

GMR in Multilayers with an Alternating In-plane and Perpendicular Anisotropy

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The magnetic properties of sputtered ($\text{Ni}_{83}\text{Fe}_{17}/\text{Au}/\text{Co}/\text{Au}$) multilayers with various thicknesses of Au ($0.5 \leq t_{\text{Au}} \leq 3$ nm), Ni-Fe ($1 \leq t_{\text{Ni-Fe}} \leq 4$ nm) and Co ($0.2 \leq t_{\text{Co}} \leq 1.5$ nm) layers were characterized. An alternating in-plane and out-of-plane anisotropy of the ferromagnetic layers was achieved for the structures ($t_{\text{Au}} \geq 1.5$ nm) showing a weak coupling between the Ni-Fe layers with an in-plane anisotropy and the Co layers ($0.3 \leq t_{\text{Co}} \leq 1.2$ nm) with a perpendicular anisotropy. For such a structure, a detailed discussion on the GMR effect is presented, relating to the magnetization reversal from a mutually perpendicular magnetic configuration at the remanence to a parallel one at the saturation. An influence of the dense labyrinth domain structure on the magnetoresistance effect is also addressed.

Key words : GMR, perpendicular anisotropy, multilayers, domain structure

1. Introduction

Giant magnetoresistance (GMR) effect is observed in various layered structures consisting of two or more ferromagnetic layers (F) separated by nonmagnetic, metallic spacer layers (S).

The main requirement to observe a GMR is a mutual rotation of the magnetizations in adjacent ferromagnetic layers, caused by a magnetic field [1]. According to this definition the resistance change of F/S/F structures versus the angle φ between magnetization directions of the ferromagnetic layers is expressed by:

$$R(\varphi) = R_P + (R_{AP} - R_P)(1 - \cos\varphi)/2 \quad (1)$$

where R_P and R_{AP} represent the resistances of the structure in the parallel and the antiparallel magnetic configuration, respectively [2]. The GMR effect is maximal for structures in which the angle φ changes from 180° to 0° (from the antiparallel to the parallel configuration). Therefore, a particular attention was focused on spin-valve structures to fulfill this requirement

[2]. However, as we have demonstrated in our previous papers [3, 4], the structures with an alternating in-plane and perpendicular magnetic configuration in adjacent ferromagnetic layers in remanence are promising for the quantitative measurements of magnetic field due to the linear and unhysteretic changes of electrical resistance with magnetic field. Moreover, the investigations of magnetic properties of the films consisting of ferromagnetic layers with an alternating in-plane and perpendicular anisotropy is important due to a possible application as a magnetic storage media with perpendicular arrangement of bit cells (see e.g. [5]). Another important aspect is the application of magnetoresistance measurements as a tool to determine the magnetic properties of ultrathin ferromagnetic layers. According to the Eq. (1), using $R(H)$ measurements, the changes of magnetization direction (in respect to a given direction, e.g., direction of H) with magnetic field $M(H)$ of one layer can be determined if the magnetization direction of the second layer is pinned or if its changes with magnetic field are well known.

To realize such a structure with an alternating in-plane and out-of-plane magnetic configuration of ferromagnetic layers at remanence we have decided to prepare the structures composed of Permalloy ($\text{Py}=\text{Ni}_{83}\text{Fe}_{17}$) and cobalt

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layers separated by a gold spacer.

Permalloy and Co were chosen since Py possesses a pronounced in-plane anisotropy whereas a thin Co layer sandwiched by two layers of Au had a strong perpendicular anisotropy [6].

