J. Nano- Electron. Phys.
3 (2011) No2, P. 26-33

PACS numbers: 72.10.Fk, 72.15.Lh, 73.40.Jn, 73.50.Gr, 73.50.Jt, 75.30.Vn

INFLUENCE OF THE ADDITIONAL Ni LAYERS ON STRUCTURE AND MAGNETORESISTIVE PROPERTIES OF THIN FILM SYSTEMS BASED ON Co AND Cu

***V.B. Loboda*[1], *Yu.O. Shkurdoda*[1], *V.N. Kolomiets*[1], *L.V. Dekhtyaruk*[2]**

[1] Sumy State Pedagogical University,
87, Romens'ka Str., 40002 Sumy, Ukraine
E-mail: shkurdoda@rambler.ru

[2] Kharkiv State Technical University of Building and Architecture,
40, Sums'ka Str., 61002 Kharkiv, Ukraine

Here, we present the investigation results of the structural-phase state, electrical and magnetoresistive properties (anisotropic magnetoresistance, giant magnetoresistance) for three-layered nanocrystalline thin film Co/Cu/Co systems with additional ferromagnetic Ni layers in an oil-free ultrahigh vacuum. Influence of Ni layers, which are embedded on the boundary between layers or within ferromagnetic Co layer, on the value of the GMR effect is shown.

Keywords: *NANOCRYSTALLINE FILM SYSTEMS, CRYSTALLINE STRUCTURE, RESISTIVITY, MAGNETORESISTANCE, GIANT MAGNETORESISTANCE.*

(Received 28 December 2010, in final form 25 February 2011, online 03 April 2011)

1. INTRODUCTION

Considerable success in the production and investigation of thin metal films, which was achieved in the end of the XX century, leaded to the discovery of new phenomena, whose study is still actual nowadays. In particular, it was established that when nanostructures consist of alternating magnetic and non-magnetic layers or quantum dots, a number of unique physical phenomena, such as spin-dependent scattering of conduction electrons, indirect exchange interaction, surface magnetic anisotropy, appear. The giant magnetoresistance (GMR) effect is the most known one, which is observed in metal multilayer structures and is conditioned by the spin-dependent scattering of conduction electrons on the type of the magnetic ordering of adjacent layers in the film [1-2]. Investigation of the initiation mechanism of this phenomenon was the reason of the creation of a number of new materials with wide capabilities for practical use: spin valves, structures with magnetic tunnel junction, spin nano-transistors, etc. Lately such materials find wider application as the mediums for ultra-high density magnetic recording media, high-sensitive magnetic field sensors, elements of magnetoresistive memory with arbitrary selection [3-4].

Investigation of nanoheterostructures is actively carried out in our country and abroad and has a complex nature. In the practical aspect, scientists are focused on the development of new systems with the specified technical characteristics, stability support of these properties and improvement of the production process. Solution of these questions is complicated by the presence of many fundamental aspects connected with the specific character of spin dependence of the conduction electron scattering on the type of the magnetic ordering and the dimensional effects as well. In particular, on the present stage of investigation of the GMR effect, influence of the impurity atoms on the effect amplitude in multilayer structures is still open and requires complementary study [5].

Based on the aforesaid, the aim of the present work is the ascertainment of the influence of additional ultra-thin ferromagnetic Ni layers embedded on the interface or within ferromagnetic Co layer on the structural-phase state, electrical conductivity and magnetoresistive properties of freshly condensed and annealed at different temperatures three-layered Co/Cu/Co film systems.

2. EXPERIMENTAL TECHNIQUE

Multilayer film systems with the layer thickness of 1-50 nm were obtained in the vacuum chamber of the plant VUP-5M at the residual gas pressure of 10^{-4} Pa [6]. Alternate film condensation was carried out by evaporation of metals with purity not less than 99,98% from independent sources (Cu from the tungsten filament; Co and Ni from the electron beam gun). Film condensation was performed at the room temperature of the substrate with the rate of ω = (0,5-1) nm/s depending on the evaporation regimes. Polished glass plates with pre-deposited contact pads were used as the substrates for the study of the electrical and magnetoresistive properties; NaCl (KBr) and carbon films were taken as the substrates for the structural-phase investigation.

Design of the substrate holder allowed us to obtain during one processing cycle two film samples with different thickness of non-magnetic sublayer and close thicknesses of ferromagnetic layers. Film geometry for the measurement of the electrical resistance was specified by the windows produced with high precision in mechanical masks made of nichrome foil.

