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Spin correlations and electronic transport in magnetic nanoclusters

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Abstract

The magnetic properties and the giant magnetoresistance (GMR) of magnetic nanoparticle assemblies are studied numerically. The equilibrium magnetic configuration of the assembly at finite temperature and under application of a magnetic field is obtained by Monte Carlo simulation. The electronic conductivity is obtained using a quantum (Kubo formalism) or a classical (resistor network) description. Results for granular systems characterized by uncorrelated positions of the nanoparticles and thin films containing magnetic nanoparticle aggregates are presented and their behavior is compared. The footprints of interparticle dipolar interactions are revealed in the field dependence of the magnetization and the GMR curves.

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Keywords: Giant magnetoresistance; Granular films; Nanoparticles; Magnetostatic interactions; Aggregation

1. Introduction

The magnetotransport properties of systems composed of magnetic nanoparticles embedded in a non-magnetic matrix currently attract a great deal of experimental and theoretical interest mainly due to the appearance of the giant magnetoresistance (GMR) effect and its technological exploitation in magnetic storage media. The GMR effect, namely the large decrease of the sample's resistivity under application of an external magnetic field, has been originally observed in exchange-coupled Fe–Cr layers [1] and later in magnetic granular films [2,3]. The common physical mechanism behind this effect is the spin-dependent scattering of the carriers off the magnetic regions (layers or nanoparticles). As the external field rotates the moments of the magnetic entities, the scattering potentials for the electrons are modified and eventually the resistivity of the sample changes. It is therefore expected that the field dependence of the magnetoresistance reflects the modifications to the micromagnetic configuration as the field varies in strength. In the early works [3], it was demonstrated that the magnetoresistance is determined solely by the long-range magnetic order of the granular metal, as the universal parabolic dependence of the MR on the sample magnetization dictates. However, flattening of the MR–M parabola observed for weak fields ($H \sim 100-1000$ Oe) has been

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subsequently attributed to the existence of short-range ferromagnetic order [4] and the grain-size distribution [5]. Magnetostatic interactions between the granules are the dominant forces that lead to ferromagnetic correlations in granular metals. The interplay between the magnetostatic energy and single particle anisotropy determines the magnetic configuration of the system.

In this paper, we study the role of the magnetostatic interactions on the GMR effect in granular films. In Section 2 we discuss films with a magnetic fraction close to the percolation threshold, and in Section 3 films that contain chain-like aggregates of magnetic nanoparticles. Our conclusions are summarized in the last section.

2. Granular films close to percolation

The magnetic granular film is approximated by an assembly of identical spherical particles located at random on the sites of a finite cubic lattice with linear dimension L. The lattice parameter is equal to the particle diameter ($D = a$). Each particle carries a three-dimensional moment of magnitude $m_o = M_s V_o$. Strong exchange interactions are assumed between nearest neighbor particles leading to the formation of magnetic clusters. Close to the percolation threshold ($x_c \sim 0.3$ for the SC lattice), the sample contains a distribution of magnetic clusters with random shape and a wide distribution of volumes, as shown in Fig. 1. Each cluster is approximated by a single magnetic domain and a random easy axis is attributed to it. The magnetostatic energy of a pair of clusters is calculated by summation of the dipolar energy terms between all pairs of moments belonging to these clusters. Periodic boundaries and the Ewald method are used to deal with the long-range character of the dipolar forces. The total energy of the system reads

$$E = g \sum_{c,c'} \sum_{i,j} \frac{\hat{m}_c \cdot \hat{m}_{c'} - 3(\hat{m}_c \cdot \hat{R}_{ij})(\hat{m}_{c'} \cdot \hat{R}_{ij})}{R_{ij}^3} - k \sum_c n_c (\hat{m}_c \cdot \hat{e}_c)^2 - h \sum_c n_c (\hat{m}_c \cdot \hat{H}), \qquad (1)$$

