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Structure and Magnetoresistive Properties of Three-layer Film Systems Based on Permalloy and Copper

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Structural and phase composition and magnetoresistive properties of three-layer film systems based on permalloy and copper are investigated. The samples are obtained by the method of layer by layer condensation followed by heat treatment in the 300-700 K temperature range. It is shown that the spin-dependent electron scattering is realized in the layer thickness range of d_{Cu} = 6-15 nm and d_{Py} = 25-40 nm for as-deposited and annealed at 400 K samples. It is established that the maximum isotropic magnetoresistance is observed after annealing of the samples at 400 K, and annealing at 550 K leads to the emergence of anisotropic magnetoresistance.

Keywords: Multilayer film system, Crystal structure, Phase composition, Anisotropic magnetoresistance, Spin-dependent electron scattering.

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1. INTRODUCTION

The study of the properties of new magnetic materials (multilayer structures, granular nanomaterials, manganites, etc.) has made it possible to discover a number of important magnetoresistive effects: giant magnetoresistance (GMR), tunneling magnetoresistance, colossal magnetoresistance, which are of interest from the point of view of fundamental research and practical applications of these materials [1-2].

Multilayer film systems based on permalloy and copper, in which spin-dependent electron scattering occurs, are widely used as sensitive elements of magnetic-field sensors, digital magnetoresistive memory devices, automotive electronics, biomedical technologies, etc. [3]. However, there is a need for further search and experimental study of film structures based on permalloy [4], which would correspond to additional requirements (reducing sensor size, increasing its sensitivity, providing repeatability of parameters, etc.). It is also important to predict the behavior of the electrophysical and magnetoresistive properties of multilayer systems by changing metal layer thickness, structure, temperature and magnitude of the external magnetic field. The solution of these problems is only possible with the use of a complex approach to study the physical properties of film systems.

The aim of this work was to study the structural and phase state, size and temperature dependence of the longitudinal and transverse magnetoresistance (MR) as well as the electrical resistance of three-layer magnetic films based on permalloy (Py) and Cu

2. EXPERIMENTAL

Multilayer film systems with (1÷50) nm thick layers were obtained in a vacuum chamber at the residual gas pressure of 10^{-4} Pa. The alternate condensation of the films was performed as a result of evaporation of metals from independent sources (Cu – from a tungsten wire, Py – from an electron beam gun). $Fe_{0.2}Ni_{0.8}$ bulk alloy was the starting material to obtain Py layers.

Film condensation was carried out at room temperature of the substrate with a rate of ω = (0.5÷1) nm/s depending on the evaporator operating modes. Glass plates with pre-deposited contact pads were used as substrates to study the magnetoresistive properties. The design of the manufactured substrate-holder allowed to obtain in one technological cycle two film samples with different thicknesses of a nonmagnetic interlayer and with similar thicknesses of the ferromagnetic layers. The geometric sizes of the films to measure their electrical resistance were specified by windows produced with high accuracy in mechanical masks of nichrome foil.

The film thickness was determined using the micro-interferometer MII-4 with a laser light source and a computer system for recording the interference pattern that allowed to improve the measurement accuracy, especially in the thickness range of d < 50 nm (up to 20 %).

The results of the study of the chemical composition of the initial alloy and the films obtained by X-ray spectral microanalysis show that they coincide within the measurement error (it did not exceed 1.5 %).

The investigation of the crystal structure and phase composition of the films was performed by electron-microscopy and electron diffraction methods (transmission electron microscope PEM-125K)

Measurements of the longitudinal and transverse MR (magnetic field in the film plane) and thermomagnetic film treatment were carried out in a special installation described in [5] in conditions of ultrahigh oil-free vacuum (10^{-6}÷10^{-7}) Pa in a magnetic field of induction to B = 200 mT. The films were annealed according to the scheme "heating – exposure at a temperature of 400, 550 and 700 K for 15 minutes – cooling".

The magnitude of the longitudinal and transverse MR

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of the film samples was calculated by the following formula: $(R(B) – R(B_C)) / R(B_C)$, where $R(B)$ is the sample resistance in a magnetic field with induction B; $R(B_C)$ is the sample resistance in the coercivity field B_C. In the absence of hysteresis of the magnetoresistive effect, the MR was calculated by the formula $(R(B) – R(0)) / R(0)$, where $R(0)$ is the sample resistance in the absence of a magnetic field.