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

After condensation films were held at the substrate temperature during 30 min. Thermal stabilization of the samples and investigation of the dependences of their electrical resistance on the temperature were performed in accordance with the scheme "heating-cooling" with the constant rate of 2-3 K/min in the temperature range of 300-700 K. Temperature control was carried out using chromel-alumel thermocouple with the error of ± 5 K. Measurement error of the resistivity (ρ) was mainly determined by the measurement error of the film thickness: 10-15% at $d < 50$ nm and 5-10% at $d > 50$ nm. Since during the magnetoresistance (MR) study the value of sample electrical resistance (not its resistivity) is directly measured, error of these measurements is 0,02%.

Measurements of the longitudinal and transverse MR and thermal treatment of the samples were performed in a special plant in an oil-free ultra-high vacuum of 10^{-6}-10^{-7} Pa in a constant magnetic field with the strength up to H = 150 kA/m.

Structural-phase investigation of the samples was carried out using the transmission electron microscope EM-125 and the electron diffractometer.

3. EXPERIMENTAL RESULTS

3.1 Structural-phase state of the samples

Results of the treatment of the electron diffraction patterns of unannealed Co/Cu/Co films with additional Ni layers (irrespective of their arrangement) imply the presence of the fcc-Ni, hcp-Co, and, possibly, fcc-Cu, phases (see Fig. 1b). Lines, which belong to the fcc-Cu and fcc-Ni phases, are not distinguished for unannealed films on the electron diffraction patterns. Therefore, we cannot speak with confidence if lines on the electron diffraction pattern belong to the fcc-Cu phase, since they can belong to the fcc-Ni. Hcp-Co and fcc-Cu phases can be fixed for unannealed three-layered Co/Cu/Co films. Crystallite size for unannealed films does not exceed 10 nm.

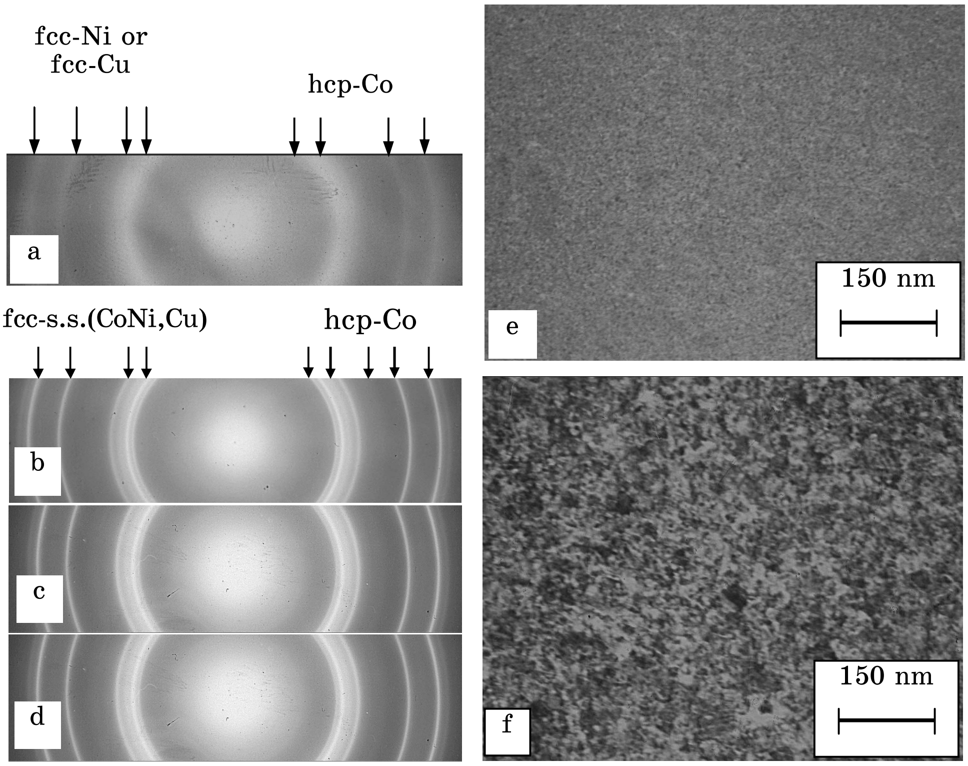

Fig. 1 *– Electron diffraction patterns (a-d) and crystalline structure (e, f) from unannealed (a, e) and annealed at T_{ann} = 700 K film systems: Ni(10)/Co(20)/Cu(8)/Co(20)/Ni(10) (a, b, e, f); Co(15)/Ni(10)/Co(15)/Cu(8)/Co(15)/Ni(10)/Co(15) (c) and Co(30)/Ni(5)/Cu(8)/Ni(5)/Co(30) (d)*

