Magnetothermopower of a $Ag_{80}Co_{20}$ granular system

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(Received 16 August 1993)

A large magnetothermopower (MTP) has been found to accompany the magnetoresistance (MR) in a $Ag_{80}Co_{20}$ granular system. The zero-field thermopower is negative and its magnitude increases as the magnetic field increases. The MTP is inversely proportional to the resistance in the temperature range where the diffusion thermopower dominates. These observations support the spin-split density-of-states (DOS) model which yields both the MTP and the MR for multilayers and granular systems, even with a spin-independent potential at the interfaces.

Giant magnetoresistance (GMR) effects, first discovered in magnetic multilayers,[1,2] have recently been reported[3,4] in granular systems such as AgCo and CuCo. The latter materials are composed of single-domain magnetic particles that have precipitated within the nonmagnetic matrix. Considerable theoretical and experimental effort[5–8] has been directed toward understanding the origin of the GMR effect, with the focus on the role of spin-dependent scattering at the interfaces between magnetic and nonmagnetic constituents. The thermoelectric power of multilayer systems was also found[9,10] to have a strong magnetic-field dependence. As we have noted[11,12] in the context of multilayers, that fact argues strongly *against* theories that attribute the GMR effect solely to spin-dependent interfacial potentials.

In this paper, we report the existence of a significant magnetic-field dependence of the thermoelectric power $S(H)$ of AgCo granular systems and show, as was the case in multilayers, that the thermopower is accurately proportional to the inverse of the resistance. Further, at higher temperatures $S(H)$ takes the sign of the magnetic constituent rather than of the matrix. We outline a model in which the matrix resistivity and that due to scattering from magnetic particles are in series; a more complete theory will be published separately.[13] The dominant relaxation mechanism is taken to be that in which s electrons scatter into the d bands of the magnetic particles. The large difference in density of states (DOS) between minority- and majority-spin bands provides a single mechanism capable of explaining the close relationship between magnetoresistance and magnetothermopower.

Granular AgCo thin films were prepared with a conventional sputtering apparatus. Co and Ag were sputtered simultaneously using two independent sputtering guns under an Ar pressure of 5×10^{-3} Torr, while the substrates were rotated at a speed of about 0.6 rps. The sputtering rates detected with quartz thickness monitors were 3.3 Å/s for Co and 30 Å/s for Ag. Thin films with a nominal concentration of 20 at. % Co (14 vol. %) were deposited up to 4300-Å thickness on thin cover-glass substrates. Samples were annealed according to published procedures[2] to obtain a large magnetoresistance ratio. The magnetization was measured with a superconducting quantum interference device magnetometer between 300 and 5 K under a magnetic field up to 50 kOe. The magnetoresistance was checked at various temperatures and magnetic fields. Thermopower and resistance were measured over a temperature range between 300 and 15 K in a magnetic field from 0 to 10 kOe on the same sample. Fine gold wires were used as electrodes and fine constantan-chromel wires as thermocouples. The apparatus and experimental details were described in a previous paper.[11]

Magnetoresistances for different annealing conditions at different temperatures were compared with each other. The behavior is qualitatively the same for all samples, but the field required for saturation and the magnetoresistance ratio depend on annealing time. The sample annealed at 300 °C for 10 min was used for the detailed magnetothermopower (MTP) study. The sample has a magnetoresistance ratio, $[R(0)-R(50\text{ kOe})]/R(0)$, about 14% at room temperature and 33% at 5 K. At 300 K, the magnetization vs magnetic-field curve does not saturate at low fields and the data can be fit reasonably well to a single Langevin function. This indicates that the Co grains in the sample show a superparamagnetic behavior; the grains are found to have an effective moment of $1450\mu_B$, which corresponds to a mean grain size of about 26 Å diam, assuming fcc Co.

Figure 1 shows the magnetic-field dependence of the

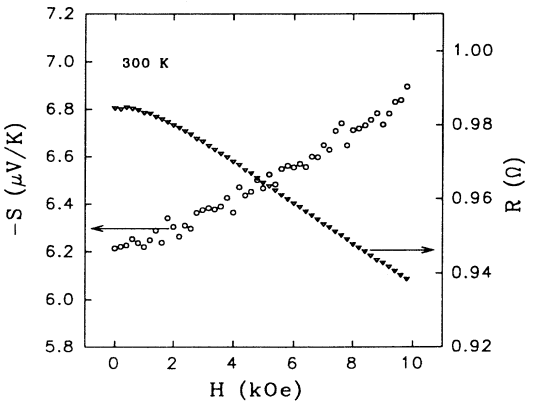

FIG. 1. Room temperature MTP (circles) and the magnetoresistance (triangles) of the granular sample $Ag_{80}Co_{20}$ annealed at 300 °C for 10 min.

