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2009 J. Phys.: Conf. Ser. 150 042063

(http://iopscience.iop.org/1742-6596/150/4/042063)

Magneto-transport flipping induced by surface oxidation in Co films

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Abstract. We have performed low temperature magnetoresistance (MR) and magnetization measurements for a series of thin Co films with different degrees of oxidation to investigate the role of surface oxidation on magnetic properties. For films with less oxidation, magneto-transport exhibits the typical anisotropic magnetoresistance behavior. However, magneto-transport flips completely in films with severe oxidation. The inverse MR is replaced by the normal MR when the applied magnetic field is along the current. The normal MR is changed to the inverse MR when the applied magnetic field is perpendicular to the current. Since the disorder will enhance the spin-orbital interaction and electron-electron interaction in thin films, we suggest that both mechanisms may be responsible for the flipping in anisotropic MR induced by surface oxidation.

1. Introduction

Giant magnetoresistance (GMR) has been extensively studied for almost two decades and is applied nowadays in several devices.[1] It has been well established that GMR arises from the spin-dependent scattering of conduction electrons in the bulk of ferromagnetic layers. Earlier works in multilayers $[F1/N/F2/N]_n$ showed that doping the ferromagnetic layers with a few percentage of impurities can affect GMR and even change its sign from negative (normal GMR) to positive (inverse GMR).[2, 3] Recently, the inverse GMR has been observed in spin-valve, tunnel junction, and binary multilayer structures.[4, 5, 6, 7, 8, 9, 10] The spin-asymmetry of bulk and interface have been taken a great deal of consideration. However, the contribution of anisotropic magnetoresistance (AMR) due to the s-d scattering has been rarely paid attention in the analysis because that it is the secondary to MR in those systems. As known, AMR plays an important role in the magneto-transport of a single ferromagnetic layer. Resistance is a maximum when all magnetic moments are along the current direction while it is a minimum when all moments are perpendicular to the current direction. In this paper, we report the flipping of MR occurs by the introduction of surface oxidation in thin Co films where AMR is the dominant mechanism.

2. Experimental details

Several 10nm thick Co thin films are fabricated on naturally oxidized (100) Si substrates using a DC sputtering system. The sputtering system is evacuated below 3×10^{-6} Torr before sputtering Co in 7.4mTorr of argon. The deposition rate is about 0.077nm/s. It has been known that Co is easy to oxide in air. By controlling the exposure time ranging from 5 to 50 hours in air for the sample, different degrees of oxidation and disorder of the sample can be achieved. Magnetization and magneto-transport measurements are performed at low temperatures. The former is obtained using a superconducting quantum interference device in PPMS. The latter is performed using a standard four-terminal ac lock-in technique in a pumped ^{4}He cryostat equipped with a 9 Tesla superconducting magnet. The sample is in a rectangular shape of 12mm×1mm and the measuring current always flows along the long axis. During MR measurements, the applied magnetic field is always oriented in the film plane and can be either parallel or perpendicular to the current direction.

3. Results and Analysis

All Co films exhibit weakly disordered behavior; the resistance increases with decreasing temperature at low temperatures. The temperature dependent sheet conductance below 20K of all films can be well described by two-dimensional (2D) weakly disordered theorem and demonstrate that $\Delta G \propto A\frac{e^2}{2\pi\hbar^2}\ell n(T)$. This behavior stems from disorder enhanced electron-electron interaction and quantum interference effects in 2D.[11] A's obtained from the linear fit of data to the above predicted form are about 1 in consistence with most 2D homogeneous thin films. Films with severe oxidation have larger slope (A) and sheet resistance than slightly oxidative samples. For all our thin Co films, sheet resistance at 15K ranges from 40 to 120 Ω and A ranges from 1.2 to 2.