2. Experimental

Three set of $(\text{Py-}t_{\text{py}}/\text{Au-}t_{\text{Au}}/\text{Co-}t_{\text{Co}}/\text{Au-}t_{\text{Au}})_N$ (N is the number of repetition of the four-layer structure) multilayers were deposited in Ar atmosphere using UHV magnetron sputtering on glass substrates:

- (i) $(\text{Py-}2 \text{ nm}/\text{Au-}3 \text{ nm}/\text{Co-}/\text{Au-}3 \text{ nm})_{15}$, $(0.2 \leq t_{\text{Co}} \leq 1.5 \text{ nm})$
- (ii) $(\text{Py-}2 \text{ nm}/\text{Au-}/\text{Co-}0.6 \text{ nm}/\text{Au-}t_{\text{Au}})_{15}$, $(0.5 \leq t_{\text{Au}} \leq \text{nm})$
- (iii) $(\text{Py-}t_{\text{py}}/\text{Au-}2 \text{ nm}/\text{Co-}0.6 \text{ nm}/\text{Au-}2 \text{ nm})_{15}$, $(1 \leq t_{\text{py}} \leq 4 \text{ nm})$

The sputtering rates were 0.06, 0.05, 0.045 nm/s, for Au, Py, and Co respectively. The multilayer structure was checked by low- and high-angle X-ray diffraction (LAXRD and HAXRD). In LAXRD pattern, both the Kiessig fringes and the satellite peaks up to the 9th order were observed (Fig. 1). This confirms a well defined periodic structure of the samples and allows us to determine the modulation wavelength and the total thickness. The HAXRD showed a rich spectrum indicating the superlattice structure of (111) texture of Au layers. The magnetization reversal processes were studied using a vibrating sample magnetometer (VSM) and polar magneto-optical Kerr effect (PMOKE). The conventional four point method was used for magnetoresistance measurements. The $GMR(H)$ dependence was determined as:

$$GMR(H) = [R(H) - R(2T)] / R(2T) \quad (2)$$

where $R(H)$ and $R(2T)$ indicate electrical resistance at

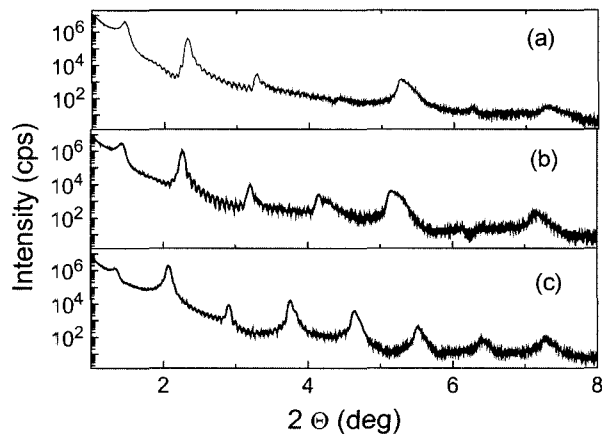


Fig. 1. Low angle X-ray diffraction spectra of $(\text{Py-}2 \text{ nm}/\text{Au-}3 \text{ nm}/\text{Co-}t_{\text{Co}}/\text{Au-}3 \text{ nm})_{15}$ with $t_{\text{Co}} = 0.3 \text{ nm}$ (a), 0.6 nm (b) and 1.5 nm (c).

magnetic field H and $H=2T$, respectively. The maximal value of $GMR(H)$ determines GMR amplitude. The remanence magnetic domain structure was investigated using magnetic force microscopy (MFM). All measurements were performed at room temperature.

3. Results and Discussion

3.1. Correlation between magnetization reversal and magnetoresistance

Before the discussion of the magnetic properties of $(\text{Py}/\text{Au}/\text{Co}/\text{Au})$ multilayers with different thicknesses of the spacer and ferromagnetic layers, we present (Fig. 2) the hysteresis loops and magnetoresistance curves measured for the sample characterized by perpendicular and in-plane anisotropy of Co and Py layers, respectively, and by a weak coupling between ferromagnetic layers. For such

