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Enhanced planar Hall voltage changes measured in Co/Cu multilayers and Co films with square shapes

C Christides†, S Stavroyiannis and D Niarchos
Institute of Materials Science, NCSR 'Demokritos', 153 10 Agia Paraskevi, Attiki, Greece

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Abstract. Enhanced planar Hall voltage changes were measured for a cross-diagonal current–voltage configuration in [Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ multilayers deposited on square 1 cm^2 Si(100) substrates by magnetron sputtering. A $(\Delta V/V_{peak}) = 4000\%$ voltage maximum change is observed with an $I_{ext} = 1$ mA current flow along one diagonal of the square and the voltage output measured along the other diagonal, for external magnetic fields H_{ext} of ±30 Oe applied in the film plane perpendicular to the edges. This result is discussed in terms of known galvanomagnetic effects in ferromagnetic thin films used in sensors.

1. Introduction

The observation of the giant-magnetoresistance (GMR) effect [1], in multilayers (MLs) or granular thin films of ferromagnetic/non-magnetic (FM/NM) elements, and the change in Ni/Co MLs recently reported [2] as 'extraordinary' anisotropic magnetoresistance (AMR), open the way for use of high-performance materials in applications of active sensors [3]. The different physical origins of GMR and the 'ordinary' AMR effect leads to different behaviour with magnetic field. The AMR is defined as $\Delta\rho = \rho_\parallel - \rho_\perp$, where $\rho_\parallel$ and $\rho_\perp$ are the saturation resistivities with $I_{ext} \parallel H_{ext}$ and $I_{ext} \perp H_{ext}$ respectively, while the GMR is defined as $(R_{max} - R_s)/R_s$ with R_{max} and R_s the resistance when the film magnetization is zero and maximum respectively.

All the FM films [4], including those exhibiting GMR, present the AMR effect with $\Delta\rho/\rho$ values from slightly different to zero up to 3–5% in permalloy or Ni films. This AMR effect is small compared to the GMR effect, that is of the order of 10–60% at RT. However, the main difference between the magnetotransport properties of most FM systems, such as Ni/Co MLs, and those exhibiting the GMR effect is that for the first the MR with $I_{ext} \parallel H_{ext}$ is positive and the MR with $I_{ext} \perp H_{ext}$ is negative while for the second both MR quantities are negative. In addition, GMR MLs present well defined maxima and minima of the GMR effect as a function of layer thickness of the non-magnetic element that correspond to antiferromagnetic (AF) and FM arrangements of magnetic moments in adjacent magnetic layers respectively. Oscillations were observed as well in resistivity and AMR of epitaxial Ni/Co(111) MLs [5], that do not exhibit the GMR effect, as a function of Ni and Co thicknesses; these were attributed to superlattice effects. However, since the GMR and AMR effects have been studied extensively in ultrathin FM/NM MLs, so far there is very little published work concerning the Hall effect in these new structures. Recent measurements of

† e-mail address: christos@cyclades.nrcps.ariadne-t.gr

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the Hall resistivity ρ_H in Fe/Cr [6] and Co(Fe)/Cu [7] MLs show an oscillatory dependence of the spontaneous Hall coefficient R_H on the Cr and Cu layer thicknesses, similar to the observed oscillations in their GMR effect, which remain unexplained so far. Very recently some more controversial results of Hall effect measurements were published in Ni/Co MLs [2, 9]. These MLs exhibit large AMR values for low saturation fields that make them possible candidates for use as MR recording sensors [8]. Hall effect measurements of Ni/Co sputtered MLs with a four-probe cross-diagonal V_H–I configuration, for films with circular shape, led to observation of a so called 'extraordinary' AMR effect of 140% at RT [2, 9]. The Ni/Co interfaces were considered to be responsible for the enhancement of this effect [10] relative to single-layer films, but the mechanism for the high-aspect-ratio achievement is unclear. Since Hall effect measurements in van der Pauw structures are sensitive to sample geometry, contact size and alignment effects [11], first of all one has to clarify whether this 'extraordinary' AMR effect is a new physical phenomenon or a geometrical effect due to the specific measurement configuration used. In order to answer these questions, it will be instructive to consider what is known in single-layer FM films.

