Large finite-size effect of giant magnetoresistance in magnetic granular thin films

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We have observed a finite-size effect of giant magnetoresistance (GMR) and extraordinary Hall effect (EHE) in $Co_{20}Ag_{80}$ and $Fe_{20}Ag_{80}$ granular thin films of varying thickness. The GMR approaches zero as the thickness approaches zero. The thickness dependence of GMR can be explained by a dimensional-confinement effect coupled with a large surface-induced spin-flipping cross section. The EHE exhibits a strong correlation with GMR, which is likely due to the surface-induced spin-orbit interaction.

Since the discovery of giant magnetoresistance (GMR) in layered structures, we have witnessed a series of developments in the understanding of the magnetotransport in heterogeneous systems.[1–5] The recent finding that GMR also exists in granular systems[5] has broadened the scope of this research area. A prevailing physical picture of GMR is that there exist two independent currents made of spin-up (+) and spin-down (−) electrons. They are scattered by the magnetic components with different scattering rates $1/\tau_+$, $1/\tau_-$ dependent on the relative orientation between the electron spin and the magnetic moment vector of a scatterer. As one increases the magnetic field (H), the magnetic moments in different layers or granules are gradually aligned. This subdues the scattering suffered by one of the spin currents, which in turn reduces the resistance of a sample due to a short-circuit-like effect. Despite the formidable structural complexities, the "two-current" model in its numerous variations[3,4] seems to account for GMR rather well in multilayers. However, there are important issues yet to be addressed in the GMR of granular systems. We do not understand what role the spin-flipping process plays in granular systems where spin-orbit interaction may be strong. The spin-flipping effect causes mixing of the two currents and is destructive to GMR. We also lack the knowledge of dimensional confinement in magnetotransport. It is believed that the essence of GMR resides in the asymmetry of two electron mean free paths (MFP), λ_+ and λ_-. Dimensional confinement allows one to probe these two important length scales and to examine their effect on GMR. Currently, there are no viable models addressing these issues in granular systems, and the models developed for multilayers are of limited utility due to the particularity of granular microstructures.

In this paper, we present measurement of thickness (t) dependence of GMR, extraordinary Hall effect (EHE), and magnetic properties for Co-Ag and Fe-Ag granular films. We have taken advantage of the fact that strong spin-orbit interaction is often dominant near the surfaces of a thin film.[6,7] By varying t we have observed a large finite-size effect of both GMR and EHE, while keeping the structure and the magnetic properties unchanged. The GMR results have been explained by a model that includes both the effect of spin-flipping and dimensional confinement. The EHE, an inseparable component of the magnetotransport, exhibits an intimate correlation with GMR, but defies conventional explanation commonly used for homogeneous alloys.

We have chosen $Co_{20}Ag_{80}$ and $Fe_{20}Ag_{80}$ (in volume fraction) granular systems for this study. They are immiscible alloys and exhibit GMR in bulk forms.[8–10] Both samples attain a maximum GMR in their alloy series. Their bulk structural and magnetic properties have been investigated previously.[9,10] Our samples were fabricated by using a high vacuum magnetron-sputtering system.[10] To achieve a high degree of consistency, films with variable t from 40 to 3200 Å were deposited during a single run on a 2×5 cm^2 SiO_2 substrate. A computerized stepping shutter swept across the substrate in ten steps according designed time intervals for each step. The total exposure time for each step determined the t of that step. All the samples used in this study were protected by a layer of photoresist immediately after deposition to avoid oxidation. The samples were not thermally annealed. The step-wedged samples were subsequently patterned using photolithography (see the inset in Fig. 1). Samples at each t consist of Hall bars for magnetoresistance (MR) and Hall effect measurement and rectangles for magnetic measurement on a SQUID magnetometer. A representative MR curve is shown in the inset in Fig. 1.

