Extraordinary Hall Effect and Giant Magnetoresistance in the Granular Co-Ag System

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We have studied the magnetotransport properties of a new metallic granular Co-Ag system. The spin-dependent electron scattering is revealed in the forms of a giant magnetoresistance and an extraordinary Hall effect, both of which are found to be strongly dependent on the Co particle size and the electron mean free path. The experimental correlation $\rho_{xy} \propto \rho_{xx}^{3.7}$ uncovered is unexpected from existing theories. Interfacial scatterings of intermediate range are required in order to account for the anomalous magnetotransport in this magnetic granular system.

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The discovery of giant magnetoresistance (GMR) in magnetic multilayers (e.g., Fe-Cr, Co-Ag) has created new opportunities for the understanding of the fundamental issue of spin-dependent electrical transport [1–5]. The recent surprising finding that a GMR also exists in granular structures brings a new dimension to this problem [6,7], and raises many new and urgent questions pertinent to the basic mechanism of the GMR. For example, how does the GMR correlate with material parameters such as the size of the magnetic particles and the electron mean free path? Another interesting question is the possible manifestation of spin-dependent electron scattering in other magnetotransport properties. Among them the Hall effect is particularly relevant, because it is very sensitive to the magnetic state of a material and because it provides crucial information on the carrier type, concentration, and mean free path if the resistivity is also known. It would be extremely beneficial to a comprehensive understanding of the GMR if many aspects of magnetotransport could be simultaneously investigated. In this Letter we present results of magnetotransport properties in granular Co-Ag films. Spin-dependent electron scattering is directly manifested in the extraordinary Hall effect (EHE), which strongly correlates with resistivity and magnetization orientation of the single-domain ferromagnetic particles. The saturation magnetoresistance attains a phenomenal value of 84% in this granular system. The correlation between EHE and resistivity cannot be described by any existing theories and is markedly inconsistent with results of homogeneous magnetic dilute or ferromagnetic alloys. Magnetic particle size and electron mean free path are found to be the two crucial parameters determining the magnitude of the GMR and EHE.

Using a two-step process, we have fabricated granular Co-Ag films by taking advantage of the fact that Co and Ag are mutually immiscible under equilibrium conditions. First we deposited by sputtering a $Co_{20}Ag_{80}$ (20 vol.%) thin film, about 2 μm in thickness, onto substrates (Si) held at 77 K using dc magnetron sputter-

ing. The high quenching rate enables the formation of a homogeneous metastable alloy. The second step was a thermal treatment, with annealing temperatures (T_A) of 200, 330, 480, and 605 °C, in a high vacuum furnace for 10 min. The annealing process induces phase separation in the metastable alloy, producing ultrafine Co particles embedded in the metallic Ag matrix. X-ray diffraction and transmission electron microscopy analysis, presented elsewhere [8], attest to the formation of a granular structure with distinct Co and Ag entities. The Co particle size, about 2 nm at $T_A = 200$ °C, increases to about 13 nm at the maximum T_A of 605 °C. We used standard photolithography and wet-etching to pattern our samples for electrical measurement. The magnetoresistivity and Hall resistivity were measured simultaneously using a dc four-probe method. We have taken precautions to eliminate measurement errors such as thermoelectric voltage and Hall-probe misalignment. A SQUID magnetometer was employed to measure the magnetic properties.

