Theory of tunneling magnetoresistance in granular magnetic films

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A mechanism for the large magnetoresistance observed recently in Co-Al-O granular magnetic films is presented. It is shown that the resistivity decreases with increasing applied magnetic field because the spin-dependent tunneling increases as the relative orientation of the magnetization between grains becomes parallel. With this mechanism we are able to account for the dependence of the magnetoresistance on the magnetization and temperature. [S0163-1829(96)50318-8]

Magnetoresistance has attracted much attention after the discovery of the giant magnetoresistance (GMR) in Fe/Cr multilayers;[1] this has provided us with a magnetotransport property in magnetic materials which has the potential for technical applications. Soon after the discovery of GMR in magnetic multilayers, granular magnetic films, such as Co/Cu and Fe/Ag, were also reported to show GMR.[2,3] The present authors have pointed out that the GMR's in the magnetic multilayers and granular magnetic films can be explained by basically the same mechanism, i.e., the spin-dependent scattering at interfaces.[4,5] However, magnetoresistance (MR) in the artificial layered structures was first observed in ferromagnetic metal–insulator–metal (FM/I/FM) junctions,[6,7] where electrons tunnel through an insulating barrier. The magnetoresistance in this case can be called tunneling magnetoresistance (TMR). Although the TMR was not large in early stages of experiments on FM/I/FM junctions, a quite large TMR, 30% MR ratio at helium temperature, has been recently reported by Miyazaki and Tezuka,[8] and Moodera *et al.*[9]

In both GMR and TMR, the resistivity decreases as the relative orientation of the magnetization between grains changes with increasing external magnetic field. The common feature of GMR and TMR in spite of the structural contrast between these materials, i.e., layered vs granular structures, allows us to anticipate that TMR appears in some granular materials. In fact, MR of the tunneling type has been recently observed in highly resistive Co-Al-O granular magnetic films.[10] The fact that the conductivity in these materials is produced by tunneling through insulating barrier between Co grains has been confirmed by the non-Ohmic character of the resistivity and its temperature dependence, i.e., $\ln\rho \propto T^{-1/2}$. Sheng *et al.*[11] have explained the characteristic dependence of the resistivity on temperature in terms of the tunneling of electrons between grains. Based on the same picture, Helman and Abeles[12] presented a theory of MR in granular magnetic films, and showed that the MR ratio depends strongly on temperature as $1/T$. However, the MR ratio in Co-Al-O films is only weakly temperature dependent; therefore the theory of Helman and Abeles is not able to account for the TMR in Co-Al-O films. The purpose of this paper is to present an alternative explanation of the TMR in the granular magnetic films that is based on the mechanism of the TMR in FM/I/FM junctions.

In the FM/I/FM junctions, the tunneling conductance G is given by[13]

$$G=\frac{e^2}{h}|\mathsf{T}|^2, \tag{1}$$

where e is the electric charge and $|\mathsf{T}|^2$ is the transmission coefficient. Let θ be the angle between the magnetizations, $\mathbf{M}_1$ and $\mathbf{M}_2$, of two FM's. Then,[7]

$$|\mathsf{T}|^2 \propto (1+P^2\cos\theta)e^{-2\kappa s}, \tag{2}$$

where

$$P=\frac{D_\uparrow - D_\downarrow}{D_\uparrow + D_\downarrow} \tag{3}$$

and

$$\kappa=\sqrt{2m^*(V-E_{\mathrm{F}})/\hbar^2}. \tag{4}$$

Here, $D_\sigma(\sigma=\uparrow,\downarrow)$ is the density of states at the Fermi energy E_{F} for electrons with spin σ, and s, m^*, and V are the thickness of the barrier, the effective mass of electrons, and barrier height, respectively. We have used the joint density of states, P^2, in Eq. (2) for the tunneling matrix element according to several theoretical works on phenomena related to spin polarization in ferromagnetic metals,[7,14,15] although the result (2) may be modified for FM/I/FM junctions if κ is not larger than the Fermi wave vectors, $k_\uparrow$ and $k_\downarrow$.[16] Another reason for adopting the expression (2) for the TMR in granular films is that the large MR ratios observed recently for FM/I/FM junctions can be semiquantitatively explained by the joint density of states.[8,9] Further discussions on this point will be given at the end.

The MR ratio is defined as

$$R_{\mathrm{MR}}=\frac{G(0)^{-1}-G(H)^{-1}}{G(0)^{-1}}=1-\frac{G(0)}{G(H)}, \tag{5}$$

where $G(H)$ is the conductance in an external magnetic field H. When the angle θ is varied from π to 0 by an external field, the MR ratio for FM/I/FM junctions is $2P^2/(1+P^2)$.