Figure 1 shows the t dependence of $\Delta\rho = \rho(0) - \rho(8\ \mathrm{T})$, the saturated H-induced change in resistivity. In all samples, $\rho(H)$ is nearly saturated at $H = 8$ T and $T = 4.2$ K. For both series, two sets of samples were made and measured, covering approximately the same t range. The data for each set, very consistent among each other, are represented by different symbols. From Fig. 1, we observe two interesting common features shared by both series: First, GMR decreases with the reducing t starting at a large t scale. Even at t as large as 1000 Å, GMR is still not fully saturated at the bulk values. Second, GMR vanishes as t approaches zero. Hence GMR cannot be sustained in the two-dimensional limit.

Before addressing the t dependence, we need to exam-

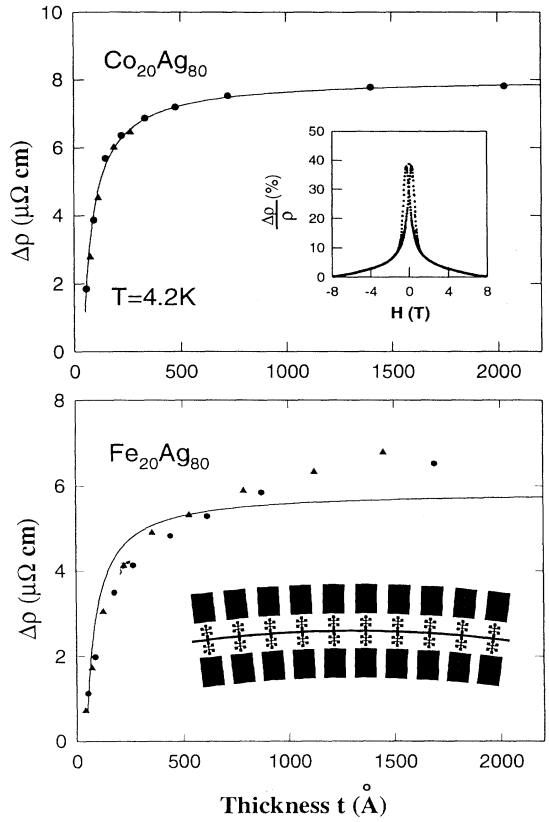

FIG. 1. Thickness dependence of the magnetically induced resistivity $\Delta\rho = \rho(0) - \rho(8\ \mathrm{T})$ at $T = 4.2$ K for the $Co_{20}Ag_{80}$ (upper panel) and $Fe_{20}Ag_{80}$ (lower panel) series. The two symbols represent data from two sets of samples for each series. The solid lines are from the model fitting (see text). The upper inset is the magnetoresistance of a $Co_{20}Ag_{80}$ sample with $t = 2030$ Å. The lower inset shows the geometry of the step-wedged samples with varying t.

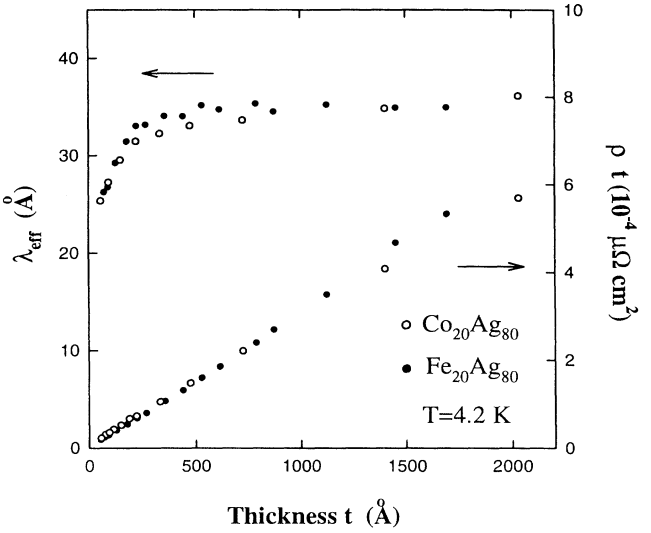

FIG. 2. Thickness dependence of the effective electron mean free path λ_{eff}, and the resistivity-thickness product $\rho \cdot t$ for the $Co_{20}Ag_{80}$ and $Fe_{20}Ag_{80}$ series.

