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Deformation and Magnetoresistive Properties of Low Entropy Functional Film Materials Based on Fe and Pd or Pt

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In the paper, the magnetoresistive properties of low entropy nanoscale film materials based on Fe and Pd or Pt as components of high-entropy film alloys are studied. The samples were formed by the method of layer-by-layer condensation followed by annealing, which made it possible to obtain systems with an atomic ordering in which the effects of anisotropic (AMR) and giant (GMR) magnetoresistance can be realized. Studies of magnetoresistive properties were carried out in three measurement geometries. The thickness of individual layers of film materials was selected in such a way that, in accordance with the state diagrams of Fe-Pd and Fe-Pt for bulk samples, the Fe_3Pd or Fe_3Pt phases; FePd or FePt and $FePd_3$ or $FePt_3$ were stabilized in the film systems, which is confirmed by electronographic studies. In this film alloys we observed abnormally small (from 1.9 to 2.2 units) values of the strain coefficient at deformation up to 2 % and GMR signs with a small amplitude from 0.1 to 0.4 %. It was established that film alloys based on Fe and Pd or Pt can be used as sensitive elements of flexible magnetic field sensors, as they are thermally stable in a wide temperature range.

Keywords: Low entropy materials, Film alloys based on Fe and Pd or Pt, Processes of ordering atoms, Abnormally low sensitivity to deformation, Magnetoresistance.

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1. INTRODUCTION

The development of flexible electronics involves the transition to new electronic devices and systems, such as, for instance, small-sized and light-weight multifunctional sensors, electronic devices of storage and displaying information, photovoltaic panels and reconfigurable antennas, flexible biological electronic implants, printed power batteries and accumulators. Distinctive features of flexible electronic devices are transparency, thermal stability, low sensitivity to deformations, elasticity. Multicomponent film alloys (including and high entropy film alloys - HEA) are formed on the basis of ferromagnetic and noble metals. For example, a single-phase fcc solid solution is formed on the basis of Pt, Pd, Rh, Ir, Cu, and Ni metal atoms, which has high stability after annealing [1]. It was established that the alloy is uniformly deformed up to ~30 % with a high strength limit – 1839 MPa.

The main directions of the development of flexible electronics is the search for new materials for sensitive elements, as well as the adaptation of known and the development of new unique methods of forming thin-film functional elements based on metals (see, for example, [2-4]). We [5] established the reason for the realization of nanoscale thickness in film materials of abnormally small values of the strain coefficient (SC), which is understood as values of SC that are smaller than a certain limit value, which corresponds to Poisson's ratio $\mu_f = 0.5$. Low sensitivity to deformation is one of the main requirements for elements of flexible electronics.

The low-entropy film alloy considered in the work is a component of HEA obtained by the method of layer-by-layer condensation with subsequent thermal annealing (see, for example, [6]). The study of low-entropy components of HEA is important, since diffusion processes that destroy the interface are possible during layer-by-layer condensation. This will provide more meaningful information about physical processes in HEA.

Abnormally small SC values were experimentally observed by us for single-layer Pd and Pt films and two-component film alloys based on Fe and Pd or Pt (SC = 1.5-2.6 at a thickness of 10-80 nm) and by other researchers for nichrome films [6] (1.5-2.2 units at d = 5-60 nm) and invar films [7] (2.0-0.9 units at d = 20-110 nm).

It should be noted that ordered alloys based on ferromagnetic and noble metals, such as, for instance, FePd and FePt, exhibit perpendicular magnetic anisotropy under certain conditions, due to which they become candidates for the creation of new superdense magnetic recording carriers with a perpendicular orientation and elements of magnetic field sensors. In addition, Pd atoms have a high paramagnetic susceptibility and a giant magnetic moment up to $10\mu_B$, being non-magnetic in compounds with Fe and Co [8, 9].

The properties of FePd and FePt films can be controlled by the magnetic field and temperature [10], which enables their wide application in sensor electronics [11].

The purpose of the work was to study the magnetoresistive (magnetoresistance, including giant magnetoresistance) properties of nanoscale film materials based on Fe and Pd or Pt with a low coefficient of strain sensitivity in terms of their application as elements of flexible electronics.

2. METHODOLOGY AND TECHNIQUE OF THE EXPERIMENT

The samples were formed by the method of layer-by-layer condensation with the heat treatment, which made it possible to obtain systems with an atomic arrangement of atoms in which the effects of anisotropic (AMR) and giant (GMR) magnetoresistance can be obtained. Si plates with a natural oxide layer were used as substrates to reduce or avoid the influence of the substrate material on the structure of film materials. The resistivity of the samples was (1-3)·10^{-7} Om·m, which would suggest the sufficient purity of the obtained samples and the minimal influence of impurity atoms on the magnetoresistive properties. The thickness of the films was monitored by the in situ quartz resonator method (accuracy 10 %).

