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Effects of dipolar interactions on the magnetic properties of granular solids

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Abstract

We use the Monte Carlo simulation technique to study the coercive behaviour of an assembly of single-domain ferromagnetic particles with random anisotropy axes directions, embedded in an insulating matrix and interacting via dipolar forces. We present results for the dependence of the coercivity on the volume packing fraction and temperature. We find a peak of the coercive field at the percolation threshold. We compare our theoretical results with experimental findings. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Granular systems; Random anisotropy; Coercive field; Percolation; Monte Carlo simulation

Granular metal solids consist of nanometer size magnetic metal grains embedded in a non-magnetic (insulating or metallic) matrix. Their magnetic properties have attracted a lot of research interest because of their important technological applications [1]. A key feature that makes these materials attractive for technological applications is their high coercive field.

Grain size effects controlled by the metal volume fraction have been used to explain the coercive behaviour of the granular materials initially [1]. However, it was recognized early on that the magnetic interactions between the grains play a crucial role in the magnetic properties of granular systems [2]. In particular, magnetic dipolar interactions appear to be the dominant ones for granules embedded in an insulating matrix. Dipolar interactions are determined by the magnitude of the dipole moments and their relative separation. The magnetic moment of each grain is proportional to its size and the packing fraction determines the average separation between the grains. Therefore these two parameters influence the role of dipolar interactions.

Previous work [3] has investigated the role of dipolar interactions for dilute granular systems ($x_v < 0.15$) at zero temperature. An important finding of this work was that the coercivity of these systems increases with increasing concentration. However, there is experimental evidence that the role of dipolar interactions becomes important for $x_v > 0.2$ [2]. The purpose of this paper is to investigate the role of dipolar interactions in the coercive behaviour of a granular material for the whole range of the metal volume fraction ($0.0 < x_v < 1.0$) at finite temperature, with the use of the Monte Carlo simulation technique.

We consider N identical spherical particles with magnetic moment m and diameter a, randomly placed on a simple cubic lattice of size L. Each particle has uniaxial anisotropy and the anisotropy axes are randomly oriented. The packing fraction x_v is defined to be equal to the percentage of occupied sites (N/L^3). The energy of the system in an external magnetic field H is

$$E = \sum_i \left[\frac{m^2}{a^3} \sum_j \frac{\hat{S}_i \cdot \hat{S}_j - 3(\hat{S}_i \cdot \hat{R}_{ij})(\hat{S}_j \cdot \hat{R}_{ij})(\hat{S}_j \cdot \hat{R}_{ij})}{R^3} - K_1 V(\hat{S}_i \cdot \hat{e}_i)^2 - mH(\hat{S}_i \cdot \hat{H}) \right],$$

where K_1 is the anisotropy energy density, V the particle volume ($V = \pi a^3/6$), R the interparticle distance, $\hat{e}_i$ the anisotropy axis direction and $\hat{S}_i$ the magnetic moment direction. We define the following dimensionless parameters: the reduced anisotropy $k = K_1 V a^3/m^2$, the reduced external field $h = Ha^3/m$ and the reduced temperature $t = k_B T a^3/m^2$. The equilibrium magnetisation of the system is calculated by the standard Metropolis algorithm. We consider an ensemble of $N = 100$ magnetic dipoles placed randomly in a cube of variable size L.

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Free boundary conditions for the cubic sample are used. To simulate the hysterisis loop of the system, the sample is initially saturated along a chosen axis by the application of a large external field which is gradually reduced. The coercive field is the applied field for which the mean component of the magnetisation along the chosen axis vanishes. To reach equilibrium for each value of the applied field we have used 2000 Monte Carlo steps per particle. Finally, for each value of the metal volume fraction an average over 15 random configurations of the particle positions was used.

