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Magnetization and impedance measurements of multilayer Co/Cu structures in millimeter waveband

D.P. Belozorov[a], V.N. Derkach[b], S.V. Nedukh[b], A.G. Ravlik[c], S.T. Roschenko[c], I.G. Shipkova[c], S.I. Tarapov[b,d,*], F. Yildiz[d], B. Aktas[d]

[a] National Scientific Center "Kharkov Institute of Physics and Technology" NAS of Ukraine, 1 Akademicheskaya St., 61108, Kharkov, Ukraine
[b] Radiospectroscopy, Institute of Radiophysics and Electronics NAS of Ukraine, 12 Ac. Proskura Street, 61085, Kharkov, Ukraine
[c] National Technical University "Kharkov Polytechnical Institute", 21 Frunze St., 61002, Kharkov, Ukraine
[d] Electronic Engineering Department, Gebze Institute of Technology, PK. 141, 41400, Gebze-Kocaeli, Turkey

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Abstract

The saturation magnetization of Co/Cu multilayer structures and individual Co films 1.0–4.0 nm thick have been studied both by electron spin resonance technique and by vibrating sample magnetometer method. The results of static and dynamic measurements are in good agreement. It was found that the saturation magnetization of Co layers imbedded in a multilayer structure was greater than the magnetization of individual films and tended to the value of bulk material. The saturation magnetization of multilayer structure with thickness of nonmagnetic Cu interlayer varying from 1.7–10 nm have been investigated for samples both with and without Fe-buffer layer. For the first case Co saturation magnetization has decreased with increase of nonmagnetic interlayer in contrast to similar samples without buffer. The measurements of impedance characteristics have been carried out for the multilayers Fe(6 nm)/[Co(t_C)/Cu(2 nm)]$_8$ ($t = 1, 2, 3$ nm) in the millimeter waveband (at $v = 44$ GHz). The transmission coefficient dependence on the external static magnetic field for a waveguide containing the multilayer specimen was found to be the same as the dependence of direct current giant magnetoresistance of the mentioned specimen.

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1. Introduction

Layered magnetic/nonmagnetic conductors (Co/Cu, Fe/Cr and others) are of interest because they show remarkable variation of DC-resistivity in the static magnetic field (giant magnetoresistance—GMR effect). They have been the subjects of intensive research since the discovery of the GMR

*Corresponding author. Radiospectroscopy, Institute of Radiophysics and Electronics NAS of Ukraine, 12 Ac. Proskura Street, 61085 Kharkov, Ukraine. Tel.: +38-0572-448-463; fax: +38-0572-441-105.
E-mail address: tarapov@ire.kharkov.ua (S.I. Tarapov).

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[1–4]. The similar phenomena take place also for alternating current (giant magnetoimpedance—GMI). In particular, it was found that the transmission coefficient of electromagnetic millimeter waves in such multilayer (ML) structures with GMR effect changed with magnetic field in a similar way to DC-ohmic resistance [5–11]. ML structures mentioned above promise to become a basis of rather compact and inexpensive elements for millimeter wavelength band technology (attenuators, switches, etc.), with very small switching time that is restricted by mainly the spin–spin relaxation time of ML materials (10^{-9}–10^{-13} s). Considerable interest of contemporary researches is focused on the possibility to use such structures in the frequency band above 20 GHz (see, for example, Refs. [6,10]). Therefore, a thorough study of ML structures in this frequency band is necessary to obtain information both about their static and dynamic characteristics. The study of various dimensional magnetization effects in ML is of special interest. In particular they include the dependence of saturation magnetization of magnetic layers on their thickness and also on the thickness of nonmagnetic interlayers.

The obtained results can be of interest not only from the industrial standpoint but they contain fundamental physical information as to magnetism of surfaces and ultrathin magnetic layers both individual and integrated into ML structure. In last years the phenomena of magnetic moment enhancement for magnetic 3d-metals have been widely discussed in literature. The enhancement takes place if ultrathin magnetic film is in the surface contact with nonmagnetic layer of noble metals or copper (see, for instance Refs. [12,13]). So, in Ref. [13] the anomalies of saturation magnetization have been detected in magnetic superlattices Fe/non-magnetic metal with ultrathin Fe-layers (0.6 nm). At some definite thickness of non-magnetic interlayers (1.0–1.2 nm for different metals from Ti, Ta, Mo, Pd group) the values of saturation magnetization M_S obtained for Fe were several times greater than the corresponding value for the bulk metal. According to Ref. [14], the monoatomic Co-layers (thickness 0.3 m) without magnetic order, became ferromagnetic, when they were included in the ML structure with Cu interlayers no more than 1.5 nm thick. One can conclude that the presence of ferromagnetic–nonmagnetic metal interface is favorable for origin of magnetism.

