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A theory of giant magnetoresistance of binary magnetic granular composites

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Abstract. A method is presented, which combines effective medium theory (EMT) with a two-channel conducting model, in order to study the giant magnetoresistance (GMR) effect of a magnetic granular composite (such as $Co_x Cu_{1-x}$) containing some superparamagnetic particles. The magnetic field and temperature dependences of GMR are investigated and the calculated results are found to be in very good agreement with recent experimental data.

1. Introduction

Since the discovery of the giant magnetoresistance (GMR) effect in a magnetic granular system [1, 2], the GMR of a variety of magnetic granular materials have been reported and a variety of technologies have been used to produce heterogeneous magnetic alloys consisting of immiscible elements. Recently, rapid quenching (meltspinning) has been shown to be a suitable method for preparing $Co_x Cu_{1-x}$ granular alloys showing bulk GMR effect [3–7]. Experimental measurements show that the magnetic granules exhibit a size distribution. If the ferromagnetic particles are sufficiently small, then these particles will become superparamagnetic (SPM). The magnetoresistance properties of such SPM particles were first studied by Gittleman *et al* [8], who indicated that the magnetoresistance should be proportional to the square of the magnetization. As a result, the GMR data for magnetic granular composites were often plotted as a function of the magnetization as it was expected that quadratic (parabolic) dependence would be observed. However, recent experimental studies have shown that the GMR of the meltspun granular sample of $Co_x Cu_{1-x}$ had almost a linear dependence on the magnetic field at high temperatures, and the quadratic (parabolic) dependence of the GMR on magnetization has not been observed [3–6]. This result was attributed to the presence of a range of sizes for very small ferromagnetic particles [9–11].

It is found that, at a given temperature, when the size of a magnetic particle is less than a critical size, the particle will become a SPM [12]. Therefore, if the magnetic particles are present in a range of sizes, only a fraction of magnetic particles will be SPM. For a magnetic granular composite consisting of very small magnetic particles, only a portion of particles become SPM above the 'blocking' temperature. The 'blocking' temperature is defined as the temperature above which some particles with a range of sizes, all less than a critical size, are SPM, and below which these particles are not SPM [13]. In other words, the 'blocking' temperature depends on the size distribution of granules so that, at any given temperature, only

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a portion of magnetic particles are SPM ('unblocked') while the remainder will not be SPM ('blocked').

In this paper, we will consider small Co particles with a packing density, p, embedded in a Cu matrix. Since the sample contains very fine ferromagnetic Co particles with a range of sizes, there will be a corresponding range of blocking temperatures. At a given temperature, a portion of the magnetic particles with packing density, p_s, are SPM, and the rest with packing density, p_n, are ferromagnetic (non-SPM). The relation $p_s + p_n = p$ should be satisfied. When there is no applied magnetic field, the magnetic moments of the non-SPM particles are randomly distributed in the non-magnetic matrix and the sample is in a demagnetized state. If a magnetic field $\boldsymbol{B}$ is applied to this sample, we assume that the magnetic moments of non-SPM particles will be aligned and saturated, and that the SPM particles will have a distribution of magnetic moments which have different angles, θ, from $\boldsymbol{B}$.

The magnetoresistance is defined as

$$MR(B) = \frac{\rho(B) - \rho(0)}{\rho(0)} \tag{1}$$

where $\rho(B) = \sigma^{-1}(B)$, and $\rho(B)$ and $\sigma(B)$ are the resistivity and the conductivity of the sample in an applied magnetic field $\boldsymbol{B}$, respectively.

Based on the two-channel conducting model, we will calculate the effective conductivity of this system. In a demagnetized state, on average, the effective conductivity $\sigma_e(0)$ of the system has no relation to spin-dependent scattering. When a magnetic field $\boldsymbol{B}$ is applied, $\sigma_e(B)$ will be related to the spin-dependent scattering, and the contribution to the conductivity of the system, arising from spin-up electrons, is different from that of spin-down electrons. Here, we will investigate the temperature dependence of the GMR effect of magnetic particles Co with a range of sizes embedded in the Cu matrix. The temperature dependence of the GMR effect is an important problem for understanding the GMR's physical origin and its practical application.

