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Magnetization and Giant Magnetoresistance of the System of Interacting Fine Particles

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Abstract

The magnetization and giant magnetoresistance (GMR) of nanosized magnetic particles embedded in a nonmagnetic metallic matrix are numerically investigated. By considering the classical dipolar interactions, we apply a Monte-Carlo simulation technique to calculate M vs H, GMR vs H, and GMR vs (M/M_s) with M, the average magnetization along the normal to the layer, M_s, the saturated magnetization, and H, the applied magnetic field. It is shown that the interfacial spin-dependent scattering of conductance electrons is dominant in GMR effect and the distance between the neighboring particles is an important parameter to obtain the GMR effect, while the size distribution only modify the shape of the curve of GMR versus H.

1. Introduction

The discovery of the giant magnetoresistance (GMR) effect in inhomogeneous alloys of magnetic and nonmagnetic metals [1] has attracted a great deal of attention to these materials. They consist of nanosized particles or clusters (e.g., Co, Fe, Ni) embedded in a nonmagnetic metallic matrix (typically Cu, Ag). The magnetic transport properties of granular metals are concerned with the size and spatial distributions of the fine particles or clusters and the interaction between the particles. Previous works studied the dependence of GMR on the size distributions [2] and successfully explained some experimental results. The interactions between particles can have a dipolar, Ruderman–Kittel–Kasuya–Yosida (RKKY), or a superexchange character, depending on the magnetic properties of the matrix. Altbir *et al.* [3] found that the classical dipolar interactions are dominant in Co-Cu systems.

2. Theory

In the present work we study the magnetization and GMR effect of the assembly of single–domain spherical ferromagnetic particles, Each particle is a saturated single domain and its magnetic moment $\vec{\mu}_i$, and the direction of its uniaxial anisotropy axis is random in space. The particles are placed in a square array consisting of 12×12 cells. The diameter of the particle i is d_i and the distance between two neighboring particles is r_0. After considering the classical dipolar interaction and crystalline anisotropy energy, the total energy of the system for a given configuration $\{\vec{\mu}_i\}$ of the magnetic moments is

$$E(\mu_i) = \sum_{i=1}^{N}[\sum_{j>i} E_{ij} + K_u V_i \sin^2 \alpha_i - \vec{\mu}_i \cdot \vec{H}], \tag{1}$$

with E_{ij}, the energy of the dipolar interaction, K_u, the effective anisotropy constant, V_i, the volume of a particle i, and α_i, the angle between the direction of the crystalline axis and $\vec{\mu}_i$. For a given temperature, the reduced magnetization $< m >$ can be calculated by averaging $m = M/M_s = |\sum_i \vec{\mu}_i|/(N\mu)$ over cluster configurations after thermal equilibrium has been reached. The crucial factor for GMR in granular system is the average value $< \cos\theta_{ij} >$, where θ_{ij} is the relative angle between the magnetic moments in sites i and j [4]. It implies that the magnetic transport properties are primarily caused by the spin–dependent scattering process of conductance electrons between magnetic particles. Namely, the spin-dependent scattering at interfaces between the magnetic particles gives rise to the GMR to a greater extent than the scattering within the magnetic particles would do [5]. In the case that the distance between the neighboring particles does not far exceed the electronic mean free path λ [6], the variation in resistivity of a granular system with the degree of field-induced magnetic order may be simply pictured as $\rho = \rho_0 - k < \cos\theta_{ij}^{(\lambda)} >$, where ρ_0 and k are constants. Assuming that there are no correlations between the magnetic moments of particles, the magnetoresistance $\Delta\rho/\rho$ can be written as $\Delta\rho/\rho = -(k/\rho_0) < \cos\theta_{ij}^{(\lambda)} >^2 = -(k/\rho_0)m^2$. Such a quadratic dependence of $\Delta\rho/\rho$ on m is actually found by some experiments [1]. However, other experiments [2] showed that $\Delta\rho/\rho$ does not vary quadratically with m because of the size distribution of magnetic particles and interaction between them.

For the system involving coupling between magnetic particles, the assumption, that the average value $< \cos\theta_{ij}^{(\lambda)} >_0$ for $H = 0$ is not vanished, gives

$$\Delta\rho/\rho = (< \cos\theta_{ij}^{(\lambda)} >_0 - < \cos\theta_{ij}^{(\lambda)} >)(Q - < \cos\theta_{ij}^{(\lambda)} >_0), \tag{2}$$

where $Q = \rho_0/k$ is the field–independent constant.