In the present article we report the results of studies of the magnetization and ferromagnetic resonance (FMR) absorption both for Co/Cu ML structure and single Co-layers. We employed the electron spin resonance—ESR (or equivalently—FMR) technique in the frequency range from 9.5 to 37 GHz. The obtained magnetization values were compared with the results obtained by the vibrating magnetometer method. It is found that the results of static and dynamic measurements are in good agreement. Though the thickness of magnetic layers and nonmagnetic interlayers in our specimens exceeded the corresponding values in Refs. [13,14], nevertheless, some peculiarities of the magnetization behavior in our experiments with Co-films can be attributed to the influence of the layer boundary and can be a starting point for future investigations. The results of FMR absorption were shown to correlate with the ML GMR data for DC. The magnetoimpedance parameters measured in the millimeter waveband are compared to that of the magnetoresistance as well.

2. Experimental

2.1. Specimens preparation

We have studied GMR multilayer structures of $Fe/(Co/Cu)_n$ with the magnetoresistance ratio up to 8–10%. The films have been produced using a combination of two methods: triode ion-plasma sputtering for cobalt and iron deposition and magnetron sputtering for copper deposition. The sputtering has been carried out in argon atmosphere at $(3–4)\times10^{-1}$ Pa. The pressure in vacuum chamber before the argon filling was $(1–2)\times10^{-4}$ Pa. The films have been deposited on uncooled glass ceramic substrates with specially prepared surface covered by amorphous oxide to diminish surface roughness. Multilayer samples under study were polycrystalline and exhibited rather well defined laminated structure according to data of X-ray diffraction and electron transmission microscopy of oblique cross-sections. In more detail the procedure of manufacturing and physical properties of such multilayers are described in Refs. [15,16]. We have investigated also the series Fe(6 nm)/[Co(t nm)/Cu(2 nm)]$_8$ and Fe(6 nm)/[Co(1 nm)/Cu(t nm)]$_{30}$ where the Fe-film played a role of a buffer layer. The Cu-interlayer thickness corresponds to the second peak of magnetoresistance ratio dependence [3] on a nonmagnetic interlayer thickness. The Co-layers thickness varied from 1 to 4 nm.

We believe that any detectable interfacial mixing in ML structures under study is absent. Co and Cu are weakly soluble in each other at the temperature range from 20°C to 400°C [17]. Because during the deposition the substrate temperature did not exceed 50°C the mixing by interdiffusion should not take place. However, it does not exclude the occurrence of separate roughness of about 1–2 atomic sizes height, which is difficult to detect experimentally.

Co-layers under consideration are continuous down to small thickness ~1 nm. The magnetic characteristics of individual Co layers prepared by the same methods, which were used, for the manufacture of ML structure support indirectly the film continuousness. The films of 1–3 nm thick have hysteresis loops with relatively small values of coercive force and saturation field (<50 Oe). This feature distinguishes them from the films broken into islands for which magnetic saturation is observed usually at far greater fields (several kOe).

2.2. Technique and installation

A high-sensitive vibrating magnetometer has been used for measuring the relative magnetization M_S/M_{Sb} of films and multilayer structures. The standard procedure was as following. In the experiment the amplitude of magnetic hysteresis loop has been measured for the sample under study. Then the obtained data have been compared with the corresponding data of thick reference film (which was supposed to have the magnetization of bulk material M_{Sb}). As we found out, the Co-film, 30 nm thick, could be used as such reference sample. For comparison with ML structures the individual Co-films deposited on a glass ceramic substrate have been used as reference specimens. In this way we obtain the value of the relative magnetization M_S/M_{Sb}. Note, that the error in M_S/M_{Sb} obtained by the method does not exceed 10–15%.

As regards the ML-structures with Fe-buffer, which contributes to a total film magnetization, the calculation of net magnetization associated with Co-layers requires the extraction of this Fe contribution. It was supposed that the magnetization of the Fe-layer 6 nm thick is close to the magnetization of a bulk Fe sample.