2. Theory

Assuming an applied field $\boldsymbol{B}$, the magnetic moments of the SPM particles will have a distribution and those of the non-SPM particles will be aligned and saturated. Therefore, for a non-SPM ferromagnetic particle, the spin direction of conductance electrons may be parallel ('spin-up') or anti-parallel ('spin-down') to the direction of the magnetic moment of the particle. For a spin-up electron, the contribution to its resistance will be high due to scattering events and thus its conductance will be low (L), while for a spin-down electron the contribution to its conductance will be high (H). σ_L and σ_H indicate the conductivities for spin-up and spin-down electrons respectively. Using the signs + (or −) for spin-up (or spin-down) and the signs n (or s) for non-SPM (or SPM) particles, we can describe the conductivities of non-SPM and SPM particles for spin-up and spin-down electrons as σ^+_{nL}, σ^-_{nH}, σ^+_s and σ^-_s, respectively. We noted that the magnetic moments of non-SPM ferromagnetic particles will be quickly aligned and saturated in the presence of a relatively small applied field. When the applied field increases, the applied field $\boldsymbol{B}$ will not have an effect on the conductivity of non-SPM particles, and the average conductivity $\langle\sigma^+_{nL}\rangle$ and $\langle\sigma^-_{nH}\rangle$ can be expressed by

$$\begin{aligned} \langle\sigma^+_{nL}\rangle &= \sigma^+_L \\ \langle\sigma^-_{nH}\rangle &= \sigma^-_H \end{aligned} \tag{2}$$

where σ^+_L and σ^-_H are independent of $\boldsymbol{B}$.

But, for σ_s^+ and σ_s^-, because the magnetic moments $\mu_s(B)$ of SPM particles are dependent on $\boldsymbol{B}$, we assume

$$\begin{aligned}\sigma_s^+ &\propto C_s^+\mu_s(B)\\ \sigma_s^- &\propto C_s^-\mu_s(B)\end{aligned}\tag{3}$$

where C_s^+ and C_s^- are coefficients.

Considering the angle, θ, between $\mu_s(B)$ and $\boldsymbol{B}$, we can write the expressions for σ_s^+ and σ_s^- according to the proposal of Gittleman *et al* [8], where the values of σ_s^+ and σ_s^- change linearly with $\cos\theta$,

$$\sigma_s^+ = \tfrac{1}{2}C_s^+\mu_s(B)(1+\cos\theta) + \tfrac{1}{2}C_s^-\mu_s(B)(1-\cos\theta)\tag{4}$$

$$\sigma_s^- = \tfrac{1}{2}C_s^-\mu_s(B)(1+\cos\theta) + \tfrac{1}{2}C_s^+\mu_s(B)(1-\cos\theta).\tag{5}$$

Taking the thermal average of σ_s^+ and σ_s^-, we get

$$\langle\sigma_s^+\rangle = \tfrac{1}{2}C_s^+\langle\mu_s(B)\rangle\langle 1+\cos\theta\rangle + \tfrac{1}{2}C_s^-\langle\mu_s(B)\rangle\langle 1-\cos\theta\rangle\tag{6}$$

$$\langle\sigma_s^-\rangle = \tfrac{1}{2}C_s^-\langle\mu_s(B)\rangle\langle 1+\cos\theta\rangle + \tfrac{1}{2}C_s^+\langle\mu_s(B)\rangle\langle 1-\cos\theta\rangle.\tag{7}$$

As is well known, $\langle\cos\theta\rangle$ equals the Langevin function, $L(\mu B/k_BT)$, and $\langle\mu_s(B)\rangle = CL(\mu B/k_BT)$, where C is a constant. Then, taking $\sigma_{sL} \equiv CC_s^+$, $\sigma_{sH} \equiv CC_s^-$, and $\alpha \equiv \mu B/k_BT$, equations (6) and (7) can be rewritten as follows

$$\langle\sigma_s^+\rangle = \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})L(\alpha) + \tfrac{1}{2}(\sigma_{sL}-\sigma_{sH})L^2(\alpha)\tag{8}$$

$$\langle\sigma_s^-\rangle = \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})L(\alpha) + \tfrac{1}{2}(\sigma_{sH}-\sigma_{sL})L^2(\alpha).\tag{9}$$