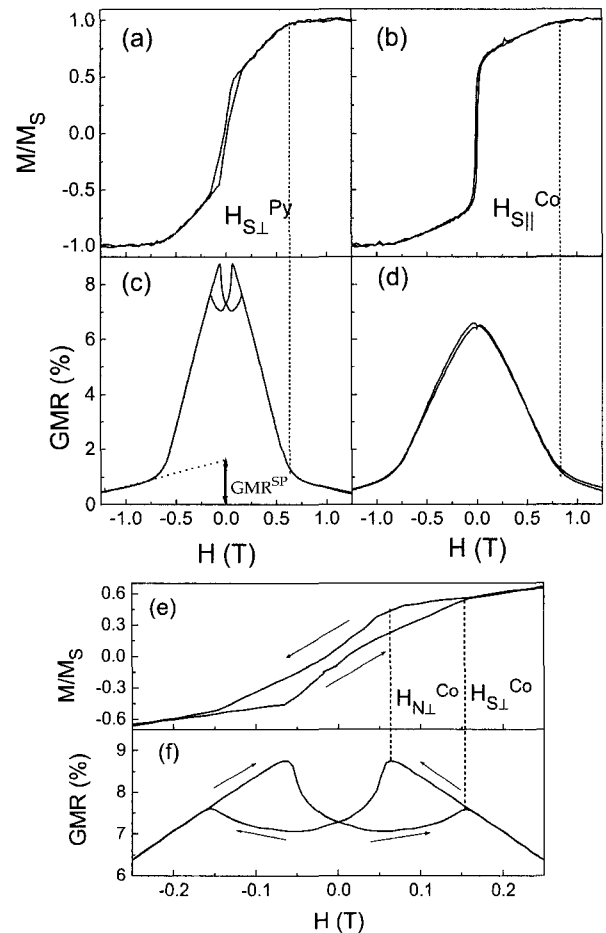


Fig. 2. Hysteresis loops (a, b) and magnetoresistance curves (c, d) of $(\text{Py-}2 \text{ nm}/\text{Au-}1.5 \text{ nm}/\text{Co-}0.6 \text{ nm}/\text{Au-}1.5 \text{ nm})_{15}$ multilayer measured with magnetic field applied perpendicular (left panel) and parallel to the sample plane (right panel). The central part of the dependences demonstrated in panels (a) and (c) are shown in (e) and (f) respectively.

sample, due to the weak coupling, the magnetization reversals of Py and Co layers are completely distinct. The central portions of the hysteresis loops (attributed to the magnetization along easy direction or in easy plane) for H_{\parallel} and H_{\perp} correspond to Py and Co reversal, respectively. Neglecting the small region of the magnetic fields, $|H_{\parallel}| \leq (H_C, H_K) \leq 10$ Oe (H_C, H_K are the coercivity and anisotropy fields of Py layers, respectively), the magnetization direction of Py layers always follows H_{\parallel} field direction (Fig. 2b). Therefore, the changes in $\cos\phi(H_{\parallel})$ and thus in $R(H_{\parallel})$ are related only to the magnetization rotation of Co layers. Hence, for investigated structures the saturation field $HS_{\parallel}^{\text{Co}}$ of Co layers for H_{\parallel} (anisotropy field of Co layers) can be determined from $R(H_{\parallel})$ measurements (Fig. 2d). For the magnetic field applied perpendicular to the sample plane (Fig. 2a, c) the part of the $M(H_{\perp})$ dependence related to Permalloy layer is unhysteretic and linear with saturation at $HS_{\perp}^{\text{Py}} \approx 4\pi M_S^{\text{Py}} \approx 0.6$ T due to the shape anisotropy [7]. The shape of the central part of the hysteresis loop (Fig. 2a, e) related to the Co layers magnetized in easy direction, i.e., for H_{\perp} is similar to those observed for (Au/Co) multilayers [8] and is typical of the films with perpendicular anisotropy and labyrinth or stripe domain structure (see also section 3.5). In $M(H_{\perp})$ and $R(H_{\perp})$ dependences, not only the saturation field HS_{\perp}^{Co} of Co layers but also the nucleation field $H_{\text{NL}}^{\text{Co}}$ where the process of domains nucleation starts on can be recognized (Fig. 2e, f).

The discussed example indicates that due to the correlation between $M(H)$ and $R(H)$ dependences (i.e., between magnetization reversal and GMR effect) some of the important magnetic properties of the Co and/or Py layers in weakly coupled Py/Au/Co/Au multilayers can be determined from magnetoresistance measurements.