For each sample we measured the resistivity ρ_{xx} and the Hall resistivity ρ_{xy}, starting from the nonmagnetized state and then increasing H to $+8T$. Afterwards, H was swept from $+8T$ to $-8T$ and back to $+8T$ again. This procedure is necessary to observe the effect of magnetic hysteresis on transport. Figure 1 shows the results of ρ_{xx} and ρ_{xy} as functions of H ($H \perp$ thin film) at $T = 4.2$ K for four samples annealed at various temperatures. The value of $\rho_{xx}(H)$ attains a maximum whenever the magnetization M is zero and ρ_{xx} approaches asymptotically to a lowest value as M reaches saturation (M_s). The GMR is observed in every sample, with saturation $\Delta\rho_{xx}/\rho_{xx}$ ranging from 50% to 84%. (Note that $\Delta\rho_{xx}/\rho_{xx}$ is referenced to the saturated resistivity at high H.) Such a large magnetoresistance is remarkable for any system, making granular structures most competitive with multilayers exhibiting the largest GMR. We note that the residual resistivity in $Co_{20}Ag_{80}$ samples is substantially larger than that (0.61 $\mu\Omega$ cm) of the sputtered pure Ag due to disorder intrinsic to the granular structure.
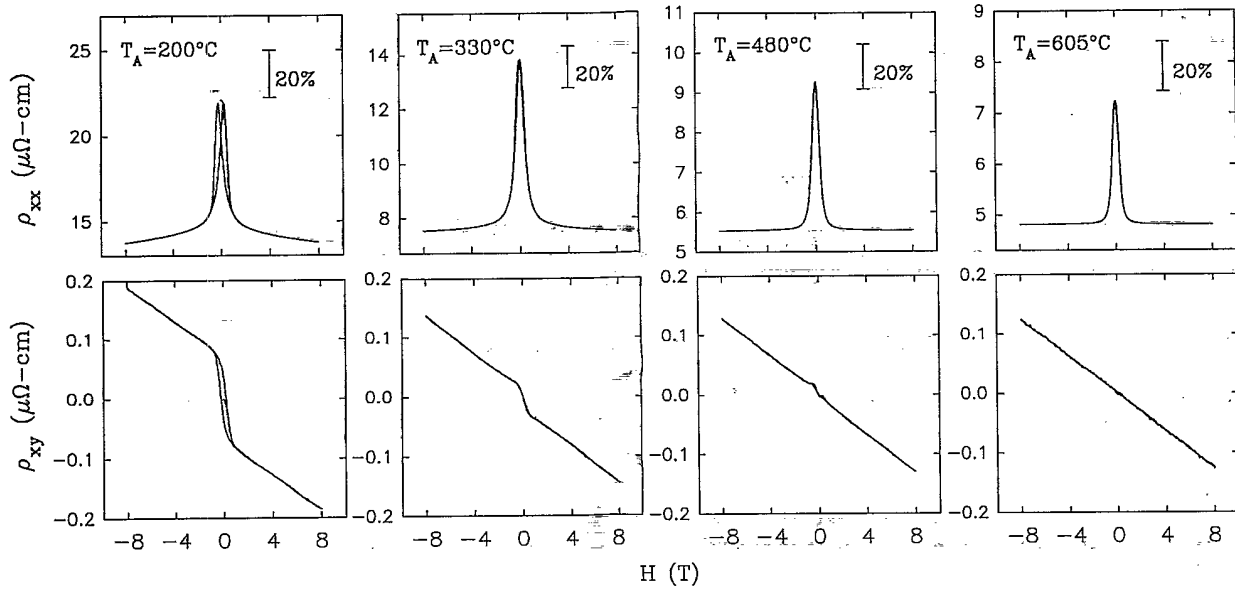
FIG. 1. Perpendicular magnetoresistivity $\rho_{xx}(H)$ and Hall resistivity $\rho_{xy}(H)$ at $T = 4.2$ K for granular Co-Ag samples annealed at various temperatures (T_A). The percentage indicators refer to $\Delta\rho_{xx}/\rho_{xx}$ as defined in the text.

There are several contributions to ρ_{xx}. The residual ρ_0 due to disorder is independent of H. At $T = 4.2$ K scatterings due to phonons or magnons are negligible. Therefore the variation in ρ_{xx} with H is solely caused by the magnetic scattering of conduction electrons by the magnetic particles. In what follows we will use $\Delta\rho_{xx}$ to represent the maximum change in ρ_{xx} between $M = 0$ and M_s. It is $\Delta\rho_{xx}$ which best characterizes the strength of the magnetic scattering responsible for the GMR. We observe that $\Delta\rho_{xx}$ decreases monotonically with T_A, and with Co particle size (see discussion below). However, the commonly used quantity $\Delta\rho_{xx}/\rho_{xx}$ does not follow a monotonic relation with T_A [$\Delta\rho_{xx}/\rho_{xx}$ =61%, 84%, 67%, and 50%, in order of increasing T_A]. This is because the denominator ρ_{xx} also contains other contributions (e.g., from disorder) which depend on T_A.

