PHYSICS AND CHEMISTRY OF SOLID STATE
V. 25, No. 3 (2024) pp. 650-655
Section: Physics
DOI: 10.15330/pcss.25.3.650-655

Vasyl Stefanyk Precarpathian National University

ФІЗИКА І ХІМІЯ ТВЕРДОГО ТІЛА
Т. 25, № 3 (2024) С. 650-655
Фізико-математичні науки

PACS: 68.37.Lp, 73.50.-h, 73.61.–r, 68.55. – a

ISSN 1729-4428 (Print)
ISSN 2309-8589 (Online)

I.M. Pazukha, O.V. Pylypenko, S.R. Dolgov-Gordiichuck, O.O. Panchoshnyi, Ya.V. Khyzhnya, A.M. Lohvynov

The effect of Ag or Au addition on electro- and magnetoresistive properties of $Ni_{80}Fe_{20}$ thin films

Sumy State University, Sumy, Ukraine, i.pazuha@aph.sumdu.edu.ua

The electrophysical and magnetoresistive properties of permalloy $Ni_{80}Fe_{20}$ (Py) and $Py_{64}Ag_{36}$ and $Py_{64}Au_{36}$ thin films prepared by the electron-beam sputtering are presented. The effect of the addition of a third element (Ag or Au) on the resistivity, temperature coefficient of resistivity and magnetoresistance of permalloy films has been described. The structural, electrophysical and magnetoresistive properties of the investigated samples showed a strong dependence on the annealing temperature.

Keywords: ferromagnetic alloy, noble metal, resistivity, annealing, temperature coefficient of resistance, magnetoresistance.

Received 5 February 2024; Accepted 17 August 2024.

Introduction

Permalloy ($Ni_{80}Fe_{20}$) film is attracting increasing attention for its applications in spintronic devices, particularly magnetic sensors [1, 2]. This is due to its favorable properties such as high permeability and low coercive field [3-5]. This makes it a promising choice as a soft magnetic material. Permalloy with noble metal impurities is a system that exhibits giant magnetoresistance. In this scenario, the giant magnetoresistance value depends on various factors, including the preparation method, thin film system composition, matrix properties, heat treatment conditions, substrate material, and so on [6, 7]. Resistivity is also a fundamental parameter influencing electronic, magnetic, and other properties of thin films [8-10]. In addition, the thermal stability of these materials remains an important area of research [11, 12].

This report presents an experimental investigation of the structure, electro- and magnetoresistive properties of three types of structures composed of permalloy (Py) thin films prepared by electron-beam evaporation. The focus of the study is on the incorporation of non-magnetic (Ag and Au) third elements at a concentration of 36 at.%. This concentration was deliberately chosen because the film systems based on permalloy and metals such as Au or Cu are characterised by maximum GMR values at concentrations close to 30 at.% [13, 14].

I. Experimental detail

Three series of samples with thickness of 45 nm were prepared: samples of A type are thin permalloy (Py) films with composition $Ni_{80}Fe_{20}$; samples of B type are $Py_{64}Ag_{36}$ thin films; samples of C type are $Py_{64}Au_{36}$ thin films. The samples of A type were deposited on amorphous glass-ceramic substrate by electron-beam sputtering at room temperature (RT) using 79 NM (79 -80 wt.% nikell, 2 - 5 wt.% molybdenum, 13 - 16 wt.% iron) in HV chamber with a base pressure of 10^{-4} Pa. The samples of B and C types were received by co-evaporation technique using permalloy 79NM and Ag or Au independent sources respectively. The thickness of each layer was monitored in real time using the built-in quartz-based thickness monitor with an accuracy of 10 %.

The samples elemental compositional were determined using an energy-dispersive X-ray (EDX) detector (Oxford Instruments) of scanning electron microscope (SEO-SEM Inspect S50-B). The EDX method allowed us to verify the concentration of components after deposition. The deviation of the concentration over the volume of the films is not more 1 at. %. Calculated data correspond to EDX-results, which revealed the material composition of the sputtered films: A type is 80 at.% nickel and 20 at.% iron; B type – 48 at.% nickel, 16 at.% iron and at.% silver; C type – 48 at.% nickel, 16 at.% iron and 36 at.% gold.