In FM films with thickness d, when a current I passes through a sample in the presence of a magnetic field, a potential is developed in the direction perpendicular to the current flow. This Hall voltage is given by [12]

$$V_H = \frac{R_s I}{d} M_s \cos\theta + \frac{kI}{d} M_s^2 \sin^2\theta \sin 2\phi \qquad (1)$$

assuming that the Hall voltage due to the Lorentz force is negligibly small in magnetic metals [13]. The first term is the spontaneous (or extraordinary) transverse V_H^s voltage which arises from asymmetric scattering of the conduction electrons from the magnetic moments in the sample and is proportional to the $M_s \cos\theta$ component in the film normal direction z. The second term is the planar (or pseudo-Hall) V_H^p voltage [14], which arises because the electric field E and current density J are not always parallel in the plane of the film. Thus, if there is a component of magnetization, $M_s \sin\theta$, in the plane of the film, then because of AMR ($\Delta\rho/\rho \propto \kappa M_s^2 \sin^2\theta$), the equipotentials may not be perpendicular to the current, and a Hall voltage is detected that exhibits a maximum when the angle ϕ between I and M_s is 45°. Since both terms in (1) are inversely proportional to d, the Hall effect is a very sensitive method of detecting the magnetization of thin films. It is well known that the direction of M_s in thin films is strongly affected by shape anisotropy. However, for circular or square films with sizes greater than a few square millimetres the direction of M_s, away from the edges, is determined from the intrinsic anisotropy and texture of the deposited film. Therefore, in galvanomagnetic measurements the film (and not the geometrically induced) properties can be seen in relatively large substrates. A geometrical form with a cross shape of four equal arms is frequently used in galvanomagnetic devices, based on permalloy, for pseudo-Hall effect (PHE) measurements [13]. This cross configuration can be well approximated with a square shaped film when the I flows along one diagonal while the V_H is measured along the other diagonal.

In this work we report MR and V_H versus H (V_H–H) hysteresis loops along the edges and square diagonals of a [Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ ML film and of a Co single-layer, 100 nm thick film, deposited on square 1 cm^2 Si(100) and MgO(100) substrates respectively by magnetron sputtering. Our purpose is to investigate whether the results published by Prados *et al* [2] are due to the specific diagonal configuration used or are intrinsic to Co/Ni MLs only.

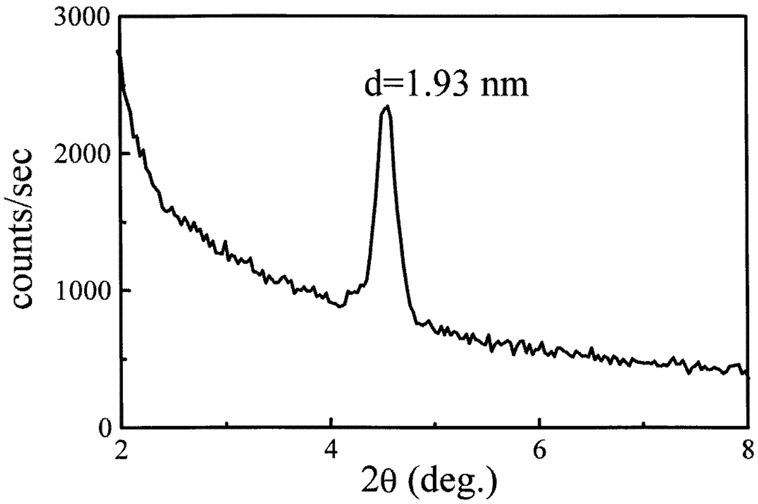

Figure 1. The low-angle x-ray pattern of the Si(100)/[Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ MLs, indicating the Co/Cu bilayer thickness observed at the first superstructure Bragg peak.