Now we turn our attention to the Hall effect as shown in Fig. 1. A few interesting features deserve attention. Unlike in nonmagnetic metals, ρ_{xy} is not linear in H. The behavior of ρ_{xy} is intimately related to that of ρ_{xx}. In particular, at low fields, ρ_{xx} changes the most, and so does ρ_{xy}, which has a large initial slope against H. Then, as ρ_{xx} approaches saturation, ρ_{xy} becomes linear with H but with a much smaller slope. Perhaps the most important feature of the Hall effect is that the magnitude of the initial change in ρ_{xy} decreases rapidly as the Co particles become larger. Furthermore, the hysteresis behavior of ρ_{xy} resembles closely the M vs H hysteresis loop, indicating that the initial ρ_{xy} is of magnetic origin. The results for ρ_{xy} in Fig. 1 can be treated phenomenologically as the sum of two components [9]

$$\rho_{xy} = R_0[H + 4\pi M(1 - D)] + R_s 4\pi M , \qquad (1)$$

where the first term is due to the ordinary Hall effect

(OHE) (D is the demagnetization factor), and the second term, resulting from a magnetic scattering, is the extraordinary Hall effect (EHE) [9] proportional to M. In what follows we will denote $\rho_{xy}^M = R_s 4\pi M$. Note that R_s is negative in every sample. Figure 2 shows the hysteresis measurement of both ρ_{xy}^M and M as a function of H. Clearly there is a one-to-one correspondence between ρ_{xy}^M and M, indicating that relation (1) provides an excellent description of the Hall effect. To characterize the
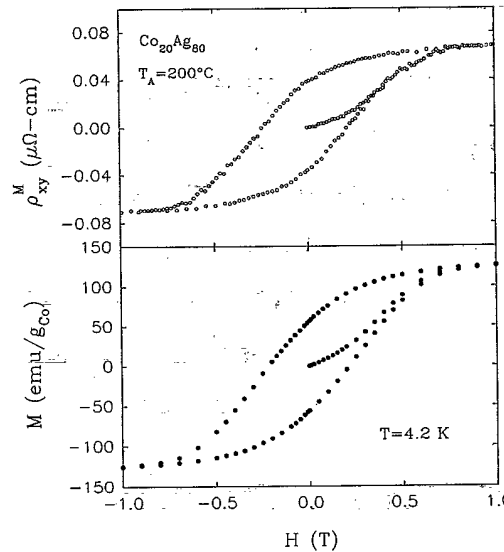

FIG. 2. The hysteresis loops at $T = 4.2$ K of the extraordinary Hall resistivity ρ_{xy}^M and the magnetization M for the Co-Ag sample annealed at $T_A = 200\,^\circ$C. Note the one-to-one correspondence between ρ_{xy}^M and M. ρ_{xy}^M has been inverted in sign to compare with M.

EHE, it is best to extract the spontaneous Hall resistivity $\rho_{xy}^{M_s}$ from Fig. 1 by extrapolating $\rho_{xy}(H)$ from the linear portion at high H to $H = 0$. In all of our samples $M_s = 142.9$ emu/g within an error of $\pm$ 5% at $T = 4.2$ K. Therefore, $\rho_{xy}^{M_s} \propto R_s$.

It is generally held that the GMR observed in layered structures is caused by the spin-dependent scattering of the conduction electrons by magnetic entities (interface and/or bulk) [10–13]. The external magnetic field affects the effective electron scattering rate through reorientation of the magnetization vectors, generating a large variation in magnetoresistance. All models proposed so far are based on planar structures only [10–13]. It is not clear whether the extension of these models to granular structures is straightforward, because of the complexity and the three-dimensional nature of the granular configuration. To clarify the mechanism of the GMR in granular solids the questions that are particularly relevant are the relative contributions of the interfacial and the bulk magnetic scattering, the particle size dependence of the GMR, and the role of the mean free path on the GMR.

In all of our samples the Co volume fraction remains constant at 20%. The effect of annealing is to change two crucial lengths: (1) the Co particle radius (r_{Co}), and (2) the effective mean free path (λ_{eff}). Indeed samples annealed at higher T_A have smaller ρ_{xx}, and correspondingly, larger λ_{eff}. Using the Drude expressions $\rho_{xx} = m^* v_F / ne^2 \lambda_{eff}$, $R_H = 1/nec$, and $v_F = \hbar \sqrt[3]{3\pi^2 n}/m^*$, we have determined λ_{eff} (4.2 K) for each sample at $H = 0$ where ρ_{xx} is at maximum. The carrier concentration n was found to be 4.1×10^{22} cm^{-3} independent of annealing temperature. It is noted that λ_{eff} depends both on ordinary disorder scattering and on spin-dependent magnetic scattering. From the transmission electron micrograph of each sample we have obtained the particle size r_{Co}, which ranges from 20 to 130 Å. As an independent check, we also used a method described in Ref. [14] to calculate r_{Co} from the temperature dependence of the susceptibility in some superparamagnetic samples. Both methods give consistent results. The values of λ_{eff}, ranging from 50–150 Å, are close to the average Co particle size. In fact, at 20% volume fraction r_{Co} is a length scale not only specifying the Co particle size, but also representing the average interparticle distance. Near the matrix-particle interface the electron scattering is expected to be strong because of the large irregularities and discontinuities in crystalline structure and magnetization. Hence, at T = 4.2 K, where inelastic scatterings are rendered ineffective, λ_{eff} should be of the order of both r_{Co} and the interparticle distance.

