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# Electrical Conductivity and Magnetoresistive Properties of Film Alloys Based on Permalloy $Fe_{0.5}Ni_{0.5}$ and Copper

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The paper shows the experimental results of the study of magnetoresistive properties and electrical conductivity of medium-entropy alloys based on permalloy $Fe_{0.5}Ni_{0.5}$ and Cu with a copper concentration from 5 to 50 % obtained by the technique of simultaneous deposition in vacuum. It is shown that the phase composition of both recently deposited and thermostabilized at a temperature of 700 K film alloys with a thickness $d = 20$-100 nm corresponds to the FCC phase of s.s. FeNi(Cu) with the lattice parameter $a = 0.360$-0.361 nm. After annealing the samples at a temperature of 700 K, an increase in the size of the crystallites to 50 nm is observed. For all samples, the anisotropic nature of the field dependences of the magnetoresistance with an amplitude value of 0.02-0.3 % depending on the thickness and Cu content is observed. With increasing concentration of copper, the value of the longitudinal and transverse magnetoresistance decreases. Annealing of film alloys leads to an increase in the value of the anisotropic magnetoresistance by 1.5-3 times. Reducing the measurement temperature to 120 K increases the value of the magnetoresistance by 1.3-1.5 times. For films with $d = 20$-30 nm and copper content of 5-30 %, the resistivity is $(40\text{-}55)\cdot 10^{-7}$ Ohm·m, and for films with $d_F = 40$-70 nm and copper content of 40-50 % – $(10\text{-}20)\cdot 10^{-7}$ Ohm·m. Annealing of the samples at a temperature of 700 K leads to an irreversible decrease in resistivity by 1.5-2 times depending on the thickness and Cu content. Typical metal temperature dependences of resistivity are observed for all thermostabilized samples.

**Keywords:** High-entropy alloys, Solid solutions, Anisotropic magnetoresistance, Resistivity, Electrical conductivity.

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## 1. INTRODUCTION

At the present stage of development of materials science, methods of formation and results of studies of the physical properties of high-entropy alloys (HEA) are of great interest. Of particular note are alloys, which can form disordered solid solutions. This is due to the study of the reasons for the formation of such structures, and the possibility of creating functional elements with improved performance.

The authors of [1] found that structures of simple solid solutions can be formed in alloys of equimolar composition. Such multicomponent alloys have a number of improved properties (hardness, strength, plasticity, corrosion resistance, etc.) [1-5]. But in [2, 3] it was shown that ordered solid solutions and/or intermetallic phases are formed in many HEA. Therefore, when obtaining a HEA with specified properties, their phase composition and microstructure should be taken into consideration.

It should also be noted that at present a large volume of both experimental and theoretical studies of the phase composition and mechanical properties of multicomponent alloys in the massive state has been accumulated, although studies of electrophysical and magnetoresistive properties have been initiated [4, 5] only for thin film samples.

Therefore, the aim of the work was to study the structural-phase composition, electrical conductivity and magnetoresistive properties of medium-entropy film alloys based on permalloy and copper, which in the future will be used as a basis for HEA.

## 2. EXPERIMENTAL DETAILS

Three-component film alloys based on permalloy ($Fe_{0.5}Ni_{0.5}$) and Cu with a layer thickness of 30-70 nm and $C_{Cu} \le 50$ at. % were obtained in a vacuum chamber at a gas pressure of a residual atmosphere of $10^{-4}$ Pa. Simultaneous deposition of the films was carried out as a result of evaporation of metals from independent sources (Cu – from tungsten tape, $Fe_{0.5}Ni_{0.5}$ – electron beam gun). The film thickness was determined using a quartz resonator with a measurement error of ± 5 %.

The components concentration was calculated based on ratio:

$$C_i = \frac{d_i \rho_i \mu_i^{-1}}{\sum\limits_{i=1}^{n} d_i \rho_i \mu_i^{-1}}, \qquad (1)$$

which takes into account the effective thickness of the metal layers [6]:

$$C_i = \frac{d_i \rho_i \mu_i^{-1}}{d_1 \rho_1 \mu_1^{-1} + d_2' \rho_2 \mu_2^{-1} + d_2'' \rho_3 \mu_3^{-1}}, \qquad (1')$$

where $d_1$ is the Cu film thickness; $d'_2$ and $d''_2$ are the effective layer thicknesses that can be formed based on Fe and Ni atoms which are part of the permalloy film; $\rho_i$ and $\mu_i$ are the density and molar mass of elements.