For simplicity, we assume that the contribution to magnetization, arising from the particles whose magnetic moment lies between μ_s and $\mu_s + \mathrm{d}\mu_s$, is independent of μ_s, up to a maximum of μ_m [3]. Then we take the integral over each term in equations (8) and (9) to obtain

$$\begin{aligned}\langle\sigma_s^+\rangle &= \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})\frac{1}{\mu_m}\int_0^{\mu_m} L(\alpha)\,\mathrm{d}\mu + \tfrac{1}{2}(\sigma_{sL}-\sigma_{sH})\frac{1}{\mu_m}\int_0^{\mu_m} L^2(\alpha)\,\mathrm{d}\mu\\ &= \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})I_1(B) + \tfrac{1}{2}(\sigma_{sL}-\sigma_{sH})I_2(B)\end{aligned}\tag{10}$$

$$\begin{aligned}\langle\sigma_s^-\rangle &= \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})\frac{1}{\mu_m}\int_0^{\mu_m} L(\alpha)\,\mathrm{d}\mu + \tfrac{1}{2}(\sigma_{sH}-\sigma_{sL})\frac{1}{\mu_m}\int_0^{\mu_m} L^2(\alpha)\,\mathrm{d}\mu\\ &= \tfrac{1}{2}(\sigma_{sL}+\sigma_{sH})I_1(B) + \tfrac{1}{2}(\sigma_{sH}-\sigma_{sL})I_2(B)\end{aligned}\tag{11}$$

where

$$I_1(B) = \frac{1}{\alpha_m}\ln[\sinh\alpha_m/\alpha_m]\tag{12}$$

$$I_2(B) = -\frac{L(\alpha_m)}{\alpha_m} + \frac{1}{\alpha_m}\int_0^{\alpha_m}\left(1-\frac{2L(\alpha)}{\alpha}\right)\mathrm{d}\alpha\tag{13}$$

$$\alpha_m = \frac{\mu_m B}{k_BT}\tag{14}$$

and μ_m is the largest magnetic moment of the Co particle which is an unblocked SPM particle at temperature T.

As the ferromagnetic particles are randomly embedded in the non-magnetic matrix, we use effective medium theory (EMT) [14, 15] to get the effective conductivity σ_e^+ and σ_e^- for spin-up and spin-down conductance electrons respectively. Then, the effective conductivity σ_e^+ and σ_e^- can be obtained from the following equations

$$p_s\frac{\sigma_s^+-\sigma_e^+}{\sigma_s^++2\sigma_e^+} + p_n\frac{\sigma_L^+-\sigma_e^+}{\sigma_L^++2\sigma_e^+} + (1-p)\frac{\sigma_h-\sigma_e^+}{\sigma_h+2\sigma_e^+} = 0\tag{15}$$

$$p_s\frac{\sigma_s^--\sigma_e^-}{\sigma_s^-+2\sigma_e^-} + p_n\frac{\sigma_H^--\sigma_e^-}{\sigma_H^-+2\sigma_e^-} + (1-p)\frac{\sigma_h-\sigma_e^-}{\sigma_h+2\sigma_e^-} = 0\tag{16}$$

where σ_h is the conductivity of the Cu matrix. Using the two-channel conducting model, we obtain the system's effective conductivity, $\sigma_e(B)$,

$$\sigma_e(B) = \sigma_e^+ + \sigma_e^-. \tag{17}$$

Then, according to the definition of magnetoresistance (MR), we have

$$MR = \frac{\sigma_e^{-1}(B) - \sigma^{-1}(0)}{\sigma^{-1}(0)}. \tag{18}$$

It should be noted that, when the packing density, p, of Co particles is given, the packing densities, p_s and p_n, are dependent on temperature T. Due to a progressively larger fraction of the ferromagnetic Co, particles will become 'unblocked' SPM particles with an increase of in temperature.

3. Comparison with experiments

In order to calculate the value $\sigma_e(B)$ and then *MR(B)*, we need to select the magnitudes of some parameters. For the sample Co_{13} Cu_{87}, the atomic percentage of Co particles is 0.13, while the packing density p of magnetic particles will be 0.122, which we use in our calculation. Based on the fact that p_s will increase and p_n will decrease with the increase in temperature, we selected the following values of p_s/p_n: 2.05, 3.07 and 3.69 corresponding to three different temperatures 7, 50 and 150 K, in order to fit to the experimental data. For simplicity, the conductivities of materials Cu and Co in the absence of an applied magnetic field are taken as 3.9 and 1.0 (arbitrary units). The parameters σ_{sH} and σ_{sL} are related to temperature and applied magnetic field. According to the experimental report in [3], we can select these magnitudes of σ_{sH} and σ_{sL} so that they can be determined by a non-linear least-squares fit to the *MR(B)* experimental curves in [3]. For example, the change of σ_{sH} and σ_{sL} with the applied magnetic $\boldsymbol{B}$ at given temperature 50 K, is listed in table 1. Based on the fact that the conductivity σ_L^+ of non-SPM particles for spin-up electrons is much less than the conductivity σ_H^- for spin-down electrons, the ratio value σ_H^-/σ_L^+ is a large value. In order to fit the experimental *MR(B)* curve [3], for $p = 0.122$, we chose σ_H^-/σ_L^+ to be 18.46. The maximum magnetic moment, $\mu_{max}(T)$, of SPM particles was determined from the experimental magnetization curve *M(B)*, and we selected the $\mu_{max}(T)$ from table 1 in [3]. The values of the two parameters σ_s^+ and σ_s^- can be determined by the use of equation (10) as given by the magnitudes σ_{sH} and σ_{sL}. On the other hand, we should note that σ_s^+ will be decreased but σ_s^- will be increased when the applied field $\boldsymbol{B}$ is strengthened. We have obtained the information needed for the theoretical calculation from experimental data, from which we can calculate the dependence of MR on the temperature and the applied magnetic field by use of our model and method.

