Dimensional Effects in Magnetoresistive Properties in Three-Layer Films

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Dimensional effects in the magnetoresistive properties of $Fe_xCo_{1-x}/Cu/Fe_xCo_{1-x}$ three-layer magnetically ordered films obtained by layer-by-layer metal condensation followed by heat treatment in the temperature range of $300\div550$ K were studied experimentally and theoretically using generalized Dieny formulas [1, 2]. It is shown that in the case when the cover layer thickness is much smaller (greater) than the base layer thickness, the numerical value of the magnetoresistance ratio δ is negligible due to shunting of the covering layer resistance by the resistances of the base layer and the nonmagnetic layer by the resistance of the covering magnetic layer). If the thickness of the base and cover layers are the same, the value of δ becomes maximum due to the absence of the shunting effect. If the thickness of the non-magnetic layer increases, provided that the thicknesses of the base and magnetic layers of the metal do not change, the magnetoresistance ratio monotonically decreases with increasing spacer thickness.

Keywords: Dimensional effect, Magnetoresistance ratio δ , Dieny formula, Resistance shunting.

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1. INTRODUCTION

The development of modern electronics requires the development and implementation of new functional elements based on magnetically inhomogeneous film materials. This stimulates intensive studies of the physical properties of multilayer structures, granular films, composite materials, manganites, etc. [3-6]. Structures with spin-dependent scattering of polarized electrons in the volume of magnetic metal layers and at conductor interfaces are of particular interest due to their wide applications [7-10].

Multilayer film systems based on Fe_xCo_{1-x} ferromagnetic alloy and copper, in which spin-dependent scattering of charge carriers is implemented, are widely used as effective magnetic field sensors, in digital magnetoresistive memory devices, automotive electronics, biomedical technologies, etc. Despite this, there is still a need for further experimental and theoretical studies of film structures that meet additional requirements (minimum size, high sensitivity, high reproducibility of samples, etc.). It is also important to predict the behavior of the magnetoresistance of multilayer magnetically ordered systems with a change in the metal layer thickness. The solution of such problems is possible only using a comprehensive approach to study the physical properties of film systems.

The goal of this work is to experimentally and theoretically study the dimensional dependence of the giant magnetoresistance (GMR) value in three-layer magnetic films (sandwiches) based on Fe_xCo_{1-x} alloy and copper using generalized Dieny formulas [1, 2].

2. METHOD AND TECHNIQUE OF EXPERIMENT

Multilayer film systems with layer thicknesses of 1-50 nm were obtained in the vacuum chamber at a

residual atmosphere gas pressure of 10^{-4} Pa. Layer-bylayer film condensation was carried out by metal evaporation from independent sources (Cu from a tungsten ribbon, Fe_xCo_{1-x} from an electron-beam gun). The source materials for obtaining Fe_xCo_{1-x} layers were massive alloys of the appropriate composition.

The results of the study of the chemical composition of the initial alloy and the obtained films showed that they coincide within the measurement error (measurement error did not exceed 2 %).

Film condensation was carried out on the substrate at room temperature with a speed $\omega = 0.5 \cdot 1$ nm/s, depending on the operating conditions of the evaporators. Glass plates with pre-deposited contact pads were used as substrates for research of the magnetoresistive properties. The construction of the manufactured substrate holder allowed to obtain in one technological cycle two film samples with different thickness of the nonmagnetic layer (spacer) and with commensurate thicknesses of ferromagnetic metal layers. The geometric dimensions of the films for measuring their electrical resistance were specified by windows in nichrome foil mechanical masks, which were produced with high accuracy.

The film thickness was determined using a quartz resonator with a measurement error of \pm 5 %.

Measurements of the longitudinal and transverse magnetoresistance and thermomagnetic treatment of the films were performed on a special installation under conditions of ultrahigh oil-free vacuum 10^{-6} - 10^{-7} Pa in a magnetic field with induction up to B = 150 mT.

