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Structure, Electrical Conductivity and Magnetoresistive Properties of Binary Film Alloys Based on Fe, Co and Ni as a Component of Heisler Alloys

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The phase state, electrical conductivity and magnetoresistive properties of binary film alloys based on Fe, Co and Ni prepared by the method evaporation in high vacuum in the temperature range 100-700 K have been studied. It has been shown that the *fcc*-phase is observed for as-deposited and annealed up to 700 K $Fe_{0,50}Ni_{0,50}$ thin films with a thickness d = 10-100 nm, the *fcc*-phase is observed. For $Fe_{0,67}Ni_{0,33}$ $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ thin films, the *bcc*-phase is fixed. It was found that the size dependences of the resistivity, temperature coefficient of resistance and magnetoresistance are realized for all structurally continuous film alloys. The character of size dependences is a like the corresponding dependences for the pure metal films. It has been shown that for $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ the size dependences of both longitudinal and transverse magnetoresistance appear much weaker than for $Fe_{0,50}Ni_{0,50}$ thin alloy. For $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ thin alloys, the value of both longitudinal and transverse magnetoresistance is weakly depending on the temperature.

Keywords: Permalloy, Crystal structure, Phase state, Magnetoresistance, Concentration effect, Temperature effect.

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1. INTRODUCTION

Heisler ferromagnetic alloys attract considerable attention from researchers due to their interesting physical properties and wide use in devices for reading and recording information, sensors, actuators, etc. [1-3]. Alloys with a Geisler structure belong to classes of functional materials, in particular, such as spin-polarized current injectors, shape memory alloys, magnetic cooling materials, alloys with giant deformation in a magnetic field, topological insulators, and others. It is worth noting that Heisler's film alloys are characterized by high spin polarization [4]. Therefore, it is relevant to study the peculiarities of the magnetic characteristics and giant magnetoresistance of nanogranular and multilayer systems based on Heisler alloys. The study of the conditions for the occurrence of certain magnetic states in a system of ferromagnetic nanoparticles or layered structures is extremely important for applications, especially in spintronics and for devices for writing and reading information, for example, magnetoresistive random-access memory (MRAM).

Fe_2CoSi (Ge, Al) and Fe_2NiSi (Ge, Al) alloys are interesting from the point of view of their magnetic characteristics. The results of the study of such alloys show very high saturation magnetization (M_s), low coercive force (H_c), high Curie temperature (T_c) and low hysteresis losses [5, 6]. A thin Fe_2CoSi film with a thickness of 75 nm has the lowest coercive force (H_c = 7 Oe). In general, Fe_2CoSi Heisler alloys have a high Curie temperature due to the strong exchange interaction between Fe and Co atoms. The saturation magnetization decreases with the increasing thickness of the thin Heisler alloy film. Such magnetic characteristics make these alloys especially valuable for the creation of functional elements in spintronics. Therefore, studies of the features of the structural phase state, magnetoresistive and magnetic properties of Fe_2CoSi (Ge, Al) and Fe_2NiSi (Ge, Al) alloys are relevant today.

For better understanding of the peculiarities of the crystal structural, phase state and magnetoresistive properties of Fe_2CoSi (Ge, Al) and Fe_2NiSi (Ge, Al) film alloys, similar studies were conducted on the binary film alloys $Fe_{0,67}Co_{0,33}$, $Fe_{0,50}Co_{0,50}$, $Fe_{0,67}Ni_{0,33}$, and $Fe_{0,50}Ni_{0,50}$, which are the basis for semi-alloys and Heisler alloys.

2. EXPERIMENTAL DETAILS

Condensation of film alloys was carried out by evaporation of massive alloys of the appropriate composition using an electron beam gun in a high vacuum. For this purpose, a series of massive samples of $Fe_{0,67}Co_{0,33}$, $Fe_{0,50}Co_{0,50}$, and $Fe_{0,67}Ni_{0,33}$ alloys were prepared, and the classic permaloy alloy $Fe_{0,50}Ni_{0,50}$ was used. The consistency of the composition of the samples and thin-film alloys was checked by X-ray microanalysis based on X-ray photon energy analysis with an energy dispersive spectrometer. The microanalysis data indicate that the composition of the specimens corresponds to the calculated values with an accuracy of 1%, and of the film alloys to 2 %.

