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Magnetothermal transport properties of granular Co-Ag solids

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Thermal conductivity and thermoelectric power have been measured between 2 K and 300 K on granular $Co_{20}Ag_{80}$ solids. As in ordinary metals, thermal conductivity is largely dominated by the electronic contribution. The Wiedemann-Franz law is found to be satisfied over the whole temperature range investigated, which is mainly ascribed to dominant large-angle elastic scattering processes. Giant magnetothermal conductivity has been observed in accordance with the giant magnetoresistance effect. The magnetothermoelectric power is negative and increases with temperature.

Much attention has been focused on magnetic multilayers in recent years, especially since the discovery of the oscillatory interlayer exchange coupling and the giant magnetoresistance (GMR) in certain multilayers with an antiferromagnetic alignment of the ferromagnetic layers.[1–5] More recently, GMR has been uncovered in granular media consisting of single-domain ferromagnetic particles (Co, Fe, or Ni) in a nonmagnetic noble metal matrix (Cu or Ag).[6,7] This observation provides a new perspective towards the understanding of the intriguing GMR effect first observed in multilayers.

To date, virtually all transport studies on multilayer and granular systems are exclusively on electronic transport, one of the several transport phenomena which are essential for a complete understanding of the intriguing transport properties in these magnetically inhomogeneous materials. The main interest of this work is to report results of the thermal transport measurements (thermal conductivity and thermoelectric power) in a granular magnetic system that exhibits GMR and to compare thermal transport with electronic conduction properties. Thermal conductivity and electrical resistivity are directly related by the Wiedemann-Franz law in a large temperature range of 2–300 K, implying that the transport phenomena, both electrical and thermal, are dominated by elastic and large-angle inelastic scattering events. We have also observed giant magnetothermal conductivity (GMTC) and giant magnetothermoelectric power (GMTEP) effects correlating with GMR. These unusual transport properties of granular solids are different from those of ordinary metals and alloys.

The lack of thermal transport studies of thin films to date stems in part from experimental difficulties. While for resistivity measurements a thin metallic film sample deposited onto an insulating substrate would suffice, these samples are unsuitable for thermal conductivity measurements. First of all, the heat transfer through the much thicker substrate would not be easily separated from that through the thin film; in fact, the former would overwhelm the latter. Furthermore, to achieve sufficiently high accuracy in the thermal conductivity measurements, film thicknesses of several tens of microns would be required, so as to obtain samples with substantial thermal conductance. Such thicknesses are nearly two orders of magnitude larger than those of most available thin films. Consequently, due to these stringent requirements, there have been no thermal conductivity measurements on thin multilayer or granular films to date, despite the fact that they have already displayed very interesting electrical transport properties. For our thermal transport measurements, we have used high-rate sputter deposition with multiple deposition sources to fabricate very thick (~ 100 μm) and substrate-free films, which eliminate the experimental problems mentioned above. The thick samples of $Co_{20}Ag_{80}$ (20 vol %) have been sputtered at room temperature, and subsequently annealed at $T_A = 480\,^{\circ}C$ and $T_A = 605\,^{\circ}C$ for 10 min in a high-vacuum furnace to create a medium of granular Co, a few nm in size, embedded in Ag. The granular nature of the samples has been confirmed by both x-ray diffraction and transmission electron microscopy. The detailed sample characterizations have been described elsewhere.[8] In granular Co-Ag samples, GMR, as much as 80%, has recently been observed.[8]

For the thermal transport measurements, we have used two experimental setups. Thermal conductivity and thermoelectric power under a magnetic field (up to 20 kG) between 2 K and 120 K have been measured using a traditional static heat-sink four-probe method with a sample holder (A) previously described.[9,10] The samples were not saturated by an external field before the $H = 0$ measurements. In a different setup, thermal transport properties up to room temperature in zero magnetic field have been measured in a special low-loss sample holder (B) specially designed for thin films and fibers. The details of these sample holders and measurements have been described previously.[11] In the figures, the data obtained using sample holders A and B are represented by circles and diamonds, respectively.