3.2. Interlayer coupling between cobalt and permalloy across gold spacer

The crucial point for GMR enhancement in all layered structures is a thickness of a nonmagnetic spacer. For example in spin-valves (SV), too thick spacer layer may decrease GMR due to the increasing shunting of the current in the spacer. On the other hand, too thin spacer gives rise to increase in the ferromagnetic coupling between free and pinned ferromagnetic layers [2]. In consequence the simultaneous magnetization reversal of both ferromagnetic layers take place ($\varphi=0$ in the whole range of H) and GMR effect disappears [2]. Similarly to SV, for the investigated structures the 1.5 nm thick gold spacer is optimal to assure the maximum in GMR amplitude (Fig. 3). A decrease of GMR for $t_{\text{Au}} < 1.5$ nm correlates with the increase of HS_{\perp}^{Co} and is caused by an

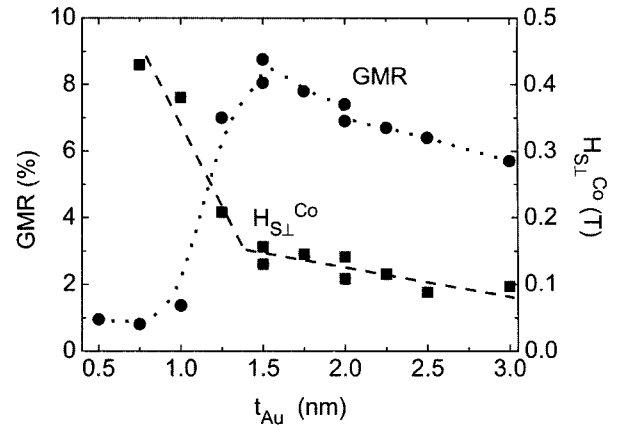


Fig. 3. GMR amplitude and saturation field of Co layers (determined for magnetic field applied perpendicularly to the sample plane) versus thickness of gold spacer of (Py-2 nm/Au- t_{Au} /Co-0.6 nm/Au- t_{Au})₁₅ structures.

increasing ferromagnetic coupling between Co and Py layers leading to their simultaneous magnetization reversal. The $HS_{\perp}^{\text{Co}}(t_{\text{Au}})$ dependence with a kink at $t_{\text{Au}} \approx 1.5$ nm suggests that two different mechanisms are responsible for the observed changes in interlayer coupling. The first one is the magnetostatic coupling, important in the whole range of t_{Au} . The second one is probably caused by pinholes and dominates for $t_{\text{Au}} < 1.5$ nm. The influence of the oscillatory, RKKY-like coupling on magnetization reversal was not observed in our investigations.

3.3. The structure and magnetic properties of Co layers

The anisotropy of Co layers sandwiched by Au is mainly determined by the shape and surface anisotropy terms. Therefore, the effective anisotropy perpendicular to the sample plane is observed only in the limited t_{Co} range and for films with the (111) textured gold layers. In our previous paper [9] we have demonstrated that these conditions are fulfilled for

Py/Au/Co/Au multilayers with $t_{\text{Co}} \leq 1.2$ nm. Here, we demonstrate that the analysis of the magnetoresistive curves (Fig. 2c, d, f) can be very helpful in the study of the microstructure and magnetic properties of Co layers depending on their thickness.

Fig. 4 shows the $GMR(t_{\text{Co}})$ dependence of (Py-2 nm/Au-3 nm/Co- t_{Co} /Au-3 nm)₁₅ ($0.2 \leq t_{\text{Co}} \leq 1.5$ nm) multilayers. Due to the thick Au spacer ($t_{\text{Au}}=3$ nm), the interlayer coupling between Co and Py is very weak. Thus, the magnetic properties of Co layers in investigated structures are not influenced by the interaction with Py layers. Four distinct ranges of Co layer thicknesses seen in Fig. 4 (indicated in the upper part of Figs 4-6) result from different structure and magnetic properties of Co layers