The thicknesses of the samples were controlled in situ by the quartz resonator method (accuracy 10 %), for which an industrial resonator of the РГ-08 type with a frequency of 10 MHz and a generator of electrical oscillations were used. Comprehensive studies of the obtained samples were carried out by electron microscopy, electronography, microrentgenospectral analysis and 2-pin resisometry.

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3. CRYSTAL STRUCTURE AND PHASE STATE

First, consider the crystal structural and phase state of film alloys containing $Fe_{0.67}Ni_{0.33}$ and $Fe_{0.50}Ni_{0.50}$. Based on the previous research on the phase composition of the bulk Fe_xNi_{100-x} alloy, as presented in references [7], we can identify three distinct concentration zones where permalloy phases (Ni_3Fe and NiFe) are stabilized at different Fe atom concentrations (c_{Fe}):

1. Up to 50 at. % Fe atoms, where permalloy phases (Ni_3Fe and NiFe) are stable.
2. Close to 64 at. % c_{Fe}, where invar phase is observed.
3. Around 75 at. % c_{Fe}, where the α–phase (NiFe) is predominant.

According to the data presented in Refs. [7, 8], the Ni_3Fe and NiFe phases have a face-centered cubic (*fcc*) lattice with lattice parameters of a = 0.354-0.359 nm and 0.359-0.361 nm, respectively. The α-phase (NiFe) has a body-centered cubic (*bcc*) lattice with $a \approx 0.286$ nm, which is very similar to the *bcc* lattice parameter of iron ($a = 0.284$ nm). It is conceivable that the chemical composition of this phase corresponds to invar Fe_3Ni, but this matter remains unresolved currently.

The study of the phase composition of the films of the electron diffraction method showed that in all as-deposited and annealed $Fe_{0,50}Ni_{0,50}$ films at a temperature of 700 *K*, as well as in bulk samples of the corresponding composition, an *fcc* phase with a lattice parameter $a = 0.361$ nm is observed (Fig. 1a).

The phase composition of as-deposited samples $Fe_{0,67}Ni_{0,33}$ corresponds to *bcc*-(Fe, Ni). The *bcc* lattice parameter is a = 0,285 – 0,286 nm. Annealing at a temperature of 700 K does not lead to a change in the phase composition (Fig. 1b). The lattice parameter does not change significantly, either.

According to the data of electron microscopic and electronographic studies for both as-deposited and annealed at a temperature of 700 K, single-layer $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ samples with a thickness of d = 10 – 100 nm, their phase composition corresponds to *bcc*-Fe_xCo_{1-x} with the lattice parameter a = 0.292-0.293 *nm* (Fig. 1c, d) [9].

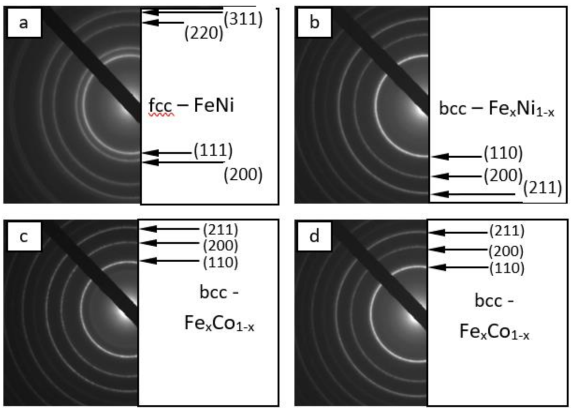
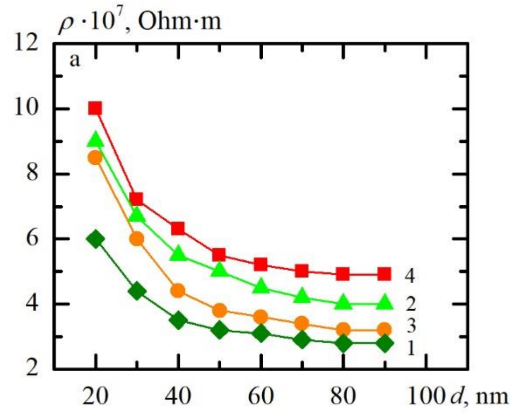


