Conditions for optimum giant magnetoresistance in granular metals

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The dependence of the giant magnetoresistance (GMR) of a metallic granular system on the concentration of magnetic particles is studied numerically. The effect of particle coalescence and dipolar interactions between the particles on the value of optimum GMR and the shape of the concentration dependence curve are discussed. The micromagnetic configuration of the system is obtained by a Monte Carlo algorithm that involves short-range effective exchange couplings and long range dipolar interactions. The conductivity is obtained using Kubo's formula for a tight binding Hamiltonian. A comparison of our results to experiments on metallic granular films is made.
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Magnetic granular systems have been extensively studied during the past few years in relation to their anomalous magnetotransport properties and in particular because they display giant magnetoresistance.[1,2] The observed large decrease of the resistivity of magnetic granular metals under application of a magnetic field is attributed to spin-dependent scattering of the conduction electrons off the magnetic particles. The microscopic origin of the scattering mechanism remains an open problem, but it is generally believed that spin-dependent scattering at the interface between the matrix and the magnetic particles is the most probable candidate.[3–5]

The complexity of the granular systems introduces certain important factors that determine the strength of the giant magnetoresistance (GMR) effect. In particular, early observations[2] indicated that there exists an optimum particle size at which the GMR is maximized. Various authors have investigated different combinations of matrix and magnetic particle materials, such as Ag–Co, Cu–Co, Cu–Fe,[2] and Fe–Cr (Ref. 6) in an attempt to maximize the effect. However, a comparative study of Ag–Ni with Ni–SiO_2 (Ref. 7) suggested that the nature of the matrix affects to a major extent the value of the sample resistivity but it does not affect the value of the normalized resistivity variation, namely the GMR. Background magnetic impurities play a crucial role in degrading the GMR effect as previous theoretical[3] and experimental studies[5,8] have demonstrated. The magnetic configuration of the system is also an important factor, as one would expect from the physical origin of the phenomenon. In this context, it has also been demonstrated both experimentally[9] and theoretically[10] that the presence of dipolar interactions between the magnetic particles degrade the GMR values and they manifest themselves through the flattening they cause to the GMR versus total magnetization parabola close to zero field values. Moreover, many experimental studies have appeared regarding the dependence of the GMR on the concentration (or volume fraction) of the magnetic particles.[4,5,11] In these works, samples have been grown by either traditional ultrahigh vacuum deposition techniques (sputtering, coevaporation) or by the more advanced technique of low-energy cluster beam deposition.[4,5] The major advantage of the latter is that extremely narrow particle size distributions are produced as well as matrices free of magnetic defects.[5] A common finding in all these studies has been that an optimum concentration is observed, close to the theoretical percolation threshold, for a maximum GMR value. However, the exact value of the optimum concentration and the shape of the magnetoresistance (MR) versus concentration curve varies.

The purpose of this work is to study the concentration dependence of MR and investigate the factors that determine the optimum concentration value and the shape of the concentration dependence curve. To this end we have developed a numerical model that includes two basic ingredients that are important in the study of granular systems. Namely, the effect of coalescence of particles at high density and the dipolar interactions between them. It is shown that the coalescence of the particles is the necessary process in order to obtain a maximization of GMR at intermediate concentrations, i.e., close to the percolation threshold. We show that the interplay between the interparticle exchange interactions and the dipolar interactions modify both the position of optimum concentration and the maximum value of GMR.

We model the magnetic structure of the granular system by an ensemble of classical spins (magnetic moments) located at random on the sites of a simple cubic lattice. Each spin represents a magnetic nanoparticle and the total energy of the system reads

$$E=\sum_i \left[g\sum_j \frac{\hat{m}_i\cdot\hat{m}_j-3(\hat{m}_i\cdot\hat{R}_{ij})(\hat{m}_j\cdot\hat{R}_{ij})}{R_{ij}^3}-h(\hat{m}_i\cdot\hat{H}) - J_{\mathrm{eff}}\sum_{\langle j\rangle}\hat{m}_i\cdot\hat{m}_j \right] \quad (1)$$

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with m_i the magnetic moment (spin) of the ith grain, g the dipolar strength, h the Zeeman energy, and R_{ij} the intergranular distance. Hats indicate unit vectors. The energy parameters in Eq. (1) are measured in units of the dipolar strength ($g=1$), while distances are measured in units of the particle diameter. In a previous study[12] we have shown that for temperatures above the blocking temperature of the isolated particles and for a wide range of particle concentrations (up to ~ 0.8) the interparticle dipolar interactions have a ferromagnetic character. In this regime, the single-particle anisotropy is immaterial to a first approximation and we therefore do not include the corresponding terms in Eq. (1). The last term in Eq. (1) is a short range (first nearest neighbors) ferromagnetic term and describes the effect of interparticle exchange coupling across their contact surface. In the limit of infinite J_{eff}, the magnetic moments of neighboring particles are aligned at all field values and are forced to rotate coherently under the influence of the external field. This limit describes the case of coalesced particles. On the other hand, for $J_{\text{eff}}=0$, neighboring particles are well-separated with negligible interparticle exchange. Therefore the model of magnetic structure, Eq. (1), serves as an interpolation scheme between systems containing well-separated particles and systems with coalesced particles.

