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Thickness dependence of magneto-transport in Cu-Co granular thin films

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This work explores the thickness dependence of magneto-transport properties in $Cu_{80}Co_{20}$ granular thin films with different thickness. These results are compared with silver-based film series studied earlier. It was observed that the thickness dependence of the GMR effect was sensitive to the surface chemistry of the films. The extraordinary Hall effect (EHE) in these films was measured and found to be different from the Ag-based system. In the Cu-based system, the EHE is a weak function of film thickness over the range studied. When the variation of the spontaneous magnetization is taken into account the effective EHE has a universal thickness dependence. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1664171]

I. INTRODUCTION

Recent research interests of several groups have focused on examining magneto-transport properties in magnetic granular thin film.[1–5] The magneto-transport properties generally can be separated into longitudinal part and transverse part. The longitudinal resistivity corresponds to the giant magnetoresistance (GMR) effect,[1,2] which has drawn much attention. This is largely due to fundamental interest in the basic mechanisms of spin-dependent scattering responsible for the GMR effect and in using the GMR effect in technological applications, such as magnetic recording and nonvolatile memory devices. One often-neglected aspect is the transverse magneto-resistivity, i.e., the Hall effect in heterogeneous materials, partly due to the Hall effect being a second order scattering process. This makes the Hall effect inherently harder to understand.[3–5]

Since its discovery by Hall in 1879, the Hall effect was among the earliest physical phenomena to be applied to technological applications, such as sensors.[6–8] In heterogeneous magnetic materials, the Hall effect can be broken down into two parts:

$$\rho_{xy}=\rho_{xy}^{0}+\rho_{xy}^{S}, \tag{1}$$

where the first term, the normal Hall resistivity, ρ_{xy}^{0}, is the same as in other metallic systems and is a measure of carrier density of the material. The second term is the so-called extraordinary Hall resistivity, and, in magnetic materials, is given by[6]

$$\rho_{xy}^{S}=4\pi R_S M. \tag{2}$$

Thus, it is proportional to the magnetization. This describes the extraordinary Hall effect (EHE) and, in general, the EHE contains contributions from two different scattering mechanisms: (1) skew-scattering and (2) side-jump.[9] The extraordinary Hall coefficient, R_S, is dependent on the materials, microstructure, and dimensional constraints.

In an earlier work, an interesting correlation was uncovered between the EHE resistivity and dimensional confinement in Ag–Co granular films:[10] The EHE smoothly varied with the film thickness in the range of 10–200 nm. Also, the GMR effect varied with the film thickness, and the two effects have almost perfect square correlation with each other, indicative of side jump scattering mechanism.

This work reports a systematic study of magneto-transport with an emphasis on the EHE as a function of thickness for another granular series, Cu–Co.

II. EXPERIMENT

Thin films of $Co_{20}Cu_{80}$ were deposited with thickness values ranging from 10 to 150 nm on 3-in. silicon wafers by dc magnetron sputtering using a composite $Co_{20}Cu_{80}$ target, carried out under a base pressure about 1.4×10^{-7} Torr. Two series of films were made under identical conditions: For the first series no attention was paid to protection of the surfaces; for the second series, once films were sputtered they were placed in a desiccator and subsequently transported to cleanroom and patterned into Hall-bar using standard photolithography for magneto-transport measurement. Square samples for magnetic measurements were processed simultaneously. Magnetic properties were measured using a SQUID magnetometer (Quantum Design). Magneto-transport properties including the GMR effect and EHE were carried out in a Physical Property Measurement System. Hall resistance was

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obtained by measuring the transverse Hall voltage in magnetic fields ranging from −60 000 Oe to 60 000 Oe.

III. RESULTS AND DISCUSSION

The GMR effect for a thick film has a typical value of 7% at low measurement temperature, $T = 10$ K, in an applied field of 60 000 Oe. In the Hall effect, the linear normal Hall slope in the high field range ($>$3000 Oe) corresponding to a carrier density of $5\times10^{22}/\mathrm{cm}^3$, in agreement with the literature value for transition metals. From these measurements one can obtain another transport parameter, the electron mean free path (MFP), λ_{eff}, using the following formula:

$$\lambda_{\mathrm{eff}} = \hbar(3\pi^2)^{1/3}/(e^2 n^{2/3}\rho_{xx}). \tag{3}$$

It was determined that the MFP for thicker samples is 2.0 nm, comparable to the typical microscopic length scale. From magnetic measurements as reported previously,[11] the average particle size is about 3.0 nm. At 20% volume fraction, the average interparticle distance is comparable to the particle size. Thus, the measured mean free path is consistent with a microscopic picture of segregated magnetic phase with nanoscale phase boundaries limiting the scattering length scale.