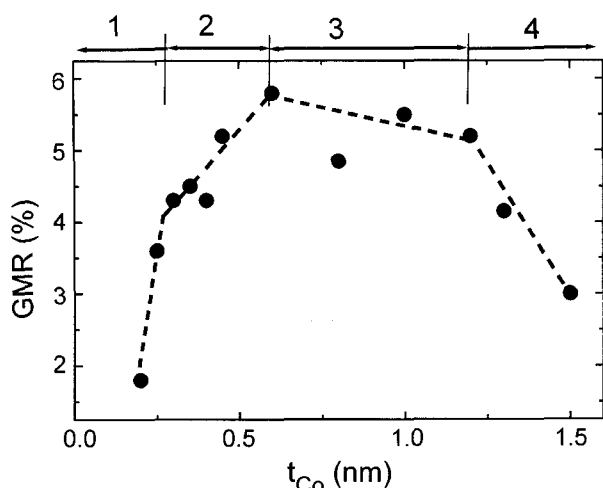


Fig. 4. GMR amplitude (determined for magnetic field applied perpendicularly to the sample plane) of (Py-2 nm/Au-3 nm/Co- t_{Co} /Au-3 nm)₁₅ multilayers with different Co layer thicknesses.

and will be discussed in details.

The first range with <0.3 nm is characterized by a superparamagnetic behavior of Co clusters. The zero value of remanence magnetization ($M_R=0$) for H_{\perp} , small value of a GMR amplitude, and nearly linear and unsaturated $GMR(H)$ dependences support this interpretation [3, 10, 11].

The second range $0.3 \text{ nm} \leq t_{Co} \leq 0.6 \text{ nm}$ is characterized by coexistence of the superparamagnetic and ferromagnetic grains of Co. The presence of the ferromagnetic Co clusters with perpendicular anisotropy is manifested by the characteristic hysteresis loops observed in the central part ($|H_{\perp}| \leq 0.15 \text{ T}$) of $M(H_{\perp})$ dependences and in consequence by $M_R > 0$ (Fig. 2a, e). $GMR(H)$ curves, both for H_{\parallel} as well as for H_{\perp} , consist of two nearly linear parts with different slopes (Fig. 2c, d). The high field parts are mainly related to the spin dependent electron scattering on superparamagnetic entities (GMR^{SP}) of Co and/or Py clusters. This effect was previously observed in various multilayers, e.g., Co/Cu, Py/Cu and its physical origin was explained in [10, 12]. Due to a relatively high efficiency of electron scattering events on superparamagnetic clusters their presence can be sensitively detected by magnetoresistance measurements. Therefore, GMR^{SP} effect was found not only in structures with discontinuous structure of ferromagnetic layers (granular structures) but also in polycrystalline multilayers with continuous ferromagnetic layers. In such films a certain amount of superparamagnetic clusters (magnetically decoupled from ferromagnetic layers) can be formed in the interfacial region during the deposition process. In our samples, due to a relatively high and constant thickness of Py layers $t_{Py}=2$

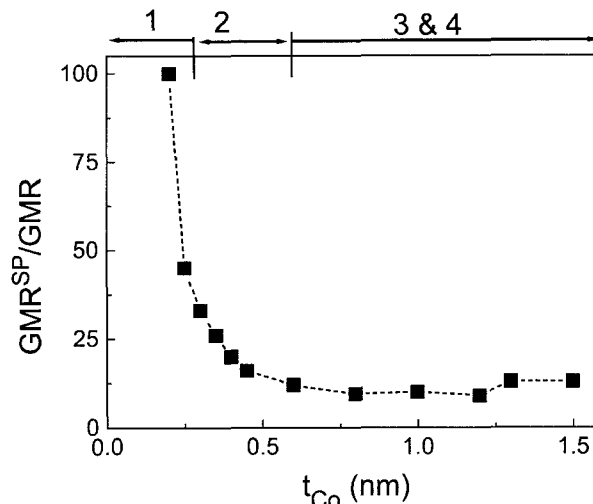


Fig. 5. Superparamagnetic contribution to GMR effect of (Py-2 nm/Au-3 nm/Co- t_{Co} /Au-3 nm)₁₅ multilayers (determined for magnetic field applied perpendicularly to the sample plane) as a function of cobalt layer thickness.

nm the observed changes in GMR^{SP} are mainly related to Co layers. The values of GMR^{SP} were determined by extrapolation of the high field parts of $GMR(H)$ dependence to $H=0$ (Fig. 2c). With increasing t_{Co} the relative contribution of GMR^{SP} to the effective value of GMR decrease (Fig. 5) and for $t_{Co} \approx 0.6 \text{ nm}$ saturates. This suggest that for $t_{Co} \geq 0.6 \text{ nm}$ cobalt layers show nearly continuous structure.

