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Magnetoresistive Effects and Magnetic Parameters of Nanocrystalline Films Based on Co, Fe, Ag, and Cu

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Here the results of the experimental studies of the magnetoresistive effect in three-layer nanocrystalline magnetic films based on Co/Ag/Fe/S and Co/Cu/Fe/S are presented. The influence of the heat treatment on the value of their magnetoresistive rate is discussed. It is established that the maximum value of the effect is observed in the presence of ultrathin layer between the magnetic layers with comparable thicknesses.

Keywords: Giant magnetoresistance, Anisotropic magnetoresistance, Coercivity, Saturation field.

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1. INTRODUCTION

Physical effects in nanosize structures [1-3], such as, in particular, magnetic multilayer nanocrystalline films (multilayers) – periodic systems consisted of alternately deposited ferromagnetic and non-magnetic metals, are of a great scientific interest during last decades. The giant magnetoresistance (GMR) effect [4, 5] is realized in the mentioned structures. On the one hand, this effect is the basis for the formation of the element base of spintronics, and, on the other hand, it allows to expand functional possibilities of the already existed devices of microelectronics.

In spite of the fact that currently a number of peculiarities of the GMR effect is experimentally studied in detail and there is a considerable quantity of publications with theoretical justification of the specified effect, there appears a need in further search and experimental investigation of multilayer film structures, where the GMR effect is realized and which could satisfy additional requirements (minimal geometry, low saturation fields, high reconstruction of the samples, etc.).

The aim of the present work consists in the experimental investigation of magnetoresistive properties and study of the behavior of magnetic parameters which characterize freshly condensed and annealed at different temperatures three-layer asymmetric film systems based on Co and Fe with non-magnetic Ag and Cu sublayers (spacers).

2. EXPERIMENTAL TECHNIQUE

Multilayer film systems with the layer thickness of 1-50 nm were obtained in a vacuum chamber of the plant VUP-5M at the residual gas pressure of 10^{-4} Pa [6]. Alternate condensation of the films was performed by the evaporation of metals of the purity not worth than 99,98% from independent sources (Ag, Cu – from the tungstic tape, and Co, Fe – from the electron-beam gun). Condensation of the films was carried out at the room temperature on the substrates with the rate of $\omega = 0{,}5$-1 nm/s depending on the evaporation modes.

Polished glass plates with pre-deposited contact areas were used as the substrates to study magnetoresistive properties. Design of the produced substrate holder allowed to obtain during one technological cycle two film samples with different thickness of non-magnetic layers. Geometry of the films for measurement of their electrical resistance was specified by windows produced with high precision in mechanical masks of nichrome foil.

Film thickness was measured using the microinterferometer MII-4 with laser light source and computer registration system of interference picture that allowed to increase the measuring precision especially in the thickness range of $d < 50$ nm.

Measurements of the longitudinal and transverse magnetoresistance (MR) and thermal treatment of the films were realized in specially produced plant in the conditions of ultrahigh oil-less vacuum (10^{-6}-10^{-7} Pa in magnetic field with the strength up to $H = 150$ kA/m.

3. MAGNETORESISTIVE PROPERTIES OF MULTILAYER FILMS

For a deeper understanding of the features of the magnetoresistive effect in three-layer Co/Ag/Fe/S and Co/Cu/Fe/S films, the detailed investigations of MR in two-layer magnetic Co/Fe/S films were performed.

Anisotropy of MR which is typical for uniform ferromagnetic materials is observed in all investigated samples. The reason of anisotropic MR in ferromagnets is the interaction of conduction electrons with internal electrons, whose spin moments condition spontaneous magnetization [7]. In Fig. 1 we present the typical dependences of the longitudinal and transverse MR on the external magnetic field strength for unannealed (see Fig. 1a) and annealed at the temperature of 700 K (see Fig. 1b) two-layer Co/Fe/S film ($d_{Fe} = 50$ nm and $d_{Co} = 50$ nm). The value of MR in unannealed films (Fig. 1a) is equal to 0,04-0,06% for the longitudinal and 0,14-0,16% for the transverse MR.

