# Origin of the temperature dependence of the giant magnetoresistance in magnetic granular solids

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The temperature dependence of the giant magnetoresistance (GMR) and magnetic properties have been measured for granular solid $Co_{20}Ag_{80}$. From the measured GMR, we have derived the temperature dependence of the effective spin $S'$ of the magnetic scatterers experienced by the conduction electrons. It is found that $S'$ shares the same temperature dependence as the spontaneous magnetization $M_s(T)$ over the whole temperature range (5–310 K) of our measurement. Our results suggest that the origin of the temperature dependence of GMR is the reduced magnetization as a result of spin-wave excitations in magnetic granules.

The effort in understanding the anomalous magnetotransport properties in heterogeneous magnetic structures has been intensified in recent years, particularly after the discovery of the giant magnetoresistance (GMR) effect in multilayers and granular materials.[1–5] The challenge in understanding this effect in granular solids is underscored by the inherent complexities and uncertainties in geometrical parameters. Recent studies have resulted in some clarifications of the underlying mechanism. In magnetic multilayers, it has been found that spin-dependent scatterings both at the interfaces and in the bulk layers are responsible for the strong field dependence of the magnetoresistance (MR).[4] In systems with a granular structure, experimental evidence has indicated that the interface scattering dominates the bulk one because of the relatively large surface-to-volume ratio of the magnetic component.[6,7] The reason that the granular interfaces are effective regions of magnetic scatterings is the existence of abundant irregularities or disorders at the particle-matrix interfaces.

In a recent work, we have systematically studied the GMR effect in a series of granular systems of different material combinations, volume fractions, and thermal treatment histories.[7] An attempt was made to account for the field dependence of the MR using a modified effective exchange interaction (MEEI) model based on a spin-dependent potential. Very good agreement was achieved for the field dependence. It was shown that MR in granular materials can be described by a simple relation involving two parameters. These parameters are intrinsic in that they reflect the internal states of the system, e.g, the magnetization of the particles and the effective exchange coupling between electrons and magnetic spins. The external influences, the applied magnetic field $H$ and temperature $T$, play their roles indirectly through their effects on these intrinsic parameters.

An important aspect of the GMR effect is its dependence on temperature. A careful study of the $T$ dependence is not only important in gaining a deeper understanding of the GMR mechanism, but also highly beneficial to potential applications where a small temperature coefficient at ambient conditions is crucial. There are several possible mechanisms that cause the $T$ dependence of the GMR. First, thermal excited spin waves will reduce the magnetization, which, in turn, suppresses the GMR effect. Second, the electron-magnon interaction may lead to a decreased MR at higher temperatures, both by the spin-preserving and spin-flipping scattering processes. Studies in multilayers have attributed the $T$ dependence of the MR to this interaction due to the existence of localized magnon excitations at the interfaces.[8] Third, the electron-phonon interaction may also complicate the $T$ dependence. Currently, there is a lack of careful analysis of the temperature effect in granular systems.[9] In our previous work, we measured the GMR effect of a granular system at only three temperatures (4.2, 77, and 300 K). The MEEI model provided a reasonable explanation to our data. We found that the $T$ dependence is primarily caused by the change in the magnetization of the magnetic particles at various temperatures. Other mechanisms are less important. However, our conclusion is severely limited by a lack of sufficient data at various temperatures. Because of the fundamental significance of the problem, we have since studied in detail the evolution of the GMR and magnetization as temperature is smoothly varied from 4.2 to 300 K. In this paper we will show that spin-wave excitations in the magnetic granules are the unambiguous cause of the $T$ dependence of the GMR effect. The MEEI model gives an excellent description of the $T$ dependence. But, contrary to the conventional belief, spin-wave excitations do not affect the GMR through the electron-magnon interaction, but rather they reduce the effective exchange interaction strength by decreasing the effective spin experienced by the carriers. To our knowledge, the origin of the $T$ dependence of the GMR effect has not been identified in granular systems before this study.