Crystalline structures of the samples were investigated by transmission electron microscopy (TEM-125K).

A four-point probe instrument was used to measure the resistance values of the samples in automated mode. All samples were annealed in a vacuum chamber at 10^{-4} Pa through two cycles of "heating ↔ cooling". The annealing temperature (T_{ann}) was 500 and 700 K respectively. The resistivity value was calculated considering the total thickness of the samples. The value of the temperature coefficient of resistivity (TCR) was calculated from temperature dependence of the resistivity by the equation: $\beta = (1/\rho_{in})\cdot(\Delta\rho/\Delta T)$, where ρ_{in} is the initial value of the resistivity, $\Delta T = T_{ann} - RT$.

The magnetoresistive properties were measured using software-hardware complex with current-in-plane geometries in an external magnetic field in the range from 0 to 500 mT at room temperature. The measuring current was $I = 1$ mA. The value of transverse magnetoresistance (TMR) and longitudinal magnetoresistance (LMR) has been calculated by equation $MR = (R(B) - R(B_0))/R(B_0)$, where $R(B)$ is the current value of resistance in the magnetic field B; $R(B_0)$ is the resistance of the sample in the field of the B_0.

II. Results and discussion

The studies of crystal structure and phase state for samples of type A, B and C were carried out for the correct analysis of their electro- and magnetoresistive properties.

Figure 1 shows the bright-field TEM images and diffraction patterns for the investigated samples before and after annealing up to 700 K. The phase state of the $Ni_{80}Fe_{20}$ thin film (A-type sample) corresponds to the