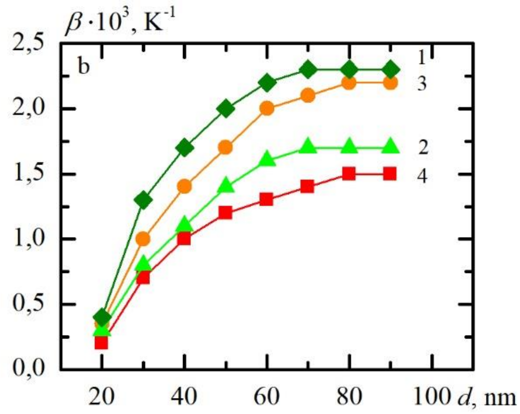
Fig. 1 – Diffraction patterns of $Fe_{0,50}Ni_{0,50}$ (a), $Fe_{0,67}Ni_{0,33}$ (b), $Fe_{0,50}Co_{0,50}$ (c) $Fe_{0,67}Co_{0,33}$ (d) film alloys annealed at 700 K

The resistivity ρ of as-deposited film alloys at T = 300 K ranges from 30 · 10 $^{-7}$ Ohm · m to 50 · 10 $^{-7}$ Ohm · m (depending on the thickness and concentration of components). High values of resiitivity can be explained not only by the size factor, which is significantly manifested at d < 10 nm, but also by the defective structure of the films themselves (relatively high concentration of vacancies and packing defects) and very small crystallite sizes.

Fig. 2 shows the typical size dependence of resistivity and TCR for all studied film alloys. As can be seen from the figure, for all the studied film alloys, the size dependence of the studied values is realized, and the nature of the size dependence is like the corresponding dependence for pure metal films obtained in Ref. [10]. The value of ρ decreases monotonically with increasing thickness and approaches the asymptotic value corresponding to ρ_∞. On the contrary, the value of TCR the contrary, increases monotonically and, at large thicknesses, reaches saturation β_∞. As the thickness decreases, the TCR value approaches zero ($d \approx 20$ nm). With a further decrease in thickness, the TCR value can have a negative value, which is due to the fact that films with small thicknesses are structurally discontinuous, and tunnel conduction is realized in them. In the following, the research results will be presented only for structurally continuous films.



(a)



(b)

Fig. 2 – Dimensional dependences of the specific resistivity ρ (a) and TCR β (b) for the alloy films of the $Fe_{0,50}Ni_{0,50}$ (1), $Fe_{0,67}Ni_{0,33}$ (2), $Fe_{0,50}Co_{0,50}$ (3) $Fe_{0,67}Co_{0,33}$ (4)

It is believed that the main factors that determine the existence of size dependence ρ and β are the contribution of additional scattering of charge carriers on the outer surfaces of the film and intergranular boundaries.

Fig. 3 shows the typical temperature dependence of electrical resistivity for film alloys with the same thicknesses, thermostabilized at a temperature of 700 K. In the temperature interval of 100 – 700 K, three characteristic areas can be indicated on the dependence of $\rho(T)$: 100 – θ_1 (**I**), θ_1 – θ_2 (**II**), θ_2 – 700 K (**III**). Extrapolation of the site **I** to 0 K makes it possible to estimate the value of the residual resistance ρ_{res}, which is associated with the scattering of conduction electrons on defects in the crystal structure and grain boundaries. Since these electron scattering mechanisms are not sensitive to temperature, section **I** is almost a horizontal line, and it is extrapolated to the value $\rho(0) \cong (2\text{-}6) \cdot 10^{-7}$ Ohm · m. Section **III** is associated with the high-temperature electron-phonon interaction since, at $T \to 0$ K it is extrapolated, like the section of the low-temperature electron-phonon interaction, to 0 K. Section **II** on the dependence $\rho(T)$ can be logically associated with magnetic interaction. Attention is drawn to the fact that the angular coefficient $\partial\rho/\partial T$ in section **III** depends more on the thickness of the films, and not on the phase composition. It is worth noting that the value of residual resistance is relatively large and depends on the concentration of components in alloys, which is typical for thin films. These results can also be explained from the point of view of strengthening the electron-phonon interaction in thin films.