3. STRUCTURAL AND PHASE COMPOSITION OF THE SAMPLES

Electron microscopic studies have shown that all unannealed single-layer permalloy films are polycrystalline with an average grain size of about 5 nm (Fig. 1a). Annealing at the temperature of 700 K leads to a 5÷6-fold increase in the crystallite size (Fig. 1b). The phase composition of as-deposited and annealed single-layer permalloy films (Fig. 1a, b) corresponds to the fcc-$FeNi_3$ phase with the lattice parameter a = 0.354-0.356 nm. These data agree well with the results given in [6, 7].

As a result of electron microscopy and electron diffraction studies of three-layer Py/Cu/Py films, it was established that all as-deposited films are polycrystalline with grain sizes less than 5 nm. Electron microscopic images of the structure of film systems (Fig. 1c-f) are similar to the structure of single-layer permalloy films.

For Py/Cu/Py films with a thickness of Cu interlayer of 5÷20 nm in the as-deposited state, the fcc-phase with the lattice parameter a = 0.353-0.357 nm is observed (see Fig. 1c, e). The lines belonging to the fcc-phase of permalloy and fcc-Cu are not separated in the electron diffraction patterns due to similar interplanar distances. The implementation of spin-dependent electron scattering in three-layer structures is the confirmation of identity preservation of Py and Cu layers.