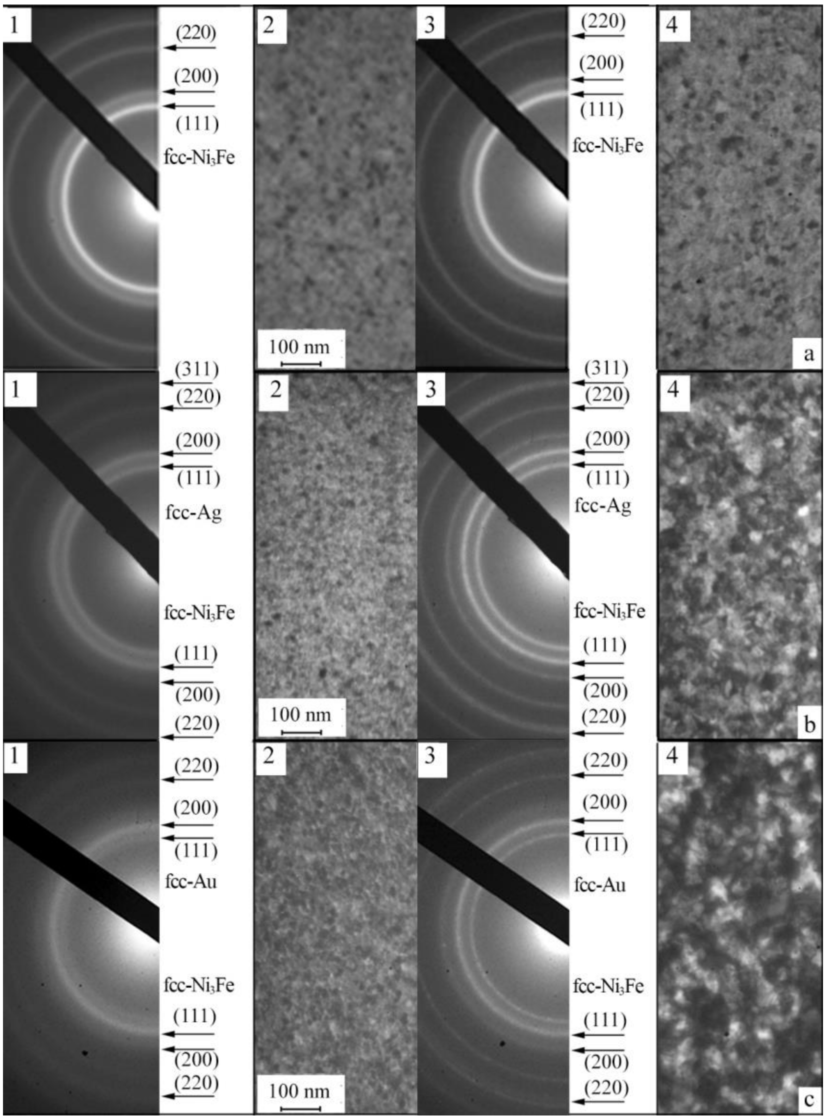

Fig. 1. Bright-field TEM images (2, 4) and diffraction patterns (1, 2) for Py (A-type) *(a)*, $Py_{64}Ag_{36}$ (B-type) *(b)* and *$Py_{64}Au_{36}$ (C-type) (c)* thin films after deposition (1, 2) and after annealing up to 700 K.

face-centred cubic (fcc) Ni_3Fe and remained unchanged after annealing up to 700 K (Figure 1,a, positions 1 and 3). The crystal structure (Figure 1a, positions 2 and 4) is typical of ferromagnetic metals, namely a highly dispersed labyrinthine structure [15]. After the addition of Ag (or Au) the phase state of the thin film samples becomes two-phase. In the case of the B-type sample, it corresponds to fcc-Ni_3Fe+fcc-Ag (Figure 1b, position 1). In the case of C-type sample, it corresponds to fcc-Ni_3Fe+fcc-Au (Figure 1,c, position 1). The annealing process does not affect on the phase state of the B and C type thin films. It stayed unchanged (Figs. 1,b and 1,c, position 3). B and C types of samples have a nanoscale structure after condensation (Figs. 1,b and 1c, position 2). The analysis of bright-field TEM images of $Py_{64}Ag_{36}$ and $Py_{64}Au_{36}$ thin films after annealing up to 700 K structure (Figs. 1,b and 1c, position 4) revealed the stognation of Ag (or Au) grain growth as a result of their isolation in the ferromagnetic matrix. These results were consistent with our previous works [16, 17].

Only when their behaviour is predictable and stable can thin films be used to miniaturise electronic devices. Therefore, from a practical point of view, it is important to understand the changes in resistivity of nanostructured thin film systems during annealing. Another important characteristic of thin film functional elements is the temperature coefficient of resistance. For the efficient operation of electronic circuits, functional elements should have adequate stability against temperature variations. It should also be noted that it is possible to understand the mechanisms of electron scattering in magnetic thin film systems by studying their electrophysical properties. Additional information for interpreting magnetotransport properties can be obtained from this.

Table 1 shows calculated values of resistivity and TCR for permalloy and permalloy based thin films after deposition and after annealing to 500 K and 700 K respectively.

Table 1.

Calculated values of resistivity and TCR for permalloy and permalloy-based thin films

	ρ, 10^{-7} Ohm·m			β,·10^{-3} K^{-1}	
	300 K	500 K	700 K	500 K	700 K
Py (A-type)	10.78	6.83	5.96	3.00	1.60
$Py_{64}Ag_{36}$ (B-type)	5.80	1.97	1.76	2.20	2.00
$Py_{64}Au_{36}$ (C-type)	4.74	2.91	2.01	1.27	1.02

We received that the values of resistivity of Py thin film is $10.78·10^{-7}$ Ohm·m. The adding of Ag or Au leads to decrease of resistivity to $5.80·10^{-7}$ Ohm·m and $4.74·10^{-7}$ Ohm·m, respectively. For type A, B and C samples, annealing to 500 K causes an irreversible reduction in resistivity. The main reasons for the reduction in resistivity are the healing of defect healing and grain size growth. Growth at annealing temperatures up to 700 K results in a further reduction in resistivity, but not as significant. This indicates that the electrophysical