For in situ measurement of the magnetization of individual magnetic components entering the multilayered specimens, we used ESR-technique (in our case—FMR-technique). Standard Bruker EMX-10 model ESR spectrometer have been used for 10 GHz band. Several special electrodynamical modules have been designed [10,18] for 20–40 GHz band. The modules allow extending the experimental possibilities of Bruker spectrometer by scanning the working frequency in the millimeter wavelength band. Note that we used here a few kits of cavity resonator [19], as well as open resonator structures [20]. The detailed discussion and analysis of the experimental technique and the installation are in above mentioned papers. For 30–40 GHz band we used the open resonator structure. This structure demonstrates the high sensitivity and informativity of FMR experiment in comparison with standard cavity. In particular, it allows measuring of FMR response from the samples of single Co-layers 1 nm thick that is impossible by means of cavity resonators.

3. Experimental results and discussion

3.1. Ferromagnetic resonance measurements

The advantage of the magnetoresonance method over the static ones consists of the possibility to measure the magnetization of separate multilayer components, whereas the static methods give only the magnetization averaged over the sample volume. This may be important if buffer layer is also made of magnetic material. In our case their own resonance peaks characterize both Fe and Co multilayer components. The typical FMR absorption lines for frequencies 20.5–26.0 GHz [10] are presented in Fig. 1. Note that according to Ref. [10] the "sharp" peaks correspond to the Fe buffer layer, and the "weak", satellite, ones—to the Co multilayer subsystem.

Our experiments correspond to the "planar geometry", i.e. both static external magnetic field vector (H) and high-frequency magnetic field vector (h) lie in the plane of the film. As every magnetic multilayer component (Fe and Co) produces its own resonance peak, we find the magnitude of saturation magnetization for each component with the help of known Kittel formula

$$2\pi\hbar\nu_{\mathrm{res}} = g\beta\sqrt{H_{\mathrm{res}}(H_{\mathrm{res}} + 4\pi M_{\mathrm{s}})}, \qquad (1)$$

where ν_{res} is the resonance frequency; H_{res} is the applied external static magnetic field; β is Bohr magneton; $\hbar$ is Planck constant; g is the g-factor (the spectroscopy splitting factor); M_{S} is the saturation magnetization. Supposing g-factor value for our specimens to be $g \approx 2.1 \pm 5\%$ (what is true for majority of low-dimensional ferromagnets of this type) we can determine the saturation magnetization M_{S}.

Note that the possibility to use Kittel formula (1) neglecting anisotropy fields is supported in our case by results of static measurements of hysteresis loops both in plane of ML structures and in the perpendicular direction. First of all the films proved to be magnetically isotropic in plane, that is the uniaxial planar anisotropy was absent. In the second place all attempts to magnetize the films in the perpendicular direction up to maximal fields obtainable in our experiments (10–13 kOe) revealed no hysteresis loop. This indicates that the perpendicular anisotropy is small if ever occurs.

In order to study the influence of the multilayer structure on the Co magnetization the identical measurements have been carried out with three various sets of multilayer specimens:

1. Fe(6 nm)[Co(t nm)/Cu(2 nm)]$_8$ ($t = 1, 2, 3$ nm)
2. Co(t nm) ($t = 1, 2, 3, 4$ nm)
3. (a) Fe(6 nm)[Co(1 nm)/Cu(t nm)]$_{30}$ ($t = 1.7, 2.0, 2.9, 4.2, 7.4$ nm)
   (b) [Co(1 nm)/Cu(t nm)]$_{30}$ ($t = 1.7, 2.0, 2.8, 4.2, 7.4$ nm)

Here t is the width of the corresponding layer.

Typical "resonance frequency–resonance field" ($\nu_{\mathrm{res}} = f(H_{\mathrm{res}})$) dependencies for Fe(6 nm) [Co(t nm)/Cu(2 nm)]$_8$ specimens and single Co-layers are presented in Figs. 2 and 3. In Fig. 2 the