The changes of the saturation fields of the ferromagnetic Co particles with t_{Co} determined from $R(H)$ measurements

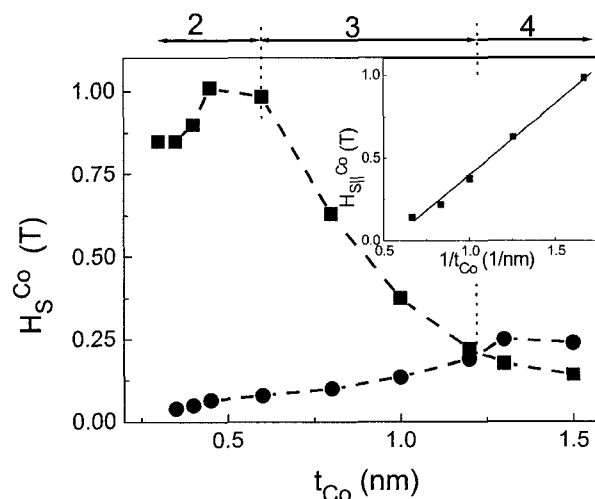


Fig. 6. Saturation field of Co layers in (Py-2 nm/Au-3 nm/Co- t_{Co} /Au-3 nm)₁₅ multilayers, determined for magnetic field applied in-plane HS_{\parallel}^{Co} (■) and perpendicular HS_{\perp}^{Co} (●) to the sample plane as a function of cobalt layer thicknesses. Inset; HS_{\parallel}^{Co} versus $1/t_{Co}$ for $0.6 \leq t_{Co} \leq 1.5 \text{ nm}$.

performed for H_{\parallel} and H_{\perp} are demonstrated in Fig. 6. The linear decrease of the effective anisotropy field of Co layers with the reciprocal of t_{Co} ($HS_{\parallel}^{\text{Co}} \propto 1/t_{\text{Co}}$) observed for $t_{\text{Co}} > 0.6$ nm (inset of Fig. 6), clearly indicates the crucial role of the surface anisotropy. On the other hand a nearly constant value of $HS_{\parallel}^{\text{Co}}$ observed for $0.3 < t_{\text{Co}} < 0.6$ nm indicates that the thickness of Co clusters is also independent on nominal thickness of cobalt layers. This means that during growth of the Co layers with $t_{\text{Co}} < 0.6$ nm only the lateral dimension of clusters increases but their thickness remains constant. Similar growth process was observed and well documented for MBE deposition of cobalt layers on Au(111) substrates [13, 14].

The third and fourth ranges ($t_{\text{Co}} > 0.6$ nm) are related to the continuous ferromagnetic Co layers. Magnetization reversal of such multilayers is similar to those discussed above. However, in contrast to the behavior observed in the second range, the GMR^{SP}/GMR factor remains nearly independent of t_{Co} (Fig. 5). Simultaneously, the typical for surface anisotropy decrease in $HS_{\parallel}^{\text{Co}}$ with t_{Co} is observed (Fig. 6). For $t_{\text{Co}} \approx 1.2$ nm the saturation fields of Co layers for H_{\parallel} and H_{\perp} are equal ($HS_{\parallel}^{\text{Co}} \approx HS_{\perp}^{\text{Co}}$) indicating the transition from perpendicular to in-plane anisotropy of Co layers, i.e., transition from the 3rd to 4th range of t_{Co} .