2. Experimental details

Metallic discs with diameter 5 cm and 99.99% pure elements were used as target materials in a high-vacuum Edwards E360A sputtering system with a cluster of Atom Tech 320-SE planar magnetron sputter sources. The samples were deposited, in a cryogenically pumped chamber with base pressure of 6×10^{-7} Torr, under an Ar (99.999% pure) pressure of 3 mTorr. An rf magnetron gun operating at 30 W with a deposition rate of 0.09 nm s^{-1} for Co and dc sputtering at 5 W resulting in 0.05 nm s^{-1} Cu was used. X-ray diffraction measurements, performed with Cu Kα radiation in a Siemens D500 diffractometer, reveal the superlattice Bragg reflections indicative of the ML Co/Cu structure (figure 1) at low angles and the fcc (111) preferred orientation at higher angles while the Co single layer is found to be hcp. It is worth noting that the used XRD set-up was not sufficient to resolve the expected reflectivity fringes from the multilayered structure. However, cross-section transmission electron microscopy (TEM) micrographs reveal the alternating stacking of the Co/Cu layers. Magnetic hysteresis loops were measured at 300 K with a Quantum Design MPMSR2 superconducting quantum interference device (SQUID) magnetometer. The isothermal magnetic measurements were recorded in the field range ±3 T and the displayed magnetic curves are normalized to the maximum $M_{max}(H)$ value obtained at $H = 3$ T. MR measurements were performed at RT with the van der Pauw method for the configurations shown in figure 2. The size ratio of the In contacts to film dimensions was less than 10^{-4} and the contacts were perfectly aligned in the corners of the square diagonal. The MR values were estimated by first subtracting from $V(H)$ and then dividing by the voltage observed at the peak position. All measurements were performed at room temperature by first applying the maximum positive field H parallel to the film plane and then finishing the loop.

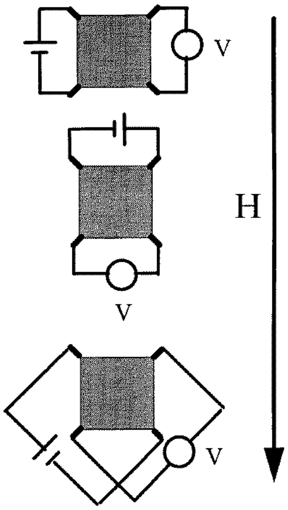

Figure 2. The three different configurations that were used in the MR measurements. From top to bottom is MR for $I_{ext} \parallel H$, MR for $I_{ext} \perp H$ and diagonal MR with the H direction forming an angle $\phi = 45^\circ$ relative to the square diagonals.

3. Experimental results

Figure 3 shows the MR curves of Si(100)/[Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ MLs observed with $I_{ext} = 1$ mA for $I_{ext} \parallel H_{ext}$ and $I_{ext} \perp H_{ext}$ and the M/M_s versus H hysteresis loops with the field direction parallel or vertical to film plane. The hysteresis loops are evident for the FM arrangement of the Co layer magnetic moments, even at zero applied field, with the average magnetization lying mainly in the film plane. The observed $M_r/M_s(\parallel) = 0.6$ value at the remanence point ($H = 0$) is more than the estimated average residual magnetization of $0.5M_s$ for a stressed material with an isotropic distribution of easy axes and closer to $\frac{2}{3} \times 0.832M_s$ for cubic anisotropy with $\langle 100 \rangle$ easy axes or to $\frac{3}{4} \times 0.866M_s$ for cubic anisotropy with $\langle 111 \rangle$ easy axes [15]. It is evident that the coercive field $H_c = 6$ and 50 Oe, for the parallel and vertical field configuration respectively, is different from the $H_{peak} = \pm 12$ Oe in the MR measurements. These AMR measurements are independent of the magnitude of I_{ext}. In figure 4 are plotted as a function of I_{ext} the V_H–H loops that are measured with the current flow applied along one diagonal and the voltage drop detected in the other diagonal in a field forming an angle ϕ of 45° relative to square diagonals. It is shown that the measured voltage along the diagonal is an order of magnitude less than that measured in AMR loops for $I = 1$ mA, while at $\pm H_{peak}$ the voltage approaches zero. This low V_{peak} value gives the $\Delta V/V_{peak} \approx 4000\%$ effect, which represents the order of magnitude voltage change between the saturating and switching field magnetic moment configurations. The observed V_H effect is found to (i) decrease on increasing I_{ext} and (ii) decrease from a maximum at $\phi = 45^\circ$ to $\phi = 0^\circ$. The 0.7 nm Cu layer thickness in the MLs corresponds to the first GMR minimum (FM configuration) located prior to the observed first AF GMR maximum at 0.9 nm [1]. It is worth noting that our Co/Cu MLs do not exhibit a GMR effect for the Cu thickness corresponding to the first AF maximum (0.7 nm $< t_{Cu} <$ 1.1 nm) because during film deposition there possibly takes place the formation of pinholes, that bridge adjacent Co layers, resulting in FM alignment of Co layer moments. Remarkably, the measured AMR and V_H values fall into the noise level signal for Cu layer thicknesses