In Fig. 3 we present the dependence of $\Delta\rho_{xx}$ and $\rho_{xy}^{M_s}$ on λ_{eff}. Both decrease with increasing λ_{eff} but at different rates. In the inset of Fig. 3, $\Delta\rho_{xx}$ is plotted against $1/r_{Co}$, a linear relation $\Delta\rho_{xx} \propto 1/r_{Co}$ is evidenced. Since in granular systems, the interfacial area per unit volume is proportional to $1/r_{Co}$, this linear relation suggests that the magnetic scattering at the interfaces is responsible for
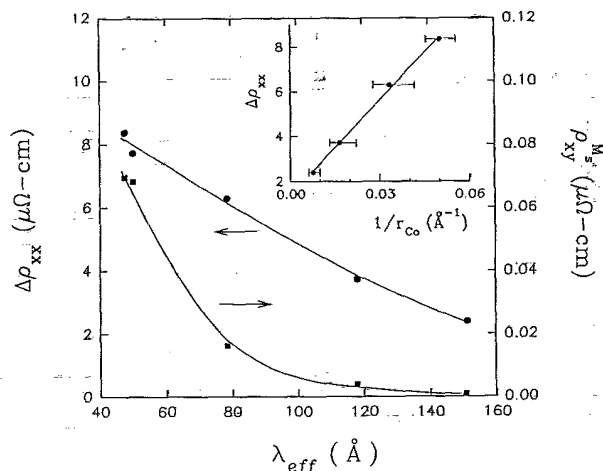

FIG. 3. $\Delta\rho_{xx}$ and $\rho_{xy}^{M_s}$ as functions of the effective mean-free-path λ_{eff} at $T = 4.2$ K. Inset: $\Delta\rho_{xx}$ vs the inverse of the Co particle size, $1/r_{Co}$.

the GMR. We believe that the data presented in Fig. 3 represent the most important features of granular systems with GMR. An understanding of the anomalous transport properties may come from the clarification of the r_{Co}—and λ_{eff}—dependence of $\Delta\rho_{xx}$ and $\rho_{xy}^{M_s}$.

In addition to the GMR, the EHE in the Co-Ag granular system also differs significantly from the EHE observed in magnetic dilute or ferromagnetic alloys. As seen in Fig. 3, $\rho_{xy}^{M_s}$ decreases with λ_{eff}. This is understandable only qualitatively. EHE is caused by spin-orbit interaction between conduction electrons and disorders such as impurities, phonons, etc. Spin-orbit interaction results in two distinctive mechanisms affecting the Hall effect [9,15,16]. The first one is the skew scattering which causes the electron trajectory to deflect asymmetrically from its original path [9]. The second mechanism, a quantum mechanical effect, is the so-called side jump [9,15,16], in which the electron trajectory is displaced transversely by a distance Δy ($\sim$ 0.1–1 Å) while the direction remains intact. Both effects rely on the electron scattering rate [9]. This is why $\rho_{xy}^{M_s}$ decreases with λ_{eff} as shown in Fig. 3. To find out whether the skew scattering or the side jump is the main driving force for the EHE, one may check the correlation between $\rho_{xx}^{M_s}$ and ρ_{xx} [9,15,16]. It has been shown both experimentally and theoretically that $\rho_{xy}^{M_s} \propto \rho_{xx}^n$ with exponent $n = 1$ for skew scattering and $n = 2$ for side jump [9,15]. In Fig. 4 we plot $\log_{10} \rho_{xy}^{M_s}$ vs $\log_{10} \rho_{xx}$, a straight line is obtained over two decades of $\rho_{xy}^{M_s}$. The exponent derived from the least-squares fit is $n = 3.7 \pm 0.2$, which is well beyond the expected value for either the skew scattering or the side jump. Moreover, this large n is obtained at $T = 4.2$ K. In magnetic dilute or ferromagnetic alloys, the skew scattering, in general, dominates at low temperatures, while the side jump becomes important at high temperatures [9,15,16]. In these systems as temperature increases, the exponent n changes from 1 to 2. In Co-Au superlattices,
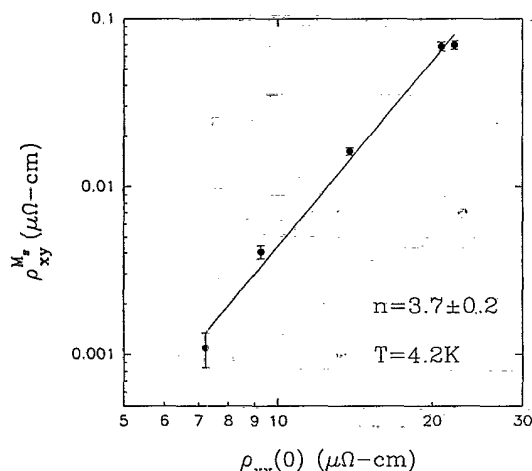
FIG. 4. Correlation between $\rho_{xy}^{M_s}$ and ρ_{xx} for all samples at $T = 4.2$ K. n is the exponent in $\rho_{xy}^{M_s} \propto \rho_{xx}^n$.