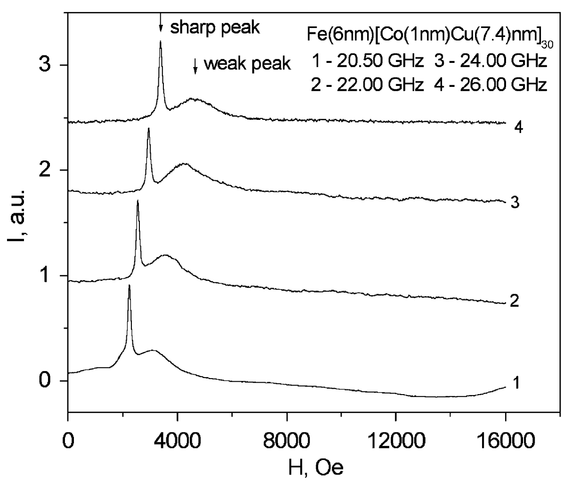

Fig. 1. Typical FMR absorption lineshape for Fe-buffered MLs Fe(6 nm)[Co(1 nm)/Cu(7.4 nm)]$_{30}$.

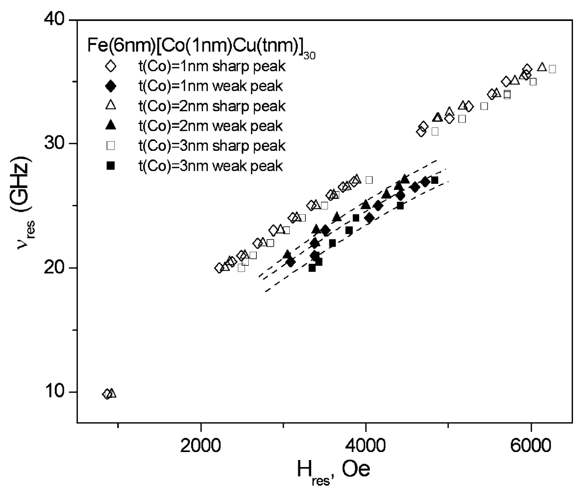

Fig. 2. MLs Fe(6 nm)[Co(t nm)/Cu(2 nm)]$_8$ ($t = 1, 2, 3$ nm). Resonance frequency-field dependence for weak (Co) and sharp (Fe) peaks in "planar geometry". The experimental error is: $\delta(\nu) \approx 1\%$; $\delta(H) \approx 1\%$.

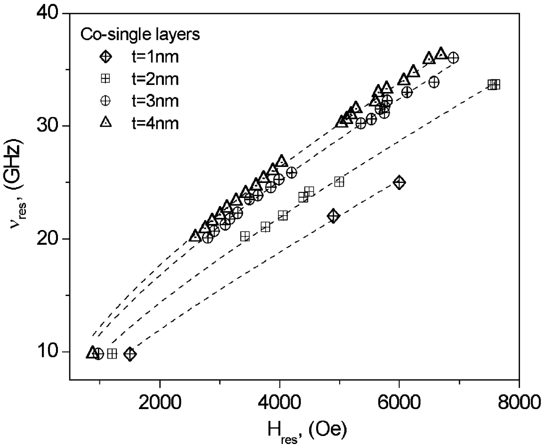

Fig. 3. Single Co-layers. Resonance frequency–field dependence for planar geometry. The experimental error is: $\delta(\nu) \approx 1\%$; $\delta(H) \approx 1\%$.

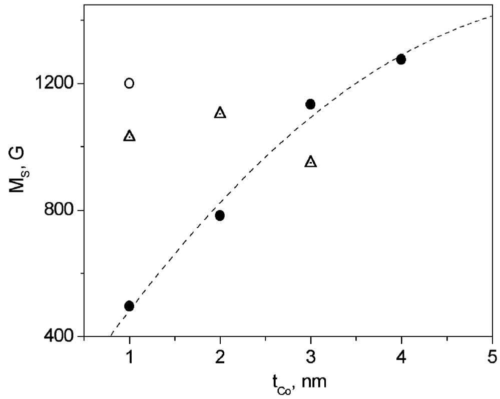

Fig. 4. ESR-technique (dynamic technique). Magnetization of saturation for single Co-layers and for Co/Cu structures versus the Co-layer thickness. The experimental error is: $\delta(M) \approx 3$–5%. ●—single Co-layers; △—Co-layers in ML structure Fe(6 nm)/[Co(t_{Co})/Cu(2 nm)]$_8$; ○—Co-layers in ML structure [Co(1 nm)/Cu(2 nm)]$_3$0.