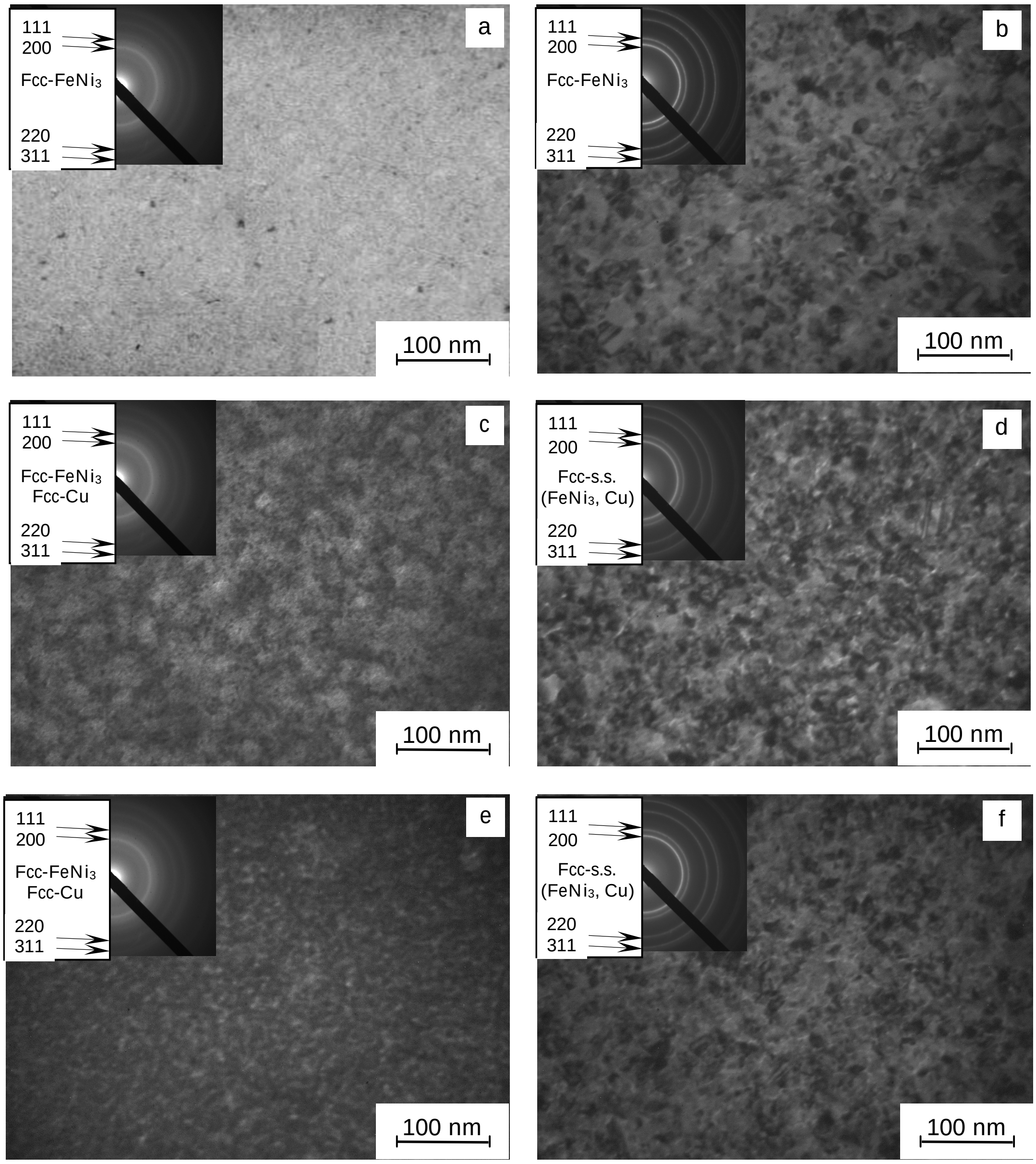

Fig. 1 – Microstructure of single-layer permalloy films (a, b)and three-layer Py/Cu/Py film structures (c-f) in as-deposited (a, c, e) and annealed at a temperature of 700 K (b, d, f) state (a, b – d_{Py} = 40 nm; c, d – $2d_{Py}$ = 60 nm, d_{Cu} =7 nm; e, f – $2d_{Py}$ = 80 nm, d_{Cu} = 5 nm). The corresponding electron diffraction patterns are illustrated on the insets

After annealing of Py/Cu/Py films at a temperature of 700 K and due to an increase in the crystallite size to 30 nm, the width of the diffraction rings decreases (see Fig. 1d, f). At that, a single-phase state is clearly fixed, and changes in the lattice parameter are not recorded in the electron diffraction patterns. A solid solution based on permalloy is probably formed in this system. This is confirmed by the data of [8, 9].

4. MAGNETORESISTIVE PROPERTIES OF SINGLE- AND THREE-LAYER FILMS

Behavior of the field dependences and the MR value of three-layer Py/Cu/Py/Sub (Sub is the substrate) films significantly depends on the thicknesses of both Cu interlayer and Py layers. We experimentally determined the layer thickness ranges, in which positive longitudinal (resistance increases when applying an external magnetic field to a demagnetized sample) and negative (correspondingly, resistance decreases) magnetoresistive effects are observed. Let us consider in detail the features of the MR of three-layer films.

A positive longitudinal magnetoresistive effect, which is a sign of the anisotropic magnetoresistance (AMR), is observed for all studied as-deposited and annealed at 400, 550 and 700 K three-layer samples with a nonmagnetic interlayer thickness of 1-5 nm, irrespective of the magnetic layer thickness [10-12]. The presence of AMR for small effective copper thicknesses (d_{Cu} = 1-5 nm) is associated with the absence of a structurally continuous nonmagnetic interlayer. Therefore, there is a rather strong direct coupling between the magnetic layers. This interaction interferes with a separate reversal of the layers, and, thus, also does not lead to the occurrence of spin-dependent electron scattering. Such a film can be approximated considered as a single-layer permalloy film with an effective thickness of $2d_F$ and inclusion of Cu islands. The AMR value for unannealed three-layer films, as in the case of single-layer permalloy films (d_{Py} = 50-100 nm), does not exceed 0.1 % at room temperature. After annealing at the temperature of 700 K, the MR increases to 0.5 % for three-layer films and to 2 % for single-layer Py films. It should be noted that the AMR is also observed for three-layer films with a magnetic layer thickness of 10-20 nm and d_{Cu} = 3-10 nm.