Table 1. Values of the parameters at 50 K.

$\boldsymbol{B}$ (T)	σ_{sH} (arb. units)	σ_{sL} (arb. units)
0.5	1.100	0.95
1.5	2.812	0.90
2.5	3.220	0.85
3.5	3.780	0.81
4.5	4.250	0.77
5.5	4.822	0.74
6.5	5.335	0.71

Figure 1 shows the calculated curves about the dependence of MR on $\boldsymbol{B}$ at three different temperatures. We can see clearly that theory and experiment agree excellently. Also we find that MR has almost a linear dependence on the applied magnetic field at high temperature. Figure 2 shows the applied magnetic field dependence of MR, where MR is expressed by $\frac{\sigma^{-1}(B)-\sigma^{-1}(B_m)}{\sigma^{-1}(B_m)}$, and $B_m = 3.0$ T. Here B_m is taken to be 3.0 T in order to compare with the experimental data [6]. When the temperature is taken as 10 K for the sample Co_{15} Cu_{85}, because the samples Co_{13} Cu_{87} and Co_{15} Cu_{85} were annealed by different annealing temperatures and the size distributions of these two samples were also different, we select the value of p_s/p_n to be 4.64 and the ratio σ_H^-/σ_L^+ to be 38.46. The theoretical result is in agreement with the experimental report in [6].

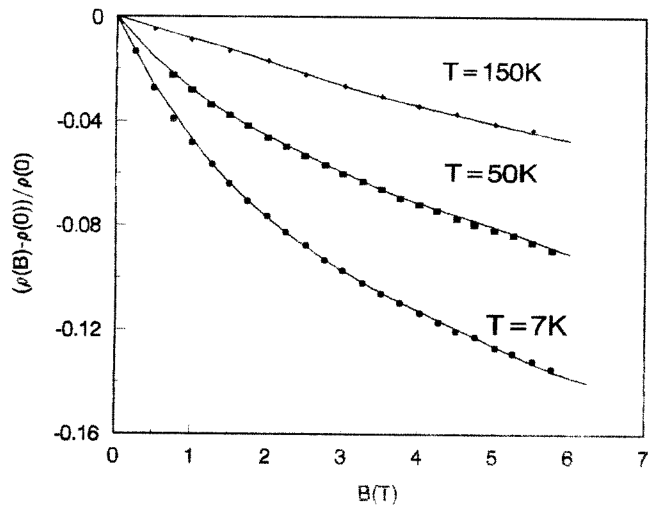

Figure 1. Applied magnetic field $\boldsymbol{B}$ dependence of GMR of Co_{13} Cu_{87} sample at three different temperatures. The packing density of Co particles is 0.122 and the ratios of p_s/p_n are 2.05, 3.07 and 3.69 when temperatures are 7, 50 and 150 K, respectively. The symbols represent the experimental data and the curves give the calculated values.

The dependence of MR (expressed by $\frac{\sigma^{-1}(0)-\sigma^{-1}(B)}{\sigma^{-1}(B)}$) on temperature T is shown in figure 3. In this figure, for the sample Co_{15} Cu_{85}, the packing density p is given by 0.141, and the density p_s of SPM, p_n of non-SPM and the conductivity without applied field $\sigma(0)$ are dependent on temperature T and determined by a non-linear least-squares fit to the experimental data in [6]. We can see that the MR decreased monotonously when the temperature increased, but the curve is not linear. This is in agreement with the experimental data reported in [6].