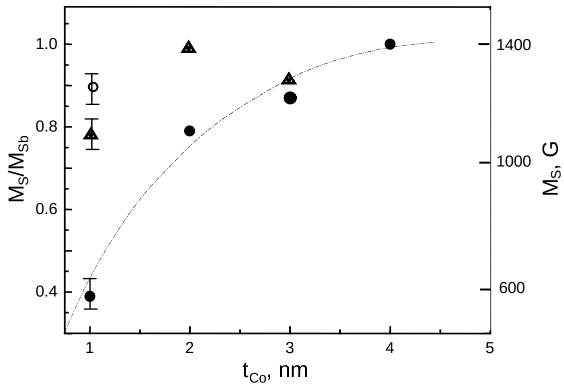

Fig. 5. Vibromagnetometer technique (static technique). Relative magnetization M_S/M_{Sb} for single Co-layers and for Co/Cu structures versus the Co-layers thickness. (M_{Sb}—is the magnetization of a reference Co-film by width 30 nm). The experimental error is: $\delta(M) \approx 10$–15%. ●—single Co-layers; △—Co-layers in ML structure Fe(6 nm)/[Co(t_{Co})/Cu(2 nm)]$_8$; ○—Co-layers in ML structure [Co(1 nm)/Cu(2 nm)]$_3$0.

experimental points for the resonance frequencies of Co-layers with thickness $t = 1, 2, 3$ nm ("weak" peaks) are interpolated with dashed lines. The resonance frequency data for single Co-layers with the thickness $t = 1, 2, 3, 4$ nm are represented in Fig. 3. As about the third set of specimens they have been measured and analyzed in Ref. [10]. And we restrict ourselves here only with the analysis of the saturation magnetization M_S dependence on the Cu interlayer thickness (Fig. 6).

In Fig. 4 the saturation magnetization data are shown as a function of the Co-layer thickness correspondingly for individual Co-layers and for Co-layers imbedded into ML structure. These data have been derived with the help of Eq. (1) from the resonant dependencies presented in Figs. 2 and 3. The results of static measurements obtained with vibrating sample magnetometer method are represented in Fig. 5. The magnetization values here are expressed in terms of M_S/M_{Sb} (where M_{Sb}—is the magnetization of bulk Co).

We see that for individual Co-layers (Fig. 4, black circles) the magnetization increases together with Co-layer thickness and approaches the value corresponding to bulk Co ($M_{Sb} =$ 1422 Oe). This result is in a good agreement with data obtained from static measurements (Fig. 5, black circles). Note that increase of magnetization with increase of Co-layer thickness is in accordance with existing theories (See, for example, Ref. [21]).

Three multilayer specimens Fe(6 nm)[Co(t nm)/Cu(2 nm)]$_8$ with thickness of Co-layers (t=1, 2, 3 nm) have been also studied. They did not show any noticeable changes in M_S as Co thickness increases. Both dynamic (Fig. 4) and static (Fig. 5) measurements give similar dependence on

Co-layers thickness. This fact shows the importance of Co/Cu interfaces and surface phenomena in the development of overall magnetization of Co-layers in the multilayer structure.

This conclusion is supported if we compare the magnetization of individual Co films with the magnetization of Co-films imbedded in the multilayer structure (Fig. 4 and 5). According to these figures, the character of dependence of M_S on thickness for individual Co-layers (black circles) noticeably differs from analogous dependence for layers, imbedded into the ML structure Fe(6 nm)/[Co(t_{Co})/Cu(2 nm)]$_8$ (triangles). As can be seen, the influence of the multilayer structure is twofold. First of all, the magnitude of Co magnetization increases together with the number of layers involved in ML structure. The results concerning magnetization of specimens with fixed Co thickness ($t_{Co} = 1$ nm) support the statement: single layer $M_S \approx 500$ Oe; eight bilayers $M_S \approx 1000$ Oe; 30 bilayers $M_S \approx 1200$ Oe. Secondly, the dependence of magnetization on Co-layer thickness for ML structure appears to be weaker than the analogous dependence for individual Co-layers. Even for structures with the thickest Cu interlayer the average value of Co magnetization still exceeds the corresponding magnetization value for individual Co-film, 1 nm thick.

It is interesting to compare the Co magnetization behavior for ML structures with and without Fe buffer layer. As can be seen from the Fig. 6 for ML structures with buffer layer the average Co magnetization decreases with increase of nonmagnetic interlayer thickness. This agrees with decrease of exchange interaction between neighbor Co-layers as the thickness of nonmagnetic layers increases.