magnetoresistance and MTP at room temperature. Neither resistance nor thermopower saturate at 10 kOe, and the resistance decreases by about 5%, while the magnitude of thermopower increases by about 10%. The thermopower value is negative (about -6μV/K at zero field), between the thermopower values of Co and Ag bulk metals (-30 μV/K for Co and 1.5 μV/K for Ag), but closer to that of Ag. An MTP (S) vs inverse resistance ($1/R$) plot for the same data is shown in Fig. 2. The thermopower is inversely proportional to the resistance. This behavior was observed in multilayers[9,11,12] and was predicted for granular systems.[13] The linear relationship between S and $1/R$ continues to hold well for $T \geq 75$ K, as Fig. 3 shows at 100 K.

In the temperature range from room temperature to about 75 K, the zero-field thermopower has a linear temperature dependence. The resistance is also linear in temperature in this range. Figure 4 shows the temperature dependence of the thermopower at zero field and at the highest applied magnetic field (9.8 kOe). The thermopower data shown in this figure have already been corrected for the temperature dependence but not the field dependence of the gold leads according to published data.[14] As the temperature is decreased below 50 K, the MTP deviates from the linear relation between S and $1/R$ in high magnetic fields and the zero-field value changes sign.

In a normal homogeneous metal, the ordinary magnetoresistance is below 1% and its origin is clear; the diffusion thermopower is not sensitive to the magnetic field,[15] and the resistance is not simply related to the thermopower. However in granular systems, the coexistence of a large MTP with a large magnetoresistance indicates that they have a common origin. In fact, in the temperature range where the diffusion thermopower is dominant, both the zero-field values and the inverse relationship between S and R can be understood within the context of the two-current model. In order to explain the unusually large MTP, we assume that the spin-dependent DOS at the Fermi surface plays an essential role in the scattering. Further, because the temperature coefficient of zero-field resistance in this temperature range is about 5.7×10^{-4} K^{-1}, an order of magnitude smaller than that of pure metals, we assume that the interfacial (with a random spin-independent potential U) and impurity scattering are the dominant mechanisms. Therefore, we consider only elastic scattering, ignoring the spin-mixing contribution caused by magnon scattering, which is a reasonable approximation as discussed previously.[5] As in the multilayer case, we choose the applied magnetic-field direction, i.e., the net magnetization direction, as the spin-quantization axis of the electrons, and use $\uparrow$ and $\downarrow$ to refer to the global spin-quantization axis. The spin-dependent DOS in the magnetic grains are given by $g_+(E_F)$ and $g_-(E_F)$ with $+$ ($-$) parallel (antiparallel) to the *local* magnetization.

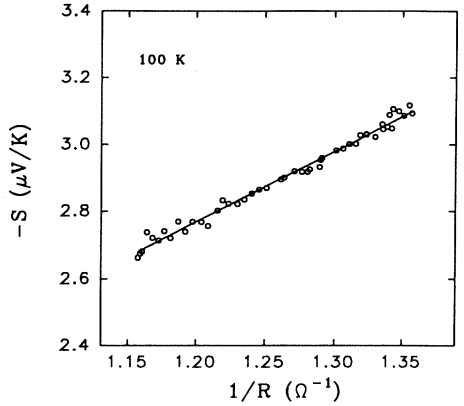

FIG. 3. $-S$ vs $1/R$ plot for 100-K data of 300 °C 10-min sample. The solid line has a slope of about 4 μV Ω/K.

Assuming that only s electrons contribute to the conduction and that s-d scattering is the principal relaxation process at the interfaces, we obtain the scattering rate for $\uparrow$ s electrons due to magnetic particles

$$\frac{1}{\tau_{\uparrow}^{p}} = \sum_{k'} P(\mathbf{k}'d\uparrow,\mathbf{k}s\uparrow) = \sum_{k'} [P(\mathbf{k}'d+,\mathbf{k}s\uparrow)+P(\mathbf{k}'d-,\mathbf{k}s\uparrow)] \,. \quad (1)$$

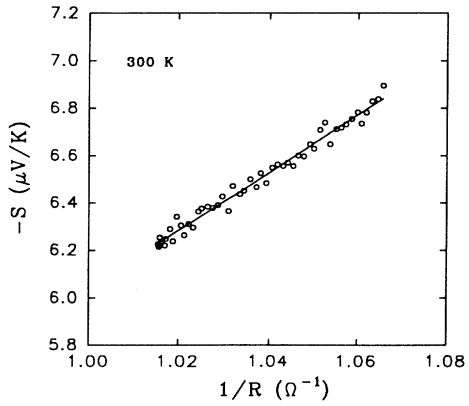

FIG. 2. The same data as shown in Fig. 1 on a $-S$ vs $1/R$ plot. The solid line has a slope of 12 μV Ω/K.