3.4. The influence of permalloy layer thicknesses on magnetic properties of Py/Au/Co/Au multilayers

Influence of t_{Py} on the magnetoresistance and saturation field of Py layers in a magnetic field applied perpendicular to the surface ($H_{S\perp}^{\text{Py}}$) was determined for $(\text{Py-}t_{\text{Py}}/\text{Au-2 nm}/\text{Co-0.6 nm}/\text{Au-2 nm})_{15}$ ($1 \leq t^{\text{Py}} \leq 4$ nm) structures (Fig. 7). The $GMR(t_{\text{Py}})$ dependence with a local maximum for $t_{\text{Py}} \approx 3$ nm is typical of all GMR structures containing Py (see e.g. [2, 15]). The increase of $H_{S\perp}^{\text{Py}}$ with t^{Py} is similar to that observed for Py/Cu

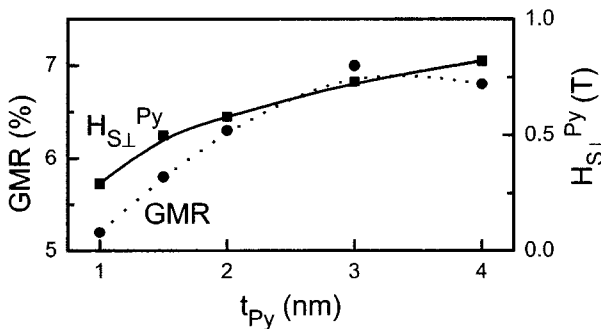


Fig. 7. GMR amplitude and saturation field of permalloy layers determined for perpendicular field configuration in $(\text{Py-}t_{\text{Py}}/\text{Au-2 nm}/\text{Co-0.6 nm}/\text{Au-2 nm})_{15}$ structures with different thickness of permalloy layers.

multilayers [7] and in the first approximation it can be interpreted as resulting from a magnetically inactive Py/Au interface layer. A strong dependence of $H_{S\perp}^{\text{Py}}(t_{\text{Py}})$ observed for the small values of t_{Py} , offers a simple way for tailoring the saturation fields of $GMR(H_{\perp})$ in our structures. Thus, for some appropriate layer thicknesses (e.g., $t_{\text{Py}}=2$ nm, $t_{\text{Co}}=0.8$ nm) the magnetoresistance response can be nearly isotropic.

3.5. The influence of the domain structure on magnetoresistance effect

Observations of remanence domain structure performed with MFM shows the labyrinth and stripe domains (domain widths of about 200 nm) (Fig. 8). Such a domain structure is typical of the films with perpendicular anisotropy and characterizes the top cobalt layer covered by 3 nm thick Au layer.

As we have mentioned in the section 3.1, the $R(H_{\perp})$ dependences show a hysteretic behavior correlated with magnetization reversal of the Co layers magnetized in the easy direction (Fig. 2f). On decreasing the magnetic field from $H_{\perp}=H_{S\perp}^{\text{Py}} \approx 0.6$ T, a linear increase of resistance, related to rotation of Py magnetization direction from perpendicular to in-plane configuration and a distinct decrease of $R(H_{\perp})$ at $H_{\perp}=H_{N\perp}^{\text{Co}}$ are observed.

There is a number of different mechanisms which link the resistance changes of ferromagnetic layers to the presence of domain structure. However, according to the results presented in the review article by Kent *et al.* [16], these mechanisms seem to be too weak to explain the resistance changes observed in our samples. One possible

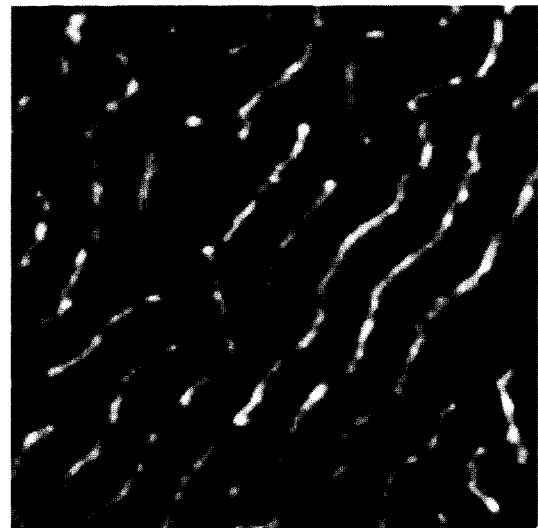


Fig. 8. MFM domain structure of $(\text{Py-2 nm}/\text{Au-3 nm}/\text{Co-0.6 nm}/\text{Au-3 nm})_{15}$ multilayer in as-deposited state. The displayed area is $5 \mu\text{m} \times 5 \mu\text{m}$.

