different magnetic domains (large grains), single-domain ferromagnetic grains, containing a single ferromagnetic domain (smaller grains) and superparamagnetic grains, in which the magnetic moment easily switches and therefore effectively have no average magnetic moment in the absence of an external field (ultra-small grains).

Theoretical models describing single-domain grains were introduced in the 1930’s [11-14]. In 1950’s several experiments [12] found that the typical size of these grains is a few hundred Angstroms. At low temperatures the direction of the magnetic moment is more or less fixed (up to small fluctuations around its preferred direction), but once temperature is high enough the magnitude of the magnetic moment remains fixed but its direction becomes random. Thus for a single domain uniaxial grain, as function of temperature, the grain changes from ferromagnetic, to a state in which it flips the magnetic moment up or down along its easy axis. The latter state was termed by Bean [15] as superparamagnetic, in which although at any given time the grain has a magnetic moment, on average it is zero [10].

For a uniaxial grain the flipping rate is given by [17]:

\[ \frac{1}{\tau} = f_0 e^{-\frac{k_B T}{\mu_0 I_s}}, \]

where \( f_0 \) is the frequency factor, \( k_B T \) is the thermal energy, \( V \) is the volume of the grain and \( K \) is an anisotropic constant (e.g. for shape anisotropy \( K = 4\pi I_s^2 \), where \( \mu_0 \) is the magnetic vacuum permeability and \( I_s \) is the saturation magnetization, i.e. the external field required to align the magnetic moment of the grain perpendicular to its easy axis).

Considering the case where the grain has an ellipsoid like shape with semi-axis \( a > b = c \), the semi major axis \( a \) is called the easy axis (denoted \( \hat{\sigma} \)). Due to this geometric property there is a magnetic shape anisotropy [10]. The Hamiltonian describing these grains is given by [18]:

\[ H = \frac{\mu_0}{2} I_s^2 \nu \sin^2 \theta - \mu_0 I_s \frac{H_0 \cdot \vec{\mu}}{\mu}, \]

\( \nu = N_a - N_b \) where \( N_a \) and \( N_b \) are the demagnetization coefficients of the ellipsoid along the a and b axes (\( \nu = 1 \) for \( N_a \gg N_b \) in MKS units), \( \vec{\mu} \) is the magnetic moment of the grain, \( \theta \) is the angle between \( \vec{\mu} \) and \( \hat{\sigma} \) (see Fig. 2) and \( H_0 \) is an external magnetic field.

For a system composed of many grains, magnetic interactions between the grains must be considered. Those take the form of dipole-dipole interactions [17]. The

![Fig. 1: SEM image of a granular material. A-F are single grains in the system, the average size is a few nanometers. The image was taken by CABL-9000C High Resolution Electron Beam Lithography System CRESTEC.](image-url)
magnetic field of a magnetic moment $\mu$ belonging to grain $i$ at a distance $R$ from the dipole is:

$$
\vec{B}_i = \frac{\mu_0}{4\pi} \frac{|\vec{\mu}|}{R^3} (2 \cos \phi \hat{e}_R + \sin \phi \hat{e}_\phi),
$$

where $\phi$ is the angle between $\vec{\mu}$ and $\vec{R}$, $R$ is the magnitude of $\vec{R}$, $\hat{e}_R$ is a unit vector in the $\vec{R}$ direction and $\hat{e}_\phi$ is a unit vector in the $\phi$ direction (perpendicular to $\hat{e}_R$ on the $\vec{\mu} - \vec{R}$ plane). Summing up the contribution of all other grains to the magnetic field at the location of the $i$-th grain ($\vec{B}_{tot}$) results in the following Hamiltonian (see Eq. 2) for the $i$-th grain:

$$
H_i = \frac{\mu_0}{2} I_s \nu \sin^2 \theta - \mu_0 I_s \frac{(\vec{H}_0 + \frac{\vec{B}_{tot}}{\mu_0}) \cdot \vec{\mu}}{|\vec{\mu}|},
$$

where the system’s Hamiltonian is the sum of the individual grain Hamiltonians: $H = \sum_i H_i$. Solving this Hamiltonian in temperature is a complex many-particle problem since the magnetic moment of each grain depends on the other grains moments.

In this paper we study the transition from the blocked phase (where grains do not flip their magnetic moment) at low temperatures to the unblocked phase (where the grains magnetic moment flip frequently) at higher temperatures. Our results suggest that the crossover between those two phases follows the behavior expected from a macroscopic second order percolation type phase transition. This is shown by studying critical exponents and the fractal properties of the clusters of blocked/unblocked grains in the vicinity of the transition [13, 20].