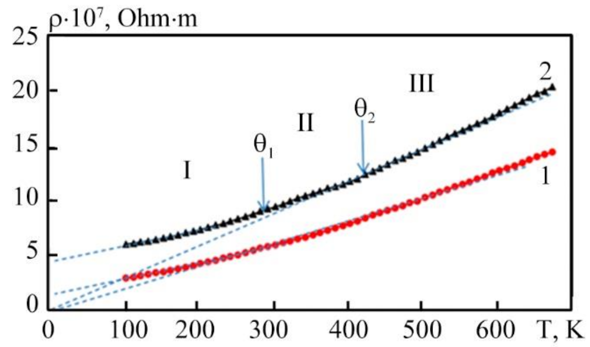


Fig. 3 – Temperature dependence of resistivity for $Fe_{0,50}Ni_{0,50}$ (1), $Fe_{0,50}Co_{0,50}$ (2) film alloys with a thickness of 25 nm

Fig. 4 shows the size dependence for the longitudinal and transverse magnetoresistance of $Fe_{0,50}Ni_{0,50}$ and $Fe_{0,50}Co_{0,50}$ film alloys as an illustration. In Figs. 4 and 5, the "–" sign for transverse magnetoresistance shows that the resistance of the demagnetized sample decreases when it is placed in a magnetic field. Note that the field dependence of magnetoresistance for all studied two-component film alloys has an anisotropic character, which is typical for magnetically homogeneous massive and film materials [11, 12]. As we can see from Fig. 4, the dimensional dependence of the value of MR for annealed structurally continuous films (d > 20 nm) of $Fe_{0,50}Ni_{0,50}$ and $Fe_{0,50}Co_{0,50}$ alloys are monotonic in nature. The value of the longitudinal and transverse MR only increases when the thickness of the samples increases, and for film alloys $Fe_{0,50}Co_{0,50}$, it reaches values of 0.6 % and 1.2 % at room temperature at a thickness of about 90 nm. For single-layer films with a different concentration of components, the size dependence has a similar character. It should only be noted that for structurally continuous film alloys $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$, and $Fe_{0,67}Co_{0,33}$, the dimensional dependence of both longitudinal and transverse magnetoresistance is much weaker. Thus, for the mentioned film alloys, the value of magnetoresistance does not exceed 0.3 %. This character of the dimensional dependence of the magnetoresistance is due to the similar character of the dimensional dependence of the saturation magnetization for these film alloys.

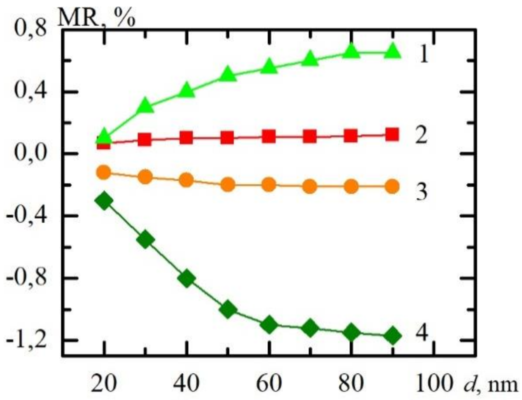


Fig. 4 – Size dependences of longitudinal (1, 2) and transverse (3, 4) magnetoresistance for $Fe_{0,50}Ni_{0,50}$ (1, 4) and $Fe_{0,50}Co_{0,50}$ (2, 3) film alloys

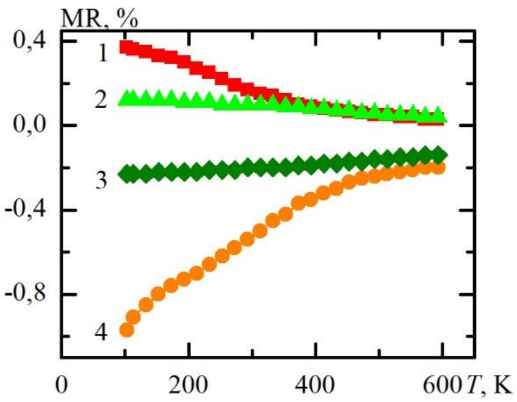


Fig. 5 – Dependence of the longitudinal (1, 2) and transverse (3, 4) magnetoresistance on the measurement temperature for $Fe_{0,50}Ni_{0,50}$ (1, 4) and $Fe_{0,50}Co_{0,50}$ (2, 3) film alloys with a thickness of 40 nm