According to [6], the thickness can be calculated as shown in ratio (2):

$$d_2' = d_2 \left( 1 + \frac{c_{Fe}}{c_{Ni}} \cdot \frac{\rho_{Ni} \mu_{Ni}^{-1}}{\rho_{Fe} \mu_{Fe}^{-1}} \right)^{-1}, \quad d_2'' = d_2 - d_2'. \qquad (2)$$

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The study of the crystal structure and phase composition of the films was carried out by electron microscopic and electrographic methods (transmission electron microscope TEM-125K). Measurements of longitudinal and transverse magnetic resistance (magnetic field in the film plane) and thermomagnetic treatment of films were performed in a special installation under conditions of ultrahigh oil-free vacuum ($10^{-6}$-$10^{-7}$) Pa in a magnetic field with induction up to $B = 200$ mT. The value of the longitudinal and transverse magnetoresistance of the film samples was calculated by (3):

$$(R(B) - R(B_C))/R(B_C), \qquad (3)$$

where $R(B)$ is the resistance of the sample in a magnetic field with induction $B$; $R(B_C)$ is the resistance of the sample in the field with coercive force $B_C$.

## 3. RESULTS AND DISCUSSION

### 3.1 Phase Composition

Let us first consider features of the structural and phase state of $Fe_{0.5}Ni_{0.5}$ film alloys. According to the results of studies of the phase composition of the massive $Fe_{0.5}Ni_{0.5}$ alloy presented in [7], when the concentration of Fe atoms ($C_{Fe}$) is about 50 at. %, the NiFe phase is stabilized. According to the data of [6, 7], the NiFe phase has a FCC lattice with the parameter 0.359-0.361 nm.

The study of the phase composition of films by electron diffraction showed that in all recently deposited and annealed at 700 K $Fe_{0.5}Ni_{0.5}$ films with a thickness $d = 20$-$100$ nm, as in massive samples of the corresponding composition, FCC NiFe phase with the lattice parameter $a = 0.360$-$0.361$ nm is observed (Fig. 1a).

The phase composition of recently condensed films based on permalloy and copper with a thickness $d = 20$-$100$ nm and $C_{Cu} = 5$-$50$ at. % corresponds to the FCC phase of s.s. FeNi(Cu) with the lattice parameter $a = 0.360$-$0.361$ nm. After annealing the films at a temperature of 700 K, due to the increase in the size of the crystallites to 50 nm, the width of the diffraction rings decreases (Fig. 1b). In this case, changes in the phase composition and lattice parameter are not electronically recorded.

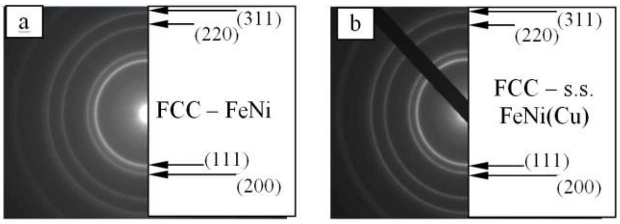

**Fig. 1** – Electron diffraction patterns of film alloys annealed at a temperature of 700 K: $Fe_{0.5}Ni_{0.5}$, $d = 40$ nm (a) and $Fe_{0.5}Ni_{0.5}$(Cu), $C_{Cu} = 20$ at. %, $d = 40$ nm (b)

### 3.2 Magnetoresistance

Let us consider the results of the study of the magnetoresistive effect in alloy films based on permalloy and copper with $d = 20 - 80$ nm and $C_{Cu} = 5$-$50$ at. %. For all samples, both recently condensed and annealed at a temperature of 700 K, the anisotropic nature of the field dependences of the magnetoresistance (positive longitudinal and negative transverse magnetoresistance) was observed. Fig. 2 shows typical dependences of the longitudinal and transverse magnetoresistance on the induction of an external magnetic field for alloy films with $C_{Cu} = 30$ at. % and $d = 40$ nm (measurement temperature $T_m = 300$ K). As can be seen from the figure, these dependences are similar to the corresponding dependences for single-layer films of ferromagnetic metals and alloys [8-10].

A common feature for all field dependences $(\Delta R/R_C)(B)$ with an anisotropic character is a sharp change in the magnetoresistance (MR) in the field interval of – 3-3 mT and a tendency to saturation in stronger fields. Note that the amplitude of the magnetoresistive effect for film alloys with a Cu concentration of less than 50 at. % is less than 0.02 %. This, in our opinion, is due to a decrease in the spontaneous magnetization of the film alloy with an increase in the Cu content. Samples with a Cu concentration of less than 50 at. % are magnetically ordered at room temperature, and their MR reflects the magnetic contribution to the scattering of conduction electrons and depends on the magnitude and orientation of the spontaneous magnetization. The reason of anisotropic MR in ferromagnets is the interaction of conduction electrons with external electrons, the spin moments of which cause spontaneous magnetization [10].