Phases, which correspond to the hcp-Co and, probably, to the fcc solid solution (CoNi, Cu) are present in Ni/Co/Cu/Co/Ni and Co/Ni/Cu/Ni/Co films after annealing at the temperature of 700 K. Lattice parameters of the hcp-Co annealed films agreed well with the corresponding parameters for Co in a bulk state (a_0 = 0,2514 nm, c_0 = 0,4105 nm [7]). We have to note that the hcp-Co, fcc-Co and fcc-Cu phases are present in Co/Cu/Co films after annealing at the temperature of 700 K.

Thus, studied film systems at the layer thickness of $d > 10$ nm in the initial state maintain layer individuality. Solid solution (CoNi, Cu) is formed after their annealing at the temperature of 700 K.

3.2 Magnetoresistive properties of multilayer samples

Positive longitudinal magnetoresistive effect (resistance increases under the action of the external magnetic field) is observed for all studied systems with the effective thickness of non-magnetic sublayer up to 2 nm (irrespective of the thickness and arrangement of ferromagnetic Co and Ni layers). This is the feature of usual anisotropic MR, which is inherent for the uniform ferromagnetic materials. Interaction between conduction electrons and external electrons of atoms, whose spin moments cause spontaneous magnetization [8], is the reason of anisotropic MR in ferromagnets.

For unannealed Ni/Co/Cu/Co/Ni films with the thicknesses d_{Cu} = 3-15 nm, d_{Co} = 20-30 nm and d_{Ni} = 5-10 nm (Fig. 2a) only the decrease in the electrical resistance irrespective of the direction of applied magnetic field, current and sample orientation (absence of the magnetoresistance anisotropy) is observed. Thus, in the given multilayer films the giant magnetoresistance (GMR) effect is realized. In the most of works GMR of multilayer films is connected with the formation of antiferromagnetic ordered regions in magnetic layers [1, 9]. In these structures the exchange coupling between ferromagnetic layers is substantially attenuated due to sufficiently thick (d_{Cu} = 3-15 nm) non-magnetic conducting sublayer. Presence of the magnetoresistive loop and small values of the saturation fields ($H_S < 20$ kA/m) [10] is the confirmation of this fact. Upper and lower magnetic layers have different magnetic properties (even at the same thickness), in particular, the coercive force. Therefore, magnetic configuration of the layers is changed from the antiparallel order of magnetic moments to the parallel one and inversely under the action of the external magnetic field. It is shown in the Ref. [11] that the GMR effect in three-layered Co/Cu/Co structures is conditioned by an asymmetry of the spin-dependent electron scattering on the grain boundaries. The GMR value for freshly condensed Ni/Co/Cu/Co/Ni films is 0,4-0,6%. The GMR value for these films is less than for three-layered Co/Cu/Co films (1-1,2%) obtained in the same conditions due to the current shunting by additional Ni layers.

When cooling samples to 150 K the shape of the hysteresis loops of the magnetoresistive effect is not almost changed (irrespective of the layer thickness). Increase in the GMR effect and peak shift on the dependence $(\Delta R/R_0)(H)$ toward the region of stronger fields are only observed.

For all obtained samples we have studied the influence of the annealing temperature on the shape of the magnetoresistive hysteresis loops and the values of the longitudinal and transverse MR. The main laws of the change of the dependences $(\Delta R/R_0)(H)$ after annealing for the systems based on Co and Cu with additional Ni layers can be seen in Fig. 2. As illustrations, in this figure we present the dependences of the MR on the external magnetic field strength for the unannealed (Fig. 2a) and annealed at the temperatures 400, 550, and 700 K Ni(10)/Co(20)/Cu(5)/Co(20)/Ni(10) film system obtained at the temperature of 300 K.