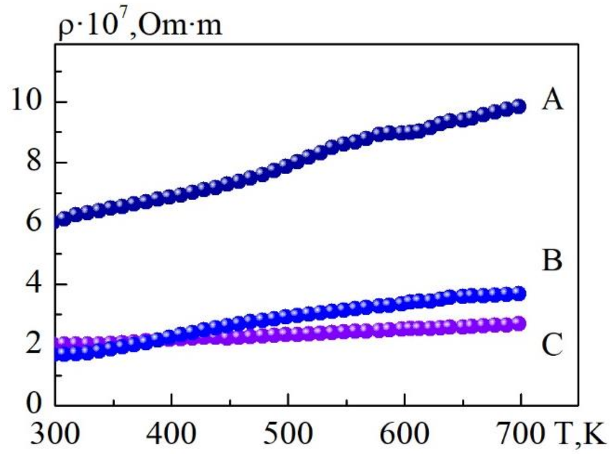

Fig. 2. Temperature dependences of resistivity for thin films of Py (A-type), $Py_{64}Ag_{36}$ (B-type) and $Py_{64}Au_{36}$ (C-type) thin films.

properties stabilized. Figure 2 shows the temperature dependence of the resistivity for the second cooling cycle for thin films of Py (A-type), Py64Ag36 (B-type) and Py64Au36 (C-type) thin films. The metallic mode of conductivity is observed for all samples. As a result, the temperature coefficient of resistance has a positive value (Table 1). A TCR order of 10^{-3} K^{-1} is typical for the permalloy thin films [4], noble metal [18] and thin film systems based on permalloy and noble metal [17].

Figure 3 shows typical magnetoresistive curves for as-deposited and annealed at 500 K and 700 K samples: Py, $Py_{64}Ag_{36}$ and $Py_{64}Au_{36}$. The results of the transverse magnetoresistance (TMR) and longitudinal magnetoresistance (LMR) calculations are summarized in Table 2.

The study of MR curves for A-type samples allows us to conclude that the value of the magnetoresistance of thin permalloy films depends on the geometries studied and the annealing temperature. For as-deposited samples (300 K), the MR(*B*) curves partially overlap in both geometries, so that a hysteretic behaviour of the magnetoresistance is observed. This is an indication of the anisotropic nature of magnetoresistance. The magnitude of magnetoresistance is relatively small (LMR = 0.14 %, TMR = 0.13 %), which is consistent with the data obtained in Refs [4, 19]. Annealing the films up to 500 K does not change the shape of the curves and results in an insignificant increase in the magnitude of the LMR and TMR. At the same time, increasing the annealing temperature up to 700 K results in a twofold increase in the magnitude of the LMR and TMR. This is due to changes in the crystal structure of the samples. The anisotropic nature of the magnetoresistance is retained after annealing to 700 K, which is typical of films of ferromagnetic alloys.

Analysis of the MR(B) curves for B-type samples shows that the addition of Ag changes the shape of the curves. LMR(B) and TMR(B) curves have no hysteresis and show a linear decrease of MR at fields up to 500 mT. Therefore, we have an isotropic character of the magnetoresistance (LMR = TMR = 0.38 %). After annealing at 500 K and 700 K, the value of isotropic magnetoresistance increases up to 0.63 % and 0.80 %, respectively. One of the reasons for this increased MR