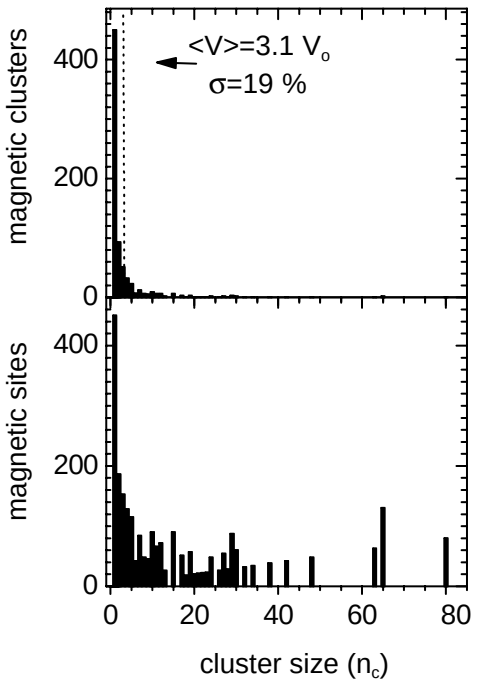

Fig. 1. Cluster size distribution of a granular film close to percolation threshold. Concentration of magnetic sites $x = 0.25$.

where c, c' denote clusters, i, j denote sites within the clusters, n_c is the number of particles that constitute a cluster, e_c is the easy axis direction and hats indicate unit vectors. The energy parameters are $g = (M_s V_o)^2 / D^3$ for the dipolar energy, $k = K_1 V_o$ for the anisotropy energy and $h = M_s V_o H$ for the Zeeman energy, while the thermal energy is denoted as $t = k_B T$. In this study, we use parameters appropriate for Co nanoparticles, namely $M_s = 1500\,\text{emu/cm}^3$, $D = 3.5\,\text{nm}$ and $K_1 = 1.2 \times 10^6\,\text{erg/cm}^3$ which result in $g/k \sim 1.0$ for the parameters of Eq. (1). The equilibrium magnetic configuration is obtained by a Monte Carlo simulation [6,7], using the standard Metropolis algorithm. Simulations were performed on a finite sample ($10 \times 10 \times 10$) for various particle concentrations and averages over 10 random arrangements of the particles were taken.

The electronic structure of the sample is described within a spin-split s-band. The conductivity of the sample is obtained by the real space

form of Kubo's formula that is expressed in terms of the Green function of the system [7]. Finally, the GMR is defined as $MR(H) = [R(H)/R_s - 1] \times 100$, where $R(H)$ is the field dependent resistance and R_s the resistance at saturation.

The blocking temperature of the system can be estimated by considering the temperature dependence of the remanence (Fig. 2). The polydisperse sample close to percolation ($x = 0.25$) is compared with a monodisperse sample containing only clusters with volume equal to the average volume of the polydisperse one ($< V > \sim 3.1 V_0$). Both types of samples have the same concentration of magnetic particles. The monodisperse sample shows an abrupt drop of the remanence at temperature $t_b \sim 0.5$ (see Fig. 2(i)), while the presence of a size distribution rounds-off this behavior through the introduction of a distribution of anisotropy energy barriers. Magnetostatic interactions establish a ferromagnetic order for temperatures above the blocking. Further on, we study the field dependence at temperature $t = 1.2$ at which the monodisperse sample contains superparamagnetic clusters only (see insert of Fig. 2(i)) and exhibits a weak remanence ($M_r/M_s \sim 0.05$) exclusively due to dipolar interactions, while the polydisperse has a substantial remanence ($M_r/M_s \sim 0.30$) and contains a mixture of blocked and superparamagnetic particles, as the insert of Fig. 2(ii) indicates.