n varies from 0.83 at 4.2 K to 1.39 at 81 K [17]. In Fe-Cr superlattices, n is 2.6 from 5 to 300 K [18]. To our knowledge, the exponent $n = 3.7$ in the granular Co-Ag system is the largest exponent ever observed. There is no mechanism, confirmed or proposed, that predicts such a large exponent.

Although the large n is suggestive of a novel mechanism, we feel that the mechanism is still the quantum mechanical side jump, but in the new context of granular structures. In the side-jump model proposed by Berger [9,15] it was found that $R_s = \rho_{xy}^{M_s}/4\pi M_s \propto \rho_{xx}^2 \Delta y$, where $\Delta y = -\lambda_{\text{s.o.}} \sigma_z k_x$ is the transverse side jump after each collision [9,15,16]. ($\lambda_{\text{s.o.}}$ is the spin-orbit coupling parameter, $\sigma_z = \pm\frac{1}{2}$ is the electron spin. k_x is the x component of the Fermi wave vector.) The side jump Δy is independent of the range, strength, or sign of the scattering potential $V(r)$ under the assumption that $V(r)$ is short ranged, i.e., $k_F R \ll 1$ with R being the radius of $V(r)$ [9,15,16]. This assumption is valid for small potential fluctuations such as phonon excitations, and most likely, for dilute impurities. Indeed, the $R_s \propto \rho_{xx}^2$ relation is followed very well over two decades of R_s for several Fe-based dilute alloys at room temperature [15]. However, the short-range $V(r)$ assumption definitely breaks down in granular structures where scatterers are much larger than atomic dimensions. The range of $V(r)$ should be of the order of a few nm, i.e., $k_F R \approx 10$. According to Eq. (17) in Ref. [15], Δy will be dependent on R away from the short-range limit, i.e., $\Delta y \propto 1/R^\alpha$ ($\alpha \leq 2$). It is reasonable to expect $\lambda_{\text{eff}} \propto R$. Therefore, $R_s \propto \rho_{xx}^2 \Delta y \propto \rho_{xx}^{2+\alpha}$, since $\rho_{xx} \propto 1/\lambda_{\text{eff}}$. From our exponent $n = 3.7$, α should be about 1.7. In order to explain the EHE and the large n observed in the Co-Ag granular system, the side-jump model must be extended beyond the short-range limit. More importantly, because of the strong correlation between Hall effect and

resistivity, it may be necessary in theory to treat both GMR and EHE on an equal footing.

To summarize, we have observed the anomalous magnetotransport properties, i.e., giant magnetoresistance and extraordinary Hall effect, in the granular Co-Ag system. Two important parameters, r_{Co} and λ_{eff}, play dominant roles in both effects. The scaling relation between $\rho_{xy}^{M_s}$ and ρ_{xx} is highly unconventional. These results provide strong constraints to a viable and comprehensive theory of both GMR and EHE.

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