The sample chosen for this study is an as-sputtered $Co_{20}Ag_{80}$ thin film of about 3100 Å thick. The percentages represent the volume fractions. The reason for using this sample is that the Co-Ag granular system has a large GMR value and its microstructure has been carefully characterized.[10] The sample fabrication process was described in Ref. 7. Specimens for transport measurements were patterned to a Hall bar configuration by the standard photolithography and lift-off technique. Magneto-

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transport measurements were performed using a variable temperature insert in a cryostat equipped with a superconducting magnet. The field dependence of the MR was measured in a sweeping field of $-8$ T $\leftrightarrow$ $+8$ T, to account for the magnetic hysteresis. The magnetic measurements were carried out in a superconducting quantum interference device (SQUID) magnetometer with a maximum field of 5.5 T.

In this work, GMR is defined as

$$\Delta\rho_{xx}(H)/\rho_{xx} = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0). \quad (1)$$

Figure 1 shows the magnetic field dependence of the MR at temperatures $T$ = 4.2 K and 300 K, and the normalized magnetization, $M(H)/M_s$, at $T$ = 5 K and 300 K. $M_s$ is the spontaneous magnetization at a particular temperature. It is observed that the MR follows closely the variation of the magnetization, $M/M_s$. At the low temperature ($T$ = 4.2 K), a large magnetic hysteresis in $M(H)$ induces a similar hysteresis in the measured MR. At $T$ = 300 K, however, the absence of hysteresis in $M(H)$ due to superparamagnetism leads to a similar lack of hysteresis in the MR. At $T$ = 4.2 K, both the MR and $M(H)$ saturate at a field of roughly 2 T. At $T$ = 300 K, the $M(H)$ saturates much more slowly due to thermal agitations. Correspondingly, the MR exhibits a slow saturation process as well.

Because of the correlation between the MR and magnetization it is helpful to probe the magnetic state of the sample at various temperatures. The magnetic susceptibility measurements provide information on the magnetic states. Figure 2 shows the data of the magnetic susceptibility, $\chi$, as a function of $T$ for the $Co_{20}Ag_{80}$ sample. Clearly, there exist two different states in the high and

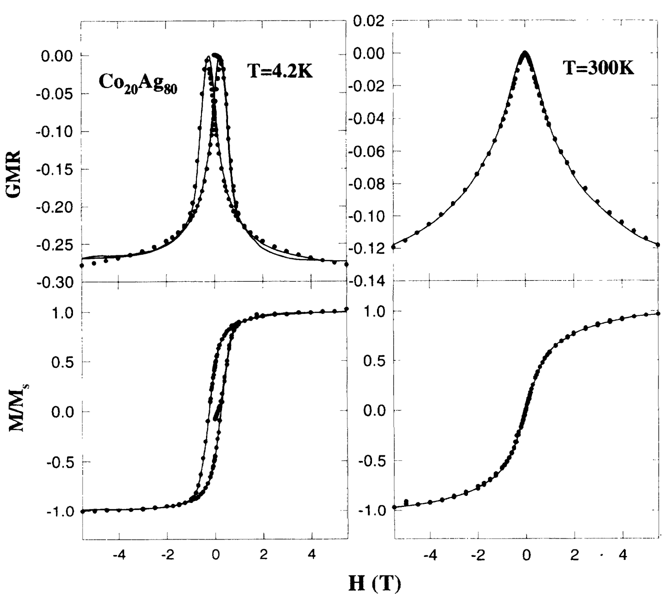


FIG. 1. Upper panels: The results of the measured GMR (solid circles) at temperatures $T$ = 4.2 K (left) and 300 K (right). The solid lines represent the fitted GMR using the MEEI model and the measured $M/M_s$. The fitted parameters are $\gamma$ = 0.330 at $T$ = 4.2 K and $\gamma$ = 0.251 at $T$ = 300 K. Lower panels: The measured normalized magnetizations $M(H)/M_s$ (solid circles) at $T$ =5 K and 300 K. The solid lines are the smooth fitting of the data points.