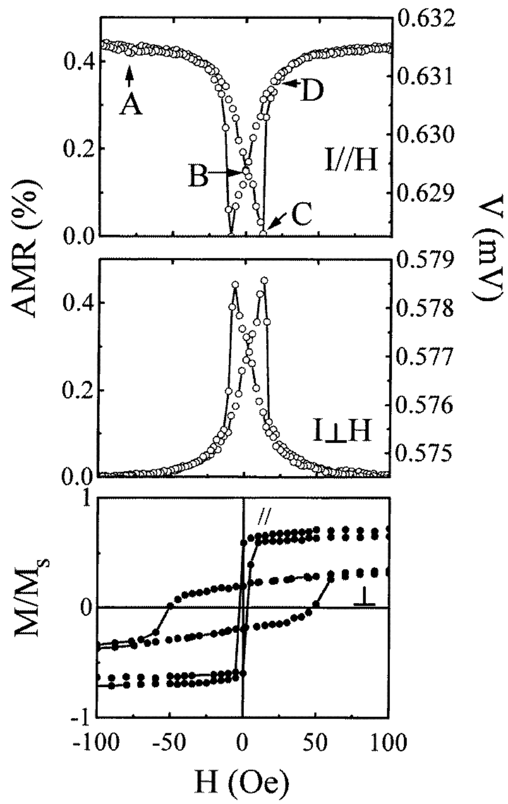

Figure 3. AMR and magnetic hysteresis loops for the Si(100)/[Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ MLs observed at RT with $I_{ext} = 1$ mA.

more than ~1.1 nm that coincide with the limit where pinhole concentration is decreasing. Since a clear AMR signal has been measured in single-layer FM films, FM/FM′ MLs, such as Ni/Co, and FM/NM$_p$ MLs, with NM$_p$ =Pd and Pt elements [16] that exhibit a large induced magnetic moment, then the observed AMR in Co/Cu MLs for $t_{Cu} < 1.1$ nm might be associated with the reported d-shell spin polarization of Cu atoms observed from x-ray magnetic circular dichroism measurements [17]. The induced Cu spin moment is found to be primarily situated at the Co/Cu interfaces, which consist of two or three atomic layers of Cu, that corresponds to 0.4–0.6 nm thickness, and its average moment is shown to fall off inversely with t_{Cu}. At the second AF GMR maximum, for 2.1 nm Cu thickness, a GMR effect of 15% was observed and it is found to remain unchanged in measurements performed with the three configurations shown in figure 2. It is worth noting that for Cu thicknesses less than 1 nm the diagonal ΔV_H effect has been found to vary from 400% up to 4000% for $I_{ext} = 1$ mA, and this variation was strongly dependent from the alignment, along the square diagonal, of the indium contacts used to connect the four Cu leads at the corners of the films.