The temperature dependence of the thermal conductivity (κ) of $Co_{20}Ag_{80}$ ($T_A = 605\,^{\circ}C$) from 2 K to 300 K, with and without a magnetic field (H), is shown in Fig. 1. Let us first discuss the results with $H = 0$. The results of κ from both sample holders A and B are in excellent

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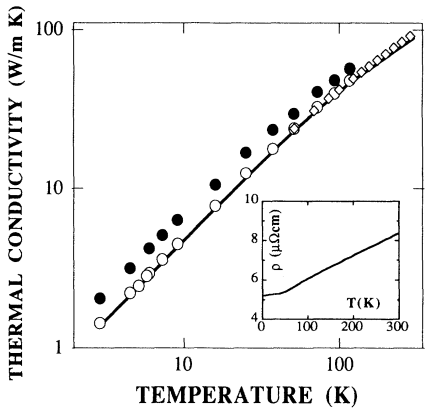

FIG. 1. Temperature dependence of the thermal conductivity κ of the $Co_{20}Ag_{80}$ sample ($T_A = 605\,^{\circ}C$) in the $H = 0$ (○) and at the saturation field H_{sat} (●) using sample holder A. The data obtained at higher temperature in zero magnetic field using sample holder B are also shown (◇). The solid line is the electronic thermal conductivity computed using the Wiedemann-Franz law [Eq. (1)] and the zero-field resistivity data shown in the inset.

agreement. It is noted that κ increases with T throughout the temperature range investigated. For comparison, the temperature dependence of the electrical resistivity is shown in the inset of Fig. 1. Before further discussion of the thermal transport properties of granular metals, it is useful to recall the relationship between the electronic thermal conductivity (κ_E) and electrical resistivity (ρ) in ordinary metallic systems. When the scattering is elastic, κ_E and ρ are related by the Wiedemann-Franz (WF) law

$$\kappa_E \rho / T = L_0 \,, \tag{1}$$

where T is the temperature and L_0 is the Lorenz number with the value of 2.45×10^{-8} V^2/K^2 for a free electron system.

For ordinary metals, where thermal conductivity is purely electronic, the WF law applies at low temperature because the scattering is entirely due to static defects and is thus elastic. On the other hand, in ordinary alloys, due to the increased impurity scattering, the WF law is valid over a more extended temperature range.[12] The WF law is also generally obeyed above the Debye temperature because the maximum phonon energy becomes small compared with $k_B T$. In this case, large-angle electron-phonon scattering dominates and the scattering can be considered quasielastic. In the intermediate temperature range, however, the WF law breaks down since the effectiveness of small-angle electron-phonon scattering in degrading electrical and thermal transport is different and the left-hand side of Eq. (1) is much smaller than L_0.

In contrast, in granular Co-Ag the WF law is found to hold *throughout* the temperature range of 2–300 K, as shown in Fig. 1. It should be noted that the solid curve in the thermal conductivity results, which is in excellent agreement with the data, is not a fit but the result of a calculation using the WF law and the resistivity data shown in the inset of Fig. 1. For some samples, we found that the coefficient L_0 can deviate by ~10% from the value of the free electron Lorenz number. However, this small discrepancy is within the uncertainties in estimating the distance between thermal and electrical contacts.

The resistivities of granular Co-Ag (e.g., 8.5 $\mu\Omega$ cm at 300 K for the sample annealed at 605 °C), although larger than those of noble metals, are more than a factor of 5 smaller than those of concentrated alloys. However, unlike those of ordinary metals and alloys, the WF law is satisfied throughout the entire temperature range studied. This result advocates both a dominant electronic thermal conductivity ($\kappa \sim \kappa_E$) and large-angle scattering events. Most of these arise from elastic scattering at the interfaces and within the two metallic media. There are also contributions to the resistivity due to scattering from phonons and other thermal excitations, although they are relatively small as indicated by the small increase of resistivity with increasing temperature (a factor of 1.5 between 4.2 K and room temperature). The validity of the WF law in granular Co-Ag above $T \sim 100$ K might be taken as an indication that the scattering events are incoherent, i.e., without conservation of **q**, giving rise to large-angle scattering. In contrast, small-angle electron scattering processes would cause deviation of the WF law though a rough estimation indicates that $\kappa\rho/T$ should not fall more than 10% below L_0 in the temperature range 100 K–300 K. Given the experimental uncertainties of our measurements, this possibility cannot be ruled out definitively.