ine some relevant transport and magnetic parameters of our samples. The resistivities at $T = 4.2$ K are 27 and 31 $\mu\Omega$ cm for bulk $Co_{20}Ag_{80}$ and $Fe_{20}Ag_{80}$, respectively. The resistivity ratio ρ (300 K)/ρ (4.2 K) is about 1.1. From the measured resistivities and Hall coefficients, we calculated the effective MFP λ_{eff} for the unmagnetized samples (at $H = 0$), shown in Fig. 2 as a function of t. It is noted that, at zero field when samples are unmagnetized, λ_{eff} for spin-up and -down electrons is the same because of the random distribution of magnetization vectors. The value of λ_{eff} remains at approximately a constant 35 Å for $t > 200$ Å. As t decreases toward zero, λ_{eff} decreases slightly because of increasing surface scatterings. The short λ_{eff} is mainly caused by the strong scatterings at the particle boundaries. As we will show later, the magnetic particle size is about 30 Å and so is the average interparticle distance at 20% volume fraction. For a continuous film with t larger than λ_{eff}, its resistivity consists of two contributions,[11] one is the bulk resistivity, ρ_B, and the other from the surface, $\frac{3}{8}\rho_B\lambda^B_{\mathrm{eff}}/t$,

$$\rho = \rho_B + \frac{3}{8}\rho_B\lambda^B_{\mathrm{eff}}/t = \frac{m^*}{2ne^2}\left(\frac{1}{\tau_B} + \frac{3}{8}\frac{v_F}{t}\right). \quad (1)$$

This expression assumes diffusive scatterings at the surfaces (the total carrier density is $2n$). In Fig. 2, we plot ρt vs t for both series. As expected from relation (1) a straight line fits the data very well. The intercepts along the vertical axis provides $\frac{3}{8}\rho_B\lambda^B_{\mathrm{eff}}$. Since λ^B_{eff} is known, we can estimate ρ_B and the results are consistent with the bulk values. The confirmation of relation (1) reveals that our thin films are continuous in the thinnest limit and t is rather uniform. It is noted that a completely different t dependence of ρ is expected for discontinuous or uneven thin films.[11]

The magnetic properties of a granular film are of paramount importance to GMR.[10] One naturally raises the question whether a possible variation in the underlying magnetic state could lead to the observed t dependence of GMR. Our magnetic measurement on different samples rules out this scenario. Figure 3(a) shows the t dependence of the spontaneous magnetization M_S at $T = 5$ K and 300 K for the $Co_{20}Ag_{80}$ series. At low T, the samples are ferromagnetic. M_S was obtained directly from the measured $M(H)$ curves ($0 \le H \le 5.5$ T). At $T = 300$ K, the samples become superparamagnetic. M_S was obtained by fitting $M(H)$ to a Langevin equation.[10] Simultaneously, the particle size was also obtained from such a fitting; the data are shown in Fig. 3(b). Clearly, both M_S and the particle size remain approximately constant over the whole t range. Therefore as t varies, the magnetic state and the microstructure are not changed appreciably and they should have little, if any, bearing on the t dependence of GMR.

To understand the t dependence of GMR, we need to focus on two major features of a thin film: the existence of surfaces and the geometrical confinement to transport. The surfaces are important spin-orbit (SO)

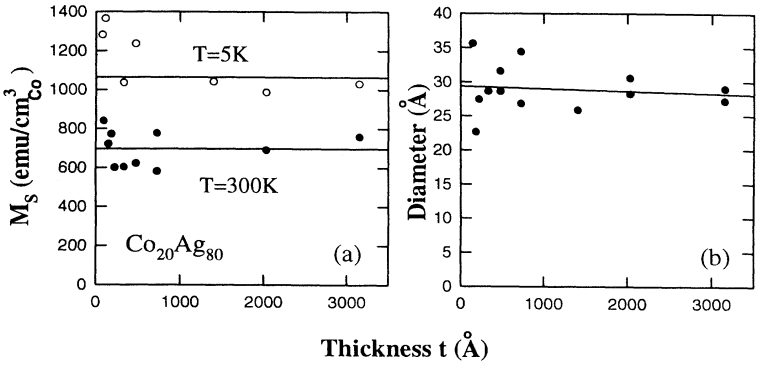

FIG. 3. Thickness dependence of (a) the spontaneous magnetization, M_s, and (b) the magnetic particle size for the $Co_{20}Ag_{80}$ series.