To examine the dependence of the observed extraordinary MR effect upon the electronic scattering at Co/Cu interfaces a single layer of MgO(100)/[Co (100 nm)] film, grown under exactly the same conditions, has been measured in the three configurations as well. Figure 5 shows the MR curves observed with $I_{ext} = 1$ mA for $I_{ext} \parallel H_{ext}$ and $I_{ext} \perp H_{ext}$ and the

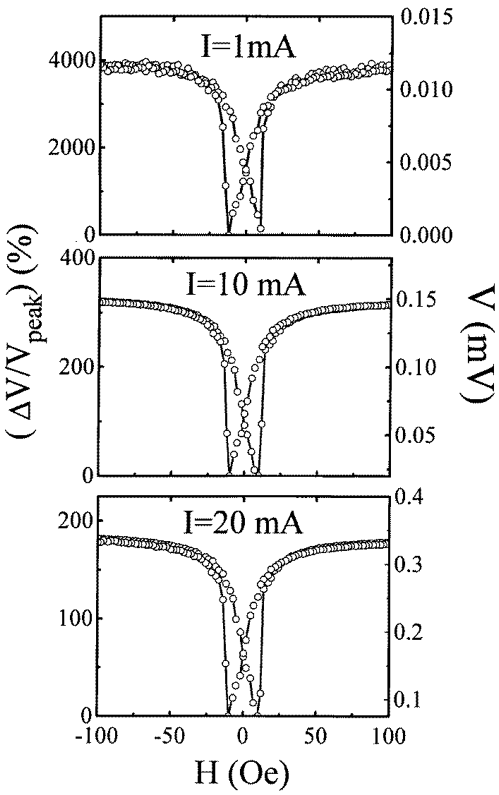

Figure 4. The PHE effect of Si(100)/[Co (1.2 nm)/Cu (0.7 nm)]$_{30}$ MLs observed for three different currents.

M/M_s versus H hysteresis loops with the field direction parallel or vertical to the film plane. It is clear from the hysteresis loops that the magnetization vector is in the film plane and a strong anisotropy is opposed to rotation of the magnetization out of the film plane. The observed $H_c = 80$ and 900 Oe, for the parallel and vertical field configuration respectively, is much larger than the $H_{peak} = \pm 8$ Oe in the MR measurements. In figure 6 are plotted the V_H–H loops as a function of three different I_{ext} values, that were measured with the current flow applied along one diagonal and the voltage drop detected in the other diagonal in a field forming an angle ϕ of 45° relative to square diagonals. For comparison similar measurements were performed in an $Ni_{81}Fe_{19}$ single layer with thickness 100 nm. MR curves with similar shapes as in the Co single layer were observed, but with an AMR effect of 2.5% and $H_{peak} = \pm 4$ Oe, for the $I_{ext} \parallel H_{ext}$ and $I_{ext} \perp H_{ext}$ configurations. In the diagonal configuration an effect of 170% was measured for $I_{ext} = 1$ mA. Although the Co single layer shows an AMR of 0.6% the corresponding diagonal effect is much larger than that of the $Ni_{81}Fe_{19}$ single layer. Thus, the observed diagonal voltage drop implies that in single layers this effect is enhanced for systems which exhibit larger M_s. Since our M–H loops are indicative that M_s is lying in the film plane, then the first term in (1) will vanish and the V_H–H loop may arise from a PHE. The shapes of these loops present similarities with loops first observed in $Ni_{90}Fe_{10}$ films that exhibit pure PHE [18].