II. THE MODEL:

We consider ellipsoid grains which have an easy-axis along the semi-major axis (see Fig. 2). These grains are arranged in our model on a two dimension square lattice, where each site represents a grain. Each grain has a dipole moment which may take any direction in the three dimensional space and a randomly chosen easy axis (which may take any direction, see Fig. 3). Disorder is added to the system by defining a random distance between the sites, composed of the Euclidean distance between the sites added to a random component drawn from a Gaussian distribution of variance $0.3a$ (where $a$ is the lattice constant). The direction of the easy axis is chosen randomly with an equal probability at any direction while its magnitude is chosen from a Gaussian distribution with an average $\langle I \rangle = 2$ and variance $0.3$.

In order to simulate the behavior of the magnetic moments of the grains we use the Metropolis Monte-Carlo algorithm. At each iteration all the grains are passed sequentially. For each grain a new random magnetic moment direction is chosen, then the total energy of the system is calculated using the Hamiltonian (Eq. 4). In cases that the energy difference between the two configuration of the systems (i.e. before and after changing the direction of the magnetic moment of the grain) is negative we accept the change, however if the energy difference is positive we accept it with probability of $p = e^{-\beta \Delta E}$. When the magnetic moment direction of the grain crosses the plane perpendicular to its easy axis it is denoted as a flip. We track the number of flips each grain undergoes, then by dividing it by the total number of iterations (i.e., time steps) we obtain the probability of the magnetic moment to flip its direction for each time step. A grain that has zero probability to flip is denoted as blocked while a grain that has a non-zero probability is regarded as unblocked.

The system is then divided into clusters of blocked and unblocked grains, where a connected component that all of its sites are blocked is denoted as a blocked cluster. We preform this process for networks with the following sizes: 900,1600 and 2500 grains arranged on a 30X30, 40X40, and 50X50 lattices. In order to get good statistics we run the simulations over 285,169 and 100 realizations respectively. For each simulation we preform 120

![FIG. 2: An ellipsoid shaped grain, with $\mu$ representing the direction of the dipole moment, $\sigma$ representing the direction of the easy axis along the semi-major axis of the ellipsoid and $\theta$ is the angle between them.](image1)

![FIG. 3: A 2D Square lattice where the arrow location represents the location of a grain in the system (only on the x-y plane) and the direction of the arrow represents the direction of the easy axis of each grain (which may point at any random direction in the 3-dimensional space).](image2)
iterations at every temperature. One can think that the monte-carlo algorithm is simple and fast, and can work on systems larger then a 50X50 lattice. However, due to these repetitions explained above this is not the case and on top of the standard monte-carlo algorithm complexity which is $n^4$ ($n$ is the number of nodes on one side of the lattice(30,40,50)) one should take into account the number of realizations, the number of temperatures, and the number of iterations. This leads to a higher complexity which prevented us to run the simulations on a larger scale.

We explore the behavior of the system by using methods borrowed from complex network and percolation theory [19, 21–29]. The probability to perform a moment flip at a given time step is considered as the percolation probability. We then consider properties such as the size of the largest and second largest blocked cluster, the clusters size distribution and their fractal dimension in order to characterize the system and to suggest that there is a second order phase transition in the system with temperature as the system is crossing from a macroscopic unblocked phase at high temperature to a macroscopic blocked one at low temperature [20, 29].

III. RESULTS:

A. Probability to Flip

The probability for a single grain to flip (Eq. 1) decays exponentially with $\beta = K_p T$, however averaging the probability of a grain to flip over the entire system, when interactions are included, the temperature dependence is different. In Fig. 4 we present the numerically averaged probability $p$ for a single magnetic moment to flip, averaged over an ensemble of different realizations of grains as a function of temperature. By dividing the number of flips a grain performs by the number of iterations in each temperature (see Sec. III) we get the probability of a magnetic moment to flip at each time step. We verify this probability by changing the number of iterations to various numbers in the range 120-720, in all cases dividing the number of flips by the number of iterations gives the same probability. This behavior hints that the interactions between the magnetic moments of the grains qualitatively changes their collective behavior.

B. Percolation of Blocked Grains

In order to deepen our understanding of the system’s behavior we plot in Fig. 5 the spatial distribution of blocked grains. The results are analogous to percolation theory where the temperature $T$ in our model plays the role of $q$, the chance of having an empty site, in the percolation model. We notice that above a given temperature, $T_c$, most of the grains are unblocked and there are only some small blocked clusters in the system. However below $T_c$ we notice that the system is dominated by blocked grains and only small unblocked clusters remain. Finally, at $T = T_c$ we notice that an almost equal number of blocked and unblocked grains, intermingled in very large clusters. This type of behavior is typical for systems close to a second order phase transition.

Moreover observing the largest cluster (see Fig. 6) in the system at the critical point $T = T_c$ which is a reminiscence of a fractal nature, suggesting the divergence of the correlation length, a typical behavior of a second order phase transition.