Another pronounced feature in the measured Hall effect is the large value of the extraordinary Hall resistivity, ρ_{xy}^S, obtained by extrapolating the high-field linear portion to zero field: Cu-based granular films tend to have a large value of EHE compared to non-Cu based granular materials. For example, EHE for $Ag_{80}Co_{20}$ is 0.06 $\mu\Omega$ cm and comparable value of EHE was found for as-sputtered $Au_{80}Fe_{20}$.[10] The sign of the EHE is also dependent on the material type: Cu–Co, Fe–Ag, Fe–Au have positive values of EHE while Co–Ag has negative values.[3]

Comparison was made in the thickness dependence of magneto-transport properties. As for the GMR effect, initial study from a series of $Cu_{80}Co_{20}$ films was carried out and apparent similar thickness dependence on the thickness as Ag-based films was reported.[11] However, for this series, there was the possibility of surface oxidation, which could have enhanced the surface effects in thinner films and made the effective thickness thinner than the actual samples.

In the new series of granular films the samples were reasonably protected by coated photoresist. Such coating is known to protect metal films from oxidizing and was successfully used before.[10] Comparison of the GMR effect for the two series of $Cu_{80}Co_{20}$ films is shown in Fig. 1, and we observe that the GMR effect is relatively constant down to 12 nm, the thinnest samples studied in the new series. Thus, it appears that surface chemistry plays an important role here.

However, what is surprising is that the EHE effect is insensitive to the surface chemistry. For both series of films, the thickness dependencies of EHE are almost identical within the margin of errors of the measurements. Figure 2 shows the measured ρ_{xy}^S versus thickness for both series of the samples. It is apparent that two sets of data display little dependence on the surface conditions, and all data points practically follows a line of constant value of 0.21 $\mu\Omega$ cm.

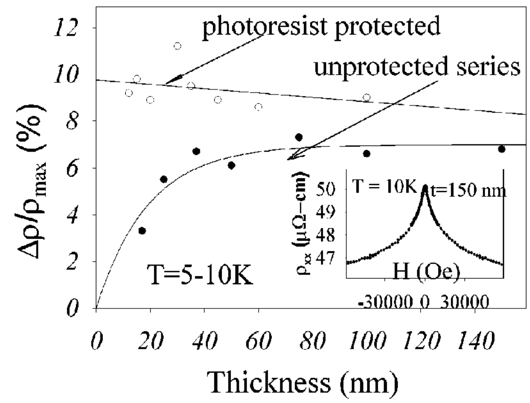

FIG. 1. Thickness dependence of the GMR effect for two series of $Cu_{80}Co_{20}$ films: Solid dots for a series without protection and subject to oxidation; open dots for the series with photoresist protection. Inset shows magnetoresistivity for a 150 nm thick film.

Representative Hall effect curves for samples of two limiting thickness values are also shown in the insets. The two arrows point to the corresponding values of ρ_{xy}^S for two different measurements on films with thickness values of 25 nm and 100 nm, respectively. The difference in low measuring temperatures should have negligible effect on the measured EHE results.

The thickness dependence of the EHE for the $Cu_{80}Co_{20}$ series displays a different thickness dependence compared to that of $Ag_{80}Co_{20}$. The extraordinary Hall resistivity of the $Cu_{80}Co_{20}$ series appears to be approximately constant throughout the range of thickness studied, whereas that of the $Ag_{80}Co_{20}$ series varied greatly with the thickness, even changing sign from a negative value in the thick limit to a positive value in the thin limit.[10] This apparent difference between Ag-based and Cu-based granular thin films can be understood in terms of the microstructure difference between the two systems.

In the Ag–Co series, it was found that the microstructure and magnetic properties remain unchanged as the thickness of the films varied. This was demonstrated by invariance of the measured particle size and spontaneous magnetization with the thickness. Thus, the variation in EHE with the film thickness can be entirely attributed to scattering from the surfaces of the films, i.e., the two surfaces of the films approach each other as the films become thinner. It can be concluded that the dimensional constraints on the EHE leads to an excess positive extraordinary Hall resistivity.

On the other hand, the apparent invariance of the extraordinary Hall resistivity with film thickness in the Cu–Co series is due to the presence of two conflicting factors, the effects of which tend to cancel each other. The first effect is the surface scattering just as in Ag–Co series. The second effect is the microstructure variation in the Cu–Co films as the thickness decreases. It was observed that, although the average particle size remained more or less constant as the thickness varied, the spontaneous magnetization, M_S, decreased with the thickness by a large fraction.[12] In fact, M_S reduces to less than half the value of the thicker films as the thickness approaches the thin limit, as shown in the inset of Fig. 3 and Ref. 12.