scattering centers, which not only affect electron mobility, but also randomize electron spin orientation. Often a phenomenological parameter ϵ is used to describe the probability of spin flipping in elastic SO scatterings,[7] $1/\epsilon$ being the average number of scattering events needed to cause spin randomization. For a thin film the spin-flipping rate, $1/\tau_{\uparrow\downarrow}$, can be separated into a bulk and a surface contribution:[7]

$$\frac{1}{\tau_{\uparrow\downarrow}} = \frac{\epsilon_B}{\tau_B} + \frac{\epsilon_S}{\tau_S} = \frac{\epsilon_B}{\tau_B} + \frac{3v_F\epsilon_S}{8t}. \quad (2)$$

In writing relation (2), we have used $1/\tau_S = 3v_F/8t$ from relation (1). Relation (2) has been confirmed in weak-localization experiments,[6,7] an efficient technique to measure $1/\tau_{\uparrow\downarrow}$. Lindelof and Wang[7] have found that on the surfaces of a thin film, ϵ_S is enhanced over ϵ_B by 1–2 orders of magnitude. In other words, spin flipping is much stronger at the surfaces than in the bulk.

Another important effect of a thin film is its geometrical confinement, which sets a physical limit to the electron MFP. The asymmetry in MFP ($\lambda_+ > \lambda_-$) is the origin of GMR. Because λ_+ is longer than λ_-, the geometrical confinement affects λ_+ preferentially. Therefore, it tends to reduce the asymmetry between λ_+ and λ_-, creating a negative impact on GMR. It should be pointed out that λ_+ and λ_- are related to magnetic scatterings only. Ordinary surfaces have little effect on them. However, for surfaces with large SO interactions, the strong spin-flipping effect does set a physical limit on MFP.

The MR of a magnetic system with spin-mixing effect can be generally described by a "two-current" model developed by Fert.[12] This model has explained the magnetotransport of some dilute and concentrated magnetic alloys.[12–14] According to this model, the H-induced resistivity is given by

$$\Delta\rho(H) = \rho_0 - \frac{\rho_\uparrow\rho_\downarrow + \rho_{\uparrow\downarrow}(\rho_\uparrow + \rho_\downarrow)}{\rho_\uparrow + \rho_\downarrow + 4\rho_{\uparrow\downarrow}}, \quad (3)$$

where $\rho_0 = m^*/2ne^2\tau_0$ is the zero-H resistivity, $\rho_\uparrow = m^*/ne^2\tau_+$ are $\rho_\downarrow = m^*/ne^2\tau_-$ are the resistivities of the two spin channels, and $\rho_{\uparrow\downarrow} = m^*/ne^2\tau_{\uparrow\downarrow}$ is due to the spin-mixing effect. We will extend this phenomenological model to account for the finite-size effect of GMR by including both the spin-mixing effect and the geometrical confinement. To do so we need to find three scattering rates, $1/\tau_\pm$ and $1/\tau_{\uparrow\downarrow}$. The t dependence of $1/\tau_{\uparrow\downarrow}$ has already been given by relation (2). As for $1/\tau_\pm$, their t dependence should include the geometrical confinement effect. In bulk samples, $1/\tau_\pm^B = (1/\tau_0)(1 \mp \Gamma)$, where Γ is a parameter characterizing the asymmetry of the two spin channels. The ratio of MFP's for bulk samples is $\lambda_+/\lambda_- = (1+\Gamma)/(1-\Gamma)$. For a thin film, $1/\tau_\pm$ also contains a contribution due to the geometrical confinement, i.e.,

$$\frac{1}{\tau_\pm} = \frac{1}{\tau_\pm{}^B} + \frac{1}{\tau_\pm{}^S} = \frac{1}{\tau_\pm{}^B} + \frac{3v_F\epsilon_S}{8t}. \quad (4)$$

The factor ϵ_S ensures that only the spin-flipping events on the surfaces can set a physical limit to λ_+ and λ_-.