Decrease in both the longitudinal and transverse MR takes place after annealing of the samples at the temperature of 400 K. And decrease in the longitudinal

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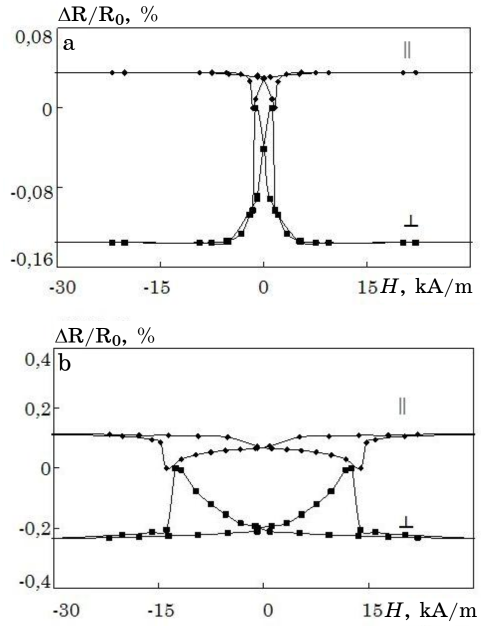

Fig. 1 – Dependence of the longitudinal (||) and transverse (⊥) MR on the magnetic field strength for unannealed (a) and annealed at the temperature of 700 K (b) Co(50 nm)/Fe(50 nm)/S film system

MR is much more (2-3 times) in comparison with the transverse MR (1,2-1,5 times). Decrease in MR, in our opinion, is conditioned by the decrease in the level of structural microstrains during thermal treatment.

Increase in the annealing temperature up to 550 K leads to insignificant increase in both the longitudinal and transverse MR, and peaks of the field dependences in this case are shifted toward the region of higher fields.

Annealing to the temperature of 700 K (Fig. 1b) leads to further increase in the value of MR (0,15-0,25% of the longitudinal MR, 0,2-0,3% – of the transverse one) and shift of the peaks to the region of higher fields. In our opinion, the cause of the increase in the value of MR at high-temperature annealing (T_{ann} = 550 K, T_{ann} = 700 K) is the increase in the crystallite sizes and interdiffusion.

Investigation results of magnetoresistive effect in unannealed and annealed at different temperatures three-layer samples have shown that for all studied systems with an effective thickness of non-magnetic sublayers d_{Ag} < 5 nm and d_{Cu} < 3 nm one can observe a positive longitudinal magnetoresistive effect (resistance increases with application of an external magnetic field) that is the indication of anisotropic magnetoresistance. The authors of [8] have shown that in the presence of ultrathin non-magnetic sublayers ($d_N << d_F$,; N = Ag, Cu; F = Co, Fe) in three-layer films there is direct exchange interaction through ferromagnetic "gangways" in non-magnetic spacer that eliminates possibility of the initiation of the GMR effect. Such film can be approximately considered as two-layer film with an effective thickness $2d_F$ with silver (copper) islands between ferromagnetic layers. In unannealed three-layer Co/Ag/Fe/S and Co/Cu/Fe/S films with an effective sublayer thickness d_{Ag} = 5-20 nm (Fig. 2a) and d_{Cu} = 3-15 nm one can observe the GMR phenomenon, whose value is equal to 0,3-1,1% and 0,3-1,2% at room temperature, respectively.