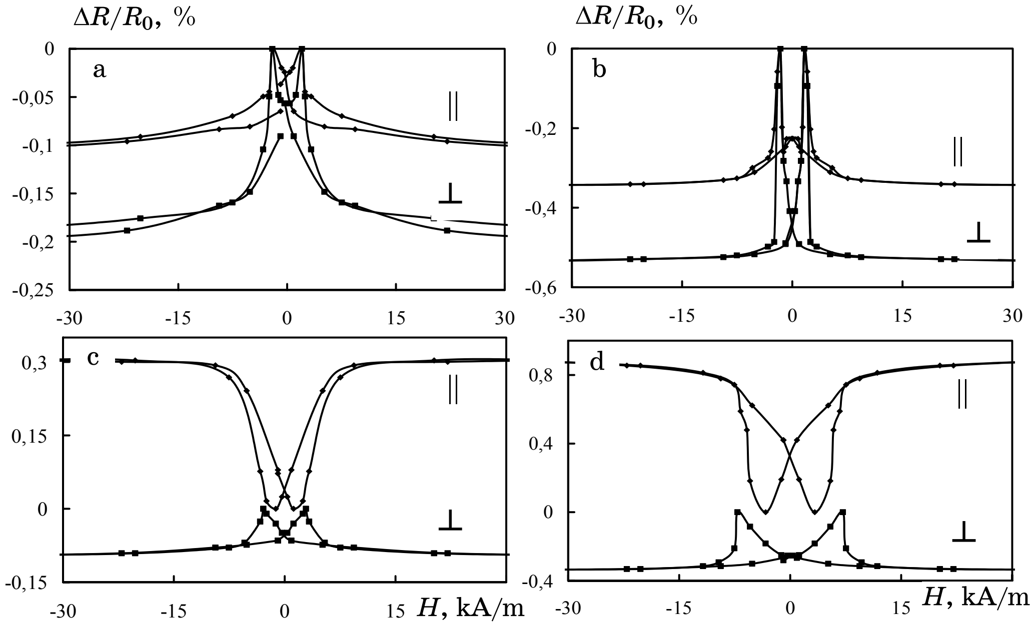

Fig. 2 *– Dependence of the longitudinal (||) and transverse (⊥) MR on the magnetic field strength for the unannealed (a) and annealed at different temperatures (b, c, d) Ni(10)/Co(20)/Cu(8)/Co(20)/Ni(10) film system; b – annealing temperature is 400 K, c – 550 K, d – 700 K*

In freshly condensed Ni/Co/Cu/Co/Ni film systems with d_{Co} = 20-30 nm, d_{Ni} = 5-20 nm and d_{Cu} = 5-10 nm the GMR effect is realized. After annealing of these samples at 400 K in magnetic field with the strength of 8 kA/m the 2-5 fold increase in the GMR value due to the increase in the value of ΔR is observed (Fig. 3).

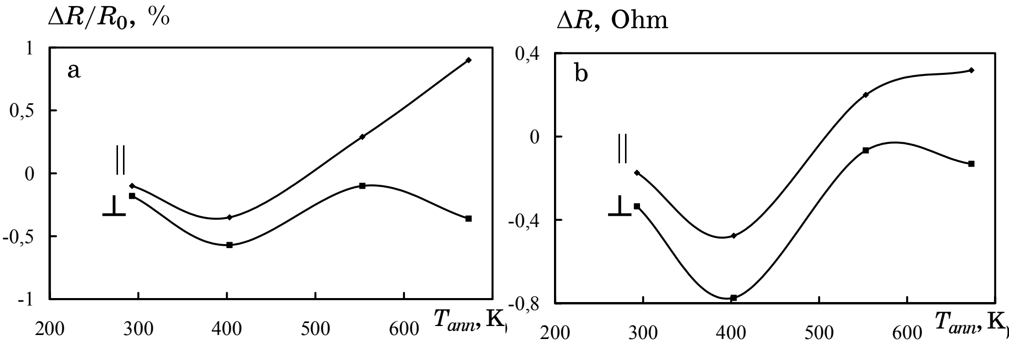

Fig. 3 *– Dependence of $(\Delta R/R_0)_{max}$ (a) and ΔR (b) on the annealing temperature for the longitudinal (||) and transverse (⊥) MR of Ni(10)/Co(20)/Cu(8)/Co(20)/Ni(10) film system*

Increase in the value of ΔR and, as a consequence, the GMR amplitude in the films with sufficiently thick copper sublayers ($d_{Cu} > 5$ nm) is possibly connected, as well as in the case of [4], with layering of Co and Cu on the layer interfaces. This leads to the increase in the reflectivity degree of the external layer boundaries, since electrons after reflection from the boundary remain the effective ones (Pippard concept of "inefficiency") due to the fact that the charge carriers do not lose their "memory". As the authors of [13] note, it is not ruled out in this situation that except of the above mentioned reasons, mechanism of the electron scattering in magnetic layers, which is connected with the roughness of the grain boundaries, also influences the GMR value behavior. Thus, with the increase in the annealing temperature diffusion depth of the intermediate Co and Ni layers increases. This causes rise of the importance of spin-dependent scattering on the grain boundaries in the GMR effect of the films. Annealing, which stimulates the diffusion processes, leads to the appearance of surplus vacancies within Co layers that is also the reason of small GMR growth since flux of spin-polarized electrons moving toward the interfaces increases.