According to relation (3), the t dependence of $\Delta\rho$ is caused by the finite-size effect on $1/\tau_\pm$ and $1/\tau_{\uparrow\downarrow}$. There are only two unknown parameters, ρ_0 and ϵ_S. All other parameters, n, v_F, Γ, and t, can be obtained from independent experiments. We have calculated the Fermi velocity, $v_F \approx 1.3 \times 10^8$ cm/s for $Co_{20}Ag_{80}$ and 1.2×10^8 cm/s for $Fe_{20}Ag_{80}$, from the carrier density n obtained from the Hall effect measurement. The parameter Γ was obtained by fitting MR data in bulk samples using a model developed for granular solids:[10] $\Gamma = 0.60$ for $Co_{20}Ag_{80}$ and 0.47 for $Fe_{20}Ag_{80}$. Based on Γ and the zero-H λ_{eff} (see Fig. 2), we have calculated the spin-dependent MFP: $\lambda_+ = 90$ Å and $\lambda_- = 23$ Å for bulk $Co_{20}Ag_{80}$; $\lambda_+ = 68$ Å and $\lambda_- = 25$ Å for bulk $Fe_{20}Ag_{80}$. With these parameters, we have used relations (3) and (4) to fit the data of the t dependence of MR shown in Fig. 1, where the best fits are represented by the solid lines. In $Co_{20}Ag_{80}$ system, the fitting quality is excellent, whereas in $Fe_{20}Ag_{80}$ system, it is satisfactory. The deviation in the latter system may be due to the fact that even at H=8 T the MR is not completely saturated in the $Fe_{20}Ag_{80}$ system.

The two parameters obtained from the fitting are $\rho_0 = 22.7$ $\mu\Omega$ cm, $\epsilon_S = 0.72$ for $Co_{20}Ag_{80}$ and $\rho_0 = 26.4$ $\mu\Omega$ cm, $\epsilon_S = 0.54$ for $Fe_{20}Ag_{80}$. The difference between the measured ρ_0 (27 and 31 $\mu\Omega$ cm, respectively) and the fitted ρ_0 is the nonmagnetic disorder resistivity. For both systems, ϵ_S is of the order of 60%. It is known that for bulk 3D solids the ratio between the spin-flipping and non-spin-flipping cross section is a few percent.[15] Therefore in granular $Co_{20}Ag_{80}$ and $Fe_{20}Ag_{80}$ the surface-

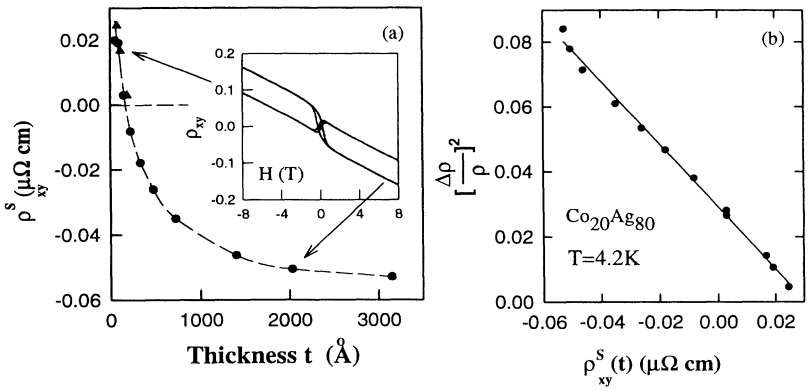

FIG. 4. (a) Thickness dependence of the spontaneous extraordinary Hall resistivity ρ_{xy}^{s} for the $Co_{20}Ag_{80}$ series. Also shown is the field dependence of the Hall resistivity ρ_{xy} in the thick- (2030 Å) and thin- (110 Å) film limit. (b) The linear correlation between $[\Delta\rho/\rho]^2$ and $\rho_{xy}^{s}(t)$.