It should be noted that the values of longitudinal and transverse MR for annealed samples are concentration-dependent in the range of $C_{Cu} \leq 50$ at. % (Fig. 3). As can be seen from the figure, both the longitudinal and transverse MR only increases with decreasing copper content. This course of dependences is due to the increase in the spontaneous magnetization of the film alloys with decreasing Cu content.

With a decrease in the measurement temperature to 120 K, no significant changes in the field dependences of the anisotropic MR are observed. The shift of the peaks in the dependences in the direction of stronger magnetic fields is fixed, which is associated with an increase in the coercive force of the films. Fig. 4 shows the dependences $(\Delta R/R(0))_{max}(T)$ for the longitudinal and transverse MR of alloy films annealed at 700 K.

For all samples there is an increase in the value of MR with decreasing temperature in the entire temperature range. It should be noted that the value of MR of annealed films does not exceed 0.5 % even at a temperature of 150 K, and at 600 K it becomes less than 0.02 %.

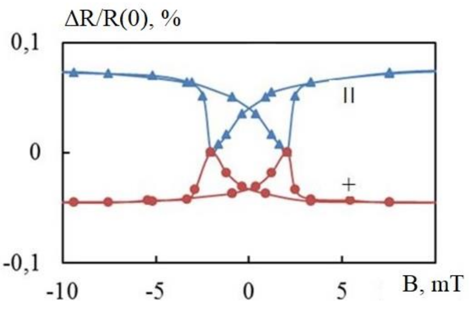

**Fig. 2** – Field dependences of longitudinal (| |) and transverse (+) MR of film alloys based on permalloy and Cu, $C_{Cu} = 20$ at. %, $d = 40$ nm

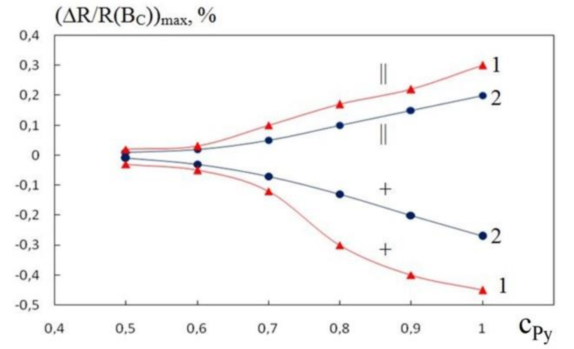

**Fig. 3** – Concentration dependence of longitudinal (| |) and transverse (+) MR for film alloys based on permalloy and copper (1 – *d* = 70 nm, 2 – *d* = 30 nm)

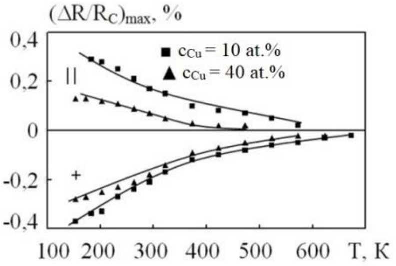

**Fig. 4** – Temperature dependence of longitudinal (| |) and transverse (+) MR for film alloys based on permalloy and copper with different concentrations of components (*d* = 50 nm)

For a more in-depth analysis of the causes of changes in the longitudinal and transverse MR, we analyzed the dependences of the resistivity $\rho$ and the value of $\Delta R_{\max}$ on temperature. As can be seen from Fig. 5, for all thermostabilized samples, typical metal temperature dependences of resistivity are observed. The course of dependences for samples with $C_{Cu} \le 30$ at. % (curves 1, 2) is due to electron scattering on defects of crystal structure, grain boundaries and interfaces (electron-magnon interaction and low-, high-temperature electron-phonon interaction) [11, 12]. The resistivity with increasing temperature from 150 K to 300 K increases by 1.15-1.75 times, depending on the concentration of components. The value of the MR is reduced by 2-2.5 times. A more significant change in the value of the MR is due to a decrease in the saturation magnetization, as a consequence of a decrease in the value of $\Delta R_{\max}$ of the samples with increasing temperature. In this case, for samples with a low copper content, the change in resistance is decisive in the temperature dependence of the MR. At higher concentrations of copper, the change in the value of $\Delta R_{\max}$ is decisive.