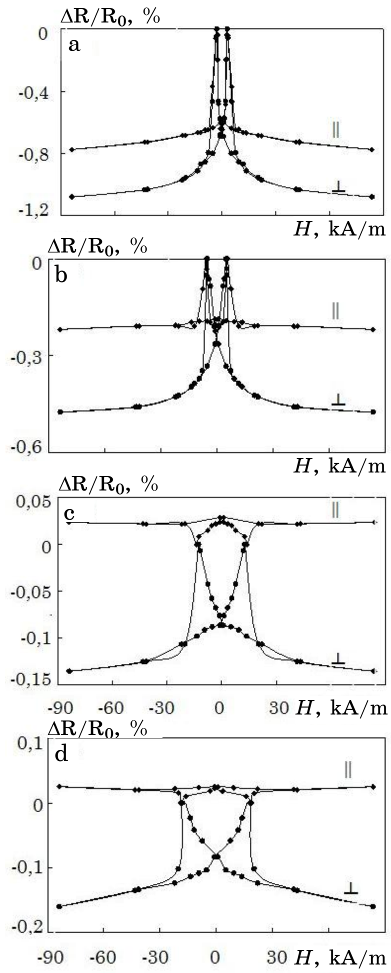

Fig. 2 – Dependence of the longitudinal (||) and transverse (⊥) MR on the magnetic field strength for unannealed (a) and annealed at different temperatures (b-d) Co(30 nm)/Ag(6 nm)/Fe(30 nm)/S film system; b – annealing temperature is 400 K; c – annealing temperature is 550 K, d – annealing temperature is 700 K

In such systems the upper and the bottom magnetic layers have different magnetic properties, in particular, coercitivity. Therefore, magnetic configuration is changed from the antiparallel ordering of magnetic moments to the parallel one and otherwise under the action of an external magnetic field that is similar, for example, to spin-valve structures [10].

At the room temperature and thickness of non-magnetic sublayers d_{Ag} ~ 6 nm, d_{Cu} ~ 3 nm the GMR value is equal to 1,1% and 1,2%, respectively. Small value of the effect at the specified thickness of spacers is conditioned by the presence of "gangways" through non-magnetic sublayer that leads to the appearance of ferromagnetic connection between magnetic layers and, as a result, to the violation of antiparallel configuration. At further increase in the thickness of non-magnetic sublayer d_{Ag} > 6 nm (d_{Cu} > 3 nm) amplitude of the mentioned effect is decreased (Fig. 3) that is conditioned by shunting of the current in high-conductivity non-magnetic sublayer and volume scattering of electrons there. When cooling the samples to the temperature of 150 K irrespective of the layer thickness, view of the hysteresis loops of magnetoresistive effect is not changed, and

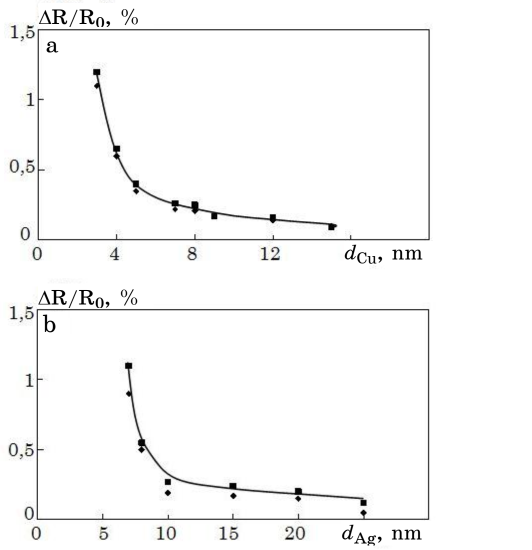

Fig. 3 – Dependence of the longitudinal (♦) and transverse (■) MR for unannealed films (a – Co/Cu/Fe/S; b – Co/Ag/Fe/S) on the thickness of Cu and Ag sublayers