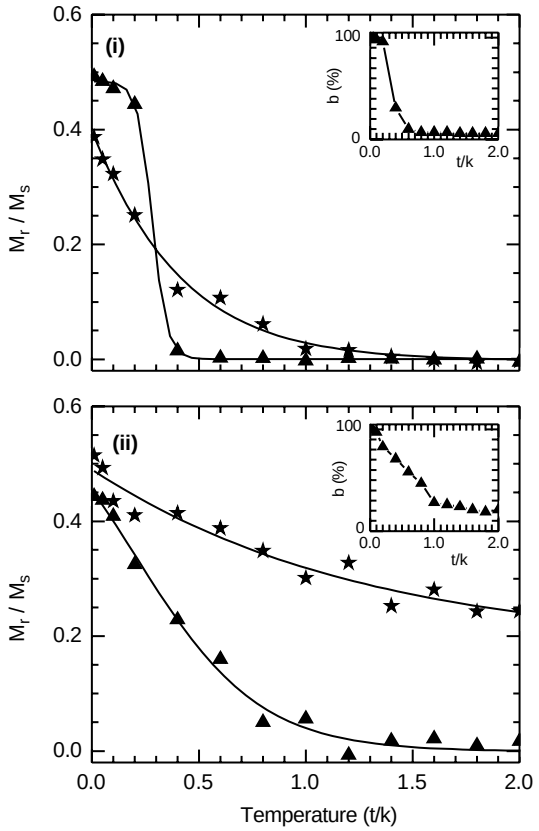

Fig. 2. Temperature dependence of remanence for (i) a monodisperse with $n_c = 3$ and (ii) a polydisperse sample with $\langle n_c \rangle = 3.1 \pm 19\%$. Insert: fraction of blocked spins as a function of temperature for the non-interacting sample ($g = 0$) at zero field ($h = 0$). Concentration of magnetic particles $x = 0.25$. Solid lines are guides to the eye. Triangles: non-interacting anisotropic clusters ($g = 0$, $k = 1$). Stars: interacting anisotropic ($g = k = 1$) clusters.

The field dependent magnetoresistance is shown in Fig. 3. Note first that the non-interacting samples in Fig. 3(i) and (ii) have very similar MR values, because the few blocked clusters that exist in the polydisperse sample are too large compared to the electron Fermi wavelength ($\lambda_F \sim D$) to produce any substantial GMR effect and the remaining smaller clusters in both samples are superparamagnetic. Evidently, magnetostatic interactions degrade the GMR effect. Their role is more dramatic in the polydisperse sample where a reduction of $\sim$5–10% can be observed with respect to the sample with non-interacting clusters. A necessary prerequisite for this large degradation is the width of the cluster-size distribution. Namely, the large clusters that exist in the polydisperse sample produce strong magnetostatic fields that lead to an efficient ferromagnetic alignment of the smaller clusters surrounding them. By switching-off the anisotropy energy terms in the sample, we observe a further reduction of the MR values at weak fields as the collective behavior caused by magnetostatic interactions between the clusters does not compete with the single-cluster behavior caused by the anisotropy term. Further indication for collective rotation of clusters in the polydisperse sample is the deviation of the MR–M curve from the parabolic dependence which is most obviously seen in the pure dipolar case (Fig. 3(ii)).

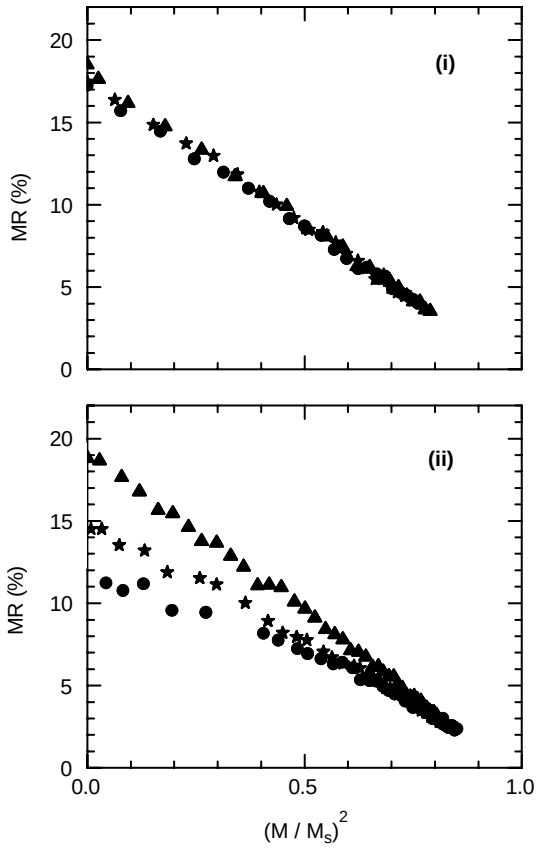

Fig. 3. MR versus sample magnetization for (i) a monodisperse and (ii) a polydisperse sample. For both samples $x = 0.25$. Triangles: non-interacting anisotropic clusters ($g = 0, k = 1$); stars: interacting anisotropic ($g = k = 1$) clusters; circles: interacting anisotropic clusters ($g = 1, k = 0$); band structure parameters: atomic potentials on the electrodes and the matrix sites $\varepsilon_0 = 0$, on the magnetic sites $\varepsilon_1 = 1$, exchange splitting of s-bands $J = 1$, hopping integral $V = 1$ and Fermi energy $E_F = 0$.