In Fig. 1 we have plotted the reduced coercive field h_c as a function of the metal volume fraction for the case of zero anisotropy strength ($k = 0$), moderate anisotropy strength ($k = 1.0$) and large anisotropy strength ($k = 10.0$). The reduced temperature in all cases is $t = 0.1$. The important feature of this figure is that for zero and moderate anisotropy the dipolar interactions lead to a maximum in the coercivity for metal volume fraction close to the percolation threshold ($x_p \approx 0.3$). We associate this behaviour with the formation of a percolating cluster. This cluster has a fractal structure [4] resulting in a chain-like arrangement of the magnetic dipoles and consequently to an enhanced anisotropy. Analogous enhancement of other physical properties (e.g. dielectric function) of a percolating cluster consisting of electric dipoles has been reported [4]. Chien and his collaborators [1] observed enhancement of the coercive field at $x_v \approx x_p$, but they attributed this behaviour to size effects of the isolated grains. In order to estimate dipolar effects and to distinguish them from the size and anisotropy effects, we have calculated the coercivity for $k = 0$ (see Fig. 1, squares). We used the parameters from Ref. [1]: particle diameter ($a = 100$ Å), saturation magnetisation ($M_s = 1200$ emu/cm^3) and temperature ($T = 300$ K). We obtain a coercive field (maximum in plot) of about $H_C \approx 120$ Oe at the percolation threshold. This result should be compared to the value of 500 Oe in Ref. [1]. We thus conclude that the dipolar interactions alone give a significant contribution to the coercive field. As we can see from Fig. 1 single-particle anisotropy modifies the behaviour of the coercive field. In the case of strong anisotropy ($k = 10.0$) an almost linear decrease of h_c with x_v is observed in accordance with Néel's prediction.

In Fig. 2 we give the temperature dependence of coercivity for a dilute system ($x_v = 0.1$) and for a system close to percolation threshold ($x_v = 0.292$), for moderate anisotropy strength $k = 0.5$. We have plotted the normalised coercivity $h_c(t)/h_c(0)$, where $h_c(0)$ is the zero-temperature-reduced coercivity, as a function of the reduced temperature t. In both cases the coercive field vanishes for temperatures higher than the ones predicted by the theory for non-interacting particles [5], which in our units is $t = 0.02$. This behaviour is attributed to dipolar interactions and as it can be seen from Fig. 2, close to the percolation threshold their influence is stronger.

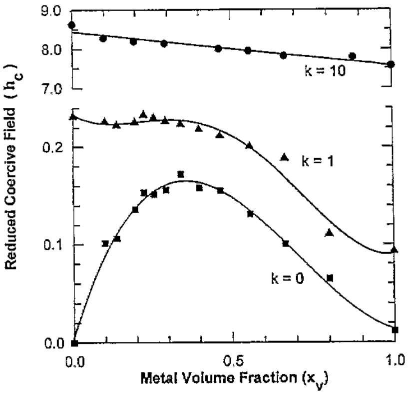

Fig. 1. Reduced coercivity (h_c) versus metal volume fraction (x_v) at $t = 0.1$ for $k = 10$, $k = 1$ and $k = 0$.

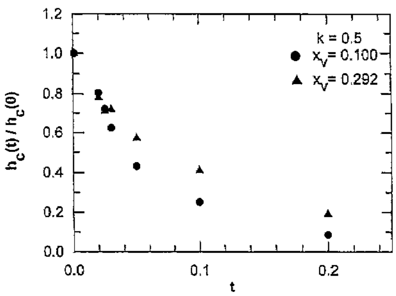

Fig. 2. Normalised coercivity versus reduced temperature for a dilute system ($x_v = 0.1$) system and for a system with $x_v = 0.292$.

In conclusion, magnetic dipolar interactions play a significant role in the coercive behaviour of granular metals embedded in an insulating matrix. Their effects are more pronounced in systems with anisotropy energy smaller than or comparable to the dipolar interaction energy and for metal volume fractions close to the percolation threshold. In our simulations we have not included grain size effects, but as we demonstrated above dipolar effects themselves have a significant contribution to the enhancement of the coercive field at x_p. Modelling of a system with interacting grains of variable size is in progress.

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