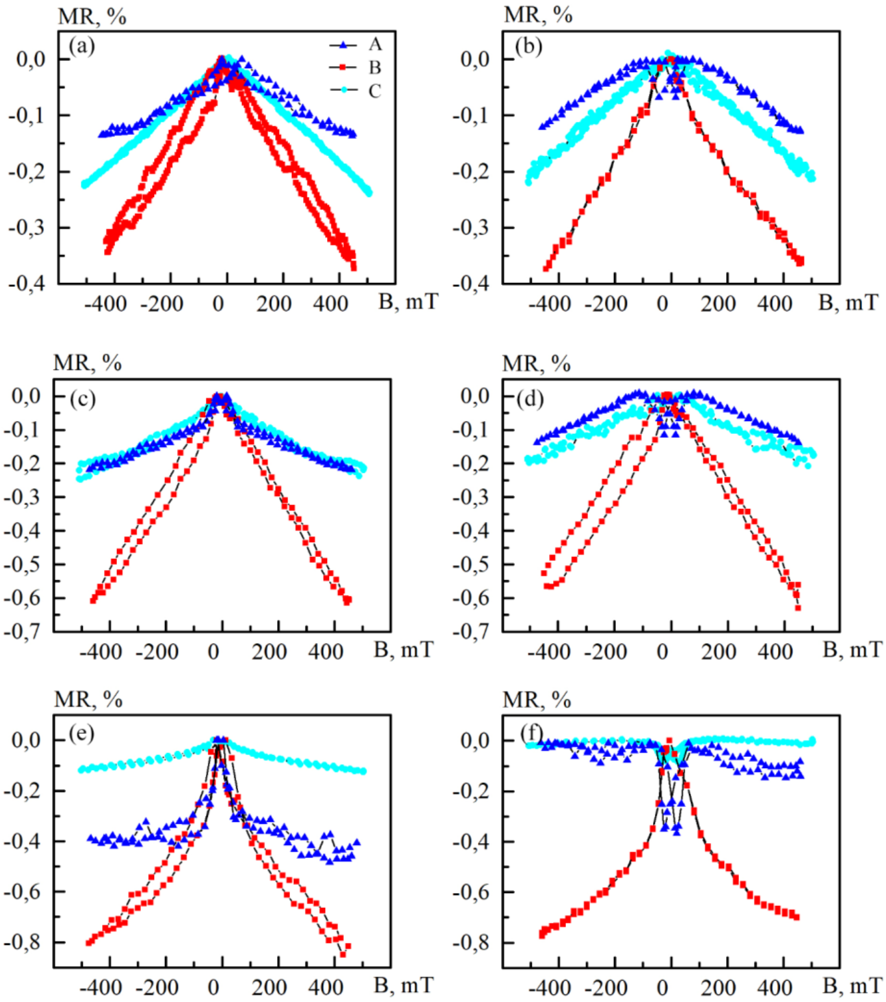

Fig. 3. Field dependence of transverse (a, c, e) and longitudinal (b, d, f) magnetoresistance for thin films of Py (A-type), $Py_{64}Ag_{36}$ (B-type) and $Py_{64}Au_{36}$ (C-type) thin films after deposition (a, b) and annealing to 500 K (c, d) and 700 K (e, f).

Table 2.

Calculated values of magnetoresistance for permalloy and permalloy-based thin films

	TMR, %			LMR, %		
	300 K	500 K	700 K	300 K	500 K	700 K
Py (A-type)	0.14	0.22	0.40	0.13	0.14	0.38
$Py_{64}Ag_{36}$ (B-type)	0.38	0.63	0.8	0.38	0.63	0.79
$Py_{64}Au_{36}$ (C-type)	0.21	0.22	0.12	0.21	0.22	0.10

effect is that the magnetic grains become larger, creating more ferromagnetic regions in the microstructure. This enhances the probability of spin dependent electron scattering and hence the magnitude of the MR effect.

The study of the magnetoresistive properties of C-type samples showed that the $Py_{64}Au_{36}$ thin film alloy also exhibits isotropic magnetoresistance. But their magnitude in 1.7 times smaller. No increase in the magnitude of the effect is observed after annealing to 500 K. By increasing the annealing temperature up to 700 K, the isotropy of the magnetoresistance has completely disappeared. Anisotropic magnetoresistance behaviour observed. Grain growth during annealing increases the probability of the electron-phonon scattering process, prevents electron transfer from one ferromagnetic grain to another. This reduced the probability of spin-dependent electron scattering.

Conclusion

In summary, we have studied the structure, electro- and magnetoresistive properties of three types of structures composed of permalloy thin films prepared by electron-beam evaporation. The metallic behaviour of the temperature dependence of resistance with a positive TCR value was observed regardless of the addition of Ag or Au. At the same time, studies of the magnetoresistive properties show that the nature and magnitude of the MR effect depends on the composition of the samples and the heat treatment conditions.