Slightly different results on the study of the MR are observed for three-layer films in the thickness range of d_{Cu} = 6-15 nm and d_{Py} = 25-40 nm. Isotropy of the field dependences, which is a sign of the giant MR (GMR) effect (Fig. 2a, b), takes place for unannealed and annealed at 400 K films. A negative MR value in Fig. 2-Fig. 4 shows that the electrical resistance of a demagnetized sample decreases, when a magnetic field is applied. The cause of the isotropy of the field dependences of the MR is the implementation of the mechanism of spin-dependent scattering of conduction electrons in the bulk of the ferromagnetic layers and at the layer interfaces. It should be noted that the magnitude of the transverse MR for all samples is slightly larger than that of the longitudinal one. This is caused by the presence of anisotropic MR of the ferromagnetic layers. The MR value for as-deposited samples with spin-dependent electron scattering is equal

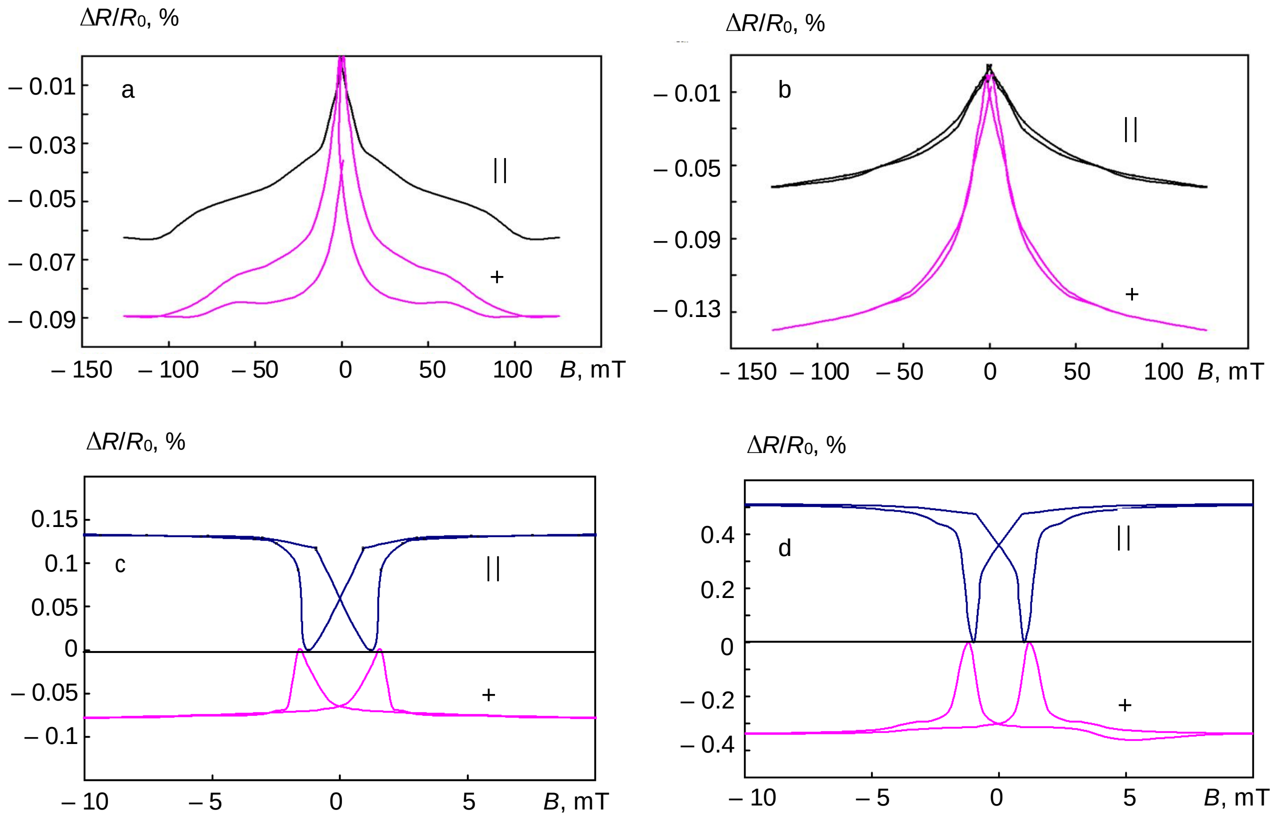

Fig. 2 – Dependence of the longitudinal (||) and transverse (+) MR on the magnetic field induction for as-deposited and annealed at different temperatures three-layer Py/Cu/Py structure with $2d_{Py}$ = 70 nm and d_{Cu} = 7 nm (a – as-deposited film; b – T_{ann} = 400 K; c – T_{ann} = 550 K; d – T_{ann} = 700 K). The measurement temperature is 300 K

