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Dipolar Interaction Effects in the Spin-Dependent Transport in Nanoparticle Systems

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The relation between the giant magnetoresistance (GMR) of a granular metal containing blocked and superparamagnetic clusters and its underlying magnetic microstructure is studied numerically. A Monte Carlo simulation is implemented to obtain the equilibrium magnetic configuration of the system, and the real space Kubo formula for a spin-dependent tight binding Hamiltonian is used to calculate the electronic conductivity. A flattening of the MR versus magnetization parabola is found at low fields ($H \approx 0$) due to the dipolar interactions between the magnetic grains. This effect is enhanced in systems containing a broad distribution of particle sizes. A maximum in the GMR value is found close to the percolation threshold and attributed to particle coalescence. Dipolar effects reduce the maximum GMR value and cause a faster decay with concentration.

The phenomenon of giant magnetoresistance (GMR) in metallic films containing transition metal grains (particles) [1] is attributed to the spin dependent scattering of conduction electrons off the magnetic grains. The asymmetry of the spin-up (majority) and spin-down (minority) conductivities in the transition metal and the spatial separation of the magnetic grains with different magnetization orientations at zero applied field are the two basic physical ingredients that lead to the GMR effect [2]. Therefore, the field dependence of the magnetoresistance reflects the evolution of the micromagnetic configuration of the system as the externally applied magnetic field varies in strength and direction.

Experimental evidence [3] that intergranular interactions play an important role in the formation of the magnetic configuration of the film is offered by the observed flattening of the MR versus sample magnetization parabola. The interactions are expected to be predominantly of magnetostatic character, due to the spatial separation of the granules. Hence, the competition between the (uniaxial) anisotropy energy of the grains and their dipolar interaction energy determines the magnetic configuration of the film.

Transition metal granules with typical diameters of a few nanometers are superparamagnetic (SPM) at room temperature and consequently, dipolar interactions offer the only mechanism for ordering of the grain moments [3]. However, as the temperature is lowered or, equivalently, as the average grain size is increased due to grain coalescence at high concentrations, blocking effects become important. In a previous work [4] we have presented a model for the study of the field dependent GMR in granular films, which is appropriate at high temperatures where all grains are SPM. In this work, we apply the same formalism to study a polydisperse sample containing a mixture of SPM and blocked grains.

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.

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The lattice parameter is equal to the particle diameter ($D = a$). Each particle carries a three-dimensional moment of magnitude $m_0 = M_s V_0$. Strong exchange interactions are assumed between nearest neighbor particles leading to formation of magnetic clusters. Consequently, at any finite concentration x of magnetic particles, the sample contains a variety of magnetic clusters with random shape and a wide distribution of volumes [4]. 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. Dipolar energy terms between moments belonging to the same cluster are neglected. 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

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

where c, c' denote clusters, i, j denote sites within the clusters, n_c is the number of particles that constitute a cluster, $\mathbf{e}_c$ is the easy axis direction, and hats indicate unit vectors. The energy parameters are $g = (M_s V_0)^2/D^3$ for the dipolar energy, $k = K_1 V_0$ for the anisotropy energy, and $h = M_s V_0 H$ for the Zeeman energy, while the thermal energy is denoted $t = k_B T$. In this study we use $M_s = 1500$ emu/cm^3, $D = 3.5$ nm, and $K_1 = 1.2 \times 10^6$ erg/cm^3, which results in $g/k \approx 1.0$, for the parameters of Eq. (1). As in our previous works [4, 5], the equilibrium magnetic configuration is obtained by a Monte Carlo simulation using the standard Metropolis algorithm. Simulations were performed on a finite sample ($10 \times 10 \times 10$) for various particle concentrations and averages over ten 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

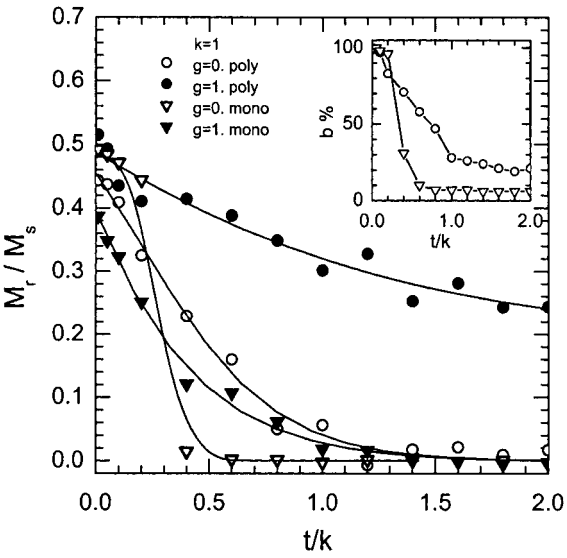

Fig. 1. Temperature dependence of remanence. The inset shows the fraction of blocked spins as a function of temperature for the non-interacting sample ($g = 0$) at zero field ($h = 0$). The concentration of magnetic particles is $x = 0.25$. The solid lines are guides to the eye

expressed in terms of the Green function of the system [4]. 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.