Note that in ML without buffer layer the average Co magnetization is almost independent of the thickness of Cu interlayer. Hence, the presence of buffer layer makes the Co magnetization more sensitive to changes in nonmagnetic layer thickness. This fact agrees with the known from literature [22] role of buffer layer. Really in the presence of the buffer layer the ML structure is more regular and perfect, so the dependence on nonmagnetic layer thickness is more pronounced. At the same time without the buffer layer the structure of Cu-layers is far less regular which leads to some averaged magnetization value and correspondingly to the absence of dependence on the Cu-layer thickness.

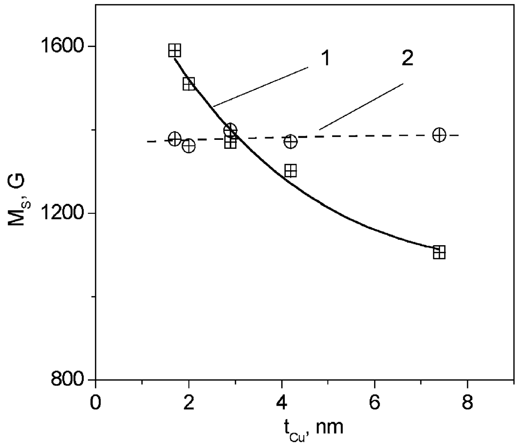

Fig. 6. Magnetization of saturation for buffered (1) and non-buffered (2) multilayers versus the interlayer thickness: 1—Fe(6 nm)[Co(1 nm)/Cu(t nm)]$_{30}$; 2—[Co(1 nm)/Cu(t nm)]$_{30}$. The experimental error is: $\delta(M) \approx 3\%$.

3.2. Magnetoimpedance measurements

Measurements of the impedance characteristics have been carried out for the multilayers Fe(6 nm)/[Co(t_{Co})/Cu(2 nm)]$_8$ ($t = 1, 2, 3$ nm) in the millimeter waveband (at $v = 44$ GHz). Fig. 7b represents the dependence of the ratio $\Delta T/T$ on the external static magnetic field (H). Here T is the transmission coefficient of the waveguide containing the specimen. The values of T are directly proportional to the impedance Z of the ML structure. Magnetic and electric components of the electromagnetic field lie in the plane of the film. The technique, installation and preliminary results of multilayer impedance study at the frequency band 40–150 GHz are presented in details in Refs. [23,10]. Here we want to stress that the shape of specific double peaks presented in Fig. 7b is reproducible in the whole frequency band mentioned above.

At the same time, these peaks copy the shape of giant magnetoresistance dependence $\Delta R/R$ (DC-case) on H. The $\Delta R/R$ dependencies shown