induced spin-flipping rate is enhanced by about 1 order of magnitude, similar to weak-localization results of thin films.[7]

Our fitting results have revealed that the direct spin-mixing effect ($\rho_{\uparrow\downarrow}$) can only account for about 10% of the reduction in $\Delta\rho$ as t is reduced, the predominant cause ($> 90\%$) of the finite-size effect is the geometrical confinement in the context of strong surface-enhanced spin flippings. To date there has been no study on the effect of spin flipping on GMR in granular systems. In studies involving multilayers the role of spin-flipping is receiving increasing attention.[15,16] In the CIP (current in plane) geometry, it is believed that the effect of spin flipping is not important.[17] However, in the CPP (current perpendicular to plane) geometry, the important length scale is the spin-diffusion length,[15,16] ℓ_{sf}. GMR is caused by a spin-dependent interface resistance due to an imbalance of the number of + and − electrons within ℓ_{sf} of an interface. Our study here has shown that the role of spin flipping in granular systems is different from that in multilayers. The direct effect of spin flipping, in terms of mixing spin channels, is too small to be important. However, because of the enhanced spin flipping near the surfaces, the spin-dependent MFP's (λ_+ and λ_-) become geometrically limited, and the confinement is preferentially more significant on λ_+ than on λ_-. The finite-size effect of GMR is, therefore, the result of a gradual equalization of λ_+ and λ_-. The comparison between granular thin films and multilayers illustrates the significant disparity in the basic physics between the two systems.

Finally, we present another anomalous magnetotransport property: the finite-size effect of the extraordinary Hall effect (EHE). Figure 4(a) shows the t dependence of the spontaneous Hall resistivity ρ_{xy}^{s} due to EHE for the $Co_{20}Ag_{80}$ series measured at $T = 4.2$ K. The total Hall resistivity ρ_{xy} vs H is presented in the inset in Fig. 4(a) for two representative $Co_{20}Ag_{80}$ samples in the thick- and thin-film limits. The value of ρ_{xy}^{s} is obtained by extrapolating the ρ_{xy} (H) curve in the high H region to $H = 0$. Quite interestingly, ρ_{xy}^{s} is negative in the bulk limit, whereas it gradually reduces its magnitude and eventually turns positive as t approaches the thin-film limit. This shows that ρ_{xy}^{s} in thin films also includes a bulk and a surface contribution differing in sign. It is well known that a SO interaction induces EHE through two mechanisms:[18] skew scatterings or side jumps. In homogeneous systems, there exists a scaling relation[18] between ρ_{xy}^{s} and ρ, i.e., $\rho_{xy}^{s} \propto \rho^{n}$, $n = 1$ for skew scatterings and $n = 2$ for side jumps. A recent theory on EHE by Zhang[19] has shown the invalidity of the above scaling relation applied to heterogeneous systems. In the current $Co_{20}Ag_{80}$ series, the conventional theory of EHE fails. One observes in Fig. 4(a) that ρ_{xy}^{s}, similar to MR, exhibits a large finite-size effect and even changes sign at a certain t. However, ρ itself changes little over a large t range due to the short λ_{eff} (see Fig. 2). A careful examination has shown that there is a new scaling relation between the GMR quantity $\Delta\rho/\rho$ and ρ_{xy}^{s}. In Fig. 4(b), we plot $[\Delta\rho(t)/\rho]^2$ vs $\rho_{xy}^{s}(t)$. The observed linear correlation between $[\Delta\rho/\rho]^2$ and $\rho_{xy}^{s}(t)$ is highly unconventional. Although the exact mechanism is not clear at the moment, we believe that the surface-induced SO interaction is the common cause of the finite-size effect of both GMR and EHE. The new correlation implies convincingly that the traditional treatment of EHE is invalid in heterogeneous systems. It is imperative in theory to treat both GMR and EHE simultaneously and self-consistently.

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