To calculate the conductance we consider an electrode–sample–electrode geometry and use a tight-binding Hamiltonian:[10,13]

$$H=\sum_{i,\alpha}\varepsilon_i c_{i\alpha}^{+}c_{i\alpha}+V\sum_{\langle i,j\rangle,\alpha}c_{i\alpha}^{+}c_{j\alpha} -J\sum_{\substack{i\in \mathrm{MG}\\ \alpha,\beta}}c_{i\alpha}^{+}(\hat{m}_j\cdot\hat{\sigma})_{\alpha\beta}c_{i\beta}, \tag{2}$$

where the on-site atomic potentials ε_i assume the values ε_{LW} in the electrodes, ε_{NM} in the nonmagnetic matrix, and ε_{MG} on the magnetic grains. Also, V is the nearest neighbor hopping integral, J is the exchange potential of the magnetic material, σ_x, σ_y, and σ_z are the Pauli matrices, and α and β are the spin indices. The energy parameters in Eq. (2) are measured in units of the hopping integral ($V=1$). The use of a single-site potential to describe the magnetic grain is justified as long as the electronic mean free path is larger than the particle diameter. The conductance of the system is given by

$$\Gamma=\frac{2e^2}{h}\mathrm{Tr}(p_z\,\mathrm{Im}\,G^{(+)}p_z\,\mathrm{Im}\,G^{(+)}), \tag{3}$$

where $G^{(\pm)}=(E_F-H\pm i\eta)^{-1}$ is the Green's function at the Fermi level and p_z is the component of the electron momentum operator along the axis of current flow (z axis). Finally, the field-dependent magnetoresistance is defined as $\mathrm{MR}(H)=[R(H)/R_s)-1]\times 100$, where the field-dependent resistance $R(H)=1/\Gamma(H)$ and R_s is the resistance of a fully saturated sample.

It is clearly seen from Eq. (2) that the configuration of magnetic moments introduces a distribution of local potentials into the sample that determines its resistance. These potentials are distributed randomly in space but their strengths are spatially correlated according to the moment–moment correlation function, the values of which are determined by the relative strength of the interaction energy strengths in Eq. (1) and the thermal energy $t=k_BT$.

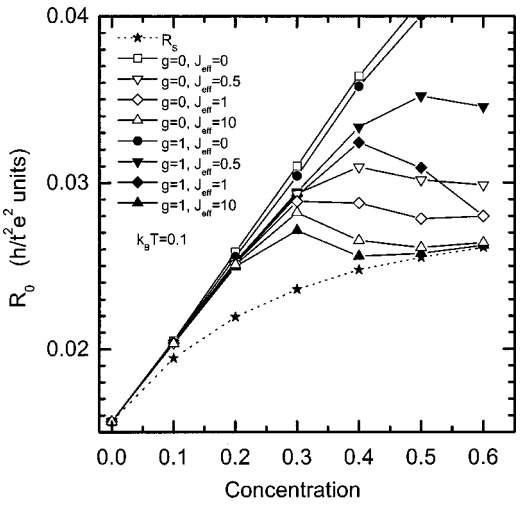

FIG. 1. Resistance as a function of magnetic particle concentration.

The Monte Carlo simulation[11,12] is performed at low temperature ($t/g=0.1$) so that ordering effects due to dipolar interactions are maximized. The effective exchange parameter between neighboring particles is also assumed to be larger than the thermal energy ($J_{\text{eff}}/t=5$, 10, and 100) because we are interested in the regime where ferromagnetic order due to interparticle exchange interactions is weakly disrupted by thermal agitation ($J_{\text{eff}}>k_BT$) and in the regime where coalescence occurs with thermal effects being negligible ($J_{\text{eff}}\gg k_BT$). For the electronic structure parameters we use $\varepsilon_{\text{LW}}=\varepsilon_{\text{NM}}=0$ so that there is no contribution to the resistance of the system from the electrode–sample contact and from the nonmagnetic matrix. Also we choose $\varepsilon_{\text{MG}}=+1$ and $J=+1$ so that the electrons in the majority spin band are less scattered by the magnetic grains than those in the minority band, when the grains are aligned ferromagnetically.[10,13] Finally, we have taken the Fermi level at the band center ($E_F=0$), so that the Fermi wavelength is comparable to the minimum particle distance or the particle size ($\lambda\sim a$).