Strain properties of metal films on teflon substrates were investigated using a deformation device constructed on the basis of a micrometer (Fig. 1).

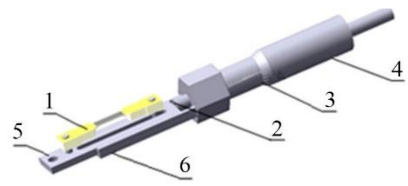

Fig. 1 – The scheme of the deformation device for CS measurement: 1 – substrate; 2 – moving rod; 3 – micrometric scale; 4 – microscrew; 5 – slider; 6 – longitudinal guide of the slider

Longitudinal deformation is determined by the ratio:

$$\Delta\varepsilon_l = \frac{\Delta l}{l_0},$$

where l_0 is the length of the film before deformation; Δl is the absolute elongation of the film, which was determined by the indicators of the microscrew. The dimensions of the film were kept constant (length l = 15 mm, width a = 5 mm).

The deformation dependences of $\Delta R/R(0)$ on the longitudinal strain ε_l were constructed for seven deformation cycles of "loading-unloading" to determine the coefficient of longitudinal strain sensitivity in the region of elastic and plastic deformation in the $\Delta\varepsilon_l$ interval,. The calculation of the value of the average coefficient of longitudinal SC was carried out by the tangent of the angle of inclination of the deformation dependences (figure 2) or by averaging the instantaneous values of SC over the entire deformation interval.

The study of the strain properties of films in the deformation interval $\Delta\varepsilon_l$ = (0-2)% (Fig. 2) was carried out using an automated system, which made it possible to perform a large number of deformation cycles in static and dynamic modes of operation at a deformation rate from 0 to 0.1 %/s.

The average and instantaneous strain coefficients were determined by the ratios:

$$\gamma_l = \frac{\Delta R}{R(0)} = \left[\frac{R(\varepsilon_l) - R(0)}{R(0)}\right], \quad SC = \frac{1}{R_i}\cdot\frac{dR}{d\varepsilon_l},$$

where $R(\varepsilon_l)$ is the resistance of the film during the ε_l deformation; $R(0)$ is the resistance of the undeformed sample.

The measurement results indicate a low value of the strain sensitivity coefficient for FePd films (from 1.8 to 2.2 units) with a film thickness from 20 to 50 nm.

Research of magnetoresistive properties was carried out in three measurement geometries, which on all dependencies have the following designations: parallel (○,||), transverse (□,┼) and perpendicular (Δ, ⊥). The value of MO was determined by the ratio

$$MR = [R(B) - R(0)]/R(0),$$

where R(B) and R(0) is the resistance of the film with a applied field and without a field.

If, under the influence of an external magnetic field, the electrical resistance of the film decreased, the inequality was fulfilled $\Delta R/R(0) < 0$, and the field dependence R had an isotropic character, then we confirm that signs of GMR are observed in such systems.

3. EXPERIMENTAL RESULTS

The thickness of the individual layers of the film materials was selected in such a way that, in accordance with the state diagrams of Fe–Pd and Fe–Pt for massive samples, different phases should be stabilized in the film systems depending on the concentration of Pd and Pt atoms and temperature. In the process of annealing in multilayers based on Fe and Pd or Pt, the following phases are formed: Fe_3Pd or Fe_3Pt (is a phase $L1_2$; $c_{Pd} <$ 50 at. % and $c_{Pt} \cong$ 14-30 at.%); FePd or FePt (is a phase $L1_0$; $c_{Pd} \cong$ 50-60 at. % and $c_{Pt} \cong$ 30-60 at. %) and $FePd_3$ or $FePt_3$ (is a phase $L1_1$; $c_{Pd} \cong$ 65-90 at. % and $c_{Pt} \cong$ 60-80 at. %). Electronographic research indicate that the phase composition of thermostabilized samples corresponds to the predicted one.