It should be noted that the resistivity of recently deposited films significantly depends on the thickness of the layers and concentration of components in the magnetic layers. Thus, for films with $d$ = 20-30 nm and $C_{Cu}$ = 10-30 at. %, the resistivity is (40-55)$\cdot 10^{-7}$ Ohm·m, and for films with $d$ = 40-70 nm and $C_{Cu}$ = 40-50 at. % – (10-20)$\cdot 10^{-7}$ Ohm·m. The high resistivity of annealed films compared to massive samples (for permalloy – $\rho_0 = 3.5 \cdot 10^{-7}$ Ohm·m, copper – $\rho_0 = 1.7 \cdot 10^{-8}$ Ohm·m) is explained for the reason of formation of high-resistance s.s. FeNi(Cu), and dimensional effects and significant defects of the films (relatively high concentration of vacancies and packaging defects).

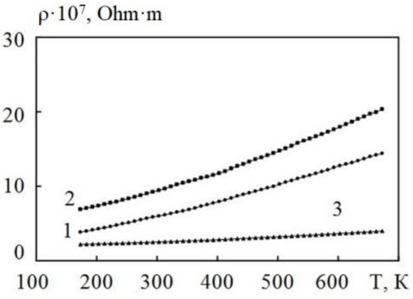

**Fig. 5** – Temperature dependence of resistivity for film alloys based on permalloy and copper (1 – *d* = 30 nm, $C_{Cu}$ = 10 at. %; 2 – *d* = 25 nm, $C_{Cu}$ = 30 at. %; 3 – *d* = 30 nm, $C_{Cu}$ = 50 at. %)

Annealing of the samples at a temperature of 700 K leads to an irreversible decrease of resistivity by 1.5-2 times. Such behavior of specific resistance at annealing is caused by "healing" of defects and increase in the sizes of crystallites.

The above-described changes in the resistivity of film alloys are the reason for the change in the value of the magnetic resistance of the samples after their heat treatment. Normally, the value of the MR of the film alloys based on permalloy and copper after annealing increases by 1.5-3 times.

## 4. CONCLUSIONS

1. The phase composition of recently deposited and thermostabilized at a temperature of 700 K film alloys based on $Fe_{0.5}Ni_{0.5}$ and Cu with thickness $d$ = 20-70 nm and copper concentration $C_{Cu}$ = 5-50 at. % corresponds to the FCC phase of s.s. FeNi(Cu) with the lattice parameter $a$ = 0.360-0.361 nm.
2. For film alloys with $d$ = 20-70 nm and $C_{Cu}$ = 5-50 at. % in the temperature range of 120-400 K, anisotropic nature of the MR with the amplitude of the longitudinal and transverse effect of 0.02-0.5 % depending on the thickness and concentration of the components is observed.
3. Reducing the measurement temperature from 300 K to 120 K leads to an increase in the value of the MR by 1.2-2.2 times depending on the concentration of the components.

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# Електропровідність та магніторезистивні властивості плівкових сплавів на основі пермалою $Fe_{0.5}Ni_{0.5}$ та міді

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В роботі представлені експериментальні результати стосовно магніторезистивних властивостей та електропровідності середньоентропійних сплавів на основі пермалою $Fe_{0,5}Ni_{0,5}$ та Cu з концентрацією міді від 5 до 50 %, отриманих методом одночасного осадження у вакуумі. Показано, що фазовий склад як свіжосконденсованих так і термостабілізованих при температурі 700 К плівкових сплавів товщиною $d$ = 20-100 нм відповідає ГЦК фазі т.р. FeNi(Cu) з параметром решітки $a$ = 0,360-0,361 нм. Після відпалювання зразків при температурі 700 К фіксується збільшення розміру кристалітів до 50 нм. Для всіх зразків фіксується анізотропний характер польових залежностей магнітоопору з амплітудним значенням 0,02-0,3 % в залежності від товщини та вмісту Cu. При збільшенні концентрації міді величина поздовжнього і поперечного магнітоопору зменшується. Відпалювання плівкових сплавів приводить до збільшення величини анізотропного магнітоопору в 1,5-3 рази. Зниження температури вимірювання до 120 К приводить до зростання величини магнітоопору у 1,3-1,5 рази. Для плівок з $d$ = 20-30 нм та вмістом міді 5-30 % величина питомого опору складає (40-55)·$10^{-7}$ Ом·м, а для плівок з $d$ = 40-70 нм та вмістом міді 40-50 % – (10-20)·$10^{-7}$ Ом·м. Відпалювання зразків при температурі 700 К призводить до незворотнього зменшення питомого опору в 1,5-2 рази залежно від товщини та вмісту Cu. Для всіх термостабілізованих зразків спостерігаються типові металічні температурні залежності питомого опору.

**Ключові слова:** Високоентропійні плівкові сплави, Тверді розчини, Анізотропний магнітоопір, Питомий опір, Електропровідність.