Pazukha I.M. – Ph.D (Physics and Mathematics), Associated Professor of the Electronics, General and Applied Physics Department;
Pylypenko O.V. – Ph.D (Physics and Mathematics), Senior Lecture of the Electronics, General and Applied Physics Department;
Dolgov-Gordiichuck S.R. – Postgraduate student, Engineer of the Electronics, General and Applied Physics Department;
Panchoshnyi O.O. – Postgraduate student of the Electronics, General and Applied Physics Department;
Khyzhnya Ya.V. – Ph.D (Medical Sciences), University Clinic, Sumy State University;
Lohvynov A.M. – Ph.D (Physics and Mathematics), Assistant of the Electronics, General and Applied Physics Department.

[1] S. Katsuaki, S. Eiji, *Spintronics for Next Generation Innovative Devices*, 255, (2015); https://doi.org/10.1002/9781118751886.
[2] L. Jogschies, D. Klaas, R. Kruppe, J. Rittinger, P. Taptimthong, A. Wienecke, L. Rissing, M.C. Wurz, *Recent Developments of Magnetoresistive Sensors for Industrial Applications*, Sensors, 15, 28665 (2015); https://doi.org/10.3390/s151128665.
[3] Y. Qianxing, M. Yaorui, C. Guoqing, F. Zinan, Z. Dong, B. Zhang, Y. Huang, *Electron beam welding with in situ heat treatment: An effective method for optimization of soft magnetic properties of permalloy*, Journal of Manufacturing Processes, 77, 642 (2022); https://doi.org/10.1016/j.jmapro.2022.03.042.
[4] O.V. Pylypenko, I.M. Pazukha, A.S. Ovrutskyi, L.V. Odnodvorets, *Electrophysical and magnetoresistive properties of thin film alloy $Ni_{80}Fe_{20}$. x*, Journal of Manufacturing Processes, 8, 03022 (2016); https://doi.org/10.21272/jnep.8(3).03022.
[5] S. Singh, L. Abelmann, H. Gao, U. Hartmann, *Influence of sputter pressure on magnetic and structural properties of Permalloy thin films*, Journal of Manufacturing Processes, 586, 171138 (2023); https://doi.org/10.1016/j.jmmm.2023.171138.
[6] Yu. O. Shkurdoda, A.M. Chornous, Y.M. Shabelnyk, V.B. Loboda, *The influence of the concentration of components in magnetic layers on the magnetoresistive properties of three-layer film systems based on Fe_xNi_{1-x} and Cu*, Journal of Magnetism and Magnetic Materials, 443, 190 (2017); https://doi.org/10.1016/j.jmmm.2017.07.078.
[7] M. Tamisari, F. Spizzo, M. Sacerdoti, G. Battaglin, F. Ronconi, *Correlation between structural and giant magnetoresistance properties of Fe–Ag nanogranular films*, J. Nanopart. Res., 13, 5203 (2011); https://doi.org/10.1007/s11051-011-0505-x.
[8] M.A. Qader, A. Vishina, L. Yu, C. Garcia, R.K. Singh, N.D. Rizzo, M. Huang, R. Chamberlin, K.D. Belashchenko, Mark van Schilfgaarde, N. Newman, *The magnetic, electrical and structural properties of copper-permalloy alloys*, Journal of Magnetism and Magnetic Materials, 442, 45 (2017); https://doi.org/10.1016/j.jmmm.2017.06.081
[9] W. Zhang, T. Wu, B. Peng, W. Zhang, *Resistivity dependence of the spin mixing conductance and the anisotropic magnetoresistance in permalloy*, Journal of Alloys and Compounds, 696, 234 (2017); https://doi.org/10.1016/j.jallcom.2016.11.274.
[10] I.Yu. Protsenko, L.V. Odnodvorets, V. Bondariev, K.V. Tyschenko, I.V. Cheshko, N.I. Shumakova, *Strain effect in film materials Ni_XFe_{1-X}/S*, Vacuum, 165, 113 (2019); https://doi.org/10.1016/j.vacuum.2019.04.015.
[11] Y.Q. Guo, H. Bai, Q.R. Cui, L.M. Wang, Y.C. Zhao, X.Z. Zhan, T. Zhu, H.X. Yang, Y. Gao, C.Q. Hu, S.P. Shen, C.L. He, S.G. Wang, *High thermal stability of perpendicular magnetic anisotropy in the MgO/CoFeB/W thin films*, Applied Surface Science, 568, 150857 (2021); https://doi.org/10.1016/j.apsusc.2021.150857.
[12] T. Yang, J. Gao, Y. Cui, J. Xu, J. Li, W. Wang, J. Luo, Meiyin Yang, *Thermal stability of SOT-MTJ thin films tuning by multiple interlayer couplings*, Journal of Magnetism and Magnetic Materials, 529, 167823 (2021); https://doi.org/10.1016/j.jmmm.2021.167823.
[13] I.O. Shpetnyi, K.V. Tyschenko, V.Ya. Pak, B.V. Duzhyi, Yu.O. Shkurdoda, I.Yu. Protsenko, *Structural-Phase State and Magnetotransport Properties of Thin Film Alloys Based on Permalloy and Copper*, J. Nano- Electron. Phys., 13, 01020 (2021); https://doi.org/10.21272/jnep.13(1).01020.
[14] I.M. Pazukha, S.R. Dolgov-Gordiichuk, A.M. Lohvynov, K.V. Tyschenko, O.V. Pylypenko, *Peculiarities of magnetoresistive properties of nanostructured $(Ni_{80}Fe_{20})_xAu_{1-x}$ thin films: concentration and annealing effects* Acta Physica Polonica A, 144, 69 (2023); https://doi.org/10.12693/APhysPolA.144.69.
[15] G. Nahrwold, Jan M. Scholtyssek, S. Motl-Ziegler, O. Albrecht, U. Merkt, G. Meier, *Structural, magnetic, and transport properties of Permalloy for spintronic experiments*, Journal of Applied Physics, 108, 013907 (2010); https://doi.org/10.1063/1.3431384.
[16] I.M. Pazukha, D.O. Shuliarenko, O.V. Pylypenko, L.V. Odnodvorets, *Concentration and heat treatment effects on magnetoresistive properties of Ag-added $Ni_{80}Fe_{20}$ film systems*, Journal of Magnetism and Magnetic Materials, 485, 89 (2019); https://doi.org/10.1016/j.jmmm.2019.04.079.
[17] I.M. Pazukha, A.M. Lohvynov, O.V. Pylypenko, *Effect of Au concentration on electrophysical properties of nanostructured $(Ni_{80}Fe_{20})_xAu_{1-x}$ thin films*, Applied Physics A, 128, 760 (2022); https://doi.org/10.1007/s00339-022-05907-0.