The tunneling mechanism can hold true even for the barriers between grains in the granular films. Because θ and s are randomly distributed in granular films, we must take an average over these quantities. Furthermore, as pointed out by Sheng *et al.*,[11] the charging energy, E_c, becomes important for tunneling in granular films; therefore another factor

$e^{-E_c/kT}$ is added to the expression of the conductance. Taking into account the factors mentioned above, the conductance may be written as

$$G = G_0 \int \int ds\, d\theta f(s) g(\theta)(1+P^2\cos\theta) e^{-2\kappa s - E_c/kT}, \tag{6}$$

where G_0 is the conductance when $P=0$, and $f(s)$ and $g(\theta)$ are distribution functions. One should note the difference between Eq. (6) and the expression used for the conductance by Helman and Abeles.[12] They included a factor of $e^{\pm E_M/kT}$, which is due to a contribution to the hopping energy from the magnetic energy E_M. This factor is neglected because E_M/kT is quite small as compared to E_c/kT. Instead, we have taken into account the angular dependence of the tunneling matrix element which plays a crucial role in the tunneling conductance. At the end, we will also give a detailed discussion on the neglect of the contribution from E_M.

As the exponential factor in Eq. (6) is independent of θ, the average over θ leads to $\langle\cos\theta\rangle$; however, we should be careful about the definition of θ and its average. When the magnetic grains behave as superparamagnets, the relative angle θ between grains can only be defined for tunneling events that occur in a period of time which is much shorter than the characteristic period of the thermal fluctuation of the magnetic moments of the grains. In this case, the average over θ means either an average of θ between one pair of grains over a period of the thermal fluctuation, or an average over θ of all pairs of grains. When the magnetic grains have stable magnetic moments, e.g., like spin glasses (or cluster glasses), θ can be defined as usual and the average is taken over θ of all pairs of grains. For both cases the average $\langle\cos\theta\rangle$ is

$$\langle\cos\theta\rangle = m^2, \tag{7}$$

where m is the relative magnetization of the system, and we have neglected the correlation between magnetic moments of neighboring grains.

The average over s is taken according to the approximation used by Sheng *et al.*[11] They assumed that the product sE_c is a constant C, and furthermore they used the steepest descent approximation, i.e., that tunneling occurs via paths which makes the exponential factor largest. The final result of the conductance is given as

$$G = G_0(1+P^2m^2)e^{-2\sqrt{2\kappa C/kT}}. \tag{8}$$

The exponential factor dominates the temperature dependence of the resistivity; this explains why the observed data goes as $\ln\rho \propto T^{-1/2}$. The MR ratio (5) is given as $P^2m^2/(1+P^2m^2) \sim P^2m^2$ for small values of P, which is proportional to m^2 and becomes P^2 for sufficiently high magnetic fields. These results explain well the observed data on magnetic granular films,[10] i.e., the m^2 dependence of the MR ratio and the weak temperature dependence. The maximum MR ratio 7.8–8.8 % observed for Co-Al-O films is smaller than the value for FM/I/FM junctions by a factor of 2.[8,9]

Because the MR ratio depends on m^2, the temperature and magnetic field dependence of MR ratio varies as the magnetic state changes. Fujimori *et al.*[10] have reported that the Co grains behave as superparamagnets above ~100 K; therefore, the MR ratio depends only weakly on temperature through the Langevin function. If a phase transition to a ferromagnetic state occurs at low temperature, the MR ratio would decrease as the magnetic field is unable to reorientate the magnetic moments of the grains.

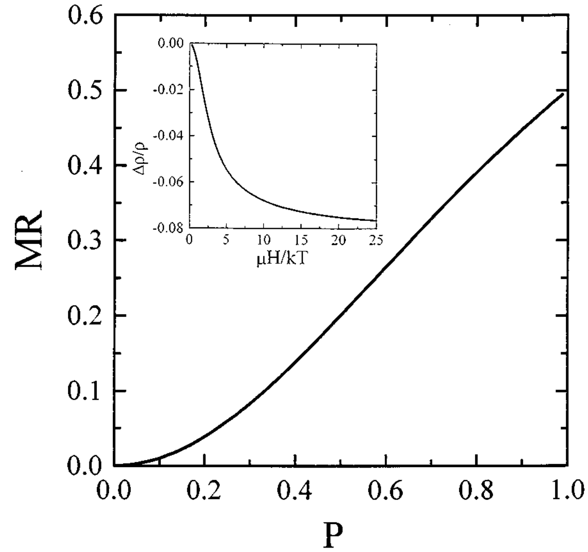

FIG. 1. Dependence of the saturated MR ratio as a function of $P=(D_\uparrow - D_\downarrow)/(D_\uparrow + D_\downarrow)$. The inset shows the H dependence of the resistivity change $\Delta\rho/\rho$ for $P=0.3$ when the superparamagnetic state is assumed.