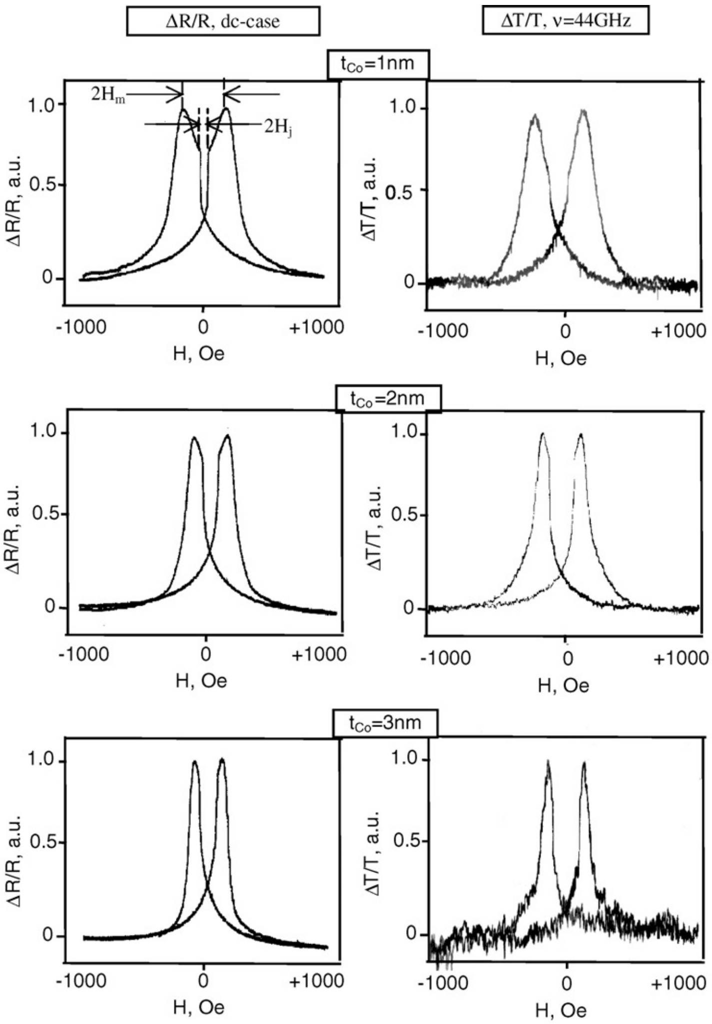

Fig. 7. Magnetoresistance (a) and magnetoimpedance $\nu = 44$ GHz response (b) for Fe(6 nm)/[Co(tCo)/Cu(2 nm)]$_8$.

in Fig. 7a represent typical GMR curves obtainable while scanning the external static magnetic field H. The intrinsic properties of these GMR dependencies for Co/Cu specimens are discussed in details in Ref. [15]. At $H = H_m$ the maximum of GMR magnitude occurs. Another characteristic field H_j, at which domain walls displacement takes place [15], is observed for each specimen too. A sharp jump-like change of the dependence $\Delta R/R = f(H)$ takes place at this field magnitude.

Almost the same situation can be seen for high-frequency case (Fig. 7b, Table 1). Magnitudes of the relative transmission coefficient $\Delta T/T$ coincide approximately with $\Delta R/R$. Characteristic fields H_m and H_j, which are well defined, are also approximately the same within the limits of experimental errors. The experimental error (δ) for various samples lie in the interval: $\delta(\Delta T/T) = 3-10\%$; $\delta(\Delta R/R) = 1-3\%$; $\delta(H) = 1-2\%$. This proves that mechanisms, which are responsible for magnetoimpedance, have relaxation time at least less than

$$\tau_{rel} \leqslant \frac{1}{\nu} = \frac{1}{44\,\text{GHz}} \sim 2.5 \times 10^{-11}\ \text{s}$$

for the given high-frequency (HF) band.

Table 1

Specimen	Static case—$\Delta R/R$			Dynamic case—$\Delta T/T$		
Fe(6 nm)/[Co(t_{Co})/Cu(2 nm)]$_8$	$\Delta R/R$ (%)	H_m (Oe)	H_j (Oe)	$\Delta T/T$ (%)	H_m (Oe)	H_j (Oe)
$t_{Co} = 1$ nm	4.7	180	26	4.69	180	35
$t_{Co} = 2$ nm	3.5	140	47	4.13	117	55
$t_{Co} = 3$ nm	3.9	120	70–80	1.90	105–110	71–85

Such very fast HF-response opens new opportunities for numerous applications of ML structures in the extra-high frequency (GHz-band) technology (millimeter-wavelength-band filters, electrically managed isolators, attenuators etc). It is necessary only that the ML-structures were built in the resonator with high enough quality factor for enhancement of the GMI response.

The nonlinear impedance dependence on H in the vicinity of H_j make it possible also to construct the nonlinear frequency multiplier in millimeter wavelength band if the high-frequency field intensity in the resonator can reach an amplitude $h \sim 1$–2 Oe.

4. Conclusions

1. The study of Co/Cu multilayer structures and individual Co-layers has been performed both by the FMR method in 10–37 GHz band and by vibrating sample magnetometer method. It was shown that the results of static and dynamic methods are in good agreement.
2. The dependence of saturation magnetization on thickness for individual ultrathin cobalt films is measured in interval from 1 to 4 nm. It was shown that saturation magnetization decreases in comparison with magnetization of a bulk material that is in accordance with the available theories.
3. The tendency of saturation magnetization increase up to level close to magnetization of bulk material is observed for ultrathin Co-layers imbedded in multilayer structure.
4. The saturation magnetization of Co-layers ($t_{Co} = 1$ nm) decreases with increase of non-magnetic interlayer thickness from 1.7 to 7.4 nm in multilayer structure with Fe-buffer. This result is not observed in similar system without the buffer.
5. High-frequency magnetoimpedance has been measured for $\nu = 44$ GHz and compared with GMR data. It is shown that relaxation times of GMI processes are less than 2.5×10^{-11} s. It makes ML structures very promising for wide scope of high-frequency technology applications.

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