4. Conclusion and discussions

So far, we have used a new method, which combines the effective medium theory and two-channel conducting model to study the GMR effect of a granular composite (such as Co_x Cu_{1-x}) which contains very small superparamagnetic particles. In fact, due to the presence of a range of sizes for magnetic particles, at a given temperature only a portion (packing density p_s) of magnetic particles lies in the superparamagnetic state, the remainder (packing density p_n) are still ferromagnetic. The ratio p_s/p_n is dependent on temperature. The temperature dependence of GMR is related to the ratio p_s/p_n. A different ratio of packing density p_s/p_n will lead GMR to different dependence on applied field $\boldsymbol{B}$. By means of the new method

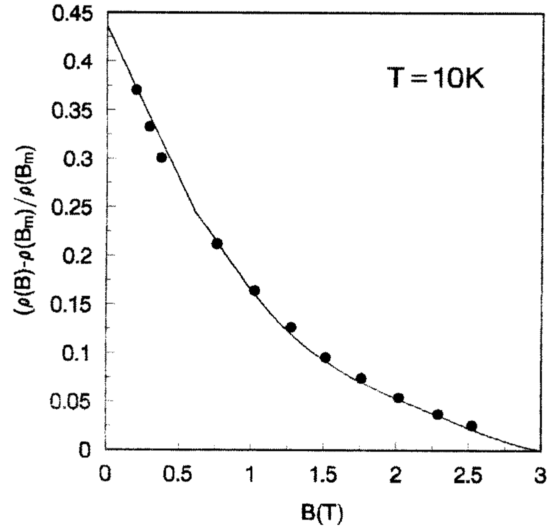

Figure 2. Applied magnetic field ***B*** dependence of the GMR at 10 K in the Co_{15} Cu_{85} sample. The packing density of Co particles is 0.141 and the density of non-SPM particles is 0.025. The symbol represents the experimental data and the curve gives the calculated values.

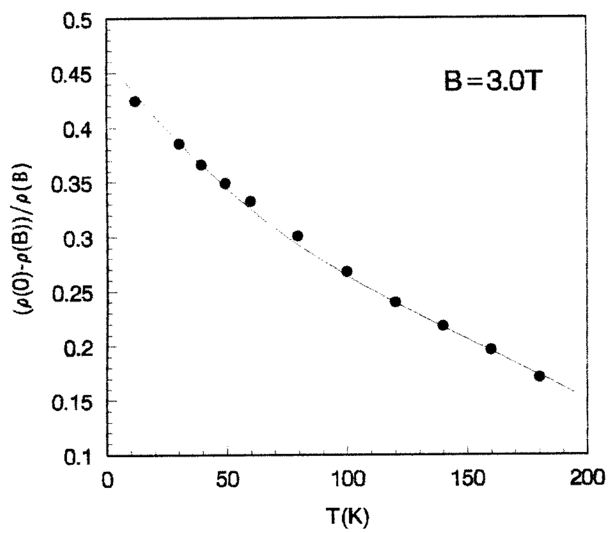

Figure 3. Temperature dependence of the GMR in Co_{15} Cu_{85} sample when given the applied field $B = 3.0$ T. The symbol represents the experimental data and the curve gives the calculated values.

and model, we have explained that GMR changes with the temperature and applied field for meltspun granular composite Co_x Cu_{1-x}. It has been found that the calculated results are in excellent agreement with recent experimental data as shown in figures 1–3. Clearly, in figure 1, at high temperature, the relation between GMR and applied field ***B*** is linear. This phenomenon is due to a progressively larger fraction of non-SPM particles changing to SPM particles when the temperature increases. The change of GMR with temperature T is complex. We have considered non-linear dependence of $\sigma(0)$ on T, and the ratio of p_s to p_n on T by use of experimental data. We know that the conductivity $\sigma(0)$ of many magnetic materials decreases when the temperature increases, and it is reasonable to assume that p_s/p_n increases with the increase in temperature. When temperature is increased, a progressively larger fraction of ferromagnetic Co particles will become 'unblocked' SPM particles, namely, the density of p_s is increased and p_n is decreased simultaneously. Having considered the above reasons, we obtained a curve of GMR on temperature in figure 3 which agrees with experimental data. In summary, the GMR of the magnetic granular composite strongly depends on the range of sizes of the particles and the temperature. The method presented is a potential approach to studying the magnetic and transport properties of magnetic granular composites containing superparamagnetic particles.

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