In Fig. 1 we show the dependence of the sample resistance at zero applied magnetic field (R_0) and at saturation (R_s) on the concentration of magnetic particles and in Fig. 2 we plot the corresponding dependence of the magnetoresistance. We note in Fig. 1 that R_0 increases linearly with the concentration for $x<0.2$ and no substantial differences are observed for systems with different strengths of interparticle exchange. This result is anticipated, as for concentrations up to about 15% the number of particle clusters (pairs, triplets, etc.) is negligibly small. Therefore interparticle exchange couplings are very rare and also the dipolar interactions have practically no effect in dilute samples. In this concentration regime, the conditions for weak scattering (dilute limit, weak potential) are fulfilled and the linearity of the R_0 with concentration is expected.[3] This behavior is also in agreement with recent observations in granular systems.[5] The linear dependence of R_0 persists for higher concentrations, only for the systems where interparticle exchange coupling is absent. In particular, for a system containing superparamagnetic or dipolar interacting but well-separated particles, the continuous increase of R_0 is a consequence of the increasing number

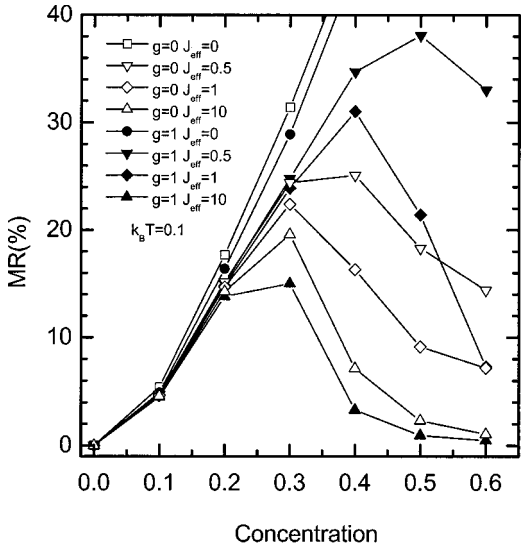

FIG. 2. Magnetoresistance as a function of magnetic particle concentration.

of scattering magnetic centers introduced in the system as the concentration increases.

For concentrations above $x \sim 0.2$ and when the interparticle exchange coupling is nonzero, the curves are drastically different. In particular, as the effective exchange is increased (open symbols in Fig. 1) a maximum in R_0 develops close to $x=0.3$ which corresponds to the site percolation threshold (x_p) for the system under consideration. This happens because at $x=x_p$ the magnetic–non magnetic interface area is maximized[5] and therefore the scattering of the carriers, which happens only at the interface within our model, is maximized too. When the interparticle exchange coupling does not overwhelm the thermal energy ($J_{\text{eff}}/t=5,10$), ferromagnetic domains occur within each cluster of particles causing extra scattering of the electrons and leading to $R_0 > R_s$ in the high concentration limit. Notice also that the fact that $R_0 \sim R_s$ for $x>0.4$ and $J_{\text{eff}}/t=100$ is consistent with the picture of coherent spins within the clusters or in other words the existence of a single magnetic domain within the clusters.

The role of the dipolar interactions (closed symbols in Figs. 1 and 2) changes depending on the strength of the interparticle exchange. In particular, dipolar interactions reduce the resistivity in the case of strong exchange ($J_{\text{eff}}/t=100$) while they have the opposite effect in the case of moderate exchange ($J_{\text{eff}}/t=5,10$). When the exchange is strong, large single domain magnetic clusters exist in the sample that produce strong dipolar fields, which result in strong ferromagnetic correlations of the cluster moments and a consequent reduction of the resistance. On the other hand, when the effective exchange is weak or moderate, dipolar interactions have an antiferromagnetic character within the clusters and reduce the magnetic ordering caused by the exchange, thus increasing the resistance.

The above trends of the resistance are also apparent in the concentration dependence of the magnetoresistance (Fig. 2). Notice that in the case of particle coalescence ($J_{\text{eff}}/t=100$) and when the dipolar interactions are active the GMR curve lies below the corresponding one for noninteracting clusters, especially above the percolation threshold. Given also that only interface spin-dependent scattering is considered within our model, the GMR curve for the system containing coherent and noninteracting clusters (open, up-triangles in Fig. 2) follows exactly the dependence of the total interface area of the clusters in the system. Therefore our data in Fig. 2 support recent magnetoresistance measurements in a Co–Ag granular film[5] where downward deviations of the GMR curve from the total surface curve were observed. Finally, it is interesting to notice in Fig. 2 that as the interparticle exchange coupling decreases the optimum GMR value occurs at higher concentrations. Also when the exchange coupling is moderate ($J_{\text{eff}}/t=5,10$) (with or without dipolar interactions) the shape of the curve around the maximum can be rather broad without a sharp drop at higher concentrations. A similar broad peak of the GMR versus concentration curve has been recently observed[14] in a $Co-Al_2O_3$ granular film and attributed to suppression of the Coulomb blockade. Our simulations indicate that the same shape arises when the formation of bulk material has not occurred but moderate exchange interparticle interactions ($J_{\text{eff}}/kT \sim 10$) are present. In conclusion, provided that the strength of the interparticle exchange can mimic the degree of formation of a bulk and magnetically saturated material, we suggest that the changes in shape and peak position of the MR versus x curve reflect this formation.

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