Results for the residual magnetization of a polydisperse system with $x = 0.25$ is plotted as a function of temperature in Fig. 1. For comparison, results for a monodisperse sample containing only clusters with size equal to the average value of the polydisperse sample ($\langle V \rangle = 3.1 V_0$) are also plotted. An abrupt drop of the remanence is seen for the non-interacting monodisperse sample at the blocking temperature $t_b \approx 0.5k$. Both the size distribution of the clusters and the dipolar interactions between the clusters introduce a distribution of energy barriers in the system and lead to a slow decay of the remanence with temperature. At temperatures above the blocking of the corresponding non-interacting system, magnetostatic interactions introduce ferromagnetic correlations between the magnetic moments, as seen in Fig. 1 for both types of samples. However, below the blocking temperature, the sample morphology is crucial. Reduction of the remanence due to dipolar interactions is seen in the monodisperse sample in agreement with previous studies [5–7], while enhancement of the remanence is observed in the system with clustered particles. This behavior originates from the presence of a few very large clusters in the polydisperse sample, that are blocked and produce a strong magnetic field which causes an efficient alignment of the small clusters. The fraction of blocked moments in a non-interacting sample and for zero field is defined as

$$b = \sum_c |\langle \hat{\mathbf{m}}_c \rangle \cdot \hat{\mathbf{e}}_c| \, n_c \Big/ \sum_c n_c \,, \tag{2}$$

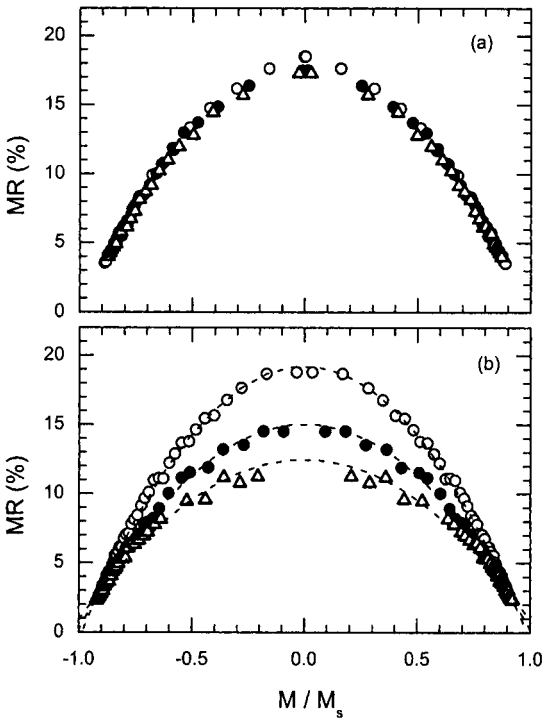

Fig. 2. GMR versus sample magnetization for a) a monodisperse sample containing clusters of size $n_c = 3$ and b) a polydisperse sample with $\langle n_c \rangle = 3.1 \pm 19\%$. For both samples $x = 0.25$. Open circles are non-interacting anisotropic clusters ($g = 0$, $k = 1$), closed circles are interacting anisotropic ($g = k = 1$) clusters, and triangles are 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 $E_F = 0$. The dotted lines are fitted parabolas

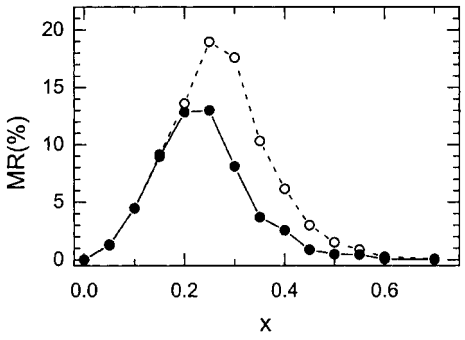

Fig. 3. Dependence of GMR on magnetic particle concentration. Open circles are non-interacting anisotropic clusters ($g = 0$, $k = 1$) and closed circles are interacting anisotropic ($g = k = 1$) clusters. The percolation threshold is at $x_p \approx 0.3$

where brackets $\langle \ldots \rangle$ indicate thermal average. Results for b are shown in the inset to Fig. 1. Furthermore, in Fig. 2 we show the GMR for the two types of samples at temperature $t = 1.2k$, at which the polydisperse sample contains a mixture of blocked and SPM clusters ($b \approx 0.25$), while the monodisperse sample contains only SPM clusters ($b \approx 0$). Both systems have similar values of GMR ($\approx$19%) when the interactions are neglected. This result indicates that the large and blocked clusters of the polydisperse sample do not contribute to the GMR effect. However, when the interactions are present, a larger suppression of the GMR effect ($\approx$25%) is observed in the polydisperse sample compared to the monodisperse sample. We therefore deduce that the large clusters, through the strong magnetostatic fields that they generate in the sample, are responsible for this effect. Further support is given to this argument by the results for the polydisperse sample in the case that the uniaxial anisotropy is neglected (Fig. 2b), which show a further flattening of the MR curve. In other words, as the ferromagnetic correlations between the moments are enhanced relative to the uniaxial anisotropy the flattening of the MR curve increases. Finally, we vary the magnetic particle concentration of the sample and we demonstrate the appearance of a maximum GMR effect just below the percolation threshold. The existence of an optimum concentration for the GMR effect is the result of the competition between the increasing number of magnetic scattering centers that enhance the effect and the increase of the average cluster size that suppresses the effect. Dipolar effects are most efficient close to percolation (Fig. 3) and produce a faster decay of the effect with magnetic particle concentration, in agreement with recent experiments [8].

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