Increase in T_{ann} up to 550 K leads to the vanishing of the GMR effect and sharp (almost 10 fold) decrease in the MR value. We have to note that in three-layered Co/Cu/Co films the GMR effect is realized even after annealing at T_{ann} = 700 K and is 4% at the room temperature [10, 12]. Additional Ni layers lead to the formation of (CoNi, Cu) solid solution during annealing, cause violations of layer individuality, and, as a consequence, the removal of spin-dependent scattering of conduction electrons. Further increase in T_{ann} leads to the growth in the AMR value, which is mainly conditioned by the increase in crystal sizes. Minimal value of MR is observed in the transition from GMR to AMR.

For Co/Ni/Cu/Ni/Co systems the GMR effect was not discovered even for freshly condensed films. In our opinion, even during deposition of Ni and Cu layers (Ni, Cu) solid solution is formed, which excludes the possibility of the antiferromagnetic ordering of Co layers. Annealing of these systems leads to the increase in the anisotropic MR value only.

3.3 Electrical conductivity of multilayer samples

We will consider the obtained experimental results of the temperature dependences of the resistivity. Resistivity ρ of the unannealed films (T = 300 K) is within the range from $10\cdot10^{-7}$ Ohm·m to $30\cdot10^{-7}$ Ohm·m (depending on the layer thickness) that is one order more than the value ρ_0 of pure metals in a bulk state ($\rho_0 = 6{,}24\cdot10^{-8}$ Ohm·m for Co, $\rho_0 = 1{,}7\cdot10^{-8}$ Ohm·m for Cu [7]).

This difference can be explained by the size factor, but, as a rule, size effects of electrical conductivity are essentially appeared at $d < 10$ nm [14]. Obviously, such considerable resistivity for unannealed films, first of all, is conditioned by the defect structure of the layers (relatively large concentration of vacancies and stacking faults), presence of defects on the interfaces, and small crystal sizes.

All obtained samples were annealed in accordance with the scheme "heating-time lag-cooling" at different temperatures. Dependence of the resistivity on the annealing temperature for three-layered films is shown in Fig. 4a. As seen from this figure, irreversible decrease in the resistance due to healing of defects and increase in the crystal size are observed during annealing. This feature is also appeared in single-layered films of these metals. In the case of single-layered films the process of defect healing is described by the Venda theory (see, for example, [15]).

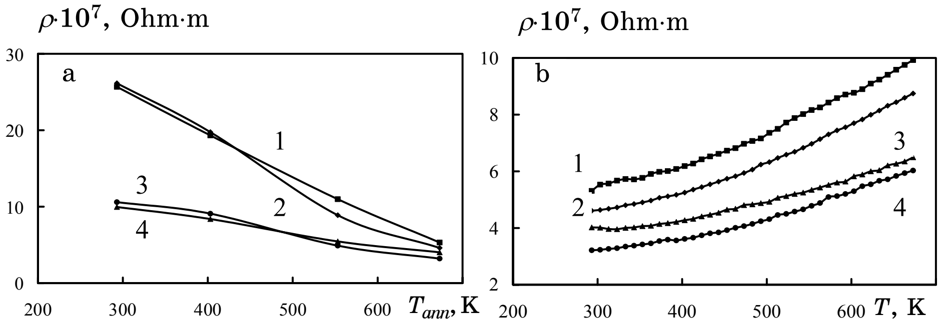

Fig. 4 *– Dependence of the electrical resistivity on the annealing temperature during 1 heating cycle (a) and temperature dependence of the resistivity (b) for Ni/Co/Cu/Co/Ni films (1 :* d_{Ni} = 10 *nm,* d_{Co} = 30 *nm,* d_{Cu} = 2 *nm; 2 :* d_{Ni} = 5 *nm,* d_{Co} = 30 *nm,* d_{Cu} = 10 *nm; 3 :* d_{Ni} = 10 *nm,* d_{Co} = 10 *nm,* d_{Cu} = 20 *nm; 4 :* d_{Ni} = 5 *nm,* d_{Co} = 30 *nm,* d_{Cu} = 30 *nm)*

We should note that for films with d_{Cu} = 2-10 nm an essential decrease in the resistivity during annealing is observed. For these films annealed at the temperature of 700 K, a 4-5 fold decrease in the resistance takes place. For films with thicker non-magnetic sublayers (d_{Cu} > 10 nm) the lesser (only 1,5-2 fold) decrease in the resistance is observed after annealing at 700 K.