For most single magnetic materials, a positive MR occurs when the magnetic field is applied along the current (I||H) while a negative MR occurs when the magnetic field is applied perpendicular to the current(I⊥H). ΔR(H) defined as R(H)−R($\mathrm{H_{saturation}}$) at both configurations, I||H and I⊥H, of a less oxidized Co film is plotted in Fig.1(a). This scenario can be attributed to the s-d scattering effect which is usually referred as AMR. Based on the AMR effect, resistance change is proportional to $\cos^2\alpha$ where α is the relative angle between magnetization and the measuring current. When the magnetic field forces all moments along the current there is a largest resistance at saturation. As the field is reduced, some moments start to deviate from the current direction and resistance decreases. Resistance reaches a minimum when field is the coercive field in the opposite direction. In contrast, when the magnetic field forces all moments perpendicular to the current there is a smallest resistance at saturation. As the field is reduced, some moments rotate to lie with the current direction and resistance increases. Moreover, hysteretic MR curves demonstrate that a thin anti-ferromagnetic CoO layer is present serving as a pinned layer and the pinned direction is along the short axis of the film. A MR curve shift by a magnetic field ($\sim$ 800Oe) is clearly seen because Co film is exchange biased by CoO layer for I⊥H configuration. Coercive field of Co film along the short axis is much larger than that along the long axis.

In a thin magnetic film, surface oxidation can become very severe and affect its intrinsic properties. For comparison, we plot MR curves for one severely oxidized Co film in Fig.1(b). What a surprise, MRs for both configurations likely to switch compared with Fig.1(a). Magneto-transport behavior flips. A negative MR occurs when current is parallel with magnetic field direction while a positive MR occurs when current is perpendicular to field direction. CoO layer still provides a strong pinning along the short axis resulting in a MR shift for I⊥H configuration. It has been known that coercive field is smaller for thicker films. Both MR and magnetization data confirm that coercive field is slightly larger for severely oxidized samples implying Co layer is slightly thinner due to oxidation.

For weakly disordered thin films, some may argue that quantum interference effects including electron-electron interaction are very important for transport behavior such as the ℓn(T) dependence of conductance. Positive MR appears due to the quantum interference effect even though the applied magnetic field is in the film plane. The size of positive MR decreases with increasing temperature. Therefore, we analyze data at T=15K to reduce the quantum

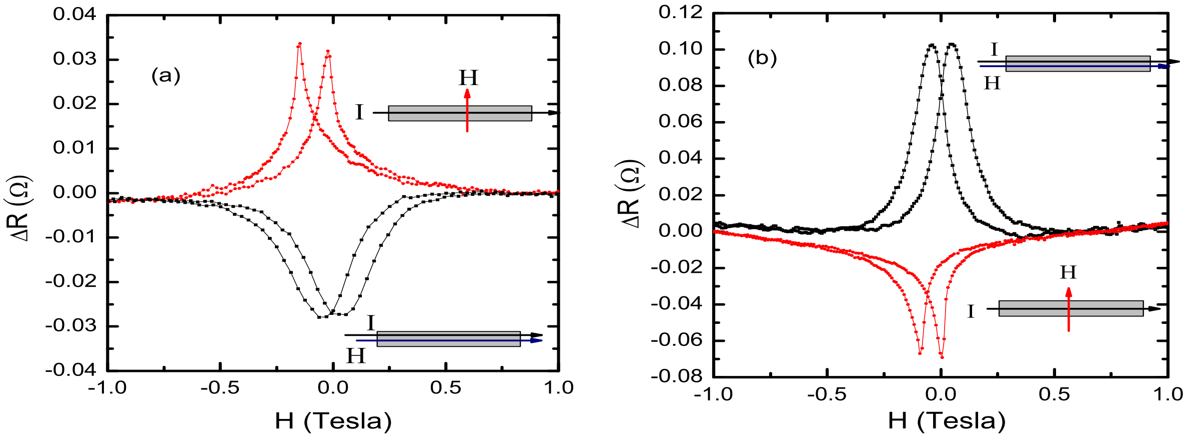

Figure 1. ΔR versus the applied magnetic field H for two Co films, (a) less oxidized one with R_{sheet}=54.8Ω and (b) severely oxidized one with R_{sheet}=117Ω, at T=15K. Black squares and red circles represent that the magnetic field is applied parallel and perpendicular to the current, respectively.

interference effect. However, our data indeed show that there is a small symmetric positive MR in all curves. It is more profound in more disordered films. For highly disordered film of R_{sheet}=117Ω shown in Fig.1(b) this positive MR background is clearly observed but the magnitude is quite small. Therefore, clear hysteresis MR results mainly from magnetic domain induced spin-dependent scattering.