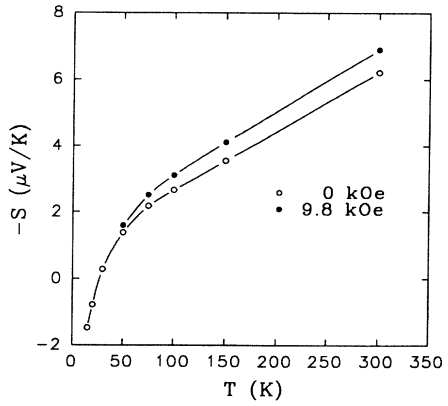

FIG. 4. The temperature dependence of the thermopower for magnetic field 0 kOe (open circles) and 9.8 kOe (closed circles).

Here, $P(\mathbf{k}'d\uparrow,\mathbf{k}s\uparrow)$ is the relaxation rate of s-d spin-preserving scattering. It is more convenient to consider partial relaxation rates, $P(\mathbf{k}'d\pm,\mathbf{k}s\uparrow)$, which are the scattering probabilities for the s electron ($\mathbf{k}s\uparrow$) to be scattered into ferromagnetic d states ($\mathbf{k}'d\pm$). In the Born approximation, these are proportional to squared matrix elements of U and to the final DOS in majority (+) and minority (−) d bands after a summation over $\mathbf{k}'$. The angle θ_p between the pth particle's local magnetization axis and the global quantization axis appears in the spinor matrix elements $\langle\pm|\downarrow\rangle$ (and $\langle\pm|\uparrow\rangle$). Total scattering rates for spin-up and -down channels can then be found by summing the scattering rate from all magnetic particles and from scattering centers in the nonmagnetic matrix. Assuming the spin-flip mean free path is significantly longer than the interparticle distance, we obtain

$$\frac{1}{\tau_{\uparrow,\downarrow}}=f_m\left[\frac{2\pi u^2}{\hbar}\right]\left[g_{+,-}(E_F)\left\langle\cos^2\left[\frac{\theta_p}{2}\right]\right\rangle+g_{-,+}(E_F)\left\langle\sin^2\left[\frac{\theta_p}{2}\right]\right\rangle\right]+\frac{f_0}{\tau_0}\ , \tag{2}$$

where

$$u^2=\left[\frac{1}{4\pi}\right]\int d\Omega|\langle\mathbf{k}'|U|\mathbf{k}\rangle|^2\ . \tag{3}$$

Here f_m is the volume fraction of the surface layer of magnetic grains, f_0 is the volume fraction of the nonmagnetic matrix, and $1/\tau_0$ is the scattering rate within the matrix for each channel.

Adding up the two parallel channel conductances and using $\langle\cos^2\theta_p\rangle=(M(H)/M_s)^2$, we can express the resistivity in any given field as

$$\rho=\left[1-\left[\frac{M(H)}{M_s}\right]^2\right]\rho_d+\left[\frac{M(H)}{M_s}\right]^2\rho_s\ , \tag{4}$$

with

$$\rho_d=\tfrac{1}{4}(R_++R_-)\ ;\quad \rho_s=\frac{R_+R_-}{R_++R_-}\ . \tag{5}$$

Here ρ_d is the resistivity in the zero-magnetic-field demagnetized state, ρ_s is the resistivity in the high-field saturated limit, and $R_\pm=f_m\rho_\pm+2f_0\rho_0$. Within this simple model, the resistivities arising from magnetic scattering are $\rho_\pm=(2\pi m^*u^2/ne^2\hbar)g_\pm(E_F)$ and that from scattering within the matrix is $\rho_0=m^*/2ne^2\tau_0$; n is the carrier density in each spin channel. In granular systems, Eq. (4) has been found to be obeyed except in very high fields.[4] We have attributed the difference between ρ_+ and ρ_-, and hence the magnetoresistance, to the spin-split DOS difference $g_+(E_F)-g_-(E_F)$. Previous researchers[5] have ignored this source of magnetoresistance, focusing rather on differences in the scattering potentials, i.e., $u_+^2\neq u_-^2$, with the final DOS always that of the matrix. As we will show below, while either or both can explain the magnetoresistance, a DOS difference is required to describe the thermoelectric power.