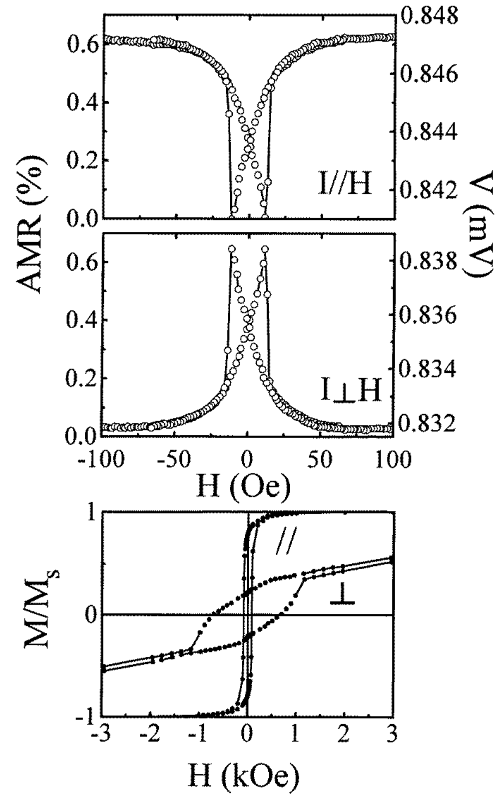

Figure 5. AMR and magnetic hysteresis loops for the MgO(100)/[Co (100 nm)] single layer observed at RT with $I_{ext} = 1$ mA.

4. Discussion and conclusions

The observed V_H–H loops, for both Co/Cu MLs and Co or NiFe single layers, indicate that a PHE may determine their properties. Strong evidence for this is the observed angular dependence of the V_H–H loop on sin 2ϕ, exhibiting a maximum for $\phi = 45°$, the angle between the I and H directions, and minima (~0) for $\phi = 0$ and 90°. In these loops the magnetic moments are aligned along the H direction because the V_H–H scans start from saturation. Two important features in these experimental results need an explanation.

(i) The H_c values observed from magnetic hysteresis loops are different from H_{peak} values in AMR and V_H–H loop measurements. However, the H_{peak} values obtained in AMR and V_H–H loops are identical. Since the demagnetizing field away from the edges of a film with thickness d and width w is [13] $H_D \propto M_s d/w$, in our samples H_D is negligible for planar film measurements. Thus, because the electronic mean free path in FM films is of the order of ~10 nm, it is expected that the switching field, where the magnetization starts to flip over the *local* easy axis direction in every domain, will be sensed by the MR and PHE signal. On the other hand, the H_c field, detected with DC magnetic loops, corresponds to an emerging magnetic domain distribution after a rotation and/or domain wall displacement process that leads to a zero macroscopic magnetization. It has already been pointed out [12] that even for coherent rotation of magnetization the coercivity measured by the Hall effect

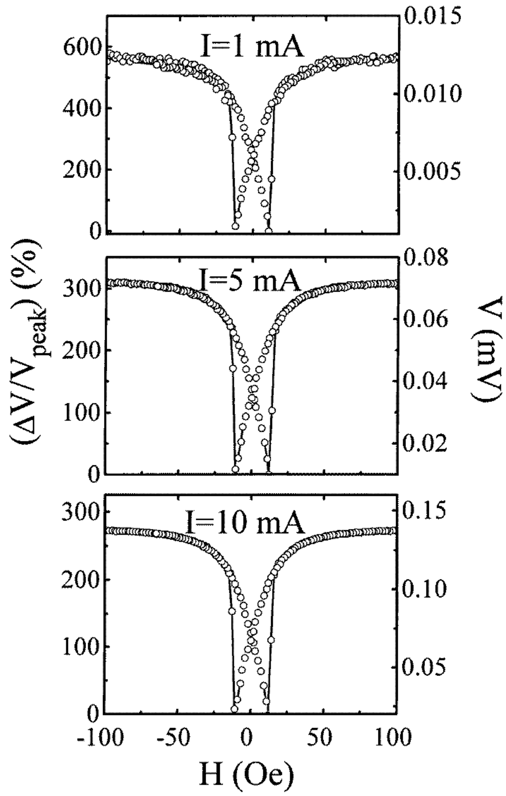

Figure 6. The PHE effect of an MgO(100)/[Co (100 nm)] single layer observed for three different currents.

is not the same as that measured with a magnetometer. Therefore, the specific distribution of easy axis directions in these films may cause the observed $H_c \neq H_{peak}$. According to this explanation, in the top panel of figure 3 the branch ABC corresponds to reversible magnetization rotation within a domain and at C (the reverse magnetic field) a jump of the magnetization occurs, which results in a sharp change of the AMR and V_H curves. Point D indicates where the reversible M rotation starts again in the opposite direction of the magnetic vector. The fact that the flip over of vector M does not occur at a unique value of H applied, but in some range ΔH, may be explained by nonuniform rotation processes in addition to uniform magnetization rotation [18]. However, in figures 5 and 6 it is shown that for the Co film the flip over jump gives a steeper galvanomagnetic effect.