The thermal averages of the system above are obtained using the standard MC procedure and the Metropolis algorithm [7]. The system is assumed to have reached thermal equilibrium after 10^4 Monte-Carlo steps per spin. Then, we are able to get the thermal averages as an arithmetic average over the accepted configurations (500 accepted configurations for ensemble averages), and to calculate the $< m >$ and GMR. Data for our MC simulation is generated and calculated as follows. Each particle is assigned a random crystalline anisotropy ($K_u = 4.0 \times 10^6 \mathrm{erg/cm^3}$) and a random direction of magnetic moment at initial state. These particles are placed in the magnetic field $\vec{H}$ applied along the normal to the array. The distance r_0 between the particles was taken as 6.0 nm (except for the up triangles in Fig. 1) which is comparable in magnitude with λ [6]. Single–domain ferromagnetic particles exhibit the phenomenon of superparamagnetism and the blocking temperature $T_b^{(0)}$ for $H = 0$ of a particle of diameter d=4 nm is equal to 38 K [8]. So we choose T=40 K close to $T_b^{(0)}$.

3. Results

First, for simplicity, we choose the same diameter d_i(=3 nm) of all the magnetic particles, which is the typical average size of particles for granular materials [6]. In Fig. 1 we plot the graph of magnetization M/M_0 vs H. Four different sets of data are shown that correspond to a system of particles with random anisotropy only (squares; r_0 = 6 nm), a moderate dipolar system with random anisotropy (circles; r_0 = 6 nm), a system with moderate dipolar interaction only (triangles down; K_u = 0, r_0 = 6 nm), and a strong dipolar system with random anisotropy (triangles up; r_0 = 3 nm). We notice that M exhibits the different field dependence, depending on the interplay of the single–particle anisotropy and the dipolar interaction effects. *E.g.*, for a

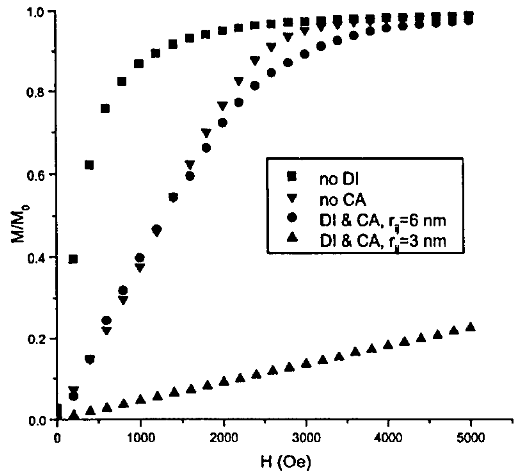

Fig.1. The dependence of M/M_s on H for T=40 K. The symbles are obtained when we do not consider the dipolar interaction(DI) (squares), or do not include the crystalline anisotropy (CA) (down triangles). The circles and up triangles are obtained by considering both the DI and CA when the r_i equals 6nm and 3nm respectively.

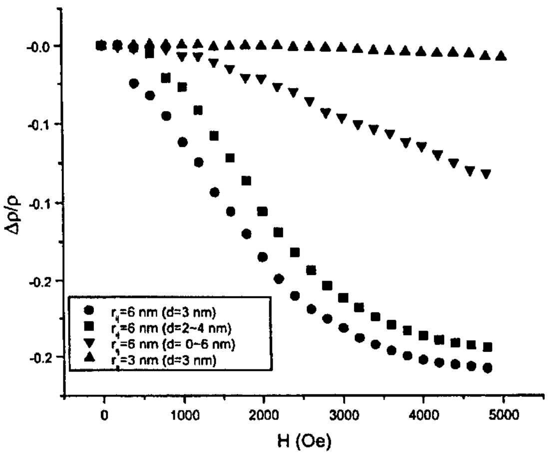

Fig.2. The dependenc of GMR on H for T=40 K. The symbles are obtained when r_i=6nm, d=3nm (circles), d=2~4nm(squares), d=0~6nm(down triangles), and r_i=3nm, d=3nm (up triangles).

system with random anisotropy only the magnetization reversal of the assembly of particles is well described by both a coherent rotation of $\vec{\mu}_i$ from the easy axes to the direction of $\vec{H}$ and by a thermally activated process over the anisotropy barrier.

A pure dipolar system ($K_u = 0$) For $H = 0$ possesses the interaction–induced anisotropy. Out–of–plane orientation $\vec{M}$ has a large energy due to the demagnetization effect. Therefore in–plane configuration of $\vec{\mu}_i$ is realized. For the free boundary conditions accepted here the ground state is antifferromagnetic due to the demagnetization effect of the lateral boundaries. This effect is not so strong compared with the demagnetization effect of the surface. Once again, if a field is applied to the array the magnetic moments $\vec{\mu}_i$ rotate coherent to the direction of $\vec{H}$.