To treat the thermopower, we apply the Mott formula[16] to Eq. (4), to obtain the relationship between the measured thermopower and resistance as functions of applied field. After some algebra we find

$$S(H)=\frac{\rho_dS_d-\rho_sS_s}{\rho_d-\rho_s}+\frac{\rho_s\rho_d(S_s-S_d)}{\rho_d-\rho_s}\frac{1}{\rho(H)}\ , \tag{6}$$

where

$$S_d=\frac{R_+\Sigma_++R_-\Sigma_-}{R_++R_-}\ ,\quad S_s=\frac{R_-\Sigma_++R_+\Sigma_-}{R_++R_-}\ , \tag{7}$$

$$\Sigma_\pm=\frac{f_m\rho_\pm(\pi^2k_B^2T/3|e|)[\partial\ln g_\pm(E)/\partial E]|_{E_F}+2f_0\rho_0S_0}{R_\pm}\ , \tag{8}$$

and where S_0 is the thermopower of the matrix. Equation (6) gives the required linear relationship between $S(H)$ and $1/R(H)$.

It is clear from Eq. (6) that the existence of MTP requires a difference in S_s and S_d. However, the conventional model, in which the density of final states always refers to the matrix, gives $\Sigma_+=\Sigma_-$ and makes $S_s-S_d=0$. There is no magnetothermopower in that model. A more complete description than given here would include the possibility of spin-dependent scattering matrix elements, but the spin-split DOS model by itself produces both magnetoresistance and magnetothermopower. The factor f_m is proportional to the surface-to-volume ratio of the magnetic particles, and causes the magnetoresistance, the demagnetized-state thermopower, and the MTP to decrease with increasing particle size at fixed Co concentration.

The presence of matrix scattering also reduces the magnitude of the magnetoresistance $\rho_d-\rho_s$ and the MTP S_d-S_s. In order to estimate its importance, we make a comparison with the thermopower of antiferromagnetic Co/Cu multilayers in the limit of thin Cu spacers, in which the Cu matrix-layer contribution is negligible. The expression for the thermopower in that case[12] is identical with S_d in the limit $f_0\to0$, which is denoted S_d^0. It is straightforward to show from the above equations that

$$S_d=S_d^0+\frac{f_0\rho_0}{\rho_d}(S_0-S_d^0)\ . \tag{9}$$

We found in previous work on Co/Cu multilayers that $S_d^0\approx-19\ \mu$V/K at 300 K. S_0 is the thermopower of the matrix, and for Ag, it is 1.5 μV/K. To obtain $S_d=-6\ \mu$V/K, as observed, from Eq. (11) requires that $f_0\rho_0/\rho_d\approx0.6$, so that the matrix resistivity is comparable to that due to embedded particles. Unlike the multilayer samples, where the spacer layers are prepared to be almost free of magnetic impurities, granular solids unavoidably have relatively higher impurity concentrations in the form of nonmagnetic grains and at grain boundaries. It is reasonable that the resistivity of the Ag matrix is significantly greater than that of pure bulk Ag, despite the insolubility of Co in Ag.

Finally, we comment on the MTP at low temperatures. For $T < 75$ K, S_0 is no longer dominated by the diffusion thermopower, but by the phonon-drag thermopower. The phonon drag[15] occurs approximately at the temperature $\theta_D/12$ (θ_D is the Debye temperature), where not only the magnitude of the thermopower is large, but also it is very sensitive to the applied magnetic field. This is the reason that the relation between the MTP and magnetoresistance deviates from the inverse linear behavior below 75 K. Below ~40 K, the phonon-drag thermopower of the Ag matrix exceeds the particle contribution.

Our approach differs from a model[17] proposed by Stearns to explain magnetoresistance effects in Fe and in multilayers. Although both models put emphasis on the spin-split DOS of the magnetic component, we treat the *s* electrons in the matrix as the charge carriers, and neglect the *d* electrons' contribution to the current. The potential scattering at interfaces gives rise to both the magnetoresistance and the MTP even though there are no antiparallel magnetic boundaries in granular systems as in Fe. Further, our model explains the M^2 dependence of the resistivity, while the magnetic boundary scattering gives a linear relationship between the magnetization and resistivity.

In conclusion, we have studied the MTP and magnetoresistance of AgCo granular systems. The MTP is inversely proportional to the magnetoresistance over the temperature range where the diffusion thermopower dominates. The zero-field thermopower value lies between those of Co and Ag but closer to that of Ag. These observations show for the first time in granular systems the evidence to support the spin-split DOS model.[13] This model provides a unified picture which explains the GMR and MTP in both multilayers and granular solids even in the absence of spin-dependent potential scattering.

J.S. and E.K. thank P. Fenimore, F. Yu, B. Everitt, K. Pettit, R. Ohigashi, and N. Tea for help and discussions. This work was supported by U.S. Department of Energy Grant No. DEFG02-91ER45439 through the University of Illinois, Materials Research Laboratory. E.K. was supported by Monbusho-Sponsored Japanese Overseas Research Fellowship. L.X. was supported by the ONR Grant No. N0001-4-89-J-1157.

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