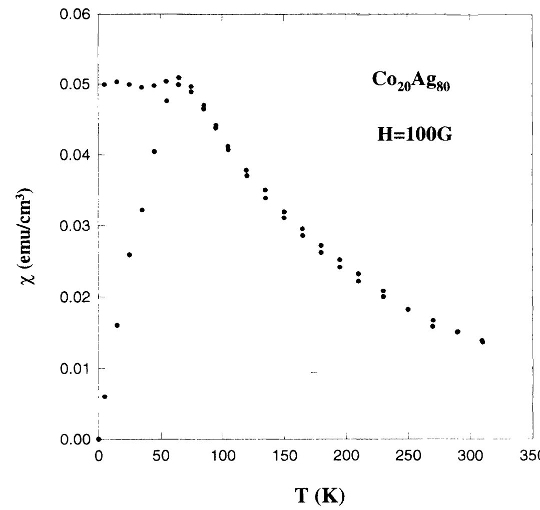


FIG. 2. The measured susceptibility $\chi$ as a function of temperature $T$ for $Co_{20}Ag_{80}$.

low $T$ regions. Below the blocking temperature, $T_B \approx 70$ K, the magnetic moments of granules are frozen along the magnetic easy axes of each individual particle. Above $T_B$, the magnetic granules are in a superparamagnetic state due to thermal agitations. In this state the susceptibility satisfies the Curie-Weiss law,

$$\chi = \frac{M_s^2(T)V}{3k_B(T - T_0)} \qquad \text{for} \quad k_BT > \mu H, \quad (2)$$

where $V$ is the volume of the magnetic particles, and $T_0$ is the effective Curie temperature reflecting the strength of the interparticle interaction.

The magnetic measurements are also among the most reliable and accurate methods for obtaining the magnetic particle size in granular solids. By fitting the $T$ dependence of $\chi$ with Eq. (2) and measuring $M_s$ at low $T$, we can obtain the volume of the particles. The results derived from the susceptibility measurement can be checked independently by measuring the magnetization curve, $M(H)$, at high temperatures and fitting it to a Langevin equation. The two methods generally yield particle sizes to within 10% of each other. For our as-sputtered $Co_{20}Ag_{80}$ samples, the average magnetic particle size is about 14 Å and the spontaneous magnetization at $T$ = 5 K is $M_s$ = 118 emu/$g_{Co}$.

The magnetic state of the small Co particles is highly susceptible to thermal agitations. Except at very low temperatures, the magnetic moment vectors fluctuate rapidly in space and change their magnitudes with temperature. This, in turn, leads to both a slow saturation in the MR in a magnetic field, as evidenced in Fig. 1, and a temperature dependence of the GMR effect. To analyze the MR data as a function of $H$ and $T$, we resort to the modified effective exchange interaction model, which was originally used to describe the magnetotransport properties of dilute magnetic alloys. The detailed description of the MEEI model can be found in Ref. 7. According to this model the spin-dependent scattering arises from an exchange interaction potential between the conduction electrons and the magnetic scatterers.

In our extension of this model to the granular systems, it is assumed that conduction electrons are scattered by some localized spin, $\mathbf{S}'$. This scattering occurs predominantly on the surface of a magnetic granule which possesses a giant spin, $\mathbf{S}$. For our sample, the average $|\mathbf{S}|$ is about 710. Because of the single-domain nature, $\mathbf{S}'$ and $\mathbf{S}$ are ferromagnetically coupled and share the same relaxation process, i.e., $\mathbf{S}' = |\mathbf{S}'|\boldsymbol{\sigma}$, where $\boldsymbol{\sigma}(\mathbf{S}) = \mathbf{S}/|\mathbf{S}|$ is the unit vector of $\mathbf{S}$. We introduce a phenomenological scattering Hamiltonian

$$H = V - 2JS'\mathbf{s}\cdot\boldsymbol{\sigma}(\mathbf{S}) = V[1 - 2\gamma\mathbf{s}\cdot\boldsymbol{\sigma}(\mathbf{S})], \tag{3}$$

where $\gamma = JS'/V$ is a dimensionless parameter. In this expression, $V$ is the spin-independent scattering potential and $J$ the exchange interaction between a conduction electron with spin $\mathbf{s}$ and a localized spin $\mathbf{S}'$ on the magnetic particle.