Upon the application of a magnetic field, very large magnitudes of magnetoresistance and magnetothermal conductivity are observed. In Fig. 1, the variation of the thermal conductivity with T at the saturation field is also shown for $Co_{20}Ag_{80}$ ($T_A = 605\,^{\circ}C$). To more clearly display their field dependences and the large magnitude of the magnetotransport properties, the results at $T = 72$ K of $\delta\rho = \rho(H) - \rho(0)$, $\delta\kappa = \kappa(H) - \kappa(0)$, and $\delta S = S(H) - S(0)$, to be discussed later, are shown in Fig. 2(a). The GMR is negative, as in all systems that exhibit GMR, with an effect size of ~33%. The GMR is even larger (~48%) at $T = 4.2$ K. The magnetothermal conductivity is positive as expected from the WF law, yielding a GMTC of ~28%. To our knowledge, this is the first report of GMTC in magnetic thin films, where the magnetic field can so substantially alter the thermal conductivity.

To illustrate the relationship among the magnetotransport properties, we plot the field dependences of thermal conductivity (κ) and electricial conductivity ($\sigma = 1/\rho$) in Fig. 2(b). Clearly both $\delta\kappa$ and $\delta\sigma$ have the same field dependence. At a given temperature, the relative variations in both quantities are the same, i.e., the magnetotransport properties also satisfy the WF law indicating that GMTC and GMR both originate from magnetic scattering of the conduction electrons.

Next we discuss the thermoelectric power results. In Fig. 3, we show the temperature dependence of the thermoelectric power $S(T)$ in zero field for $Co_{20}Ag_{80}$ ($T_A = 480\,^{\circ}C$ and $T_A = 605\,^{\circ}C$). In both cases, the thermoelectric power (S) is negative with a similar temperature dependence, except that the slope of $S(T)$ for the

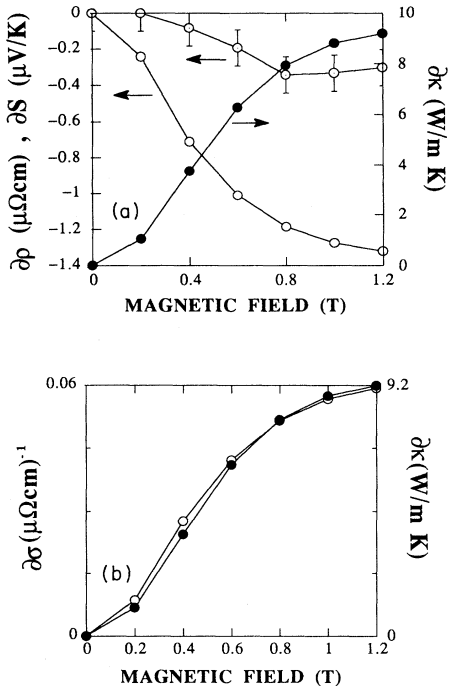

FIG. 2. (a) Changes under the magnetic field at $T=72$ K of the electrical resistivity (○), thermal conductivity (●), and thermoelectric power (□) for the $Co_{20}Ag_{80}$ sample ($T_A=605$ °C). δX (with $X=\rho,S,\kappa$) is $X(H)-X(0)$ and the magnetic field is perpendicular to the film. (b) The field dependence of the electrical conductivity σ (●) and the thermal conductivity κ (○) at $T=72$ K. Both $\delta\sigma$ and $\delta\kappa$ have the same functional dependence on H.

sample with $T_A=480$ °C is higher by about 50%. For both samples, S depends linearly on T above $T\sim100$ K, suggesting that the diffusion component is the dominant contribution. As T is reduced to below 100 K, a departure from the linear T dependence, in the form of a small negative peak, is observed. In noble metals, a positive peak in $S(T)$ due to phonon-drag effect appears around $T=60$ K.[13] However, since the phonon-drag peaks in noble metals are rapidly suppressed on alloying, the anomalies observed in $S(T)$ around $T\sim70$ K for $Co_{20}Ag_{80}$ solids are unlikely attributable to phonon drag.