If both the dipolar interaction and the random uniaxial anisotropy of particles are involved it is more difficult for $\vec{M}$ to reach saturation since these effects impede the collinear ordering of $\vec{\mu}_i$ and reduce the magnetization of the system. The anisotropy induced by dipolar interactions is very sensitive to the spatial arrangement of the particles (average distance between the particles and their size) and it is enhanced with decreasing r_0. Therefore in a strong dipolar system (triangles up; $r_0 = 3$ nm) the $\vec{M}$ is harder to be saturated in comparison with a moderate dipolar system (circles; $r_0 = 6$ nm).

In Fig. 2 we plot the field dependence of $\Delta\rho/\rho$ for T=40 K. All curves are calculated in terms of Eq. (2) with Q=5.2 after considering both the effects of anisotropy and interactions when the system reaches the equilibrium state. To investigate the influence of the distance r_0 and the particle–size distribution on the GMR four different sets of data are shown that correspond to particles of fixed diameter with strong (up triangles; d_i=3 nm, r_0=3 nm) and weak (circles; d_i=3 nm, r_0=6 nm) dipolar interactions, a narrow particle–size distribution from 2 to 4 nm (squares; $\bar{d}$=3 nm, r_0=6 nm), and a wide particle–size distribution from 0 to 6 nm (down triangles; $\bar{d}$=3 nm, r_0=6 nm) with $\bar{d}$ is the average particle size. From Fig. 2 it follows that the GMR effect depends crucially on the particle density. If the dipolar interaction are strong (d_i=3 nm, r_0=3 nm) all the particles are coupled to each other, and the negative GMR disappears. For a narrow particle–size distribution from 2 to 4 nm (squares) the field dependence of $\Delta\rho/\rho$ is closed to that for the system of particles with fixed diameter and a weak dipolar interection (circles). In this case the blocking temperature of the maximum particle $T_b \approx$38 K, and all the particles are superparamagnetic. This result was observed in [5]. For a wide particle–size distribution from 0 to 6 nm, certain of large particles are blocked at 40 K in the region of strong magnetic fields, whereas the rest small particles are still in the superparamagnetic state. Then the modulus of $|\Delta\rho|/\rho$ is substantially reduced relative to the case of a narrow particle–size distribution. This effect was observed and explained in [2] for the systems with a wide particle–size distribution. So we believe that the size distribution strongly affects the shape of the curves of GMR vs H.

In order to investigate in more detail the role of a particle–size distribution in GMR effects, we show in Fig. 3 the dependence of $\Delta\rho/\rho$ vs M/M_s for a narrow (hollow squares) and wide (up triangles) particle–size distributions. From Fig. 3 it follows that for a narrow particle-size distribution from 2 to 4 nm, the behavior of GMR vs (M/M_s) is close to the parabolic line, whereas for a wide particle–size distribution from 0 to 6 nm, it is deviated essentially from the parabolic law. It indicates that the wide distribution of magnetic particles may explain the noncompliance with the parabolic law for the GMR as a function of the $\vec{M}$ [2].

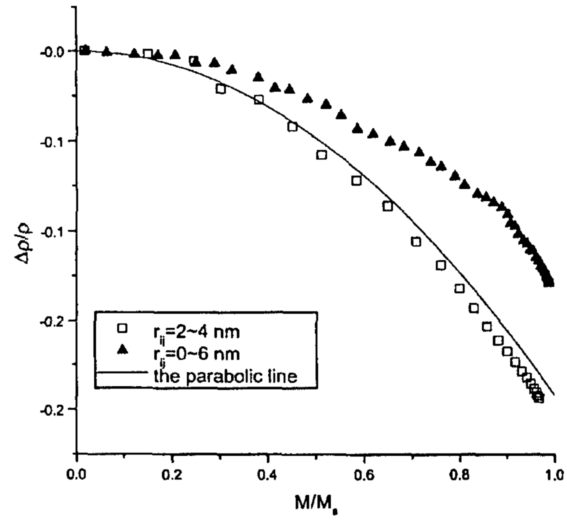

Fig.3. The dependence of GMR on M/M_s for T=40 K. The hollow squares and up triangles are obtained by varing of r_{ij} from 2 to 4 nm and 0 to 6 nm respectively. The solid line is the parabolic line.

4. Conclusion

In conclusion, we have presented the results for the field dependence of M and GMR of a granular magnetic film. We demonstrate that the magnetic properties of the system depends essentially on the particle–size distribution and the average distance between the magnetic particles. To discuss experimental data the state of single–domain magnetic particles is usually assumed to be blocked or collective at low temperatures and to be superparamagnetic at high temperatures. Within our MC approach, there is no need for making *a priori* assumptions about the particle state. However, it is likely that the high density regime favours the collective state, and the low density, wide particle–size distribution and strong magnetic field regime favours the blocked state. The collective behaviour at high particle density reveals itself in the disappearance of the negative GMR effect. A manifestation of the blocked state effects is a substantial decrease in $|\Delta\rho|/\rho$ for the low density and wide particle–size distribution in the region of strong magnetic field.

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