In Fig. 1, the dependence on P of the saturated ($m=1$) MR ratio $P^2/(1+P^2)$ is shown. The observed MR ratio corresponds to a value of $P\sim 0.3$, which may not be unreasonable for ferromagnetic Co when compared with the experimental results of the polarization of conduction electrons.[15] The inset shows a calculated result of the resistivity change $\Delta\rho/\rho$ as a function of $\mu H/kT \equiv \xi$ by assuming that m obeys the Langevin function $L(\xi)$, which is also in agreement with the experimental results at high temperatures where superparamagnetism is observed.

Now we discuss the assumptions used in the present formalism; one is the assumption that κ is larger than k_σ and the other is our neglect of the magnetic energy E_M. For the first assumption, we note that tunneling occurs between grains and probably is local in character; therefore all of the Fermi momentum vectors may contribute to the tunneling of electrons and k_σ should be taken as an average of these Fermi momenta. Therefore, D_σ may be more appropriate than k_σ to represent the tunneling phenomena in granular films. It is worthwhile to note that the high MR ratios observed recently for FM/I/FM junctions[8, 9] have been explained reasonably well by the values of P of Fe and Co metals, although the barrier height for Al_2O_3, 1–3 eV,[17] may not be sufficiently high as compared with the Fermi energy ~1 eV for conduction electrons. Furthermore, it is not evident that the relation between the MR and the ratio of κ/k_σ obtained for FM/I/FM junctions[16] can be applied to the TMR in granular films, because the translational symmetry along interfaces is lost in the granular films.

In order to discuss the second assumption, we first incorporate the magnetic energy E_M into the present formalism following the argument of Helman and Abeles.[12] In their formalism, E_M is defined as the difference between the magnetic energies of an electron as it tunnels from grain 1 to grain 2. When the spin of the tunneling electron is parallel (antiparallel) to the magnetization of the grain 1, but antiparallel (parallel) to that of grain 2, a factor $e^{-E_M/kT}(e^{+E_M/kT})$ is added to the expression of the conductance. Because we use a local axis of spin quantization for each grain, tunneling of an electron can occur between ↑ and ↓ spin states of grains 1 and 2. By taking into account the above mentioned factors $e^{\pm E_M/kT}$ into our formalism, the term $(1+P^2\cos\theta)$ in Eq. (6) is replaced by

$$a^2\cos^2(\theta/2)+a(1-a)\sin^2(\theta/2)e^{-E_M/kT}+(1-a)a\sin^2(\theta/2)e^{E_M/kT}+(1-a)^2\cos^2(\theta/2),$$

with $a\equiv D_\uparrow/(D_\uparrow+D_\downarrow)$. Then, the conductance is given as

$$G\propto 1+2a(1-a)\left(\cosh\frac{E_M}{kT}-1\right)+\left\{P^2-2a(1-a)\left(\cosh\frac{E_M}{kT}-1\right)\right\}m^2, \qquad (9)$$

from which we find that the E_M term gives always a *positive* MR. This positive MR is dominant at low temperatures because of the factor of $e^{E_M/kT}$ in Eq. (9). The term $\cosh(E_M/kT)$ also exists in the expression obtained by Helman and Abeles. In their expression, there is a term which is a linear function of the polarization of the tunneling electrons, from which the negative MR appears. On the other hand, the conductance G given by Eq. (9) is an even function of the polarization of the tunneling electrons as it should be. Because no positive MR has been observed so far above 4.2 K at least,[18] E_M must be very small. Therefore, E_M cannot be the exchange energy, which is large as compared with the helium temperature. When an electron is added to a magnetic grain, the Coulomb interaction between electrons increases both charging and magnetic energies. In order to estimate the value of E_M, we consider E_M as the energy to reverse the direction of one spin without changing the number of electrons in the grain. The energy may be given as the Zeeman energy of one spin in a magnetic field H_C by which the magnetization of the grain is increased by $1\mu_B$. Assuming that the high field susceptibility of Co grains is similar to that of Fe, Co, or Ni metal which is $(1–3)\times 10^{-4}$ emu/mol,[19] we obtain H_C is at most 10 T when the number of atoms in Co grains is 10^3–10^4. The Zeeman energy is then $\lesssim 0.3$ meV which is irrelevant for temperatures higher than 4.2 K.

Finally, in the present formalism, wide distributions of the size of grains and intergrain distance have been implicitly assumed to validate the assumption that sE_c is constant. Although no clear evidence has been given so far to justify this, with this assumption Sheng *et al.*[11] have explained the observed temperature dependence in highly resistive granular films quite well. It may be possible to interpret E_c as the charging energy of a condenser made by two grains in the tunneling event. In this case the assumption that sE_c is constant can be reasonably understood. However, further experimental studies using well-controlled samples as well as theoretical study of tunneling phenomena in heterogeneous systems are necessary.

In conclusion, the origin of the magnetoresistance in granular magnetic films of Co-Al-O can be explained in terms of a dependence of the tunneling matrix element on the relative orientation of magnetic moments between grains. The mechanism is able to account for the observed m^2 dependence of the MR ratio and its weak temperature dependence.

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