3. RESULTS OF EXPERIMENTAL RESEARCH

Experimental studies of the magnetoresistive properties of $Fe_xCo_{1-x}/Cu/Fe_xCo_{1-x}$ three-layer films made it possible to establish the effect of the thickness of both the covering magnetic layer and the nonmagnetic layer on the GMR value. It should be noted that negaYU.M. SHABELNYK, D.I. SALTYKOV ET AL.

tive magnetoresistance was registered for three-layer films unannealed and annealed at temperatures of 400 and 550 K with thickness of magnetic layers $d_m = 10$ -30 nm and thickness of nonmagnetic Cu layers $d_n = 5$ -15 nm for both longitudinal and transverse geometry of measurements (Fig. 1). In the studied structures, the change in the magnetic configuration (transition from antiferromagnetic ordering of magnetic moments to ferromagnetic ordering and reverse transition) occurred under the influence of a relatively weak external magnetic field. Due to the change in the magnetic configuration, the sample resistance decreased, i.e., the GMR effect was realized. Note that in such magnetically ordered three-layer films, there was practically no indirect magnetic interaction between the ferromagnetic layers due to the relatively large thickness of the non-magnetic layer ($d_n = 5-15$ nm).



Fig. 1 – Field dependences of GMR for as-deposited (a) and annealed at a temperature of 550 K (b) $\text{Fe}_{0.2}\text{Co}_{0.8}/\text{Cu}/\text{Fe}_{0.2}\text{Co}_{0.8}$ three-layer films ($d_{m1} = d_{m2} = 20$ nm, $d_n = 7$ mn). The measurement temperature is 300 K

Fig. 2 shows the dependence of the GMR value on the covering magnetic layer thickness d_{m2} (the layer condenses on the nonmagnetic layer) normalized to the base magnetic layer thickness $d_{m2} = \text{const}$ (the layer condenses on the substrate) in Fe_{0.5}Co_{0.5}/Cu/Fe_{0.5}Co_{0.5}/S three-layer films. As can be seen from Fig. 2, the dependence is nonmonotonic. The reasons for such behavior will be analyzed in detail in a theoretical study of the corresponding dimensional dependence. Here we only note that at a small effective thickness of the covering magnetic layer (up to 5-10 nm), nonmagnetic solid solutions can be formed, i.e., the covering magnetic layer is not formed and, accordingly, the GMR effect is not realized.



Fig. 2 – Dependence of the GMR value on the thickness d_{m2} of the covering magnetic layer normalized to the base layer thickness $d_{m2} = \text{const}$ of Fe_{0.5}Co_{0.5}/Cu/Fe_{0.5}Co_{0.5}/S three-layer film. The base layer thickness is $d_{m1} = 25$ nm, the layer thickness is $d_n = 4$ nm

Fig. 3 dependence of shows the the magnetoresistance ratio δ on the copper layer thickness for as-deposited and thermostabilized Fe0.2Co0.8/Cu/Fe0.2Co0.8 three-layer system at 400 and 550 K. The maximum (amplitude) GMR value was observed at the nonmagnetic layer thickness $d_n = 3$ -8 nm, depending on the concentration of components in the magnetic layers.



Fig. 3 – Dependence of the magnetoresistance ratio δ on the nonmagnetic layer thickness for as-deposited (1) and annealed at 400 K (2) and 550 K (3) Fe_{0.2}Co_{0.8}/Cu/Fe_{0.2}Co_{0.8}/S three-layer films ($d_m = d_{m1} = d_{m2} = 25$ nm)

It should also be noted that the effective layer thickness at which the maximum GMR value was observed depended on the heat treatment conditions. The minimum effective copper layer thickness, at which the isotropic dependence of magnetic resistance on induction was detected, was $d_n = 3.4$ nm for the as-deposited

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samples. An increase in the annealing temperature caused an increase in the minimum layer thickness to 10 nm, at which an isotropic field dependence of GMR was observed due to an increase in the width of the split boundaries (interfaces) between the metal layers. The maximum GMR value at room temperature, which was 3.5 %, was recorded for Fe0.1Co0.9/Cu/Fe0.1Co0.9 film with $d_n = 7 \text{ nm}$ after annealing at a temperature of 550 K [11]. A further increase in the thickness of copper layers led to a decrease in the magnetoresistance caused by the scattering of charge carriers in their volume. Oscillations of the GMR amplitude depending on the nonmagnetic layer thickness were not recorded. The reason for this was the large effective thickness of Cu layers. As a result, the exchange interaction between the magnetic layers was practically absent.