Studies of the magnetoresistive effect of film alloys of different temperatures in the temperature range of 100-600 *K* were conducted. Fig. 5 shows the temperature dependence of the amplitude of the longitudinal and transverse magnetoresistance of samples thermally stabilized at a temperature of 700 K for $Fe_{0,50}Ni_{0,50}$ and $Fe_{0,50}Co_{0,50}$ samples. As can be seen from the Fig. 5, the magnitude of the longitudinal magnetoresistance of $Fe_{0,50}Ni_{0,50}$ film alloys decrease by 10 times and the transverse magnetoresistance by 5 times when the temperature changes from 100 to 600 K. For film alloys $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$, and $Fe_{0,67}Co_{0,33}$, the value of both longitudinal and transverse magnetoresistance depends slightly on temperature. Thus, the magnitude of magnetoresistance decreases almost linearly with increasing temperature, by only 2-3 times. The reason for such differences may be the different course of the temperature dependence of the saturation magnetization for the respective alloys.

4. CONCLUSION

The study of the phase composition of the films on the electron diffraction method showed that in all as-deposited and annealed at a temperature of 700 K $Fe_{0,50}Ni_{0,50}$ films with a thickness of d = 10-100 nm, a *fcc* phase with a lattice parameter a = 0.361 nm is observed, for $Fe_{0,67}Ni_{0,33}$ samples, a phase composition corresponds to *bcc*-(Fe-Ni). The *bcc* lattice parameter is a = 0,285-0,286 nm. The phase composition of $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ film alloys corresponds to *bcc*-Fe_xCo_{1-x} with a lattice parameter a = 0,292-0,293 nm.

Dimensional dependence of specific resistance and thermal coefficient of resistance is realized for all studied film alloys, and the nature of dimensional dependence is similar to the corresponding dependence for films of pure metals.

In the structurally continuous films $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$ the dimensional dependence of both longitudinal and transverse magnetoresistance appears much weaker than for $Fe_{0.50}Ni_{0.50}$ films, which may be due to their phase composition.

The magnitude of the longitudinal magnetoresistance of $Fe_{0,50}Ni_{0,50}$ film alloys decreases by 10 times and the transverse one by 5 times when the temperature changes from 100 to 600 K. For the film alloys $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ and $Fe_{0,67}Co_{0,33}$, the values of both longitudinal and transverse magnetoresistance depend slightly on temperature.

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Структура, електропровідність та магніторезистивні властивості бінарних плівкових сплавів на основі Fe, Co та Ni у складі сплавів Гейслера

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У роботі проведено дослідження фазового складу, електропровідності та магніторезистиних властивостей двокомпонентних плівкових сплавів на основі Fe, Co та Ni отриманих методом конденсації парів металів у високому вакуумі у температурному інтервалі 100 – 700 *К*. Показано, що у всіх свіжосконденсованих та відпалених за температури 700 *К* плівках $Fe_{0,50}Ni_{0,50}$ товщиною d = 10 – 100 *нм*, спостерігається ГЦК-фаза, для зразків $Fe_{0,67}Ni_{0,33}$ $Fe_{0,50}Co_{0,50}$ та $Fe_{0,67}Co_{0,33}$ фіксується ОЦК - фаза. Установлено, що для усіх структурно-суцільних плівкових сплавів реалізуються розмірні залежності питомого опору, термічного коефіцієнта опору та магнітоопору а характер розмірних залежностей аналогічний відповідним залежностям для плівок чистих металів. Показано, що для плівкових сплавів $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ та $Fe_{0,67}Co_{0,33}$ розмірні залежності як поздовжнього так і поперечного магнітоопору прявляються значно слабше ніж для плівок $Fe_{0,50}Ni_{0,50}$. Для плівкових сплавів $Fe_{0,67}Ni_{0,33}$, $Fe_{0,50}Co_{0,50}$ та $Fe_{0,67}Co_{0,33}$ величина як поздовжнього так і поперечного магнітоопору слабо залежить від температури.

Ключові слова: Пермаллой, Кристалічна структура, Фазовий стан, Магнітоопір, Концентраційний ефект, Температурний ефект.