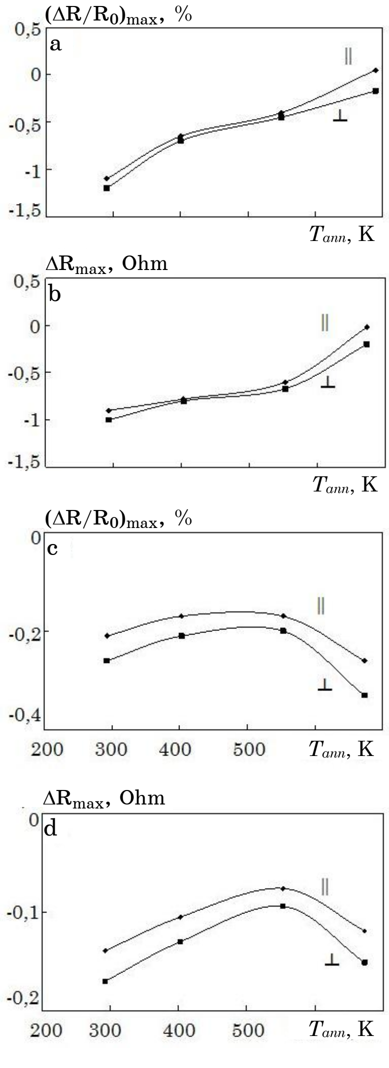

Fig. 4 – Dependences $(\Delta R/R_0)_{max}$ and ΔR_{max} on the annealing temperature T_{ann} for three-layer structures: a, b – Co(35 nm)/Cu(3 nm)/Fe(35 nm)/S; c, d – Co(30 nm)/Cu(7 nm)/Fe(30 nm)/S

increase in the GMR effect and shift of the peaks on the field dependence toward the region of higher fields are only observed.

Decrease in the effect amplitude with temperature increase is connected with electron-phonon scattering with spin flip (spin-flip processes) in non-magnetic sublayer that leads to the decrease in magnetic interaction between magnetic layers of metal due to the decrease in the probability of electron passage from one ferromagnetic layer into another [10].

It is known that thermal treatment is an effective method of impact on the magnetoresistive properties of multilayer structures [10]. In our experiment, all obtained films were exposed to a stepwise vacuum annealing at different temperatures T_{ann}. Duration of thermal treatment at each temperature was 15 min. Now we consider how the value of maximum magnetoresistive ratio $(\Delta R/R_0)_{max}$ is changed after annealing.

In Fig. 4 and Fig. 5 we represent the dependences $(\Delta R/R_0)_{max} = f(T_{ann})$ and $\Delta R = f(T_{ann})$ for Co/Cu/Fe/S and Co/Ag/Fe/S structures with different thickness of non-magnetic sublayers. As seen from the figures, behavior of the mentioned dependences is different depending on the thickness of non-magnetic sublayer.

For films with the thickness of spacers d_{Cu} = 3-5 nm dependence $(\Delta R/R_0)_{max}$ on the annealing temperature T_{ann} has a monotonic behavior. After their annealing at the temperature of T_{ann} = 400 K in magnetic field of the strength of 8 kA/m, a 1,2-1,5-fold decrease in the GMR value is observed. With the increase in the annealing temperature up to T_{ann} = 700 K, anisotropy of magnetoresistance appears.

For three-layer Co/Cu/Fe/S films with non-magnetic sublayers d_{Cu} = 6-8 nm one can separate on the dependence $(\Delta R/R_0)_{max} = f(T_{ann})$ the range of annealing temperatures, where $(\Delta R/R_0)_{max}$ decreases, and the range of higher temperatures, where GMR value gradually increases. For films with spacers d_{Cu} = 6-8 nm, increase in the GMR value is fixed only after annealing at the temperature of T_{ann} = 700 K. In this case, the shape of the field dependences $(\Delta R/R_0)_{max} = f(H)$ is not almost changed. If perform reannealing of the films at the temperatures T < 700 K, the value of $(\Delta R/R_0)_{max}$ also is not changed.