4. THEORETICAL ANALYSIS OF THE DEPENDENCE OF GMR ON COVERING MAGNETIC LAYER THICKNESS

Quantitatively, the GMR effect [12, 13] is described by the magnetoresistance ratio (MRR), which is determined by the change in the specific magnetoresistance $\left(\overline{\rho_{\rm ap}}(0) - \overline{\rho_{\rm p}}(B)\right)$ of the sandwich as a result of its magnetization reversal by the external magnetic field induction *B* normalized to the resistance $\overline{\rho_{\rm ap}}(B)$:

$$\delta = \frac{\overline{\rho_{\rm ap}}(0) - \overline{\rho_{\rm p}}(B)}{\overline{\rho_{\rm p}}(B)} = \frac{\overline{\rho_{\rm ap}}(0)}{\overline{\rho_{\rm p}}(B)} - 1, \qquad (1)$$

where $\overline{\rho_{\rm ap}}(0)$ is the specific conductor resistance averaged over the sandwich thickness in the absence of an external magnetic field, i.e., when the sandwich implements an antiferromagnetic configuration (the directions of the spontaneous magnetization vectors **M** in the magnetic layers of the metal are antiparallel), $\overline{\rho_{\rm p}}(B)$ is the specific sample resistance averaged over the thickness of the magnetically ordered three-layer film in the presence of an external magnetic field, i.e., when a ferromagnetic configuration is implemented in the sandwich (the directions of the spontaneous magnetization vectors **M** in the magnetic layers of the metal coincide).

It was experimentally and theoretically substantiated that the dimensional dependences of the transport coefficients (conductivity, resistivity, magnetoresistance, etc.) on the metal layer thickness in both nonmagnetic [14-16] and magnetic [17-20] multilayer structures depend on the ratio between the thickness of the layers of the conductor's metal. Thus, in particular, the nature of the behavior of MRR δ depending on the

change in the value of
$$d_{m2,m1} = \frac{d_{m2}}{d_{m1}}$$
, $\left(d_{m1} = \mathrm{const}\right)$ de-

pends on the sign of the inequality between the thickness d_{m2} of the covering magnetic layer and the total thickness of the nonmagnetic layer (spacer) and the base magnetic layer, i.e., $d_n + d_{m1}$.

If inequalities $l_{int}^s \ll l_{mj}^s, l_n^s$ $(l_{mj}^s, l_n^s$ are the mean free paths of spin-polarized electrons in the *j*-th magnetic

layer and in the nonmagnetic layer, respectively, $s = \pm (\uparrow \downarrow)$ are the spin indices that determine the sign of the projection of the electron spin on the direction of the spontaneous magnetization vector **M** in the magnetic layer of the conductor) and $l_{int}^s \ll \sqrt{Dt_D}$ (*D* is the mutual diffusion coefficient, t_D is the diffusion time) [20, 21] are observed for mean free paths of electrons l_{int}^s in the transition regions between the spacer and the magnetic layers of the metal, then the boundaries of separation of metal layers can be modeled using geometric planes. For this reason, the variable thickness d_{m2} of the covering magnetic layer should be naturally normalized to the total thickness $d_n + d_{m1}$. Accordingly, the approximating Dieny formula [1] can be written as [23]:

$$\delta(d_{m2,m1}) \sim \frac{1 - e^{-\frac{d_{m2,m1}}{1 + d_{n,m1}}}}{1 + \frac{d_{m2,m1}}{1 + d_{n,m1}}},$$
(2)

where $d_{n,m1} = d_n/d_{m1}$ is the thickness of the nonmagnetic layer d_n which is normalized to the thickness of the base magnetic layer d_{m1} .