The strength of the spin-dependent scattering potential is parametrized by the composite parameter $\gamma$. Under a large $H$, $\boldsymbol{\sigma}$ will be aligned along $\mathbf{H}$. The maximum MR achievable depends on $\gamma$. At moderate $H$ and $T$, the average $\langle\boldsymbol{\sigma}\rangle$ is a function of both $H$ and $T$, leading to a dependence of MR on these parameters. Using Born approximation and neglecting terms of the order of $O(1/S)$ we have obtained an expression for the MR in granular solids,

$$\Delta\rho_{xx} = \rho_0\frac{4\gamma^2}{(1+\gamma^2)^2}\left(\frac{M}{M_s}\right)^2, \tag{4}$$

where $\rho_0$ is the zero field resistivity due to magnetic scattering. Thus, the MR in granular materials can be characterized by only two parameters which depend on the intrinsic properties ($\gamma$ and $M$) of the system. Since $\gamma$ is field independent, the $H$ dependence of the MR is reflected through its dependence on $M$.

In our experiment, the dependence of both the MR and the $M$ on $H$ or $T$ can be independently measured and determined, at various fixed temperatures or fields. Relation (4) asserts that $\Delta\rho_{xx}/\rho_{xx}$ is a quadratic function of $M/M_s$. This is indeed the case for $Co_{20}Ag_{80}$ as shown in Fig. 1, where the solid lines in the GMR data are the theoretical curves resulting from a fit using relation (4). It is observed that both the field dependence of the GMR and the hysteresis effects are very well described by the MEEI model.

Next, we will concentrate on the $T$ dependence of the GMR effect, which provides another independent check of the validity of the MEEI model. According to the model, the saturated $\Delta\rho_{xx}/\rho_{xx}$ value obtained when $M/M_s \to 1$ is solely determined by $\gamma$. By measuring the $T$ dependence of the GMR, one can obtain $\gamma(T)$. For our $Co_{20}Ag_{80}$ sample, the measured saturated $\Delta\rho_{xx}$ is presented in Fig. 3(a). $\gamma(T)$ is a composite parameter involving $J$, $V$, and $S'$. Nevertheless, the $T$ dependence of $\gamma(T)$ is directly resulted from that of $S'(T)$, because both $J$ and $V$ are temperature independent. Because of the single-domain nature of the magnetic particle, $S'(T)$ and the spontaneous magnetization, $M_s(T)$, share the same temperature dependence. $M_s$ can be measured using the SQUID magnetometer. The measured $M_s(T)$ is shown in Fig. 3(b). If we can demonstrate that $S'(T)$ and $M_s(T)$ follow the same temperature dependence, the validity of MEEI model can be strongly supported. It is noted that $S'(T)$ and $M_s(T)$ are derived from two independent measurements, one from magnetotransport and the other from magnetic measurements.

One may question how $S'(T)$, which resides mainly on the surface region of a particle, can follow the $T$ dependence of the magnetic moment, $M_s(T)$, of the entire particle. It is true that the surface spins are floppier than the bulk spins, because of the weakened ferromagnetic coupling near the surface. Experiments and theories have shown that they still have the same $T$ dependence in multilayers.[11–13] The only difference is that the spin-wave constant of the surface spins is about 2–3 times larger than that of the bulk spins. If we assume that this is the case in our granular system, we can write

$$S'(T) = S'(0)[1 - B_s f(T)] \tag{5}$$

and

$$M_s(T) = M_s(0)[1 - B_b f(T)]. \tag{6}$$

Here, $B_s$ and $B_b$ are the spin-wave constants of the surface spins and the whole particle, respectively; $S'(0)$ and $M_s(0)$ are the zero temperature values; and $f(T)$ is the temperature dependence. For bulk systems, $f(T) = T^{3/2}$ from the Bloch law of the spin-wave excitations. For low dimensional systems, $f(T)$ depends on a particular system because of the physical confinement of the spin-wave excitations. We may rearrange relation (5) and (6) as follows:

$$1 - \frac{S'(T)}{S'(0)} = B_s f(T), \tag{7}$$

$$1 - \frac{M_s(T)}{M_s(0)} = B_b f(T). \tag{8}$$

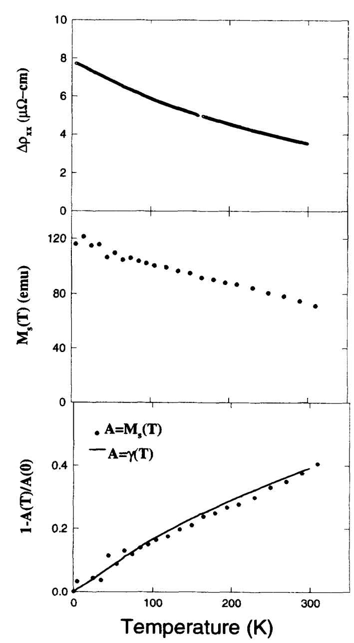


FIG. 3. (a) The net change in resistivity $\Delta\rho_{xx} = \rho_{xx}(0) - \rho_{xx}(8\text{T})$ as a function of temperature. (b) The measured spontaneous magnetization, $M_s(T)$, versus temperature. (c) The scaled $1 - M_s(T)/M_s(0)$ (solid circles) and $1-\gamma(T)/\gamma(0)$ (solid line) versus temperature. The systematic errors in $M_s$ and $\gamma$ at $T = 300$ K are both about $-15\%$.

In other words, $1 - S'(T)/S'(0)$ and $1 - M_s(T)/M_s(0)$ should scale with the same function of temperature, $f(T)$. Because of the linear relationship between $S'$ and $\gamma$, we have $1 - S'(T)/S'(0) = 1 - \gamma(T)/\gamma(0)$.

In Fig. 3(c), we plot the scaled $1 - A(T)/A(0)$, where $A = M_s$ and $\gamma$, as a function of temperature. Indeed, $M_s$ and $\gamma$ share the same $T$ dependence as expected from relations (7) and (8). This is a convincing evidence that the MEEI model is the correct one in describing the magnetotransport characteristics of granular systems. It also demonstrates, for the first time, that the $T$ dependence of the GMR effect can be explained within the framework of the modified spin-dependent scattering potential for granular systems.

Our results suggest that the origin of the $T$ dependence of the GMR is due to a reduced magnetization at finite temperature as a result of spin-wave excitations. Our study suggests a mechanism different from the electron-magnon interaction which was used to explain the $T$ dependence of the GMR in magnetic multilayers.[8] In multilayers, a finite temperature causes a variation in the population of magnons which affects the electron-magnon scattering rate. At the same time, such an interaction may also cause spin mixing which is detrimental to the GMR effect. In granular systems, the main role of magnons is to reduce the magnetization, which in turn reduces the strength of the spin-dependence interaction. The direct electron-magnon interaction is not considered crucial. It plays a less important role in determining the magnetotransport properties of the granular systems. Other evidence is the observed $T$ dependence of the MR itself. Earlier studies on dilute magnetic alloys[14] and magnetic multilayers[15,16] have shown that electron-magnon scattering leads to a power law, $T^{\alpha}$, where $\alpha$ =1.5–2, in the $T$ dependence of the MR. The MR in our granular Co-Ag sample has a $T$ dependence of $T^{0.8}$. Such a low exponent seems to rule out electron-magnon interaction as the responsible mechanism.

Our conclusion is important in the effort to search for the best materials for magnetic sensor applications. To achieve good thermal stability it is highly desirable to use ferromagnetic component with stiff spin-wave constant and high magnetic ordering temperature. The other factors of consideration are a large exchange constant $J$ in relation (3), reduced disorder and phonon contributions, and reduced anisotropies for the minimizing hysteresis effect.

In summary, we have studied the temperature dependence of the GMR and magnetization of a granular $Co_{20}Ag_{80}$ thin film. A correlation between the transport and magnetic properties has been established. It has been shown that the universal parameter $\gamma = JS'/V$ used to characterize the GMR in the MEEI model shares the same temperature dependence as that of the spontaneous magnetization $M_s$. Our results suggest that the origin of the $T$ dependence of the GMR effect is the reduced magnetization as a result of spin-wave excitations at finite temperatures.

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