For films with d_{Cu} = 10-20 nm increase in $(\Delta R/R_0)_{max}$ occurs after annealing at T_{ann} = 550 K, but the value of $(\Delta R/R_0)_{max}$ does not exceed 0,5% even after annealing at T_{ann} = 700 K. For three-layer Co/Ag/Fe/S films (Fig. 5) with the thickness of d_{Ag} = 5-12 nm, behavior of the dependence $(\Delta R/R_0)_{max} = f(T_{ann})$ is similar to the corresponding dependences for Co/Cu/Fe/S films with copper sublayers of 3-5 nm. Another situation is observed for the samples with the spacer thickness of d_{Ag} = 15-20 nm. Annealing at the temperature of 400 K leads to almost 2-fold decrease in the value of $(\Delta R/R_0)_{max}$, while during annealing at the temperature of T_{ann} = 550 K the opposite tendency, namely, more than a 2-fold increase in the GMR value, takes place. Further increase in the annealing temperature up to 700 K leads again to a 2-3-fold decrease in the longitudinal magnetoresistance, and a 1,5-2-fold decrease in the transverse magnetoresistance, but field dependences remain isotropic in this case.

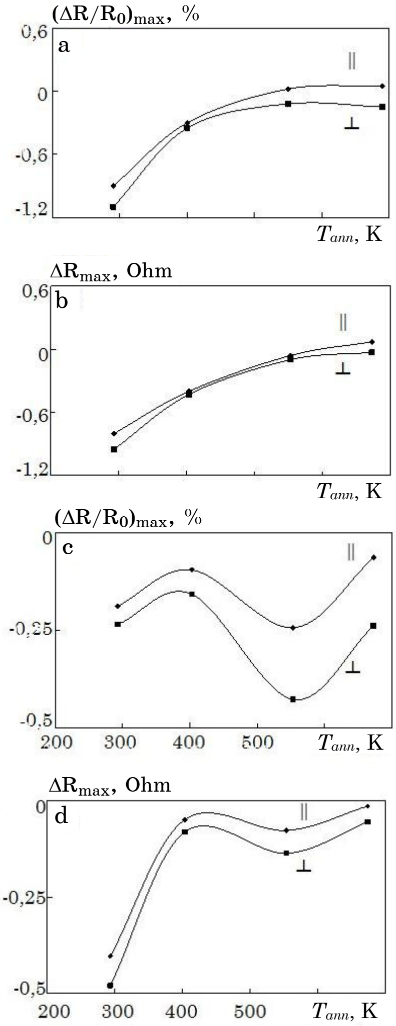

Fig. 5 – Dependences of $(\Delta R/R)_{max}$ and ΔR_{max} on the annealing temperature T_{ann} for three-layer structures: a, b – Co(35 nm)/Ag(6 nm)/Fe(35 nm)/S; c, d – Co(25 nm)/Ag(15 nm)/Fe(25 nm)/S

Increase in T_{ann} up to 550 K for Co/Ag/Fe/S films with sublayers $d_{Ag} < 15$ nm and Co/Cu/Fe/S films with spacers $d_{Cu} = 3$-5 nm (Fig. 4a and Fig. 5a) leads to the decrease in the effect amplitude and appearance of the MR anisotropy that is conditioned by destruction of the initial magnetic order in magnetic layers. Due to the recrystallization processes, interdiffusion of silver, copper, iron, and cobalt atoms, and small thickness of non-magnetic sublayer ($d_{Cu,Ag} = 3$-12 nm), continuity of the interface is violated, and, as a result, a spin-dependent electron scattering disappears. Increase in the value $(\Delta R/R_0)_{max}$ can be connected with both the increase in the difference $\Delta R = R_{max} - R_s$ (here R_{max} is the electrical resistance of the sample measured in the field $H = H_C$; R_s is the electrical resistance measured in the maximum field) and the decrease in R_s. Indeed, these both values were decreased during annealing. Moreover, both R_{max} and R_s were decreased in such a way that ratio $(\Delta R/R_0)_{max}$ was increased in this case. Decrease in the electrical resistance of the films can imply smoothing (decrease in the "roughness" amplitude) of the external boundaries of the films and, respectively, increase in the specular reflection of charge carriers [9]. Ii is more difficult to explain the increase in the difference ΔR, but this phenomenon is also connected with the processes occurring near the external boundaries and interfaces during annealing of semiconductors. As shown in the work [10], magnetoresistive ratio should achieve its maximum value at some optimal roughness of the interfaces, but not at an absolutely smooth surface. In systems where components are not mixed (three layer films studied in the specified experiment also belong to such systems), roughness of the interfaces can be considerably decreased because of smoothing during annealing. This process is possible due to the fact that decrease in the free energy of such structure is achieved at the diffusion of atoms along the interface in order to decrease its area. Change of the roughness toward optimal value takes place during annealing that gives the resulted increase in ΔR and, as a result, increase in $(\Delta R/R_0)_{max}$. We should note, increase in the probability of specular reflection of charge carriers by semiconductor surfaces leads to the increase in the effect amplitude only in the case when volume spin-dependent electron scattering is the dominant mechanism of the GMR effect. If interface spin-dependent electron scattering is the dominant mechanism of the GMR effect, then channeling effect, which decreases the effect amplitude because of the decrease in the interaction between metal layers, appears with the increase in the specular reflection of charge carriers by sample interfaces. At simultaneous action of two mechanisms of asymmetrical spin-dependent scattering in the volume of layers and on their interfaces, the GMR effect is maximum [12].