Upon measuring the sizes of the two largest blocked clusters as a function of temperature (see Fig. 7) the analogy to percolation is reinforced. In percolation theory, the largest cluster (called the giant component) is zero for high values of $q$ and approaches the system’s size when $q$ is zero [19, 21–29]. The size of the second largest cluster approaches zero for high and low $q$ values, however when approaching the critical percolation threshold the size of the second largest cluster has a sharp maxima at $q_c$, the critical percolation value. By running a BFS (Breadth First Search) algorithm [30] on the simulations results, the blocked clusters of the system are identified and their size is measured. The blocked clusters behave in a similar way to percolation clusters. The giant component’s size grows to the system’s size as $T$ approaches zero, and the size of the second largest cluster is zero for high and low temperatures and has a maximal value at $T_c$. In order to test the dependence on the system’s size we measure these sizes on a scaled unit $p_\infty$ which is the number of connected nodes in the giant component divided by the number of nodes in the system. (see

FIG. 4: The probability to flip $p$ averaged over the entire system and on the ensemble of realizations as a function of temperature $T$. Our results suggest that the system undergoes a second order phase transition behavior.
Thus, the properties of the giant and the second largest cluster seem to follow those expected from a second order phase transition. Moreover, in order to verify that these properties are not size-dependent, we analyzed the giant component of systems with different sizes. We see that all system sizes show the same behavior and the giant component emerges at the same temperature. We also note that the transition becomes sharper for larger systems which insinuates that the width around $T_c$ is due to finite size effects and for infinite size system the order parameter $p_\infty$ is expected to have a discontinuity in the first derivative (see Fig. 8).

The second largest cluster peak center also dose not change with size for our systems, and the fraction of the system covered by this cluster at $T_c$ is also similar for all system sizes but become sharper as the size of the system increases (see Fig. 9). Again this is in agreement with expectations a second order phase transition.

C. Critical Exponents

Examining the results described above (see Sec. III B) one can quantify the transition properties by analyzing the scaling properties of the blocked clusters. The emergence of scaling laws is expected from systems undergoing a second order phase transition in which the correlation
length diverges. The size distribution of blocked clusters can be used as indication for a transition, since at transition, there should be no typical cluster size, i.e., the distribution should follow a power-law. Indeed, examining the blocked cluster size distribution in our system strongly suggests the existence of a scaling law. As shown in Fig. 10, the cluster size distribution $n(s)$, is in fact a power-law $n(s) \sim s^\tau$, typical for second order phase transitions. Moreover, we can see the same slope for systems with different sizes, indicating that they all converge to the same critical exponent ($\tau = 1.63$). In addition, as we increase the system size the cut-off value increases as well, thus one may expect that for an infinite size system there will be no cut-off (see Fig. 10).

In systems that undergo a second order phase transition it is known that the correlation length diverges at the critical point. The divergence of the cutoff in the cluster size distribution seen in Fig. 10 is a signature of a diverging correlation length. The fractal nature of the clusters is another way of showing this divergence, as is well known both for percolation and Ising systems [31]. At the critical point voids with no characteristic length scales appear in the giant component, hence the correlation length which is the typical size of these voids diverges (see Fig. 6). This may be demonstrated by calculating the fractal dimension of the clusters.

In order to calculate the fractal dimension (noted $d_f$) of the blocked clusters in the system, we use the following relation [20, 27, 29, 32]:

$$M \sim R_{G}^{d_f},$$

where $M$ is the mass of the cluster and $R_G = \sqrt{\langle R - R_{cm} \rangle}$ is the radius of gyration ($R_{cm}$ is the radius of the center of mass). As we have shown above (see Fig. 6) the cluster in our system has a fractal nature. By plotting the mass of the clusters as a function of their $R_G$ on a log-log scale, we can see they are obeying a power law (see Fig. 11). The slope of the power low fit is the dimension of the clusters which are fractal and their fractal dimension is $d_f \approx 1.89$. Systems with different sizes have the same fractal dimension.

IV. SUMMARY:

In summary, by using the analogy between the behavior of blocked/unblocked grains in magnetic granular systems composed of nanometric ferromagnetic grains and percolation, we suggest that the granular system exhibits a new kind of second order phase transition. In this transition the role of the order parameter is played by the dynamics of the magnetic moment of the grain, i.e.
FIG. 11: The mass of the clusters as a function their radius of gyration at criticality. The slope represents the fractal dimension of the blocked clusters in the systems, $d_f = 1.89$, which is very close to that of percolation (1.896).

Whether the magnetic moment of the grain can flip or it is blocked. The second giant component has a peak at the critical temperature $T_c$, the cluster size distribution obeys a power law, and the clusters have a fractal dimension. All these properties can be seen for systems of different sizes which support the assumption that the transition we observe is a real thermodynamic second order phase transition.

From the experimental point of view, we believe that this transition, in principal, could be observed by direct measurements of the local magnetic field using a SQUID (superconducting quantum interference device), or an MFM (Magnetic force microscope) tip. Another method, which is probably easier from the experimental point of view, is to identify the transition by its effects on the electrical transport through these systems, especially the resistance noise. We expect that the transport noise will be strongly enhanced in the vicinity of the transition due to the divergence of the correlation length.

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