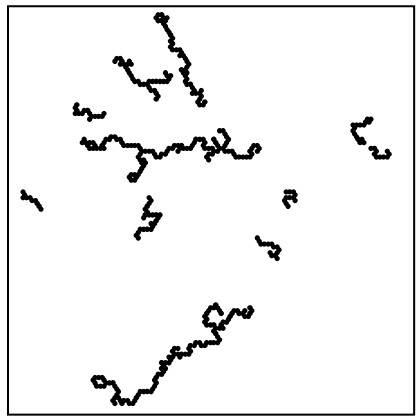

Fig. 4. Morphology of a sample containing nanoparticle aggregates.

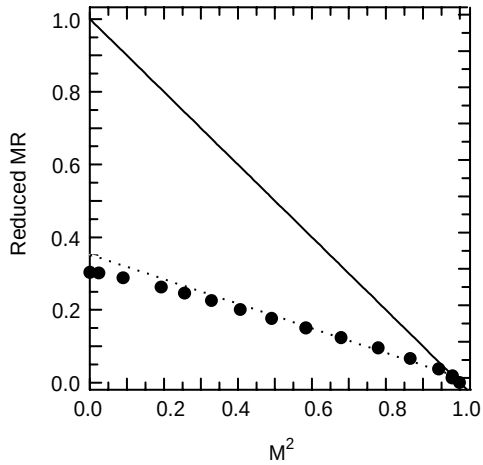

Fig. 5. Reduced MR as a function of sample magnetization. Solid line: theoretical prediction for non-interacting nanoparticles; dotted line: straight line fit; circles: results for dipolar interacting nanoparticles forming aggregates.

3. Granular films containing nanoparticle aggregates

We next consider the magnetic granular films characterized by a correlated morphology. In particular, we discuss the effect of nanoparticle aggregation on the magnetotransport properties of a thin film [8]. For simplicity, we have simulated the growth of aggregates on an inert, planar substrate [9] under the action of the interparticle dipolar interactions using a diffusion limited cluster aggregation (DLCA) model that leads to the formation of aggregates with a chain-like morphology (Fig. 4).

The magnetic structure of the thin film is obtained using the Monte Carlo technique described previously, except that of the single-particle anisotropy terms that have been neglected. Since the emphasis here is on the effect that dipolar interactions have on GMR, we have

calculated the reduced magnetoresistance [4] which is independent of the material parameters and in a resistor-network description it takes the simple expression $MR_r(H) = 1 - f(H)$, where $f(H) = \langle \hat{m}_i \cdot \hat{m}_j \rangle$ is the magnetic moment correlation function between the nearest neighbor particles. For non-interacting particles, obviously $f(H) = (M/M_s)^2$ and the well-known MR–M parabola is obtained. Deviations from the parabola indicate short-range correlations. We have calculated the reduced GMR for an external field perpendicular to the substrate and neglecting the magnetostatic coupling between separate aggregates. Results are shown in Fig. 5, where a dramatic decrease of MR ($\sim 70\%$) due to dipolar interactions is observed.

4. Conclusions

Magnetostatic interactions between nanoparticles in magnetic granular films are responsible for flattening of the MR–M parabola at weak fields. The flattening is enhanced by the presence of a size distribution of nanoparticles [4] or clusters of nanoparticles in samples close to percolation [10]. The presence of nanoparticles aggregates with a chain-like morphology enhances the effect of magnetostatic interactions due to the strong ferromagnetic correlations of the magnetic moments arising from the quasi-one-dimensional shape of the aggregates. The presence of similar short chain-like clusters of nanoparticles have been recently observed in granular films [10] in which dipolar interactions were shown to have a substantial effect on MR.

Acknowledgements

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