4. MAGNETIC PARAMETERS OF THE FILMS

Coercitivity H_C and saturation field H_S were determined using magnetoresistive hysteresis loops. Coercitivity was defined by the arrangement of loop peaks, and saturation field was taken to be equal to the field measured on the height of $0,9(\Delta R/R_0)_{max}$ on the curve $\Delta R/R_0 = f(H)$.

As known, in three layer films coercitivity is defined by the component H_C^* conditioned by the exchange coupling of ferromagnetic layers and the component H_C^0 conditioned by the domain wall pinning by structure defects, such as intercrystalline interfaces, dispersion of crystallographic anisotropy axes, surface roughness, etc. [11]

$$H_C = H_C^* + H_C^0 .$$

Dependences $H_C = f(T_{ann})$ for three-layer Co/Ag/Fe/S and Co/Cu/Fe/S films with different thickness of silver and copper sublayers are illustrated in Fig. 6a.

For all Co/Ag/Fe/S films with $d_{Ag} = 5$-20 nm dependences $H_C = f(T_{ann})$ (Fig. 6a, curve 2) are similar to the behavior of H_C in single-layer films of pure cobalt represented in the work [11] and two-layer Co/Fe/S samples (curve 1). Coercitivity of these films after annealing up to 400 K is not considerably changed, and there is a 2-4-fold increase in the coercitivity in the annealed at 550 K films and a 4-7-fold increase – in the annealed at 700 K films.

Usually, increase in the coercitivity in three-layer films is explained by the amplification of the exchange interaction between ferromagnetic layers. In our case, effective thickness of non-magnetic silver sublayer is equal to 5-20 nm that leads to the significant attenuation of the exchange interaction. Thus, we can neglect the component of coercitivity which is conditioned by the exchange coupling of ferromagnetic layers. Based on the stated considerations, one can conclude that increase in the coercitivity for Co/Ag/Fe/S films with d_{Ag} = 5-20 nm is conditioned by the same reasons that for single- and two-layer films. According to [8], in single- and two-layer films increase in the coercitivity is connected with the increase in the grain sizes and redistribution of point defects along the intercrystalline boundaries that leads to the increase in the role of volume mechanisms of the domain wall pinning.

We have to note that after annealing of the films with d_{Ag} = 6-12 nm at T_{ann} = 550 K anisotropy of MR appears.

In Co/Cu/Fe/S films with d_{Cu} = 3-5 nm (curve 3) annealing at the temperature of T_{ann} = 400 K and 550 K leads to a 1,5-2-fold decrease in the coercitivity. Decrease in the coercitivity occurs due to the decrease in the role of the domain wall pinning that is connected with the increase in the surface reflectivity. Annealing at 700 K leads to a 2-fold increase in H_C that is related to the same effects which are mentioned for the films with silver sublayers.