[18] A.I. Oliva, J.M. Lugo, R.A. Gurubel-Gonzalez, R.J. Centeno, J.E. Corona, F. Aviles, *Temperature coefficient of resistance and thermal expansion coefficient of 10-nm thick gold films,* Thin Solid Films, 623, 84 (2017); https://doi.org/10.1016/j.tsf.2016.12.028.

[19] W. Zhang, T. Wu, B. Peng, W. Zhang, *Resistivity dependence of the spin mixing conductance and the anisotropic magnetoresistance in permalloy,* Journal of Alloys and Compounds, 696, 234 (2017); http://dx.doi.org/10.1016/j.jallcom.2016.11.274.

І.М. Пазуха, О.В. Пилипенко, С.Р. Долгов-Гордійчук, О.О. Панчошний, Я.В. Хижня, А.М. Логвинов

Вплив додавання Ag або Au на електро- та магніторезистивні властивості тонких плівок $Ni_{80}Fe_{20}$

Сумський державний університет, м. Суми, Україна, i.pazuha@aph.sumdu.edu.ua

У роботі представлені електрофізичні та магніторезистивні властивості тонких плівок пермалою $Ni_{80}Fe_{20}$ (Py) і $Py_{64}Ag_{36}$ і $Py_{64}Au_{36}$, отриманих методом електронно-променевого осадження. Описано вплив додавання третього елемента (Ag або Au) на питомий опір, температурний коефіцієнт питомого опору та магнітоопір плівок пермалою. Структурні, електрофізичні та магніторезистивні властивості досліджуваних зразків показали сильну залежність від температури відпалювання.

Ключові слова: феромагнітний сплав, благородний метал, питомий опір, відпалювання, температурний коефіцієнт опору, магнітоопір.