In Fig. 4, we show the temperature dependence of the total change in thermoelectric power, $\Delta S=S(H=H_S)-S(H=0)$, where H_s is the saturation field for the $Co_{20}Ag_{80}$ samples with $T_A=480$ °C and $T_A=605$ °C. The quantity ΔS is negative and its magnitude is very small at low temperature and increases rapidly above 30 K. Recently, the first measurements of GMTEP in magnetic multilayers of Co/Cu and Fe/Cu have shown a negative GMTEP with approximately a T^2 variation at low temperatures.[9,10] Such a rapid increase of the GMTEP with T has been explained invoking a significant electron-magnon scattering (responsible for the "spin-mixing" in the framework of the two-current model) to the diffusion thermoelectric power.[9,10] For granular systems with very small particles, scattering of conduction electrons by "extended" magnons should be less important due to severe restrictions on the spin-wave wavelengths. However, incoherent scattering due to localized magnons at the Co/Ag interfaces might contribute to the electrical resistivity. Owing to the fact that the joint scattering of electrons by magnons and impurities can lead to very large thermoelectric power in ferromagnetic systems, this interpretation is also plausible. In addition, it should be pointed out that the sign of the GMTEP observed on the $Co_{20}Ag_{80}$ samples also agrees with that predicted in the above-mentioned model.

In conclusion, we have observed very unusual features in the thermal transport properties of granular magnetic systems that exhibit GMR. Thermal conductivity, which is dominated by the electronic contribution, exhibits large changes under moderate magnetic fields. Both the thermoelectric power and GMTEP are negative while the temperature dependence of the MTEP is similar to that previously reported on magnetic multilayers. These transport properties are different from those of ordinary metals and alloys. The validity of the Wiedemann-Franz law over a wide temperature range gives support for a dominant elastic scattering and suggests that the inelastic scattering processes are incoherent. These important as-

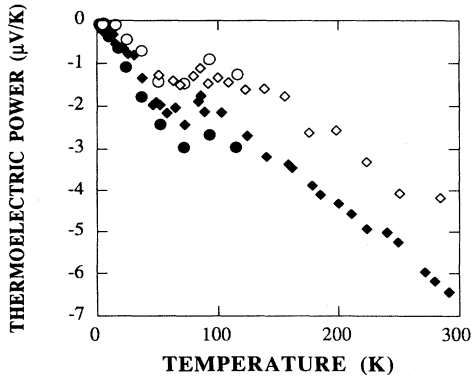

FIG. 3. Thermoelectric power S as a function of temperature in zero magnetic field for the $Co_{20}Ag_{80}$ samples with $T_A=480$ °C (●,◆) and $T_A=605$ °C (○,◇).

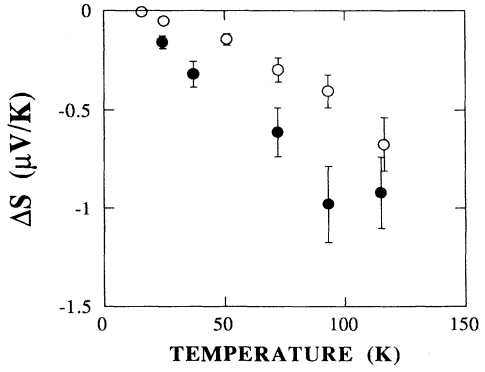

FIG. 4. Temperature dependence of the GMTEP at the saturation field ΔS for the $Co_{20}Ag_{80}$ samples with $T_A=480$ °C (●) and $T_A=605$ °C (○).

pects should be incorporated into, and addressed by, theoretical models attempting to account for the intriguing transport properties.

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