Such behavior of the resistivity during annealing for films with different d_{Cu} can be explained by different faultiness of Co, Ni, Cu films and different asymptotical values ρ_∞. With the increase in the thickness of less defective in unannealed state sublayers, the irreversible resistance drop of three-layered films decreases during annealing. Also it is necessary to take into account here the diffusive processes and presence (or absence) of non-magnetic layer continuity.

In Fig. 4b we present typical temperature dependences of the electrical resistivity for Ni/Co/Cu/Co/Ni films. Quadratic dependence $\rho(T)$ is fixed in the temperature range 150-600 K. Probably, this dependence is conditioned by the electron-magnon interaction. For all studied samples, peculiarities in the characteristic points (for Co the Debye temperature is 460-470 K, the temperatures Θ_1 and Θ_2 are 590 and 180 K, respectively) are not almost observed.

4. CONCLUSIONS

Based on the obtained experimental results we can conclude the following:

1. The fcc-Ni, hcp-Co and fcc-Cu phases are present in the unannealed Co/Cu/Co films with additional Ni layers; the hcp-Co and fcc-solid solution (NiCo, Cu) phases are fixed after annealing at the temperature of 700 K.
2. In multilayer Ni/Co/Cu/Co/Ni film systems with d_{Co} = 20-30 nm, d_{Ni} = 5-20 nm and d_{Cu} = 5-10 nm the GMR effect is realized. 3-5 fold

increase in the amplitude of the GMR effect due to the increase in $\Delta R_{\max}$ is observed during annealing of the samples at the temperature of T_{ann} = 400 K.

3. Appearance of the MR anisotropy irrespective of the thickness of Cu sublayer is fixed after annealing of the films at T_{ann} = 550 K. 2-10 fold decrease in the MR value takes place in this case.
4. For Co/Ni/Cu/Ni/Co systems the GMR effect was not discovered even in freshly condensed films. The reason of this fact is the formation of (Ni, Cu) solid solution, which excludes the possibility of the antiferromagnetic ordering of Co layers, during Ni and Cu layer deposition.

REFERENCES

1. M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Greuzet, A. Fiederich, J. Chazelas, *Phys. Rev. Lett.* **61**, 2472 (1988).
2. G. Binasch, P. Grunberg, F. Saurenbach, W. Zinn, *Phys. Rev. B* **39**, 4828 (1989).
3. Ch.P. Poole, F.J. Owens, *Nanotekhnologii* (M.: Tekhnosfera: 2006).
4. S.I. Protsenko, I.V. Cheshko, L.V. Odnodvorets, I.M. Pazuha, *Struktura, dyfuzijni protsesy i magnitorezystyvni ta elektrofizychni vlastyvosti plivkovyh materialiv* (Sumy: SumDU: 2008).
5. M. Marszalek, O. Bolling, J. Jaworski, M. Kac, R. Kruk, V. Tokman, B. Sulkio-Cleff, *phys. stat. sol. C* **12** 3239 (2004).
6. V.B. Loboda, Yu.A. Shkurdoda, V.A. Kravchenko, *Functional Materials* **14** No1, 37 (2007).
7. G.V. Samsonov, *Fiziko-khimicheskie svojstva elementov* (K.: Nauk. dumka: 1965).
8. S.V. Vonsovsky, *Magnetizm* (M.: Nauka: 1971).
9. S.T. Roschenko, I.G. Shipkova, Yu.I. Chekrygina, *Functional Materials* **16** No4, 424 (2009).
10. E.Y. Tsymbal, D.G. Pettifor, *Solid State Phys.* **56**, 113 (2001).
11. Yu.A. Shkurdoda, V.B. Loboda, L.V. Dekhtyaruk, *Functional Materials* **15** No1, 38 (2008).
12. L.A. Chebotkevich, Yu.D. Vorobjov, I.N. Burkova, A.V. Kornilov, *FMM* **89** No3, 56 (2000).
13. N.N. Svirkova, *Tech. Phys.* **49**, 296 (2004).
14. Z.V. Stasyuk, A.I. Lopatinskiy, *FKhTT* **2**, 521 (2001).
15. L. Maissel, R. Glang, *Tekhnologiya tonkih plenok* (M.: Sov. radio: 1977).