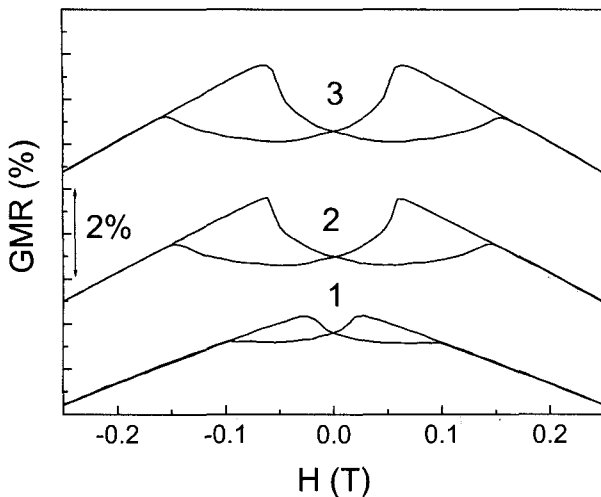


Fig. 9. The central (hysteretic) range of magnetoresistance curves determined for perpendicular field configuration and samples (Py-2 nm/Au- t_{Au} /Co-0.6 nm/Au- t_{Au})₁₅ with $t_{Au}=3$ nm (1), $t_{Au}=1.75$ nm (2), $t_{Au}=1.5$ nm (3).

origin is an abrupt change in mutual magnetic configuration of Co and Py layers (changes of $\cos\varphi$ and $R(H)$ values) caused by magnetic stray fields from domain walls. This effect, known in literature as domain wall induced coupling, increases with decreasing thickness of spacer layer [17]. In our structures the decrease in the electrical resistance at a domain nucleation field is also as stronger as thinner are Au layers (Fig. 9). The qualitative agreement between both cases suggest their common origin.

For magnetic field applied in-plane, a domain structure of Co layers gradually appears, and therefore the influence of domain nucleation on $R(H_{\parallel})$ is not substantial. The decrease of resistance caused by domain nucleation is responsible for smaller resistance values in the remanence as at $H_{\perp}=HN_{\perp}^{Co}$. This result also explains the difference between GMR amplitudes measured with H_{\perp} and H_{\parallel} (Fig. 2c, d).

4. Summary

An alternating in-plane and out-of-plane anisotropy of ferromagnetic layers was achieved in (Ni₈₃Fe₁₇/Au/Co/Au) multilayers deposited by sputtering. For such a structure, a detailed discussion on the GMR effect is presented, relating to the magnetization reversal from a mutually perpendicular magnetic configuration at the remanence to a parallel one at the saturation. The changes in GMR amplitude, relative contribution of superparamagnetic entities to the GMR effect and the saturation fields of Co and Ni-Fe layers were determined from the magnetoresistance

measurements with a magnetic field applied perpendicular and in sample plane. The analysis of the results was performed for structures with different thicknesses of constituent layers. The main results are as follows:

- For Au spacer thickness higher than 1.5 nm, the coupling between ferromagnetic layers is weak and magnetization reversal of Ni-Fe and Co take place independently.

- The changes of Ni-Fe layer thickness offers a simple way for tuning the saturation fields of $R(H)$ dependence for perpendicular configuration of magnetic field.

- In the growth process of Co layers, the following stages can be identified: (i) creation of small superparamagnetic clusters ($t_{Co}<0.3$ nm), (ii) lateral growth of the clusters—coexistence of superparamagnetic and ferromagnetic grains ($0.3 \leq t_{Co} \leq 0.6$ nm), (iii) formation of continuous ferromagnetic layers with perpendicular anisotropy ($0.6 < t_{Co} \leq 1.2$ nm) and in-plane anisotropy ($t_{Co} > 1.2$ nm).

The influence of the domain walls in Co layers on magnetoresistance effect was analyzed. It is thought that the decrease of electrical resistance correlated with the creation of dense labyrinth domain structure is caused by the domain wall induced coupling.

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