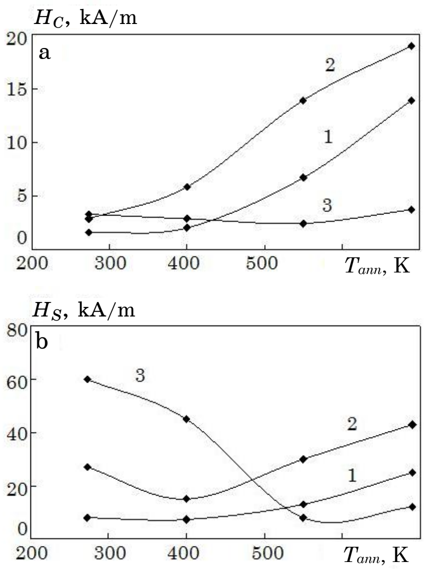

Fig. 6 – Dependence of the coercitivity H_C (a) and saturation field H_S (b) on the annealing temperature T_{ann} for the studied samples: 1 – Co(50 nm)/Fe(50 nm)/S; 2 – Co(20 nm)/Ag(10 nm)/Fe(20 nm)/S; 3 – Co(20 nm)/Cu(6 nm)/Fe(20 nm)/S

In Fig. 6b we represent the dependences $H_S = f(T_{ann})$ for two- and three-layer films with different thickness of silver and copper sublayers (H_S is the field where magnetic moments in ferromagnetic layers are oriented parallel, i.e. the saturation field).

For two-layer films (curve 1) one can observe only increase in the value of H_S (2-3-fold) after annealing at the temperature of 700 K. For three-layer Co/Ag/Fe/S films (curve 2) one can observe a 1,5-2-fold decrease in the saturation field H_S after annealing at the temperature of 400 K and a 2-3-fold increase at further increase in the annealing temperature T_{ann}. For Co/Cu/Fe/S films (curve 3) a substantial decrease (5-7-fold) in the value of H_S is observed up to the temperature of T_{ann} = 550 K. Annealing at the temperature of 700 K leads to the insignificant increase in the saturation field; minimum on the curves $H_S = f(T_{ann})$ is connected with the transition from the GMR to the anisotropy MR.

5. CONCLUSIONS

1. It is experimentally shown that at the thickness of non-magnetic sublayers (d_{Ag} = 5-20 nm, d_{Cu} = 3-15 nm) the GMR effect is realized in all studied systems. Maximum value of GMR for freshly condensed films (1-1,2%) is observed at the same thickness of magnetic layers and thickness of sublayers d_{Ag} ~ 6 nm and d_{Cu} ~ 3 nm, respectively. The value of GMR is monotonously decreased with the increase in the thickness of spacers from d_{Ag} = 8 nm to 20 nm and from d_{Cu} = 3 nm to 15 nm.

2. Investigation of the influence of thermomagnetic treatment on the magnetoresistive effect of three-layer films allowed to establish the following regularities: for Co/Ag/Fe/S films with an effective thickness of silver of 15-20 nm annealing at the temperature of 400 K leads to a 1,7-2-fold decrease in the GMR amplitude; increase in the annealing temperature T_{ann} up to 550 K leads to a 2-2,3-fold increase in the GMR value; further increase in the annealing temperature up to 700 K decreases the GMR value; for three-layer Co/Cu/Fe/S films with thickness of sublayers d_{Cu} = 10-15 nm increase in the annealing temperature up to 700 K leads only to the increase in the GMR value.

3. Obtained experimental results imply sufficiently high thermostability of the properties of film structures based on Co, Fe, Cu, and Ag that allows to recommend them for the production of magnetoresistive elements, as one of the stages of technological process – thermostabilization annealing in vacuum in the temperature range of 500-700 K directly after film deposition.

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