to 0.1-0.4 %. Small values of MR can be explained by a relatively large copper interlayer thickness, resulting in shunting of the effect and the formation at the interface of a permalloy-based solid solution that leads to "loss of information" by electrons about their spin when they are scattered. The theoretical analysis performed in [13, 14] indicates that the maximum value of the GMR effect is determined only by the value of the volume asymmetric spin-dependent electron scattering α ($\alpha = \rho^+/\rho^-$, where ρ^+ and ρ^- are the resistivity of different spin channels); and it does not matter, in which spin channel the mentioned asymmetry will be greater. The calculations carried out on the basis of experimental data show that the value of the parameter α for Py layers is small (α_{FeNi} = = 1.046). Small values of the parameter α are due to, in our opinion, the formation of a solid solution that leads to low values of the GMR effect in three-layer film systems based on permalloy and copper. A decrease in the parameter α for the Fe/Cr system was observed by the authors of [15]. They associate this change in α with the great imperfection of interfaces and an increase in the processes of diffuse electron scattering.

Behavior of the field dependences after low-temperature annealing (T_{ann} = 400 K) does not change, the magnitude of the longitudinal MR is almost constant, while the magnitude of the transverse MR increases approximately 1.3-1.5 times. The latter is probably associated with an increase in the effect of a positive longitudinal MR of the ferromagnetic layers after annealing.

Annealing at 550 K leads to the appearance of AMR with an amplitude of 0.1-0.2 % (Fig. 2c). The most important reason for the AMR appearance is the interdiffusion of atoms and the formation of a Py-based solid solution, which result in a discontinuity of the copper interlayer that is confirmed by electron diffraction patterns. With further annealing of the samples at the temperature of 700 K, the AMR remains, the magnetoresistive hysteresis loops become clearer, and the MR magnitude increases to 0.5-0.7 % (see Fig. 2d).

Let us consider in detail the reasons for the change in the MR after annealing. The phenomenon of change in the magnitude of $(\Delta R/R(B_C))_{max}$ in the annealing process can be caused by both the change in the magnitude of $\Delta R_{max} = R(B_C) - R(B_S)$ (here $R(B_S)$ is the sample electrical resistance measured in the maximum field) and the magnitude of $R(B_C)$. Indeed, in the stepwise annealing process, the magnitudes of $R(B_C)$ and $R(B_S)$ vary. However, their changes occur in such a way that the ratio $(\Delta R/R(B_C))_{max}$ can both increase and decrease. Therefore, for a better understanding of the influence of T_{ann} on the MR magnitude, the study of the dependence of the resistivity and ΔR_{max} magnitude on the annealing temperature was carried out. As seen from the data shown in Fig. 3a, an irreversible decrease in the electrical resistance as a result of healing of the crystal structure defects and an increase in the crystallite size occurs during the annealing. At that, low-temperature annealing does not lead to a significant change in the ΔR_{max} magnitude for both the longitudinal and transverse measurement geometries (Fig. 3b). Thus, the processes, which would change the conditions of spin-dependent electron scattering, in particular, a change in the roughness amplitude of the interfaces, an increase in the crystallite size, etc., do not occur in the films.