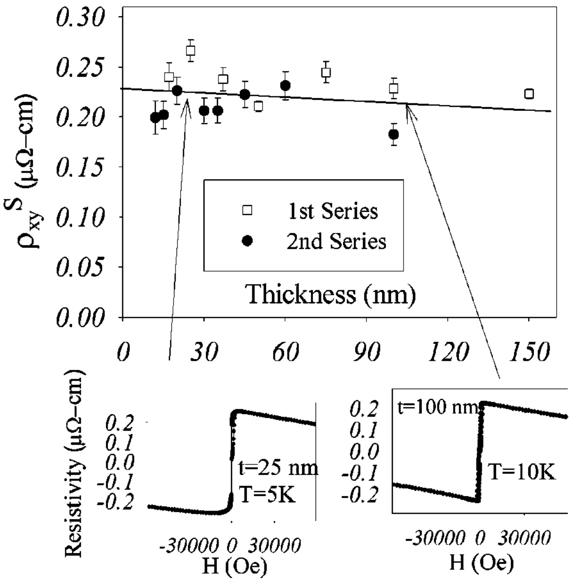

FIG. 2. Thickness dependence of extraordinary Hall resistivity for both series of $Cu_{80}Co_{20}$ films with and without photoresist protection. Insets show the corresponding Hall effect measurements for a thin (25 nm) film and a thicker (100 nm) film, respectively. The two arrows indicate the corresponding data points for the two samples.

The effect of such magnetization reduction can be understood by Eq. (2), showing the extraordinary Hall resistivity as proportional to M_S. Such variation in M_S should influence the thickness dependence of the EHE in the Cu–Co series. In particular, since the value of the extraordinary Hall coefficient, R_S, is positive in Cu–Co, as the thickness decreases, the bulk extraordinary Hall resistivitiy should decrease with the associate decrease in M_S, tending to reduce the EHE. On the other hand, if one assumes that the surface contribution to the EHE as a function of film thickness is universal for Ag-based and Cu-based films, one could expect that the thinner films tend to have more positive extraordinary Hall resistivity from surface scatterings. It is conceivable that the two contributions tend to cancel each other, resulting in relatively weak thickness dependence of the EHE.

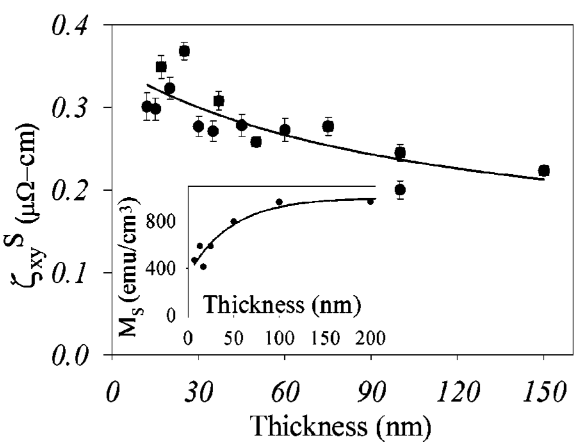

FIG. 3. Thickness dependence of the effective extraordinary Hall resistivity, $\zeta_{xy}^S(t)$, after correcting for the variation of spontaneous magnetization, M_S. Inset: variation of M_S versus thickness.

Assuming that R_S is only material dependent, i.e., it does not strongly depend on the associated microstructure changes as film thickness varies, then, the thickness dependence of ρ_{xy}^S can be written as

$$\rho_{xy}^S(t)=R_S[M_S^B-\Delta M_S(t)]+S(t). \tag{4}$$

In this equation, $\Delta M_S(t)$ is the net reduction of spontaneous magnetization, M_S^B is the bulk spontaneous magnetization for the granular films, and $S(t)$ is the universal surface contribution to the extraordinary Hall resistivity, as functions of thickness, t. The present results show that $\rho_{xy}^S(t)$ is almost independent of t, and one can set it equal to the bulk value, $R_S M_S^B$. Then, we conclude that if the microstructure does not change with the film thickness, the effective extraordinary Hall resistivity, $\zeta_{xy}^S(t)$, should be further increased with decreasing thickness as

$$\zeta_{xy}^S(t)=R_S M_S^B+S(t)=R_S M_S^B\left(1+\frac{\Delta M_S(t)}{M_S^B}\right). \tag{5}$$

Such thickness dependence can be derived from the variation of spontaneous magnetization with film thickness. For the Cu–Co films examined in this study, the bulk value of M_S is 1000 emu at the cryogenic temperature, and the function $\Delta M_S(t)$ can be derived from the interpolation of the measured data set. The effective EHE result thus derived is plotted in Fig. 3, and reveals a similar thickness dependence of the extraordinary Hall resistivity on the thickness in $Cu_{80}Co_{20}$ as in $Ag_{80}Co_{20}$. It is likely for the mechanism of the surface induced EHE to be side jump.

ACKNOWLEDGMENT

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