(ii) The most significant issue now is the voltage drop variation in the V_H–H loops for the diagonal configuration. The observed sensitivity to current flow density and alignment of electric contacts at first glance gives the impression that there is an artefact due to high sensitivity of Hall measurements from these effects. Since the contact sizes are negligibly small compared to the film surface, the observed changes of the $\Delta V_H/V_{peak}$ percentage from their alignment along the square diagonals can be attributed only to deviations from the $\phi = 45^\circ$ condition, for the angle between I and H, that gives the maximum effect. On the other hand, the observed decrease of the $\Delta V_H/V_{peak}$ percentage with increasing current intensity can be related to an increase in density of the electric field E dynamic lines along the current direction due to a change of equipotential lines. This usually causes a larger electric field gradient in the current flow direction along the square diagonal. Therefore, because the PHE is basically an anisotropic MR effect for $\phi = 45^{\circ}$, in an FM film with larger average domain sizes relative to the electronic mean path no decrease of $\Delta V_H/V_{peak}$ on increasing I is expected, unless electronic scattering at grain boundaries and interfaces is significant. In this case, on increasing I the net resistance from boundary scattering is increasing as well. To the best of our knowledge, there are no available experimental data describing the effect on resistivity of electronic scattering at magnetic grain boundaries in the presence of an external field H. Therefore, it can be speculated that a combination of the AMR effect with grain boundary scattering can cause the observed decrease of the $\Delta V/V_{peak}$ ratio. In support of this argument is the fact that in our cross diagonal measurements we could not achieve any significant decrease of the $\Delta V_H/V_{peak}$ ratio, relative to those shown in the bottom panels of figures 4 and 6, for currents as high as 60 mA. A second indication arises from cross-section and planar TEM images [19] that indicate a grain size distribution centred about ∼10 nm. These observations suggest that their magnetotransport behaviour requires micromagnetic study. In accordance, the PHE curves observed [20] in micrometre-scale Ni thin-film squares were found, from magnetic force microscopy (MFM), to be strongly affected by their magnetic domain structures, implying that each domain flipping causes a sudden change in the local resistivity of the domain regions. A third indication relies on the significant increase of the $\Delta V_H/V_{peak}$ ratio observed in Co/Cu MLs relative to magnetic single-layer films. This is in agreement with that reported in Ni/Co MLs [10], where interface effects were considered to be responsible for the enhancement of $\Delta V_H/V_{peak}$. Finally, we are inclined to exclude the possibility that these effects are caused by non-linear resistivity contact effects because we cannot observe such a behaviour in GMR films of Co/Cu or NiFe/Ag MLs.

In conclusion, it is shown that the large $\Delta V_H/V_{peak}$ value observed in the cross-diagonal Hall configuration is not associated (a) with the Ni/Co MLs only but is related to the specific measurement method, (b) with the intrinsic observed AMR effect in FM films, (c) with the so called 'extraordinary' or spontaneous Hall effect or, (d) with artefacts due to resistivity effects from the contacts. The dependence of $\Delta V_H/V_{peak}$ loops on the angle ϕ between I and H suggests a connection with a PHE mechanism that is associated with electronic scattering in magnetic grain boundaries and interfaces, possibly caused by local truncations of equipotential lines that are not perpendicular to the current and result in a Hall-like voltage. Certainly, at present this PHE mechanism is only a suggestion that needs careful investigation. In support of our observations the study of this effect in Ni thin-film squares [20] suggests that the PHE response cannot be explained by lump circuit models. Consequently the Wheastone bridge model [2] is a rather poor explanation for the observed behaviour of the PHE measurements. The present experiment reveals the important role of micromagnetic effects in FM coupled thin films, that is associated with the observed dependence of PHE response on the applied magnetic field magnitude and direction.

Acknowledgment

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