We summarize magneto-transport results of all samples in Fig.2. For a negative MR, we characterize the behavior by a positive ΔR_{peak} which is defined as the maximum value R_{peak} subtracts the minimum value $R(H_{saturation})$. On the other hand, we characterize the positive MR by a negative ΔR_{dip} which is defined as the minimum value R_{dip} subtracts the maximum value $R(H_{saturation})$. As shown in Fig.2, for I$\|$H configuration, a positive MR and a negative ΔR_{dip} appear in the less oxidized Co films. With increasing disorder (R_{sheet}), the magnitude of ΔR_{dip} shrinks to zero and then MR behavior flips from a positive MR to a negative MR. In contrast, for I$\perp$H configuration, a negative MR and a positive ΔR_{peak} appear in the less oxidized Co films. With increasing disorder (R_{sheet}), the magnitude of ΔR_{peak} shrinks to zero and then MR behavior flips from a negative MR to a positive MR. The flipping of MR behaviors for both directions occurs around $R_{sheet} \sim 70\Omega$. Three less disordered samples with $R_{sheet} \leq 70\Omega$ demonstrate the ordinary anisotropic MR behaviors. Other more disordered samples with $R_{sheet} \geq 70\Omega$ demonstrate completely opposite behaviors, inverse AMR.

Earlier works in magnetic multi-layers and exchange biased spin valves explored that MR can be inverse from a negative MR to a positive MR by changing the sign of spin asymmetry coefficients (β or γ) of either bulk or interfacial layer.[2, 3, 5, 6, 10] In their systems, it is caused by GMR rather than AMR mechanism because that their MR is independent of the relative angle between the applied magnetic field and the current. Our system is unlike theirs. Instead, AMR is the dominant mechanism in our thin Co films. No matter how long the sample exposed to air, there always exists a thin layer of CoO atop. The oxidized layer CoO is an antiferromagnetic material that may provides exchange coupling with the rest ferromagnetic Co layer. Since the MR shift by a magnetic field is clearly present at I$\perp$H configuration, the antiparallel coupling is along the short axis of the film. Although the exposure time in air does not change much in thickness and magnetic properties of CoO, it indeed promote the degree of disorder of the Co film. As to the flipping in AMR induced by surface oxidation, there are two possible mechanisms. One is that the enhanced spin-orbital interactions arose from disorder influence the original s-d

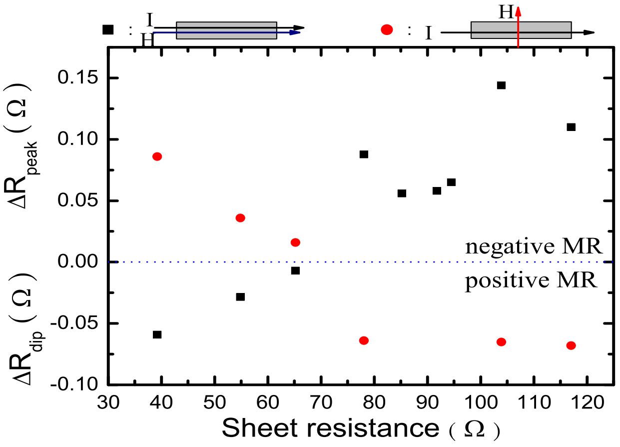

Figure 2. ΔR (peak or dip) versus sheet resistance at T=15K for a series of films. Red circles : The magnetic field is applied perpendicular to the current. Black squares: The magnetic field is applied parallel with the current.

scattering of ferromagnet which is responsible for AMR. In 2D homogeneous thin films, spin-orbital scattering can be increased by the introduction of impurity (disorder). The other is that the disorder enhanced electron-electron interactions cause the asymmetric density of state change resulting in the sign change of spin polarization. A Coulomb cusp in the tunneling density of state near the Fermi energy always appears in disordered samples.

4. Conclusions

Co thin films are fabricated by DC sputtering. The typical AMR behavior is observed in less disordered Co films. By keeping sample in air to increase surface oxidation, the film becomes more disorder and correspondingly, MR behavior can flips. The evolution of MR at both direction (I||H and I⊥H) configurations versus disorder is clearly displayed to show the occurrence of the flipping. We suggest that disorder enhanced spin-orbital and electron-electron interactions may be responsible for the flipping in AMR.

Acknowledgments

Magnetization measurements are made in superconducting and magnetism laboratory of Dr. J.Y. Lin. This work was supported by NSC grant in Taiwan under project No NSC96-2112-M-009-030-MY3 and MOE ATU program.

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