Let us investigate formula (2) for the boundary values of the thickness d_{m2} . In the region of small values of the covering magnetic layer thickness d_{m2} in comparison with the total thickness of the spacer and the base magnetic layer, i.e., in the inequality:

$$d_{m2,m1} << 1 + d_{n,m1} , \qquad (3)$$

MRR δ increases linearly with increasing thickness of the covering magnetic layer d_{m2} (Fig. 2, section (0, I)):

$$\begin{split} &\delta\left(d_{m2,m1}\right) \sim \frac{d_{m2,m1}}{1+d_{n,m1}} \approx d_{m2,m1}\left(1-d_{n,m1}\right) \equiv \\ &\equiv \frac{d_{m2}}{d_{m1}} \left(1-\frac{d_n}{d_{m1}}\right) \end{split} \tag{4}$$

and the effect, as follows from the asymptotic formula (4), is insignificant. This is due to the fact that in the specified range of thicknesses (3), the current in the sandwich is shunted by the base magnetic layer and the nonmagnetic layer. As the thickness d_{m2} increases, the value of the current in the covering magnetic layer increases, which leads to an increase in the MRR (Fig. 2, section (I, II)). It also follows from expression (4) that under conditions of a detected (constant) thickness of the covering and base magnetic layers of the metal, MRR decreases with increasing thickness of the nonmagnetic layer (for more details, see the next paragraph).

In the opposite case, in comparison with (3), the following inequality holds:

$$d_{m2,m1} >> 1 + d_{n,m1} \,. \tag{5}$$

An asymptotic formula

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$$\delta(d_{m2,m1}) \sim \frac{1+d_{n,m1}}{d_{m2,m1}} \approx \frac{1}{d_{m2}} (d_{m1} + d_n)$$
(6)

correctly describes the dimensional dependence only in the range of thicknesses $\delta(d_{m2,m1})$ $d_{m2,m1} > 1 + d_{n,m1}$, i.e., MRR decreases with increasing covering magnetic layer thickness d_{m2} since the opposite situation is observed: the resistances of the base magnetic layer and the nonmagnetic layer are shunted by the resistance of the covering magnetic layer (Fig. 2, section (II, III). In reality, as was experimentally established [4, 9, 24], in the specified range of thicknesses of the covering magnetic layer, MRR δ decreases exponentially with increasing d_{m2} (Fig. 2, section (II, IV). Such discrepancy between the experimental research results and the corresponding theoretical calculations is due to the fact that in the case of inequality (5), the covering magnetic layer of the metal becomes thick (the mean free path of spin-polarized charge carriers in the magnetic layer $l_{m2}^{s} << d_{m2}$, $s = \pm (\uparrow \downarrow)$ determines the sign of the projection of the electron spin on the direction of spontaneous magnetization). As a result, the covering

spontaneous magnetization). As a result, the covering and base magnetic layers of the metal become "independent" in the sense that there will be no interaction between them (charge carriers do not tunnel from one magnetic layer to another).

Taking into account the opposite behavior of MRR (increase in the region of small thicknesses d_{m2} and decrease in the region of large values of d_{m2}) for the boundary values d_{m2} , it is advisable to study expression (2) for the presence of an extremum. To do this, we differentiated expression (2) with respect to $d_{m2,m1}$ and equated the result to zero. As a result, we obtained the transcendental equation:

$$e^{-\frac{d_{m2,m1}}{1+d_{n,m1}}} \left(2 + \frac{d_{m2,m1}}{1+d_{n,m1}}\right) - 1 = 0, \qquad (7)$$

whose approximate solution is the function

$$d_{m_{2,m_1}}^{\text{extr}} = 1 + d_{n,m_1} \,. \tag{8}$$

In the case of equation (8), the value of MRR δ acquires a maximum (amplitude) value due to the lack of the shunting effect.

5. THEORETICAL ANALYSIS OF THE DEPENDENCE OF GMR ON NONMAGNETIC LAYER THICKNESS

During the experimental study of MRR δ , depending on the change in the thickness of the nonmagnetic layer d_n , in order to avoid the shunting effect, the thickness of the magnetic layers is usually chosen equal ($d_{m1} = d_{m2} = d_m$). Therefore, the thickness of the layer d_n in the Dieny formula [14] should be naturally normalized to a double magnetic layer, i.e.:

$$\delta(d_{m2,m1}) \sim \frac{e^{-\frac{d_n}{2d_m}}}{1 + \frac{d_n}{2d_m}} \equiv \frac{e^{-0.5d_{n,m}}}{1 + 0.5d_{n,m}}, \qquad (9)$$

where $d_{n,m}$ is the thickness of the nonmagnetic layer normalized to the thickness of the magnetic layer.