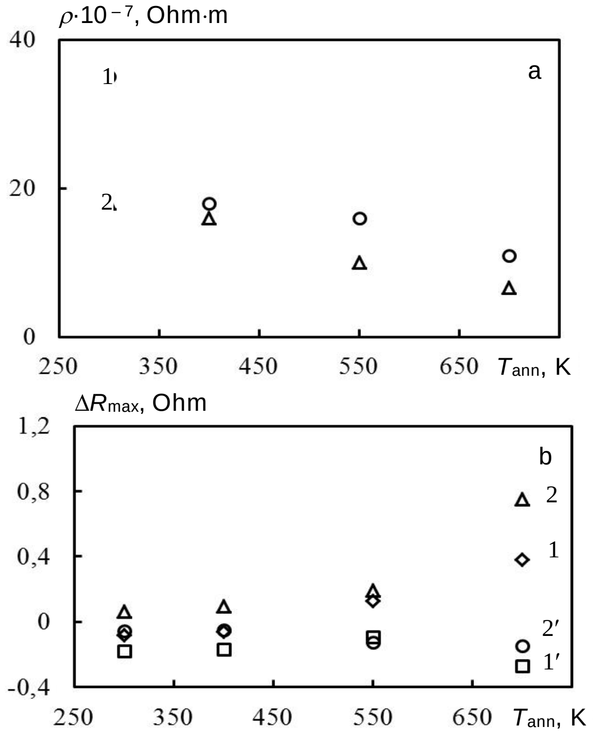

Fig. 3 – Dependence of the resistivity ρ (a) and ΔR_{max} magnitude (b) on the annealing temperature for three-layer Py/Cu/Py films with layer thicknesses of 1 – d_{Py} = 30 nm, d_{Cu} = 7 nm and 2 – d_{Py} = 50 nm, d_{Cu} = 5 nm

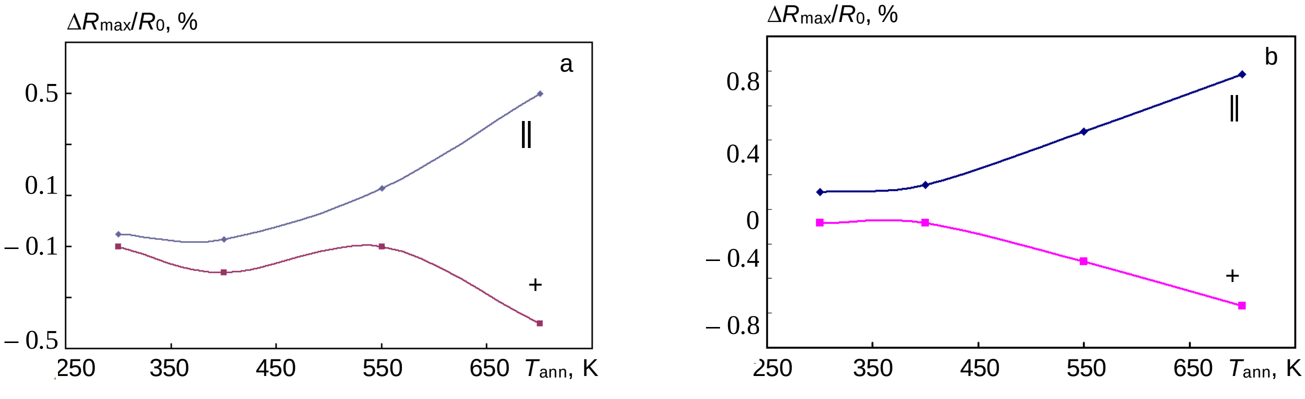

Fig. 4 – Dependence $\Delta R_{max}/R_0$ on the annealing temperature for three-layer Py/Cu/Py films with the following layer thicknesses: a – d_{Py} = 30 nm, d_{Cu} = 7 nm; b – d_{Py} = 50 nm, d_{Cu} = 5 nm

Therefore, an increase in the $(\Delta R/R(B_C))_{\max}$ magnitude (Fig. 4a) occurs only due to a decrease in the $R(B_C)$ magnitude. The MR decrease in the transition to AMR is caused by the $\Delta R_{\max}$ decrease. For all samples with AMR, the dependences of the MR on the annealing temperature are monotonic, and MR increases with increasing annealing temperature. It should be noted that for such samples in the stepwise annealing processes, there occurs an increase in $\Delta R_{\max}$ and a decrease in R_0, i.e. (B_C), that leads to a significant MR increase (5-10 times) (Fig. 4b).

5. CONCLUSIONS

1. The study of the phase composition of Py/Cu/Py films has shown that the fcc-phase with the lattice parameter a = 0.353-0.356 nm is observed in as-deposited films. A solid solution ($FeNi_3$, Cu) is formed in the given system after annealing at 700 K.

2. The spin-dependent scattering of conduction electrons, which is manifested in the isotropy of field dependences, is implemented for as-deposited and annealed at 400 K three-layer samples with d_{Py} = 25-40 nm and d_{Cu} = 6-15 nm. Annealing at temperatures higher than 550 K for the samples with initial isotropic field dependences leads to the appearance of MR anisotropy because of the destruction of a nonmagnetic interlayer.

3. The experimental results obtained indicate a possible thermal stabilization annealing of magnetoresistive permalloy and Cu based elements in vacuum at a temperature of 400 K immediately after their deposition.

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