If the inequality is met:

$$d_n \ll 2d_{n,m},\tag{10}$$

that is, when the spacer is ultrathin, the asymptotic expression of formula (9) can be written as:

$$\delta\left(d_{m2,m1}\right) \sim 1 - \frac{d_n}{d_m}, \qquad (11)$$

that is, MRR decreases linearly with increasing thickness of the nonmagnetic layer (Fig. 3, curve 3, section (I, II). This decrease is due to the fact that with increasing thickness of the nonmagnetic layer, there is a probability of scattering of spin-polarized charge carriers in the volume of the nonmagnetic layer, which leads to a decrease in the interaction between the magnetic layers of the metal and, as a result, to a decrease in the amplitude of GMR.

In the opposite case, in comparison with inequality (10), $d_n >> 2d_{n,m}$, i.e., when the spacer is thick enough. In this case, the magnetic metal layers become independent in the sense that spin-polarized charge carriers do not pass from one magnetic layer to another through the nonmagnetic layer (in point 3, the magnetic layers become independent due to the fact that the covering layer becomes thick). Therefore, the exponent in formula (9) asymptotically tends to zero. MRR also goes to zero ($\delta \rightarrow 0$), and, accordingly, the GMR effect is absent (Fig. 3, curve 3, section (II, III).

6. CONCLUSIONS

Thus, in the region of small thicknesses compared to the thickness of the base magnetic layer, i.e., when the inequality $d_{m2} \ll (d_n + d_{m1})$ is satisfied, the value of δ is negligible due to shunting of the covering layer resistance by the resistances of the base layer and the nonmagnetic layer. In the case of the opposite inequality $d_{m2} \gg (d_n + d_{m1})$, i.e., in the region of large thicknesses of the covering layer, the opposite effect is observed. Namely, the shunting effect of the resistance of the base layer and the nonmagnetic layer and the nonmagnetic layer by the covering layer resistance. If the equality $d_{m2} = (d_n + d_{m1})$ is fulfilled, the value of δ acquires a maximum (amplitude) value due to the absence of the shunting effect.

As the thickness of the nonmagnetic layer increases, provided that the thickness of the base and magnetic layers of the metal does not change, MRR monotonically decreases.

Note that the above formulas can be used to substantiate the dimensional dependence of the transport coefficients on the thickness of the metal layer in nonmagnetic multilayer structures. DIMENSIONAL EFFECTS IN MAGNETORESISTIVE PROPERTIES ...

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Розмірні ефекти в магніторезистивних властивостях у тришарових плівках

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Експериментально і теоретично з використанням узагальнених формул Дієни [1, 2] досліджені розмірні ефекти в магніторезистивних властивостях тришарових магнітовпорядкованих плівках $Fe_xCo_{1-x}/Cu/Fe_xCo_{1-x}$ отриманих методом пошарової конденсації металів з наступною термообробкою в інтервалі температур 300+550 К. Показано, що у випадку, коли товщина покриваючого шару значно менша (більша) товщини базового шару числове значення магніторезистивного відношення δ мізерно мале внаслідок шунтуванням опору накривного шару опорами базового шару та немагнітного прошарку (шунтуванням опорів базового шару та немагнітного прошарку опором верхнього магнітного шару). Якщо ж товщини базового і покриваючого шарів однакова, то величина δ набуває максимального значення в силу відсутності ефекту шунтування. У разі збільшення товщини немагнітного прошарку за умови, що товщини базового та магнітного шарів металу не змінюються, магніторезистивне відно-шення монотонно зменшується зі збільшенням товщини спейсера.

Ключові слова: Розмірний ефект, Магніторезистивне відношення б, Формула Дієни, Шунтування опору.