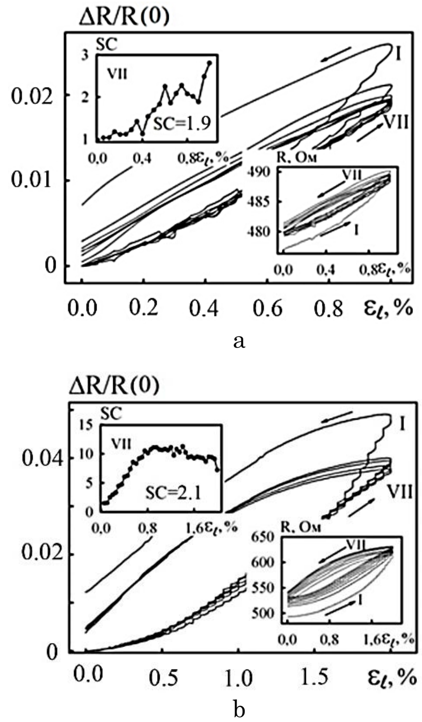

Fig. 2 – The deformation dependences for FePd/S films (30 nm) in the range of deformations (a) (0-1) % and (b) (0-2 %). S – substrate

Based on the results of studies of the crystal structure and solid-phase reactions, we proposed a scheme of the process of formation of a disordered s.s. in the process of condensation and post-condensation aging and transition to the ordered phase after annealing.

It has been established that in film systems based on

Fe and Pd or Pt due to the high diffusion mobility of atoms, caused by condensation-stimulated diffusion and the small thicknesses of individual layers that were set in advance, a s.s. is formed in the process of condensation, blurring of structural interfaces occurs, and so-called magnetic interfaces become an additional scattering mechanisms. At the same time, electrons are scattered not on the actual boundaries of the separation of layers, but on the magnetic moments of atoms of ferromagnetic and nonmagnetic metals.

In multilayers formed on the basis of Fe and Pd or Pt, additional mechanisms of electron scattering have a significant influence on their properties: structural (in the case of preserving the individuality of individual layers) or magnetic (in the case of solid solution formation) interfaces. In the second case, the spin-dependent scattering of electrons (SDSE) can be realized and the system will have antiferromagnetic ordering.

In Fig. 3 typical field dependences of MR are shown, on which GMR are observed, on the example of FePd film alloys formed based on multilayers $[Pd(1.1)/Fe(0.9\ nm)]_n/S$ (n = 3; 5; 10). The largest MR values (0.10-0.25 %) for newly condensed film materials are observed in the parallel measurement geometry, which is connected with a change in the trajectory of electron movement under the Lorentz force, when electrons, moving along a helicoid trajectory, are scattered not only at grain boundaries in the sample, but also between the metal layers. After thermal annealing of the samples to 780 K, the MR increases from 0.2 to 0.4 % (Fig.3 b, d) and peaks that arise in the process of remagnetization and correspond to the coercivity field can be observed on the dependences.

The increase of MR after annealing can be explained by some physical processes: an increase in the average size of crystallites, activation of diffusion processes and the processes of formation of an ordered hct-FePd phase, diffusion of Pd atoms into Fe layers, and an increase of spin-orbital interaction influence, which leads to an increase in ferromagnetic properties of the system after the annealing.

In the case when, under the influence of temperature, the formation of an ordered $L1_0$ structure occurs in the system, the MR values for perpendicular and transverse measurement geometries have greater values than for parallel ones.

A feature of the typical field dependences of MR for Fe_3Pt film alloys is the fact that the magnetoresistive hysteresis loops in the three measurement geometries indicate the presence of the GMR effect with a relatively small amplitude (up to 0.1%). With a gradual increase in the concentration of Pt atoms from 15 to 45 at. %, the amplitude of the effect increases slightly at first and then GMR is transformed into AMR. In FePt film alloys the GMR effect with an amplitude of 0.20 to 0.45 % is observed.

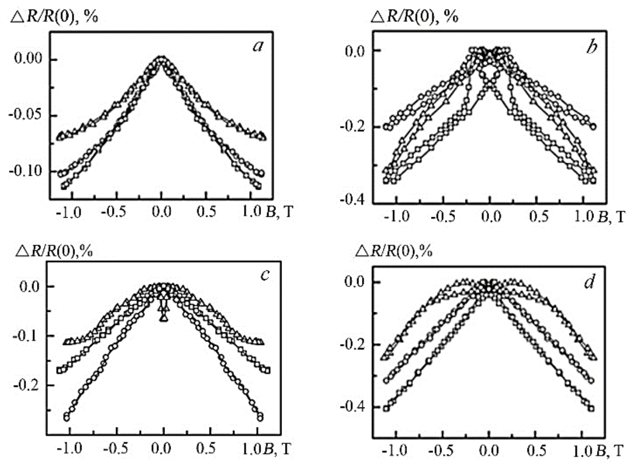

Fig. 3 – The deformation dependences for FePd/S films (30 nm) in the range of deformations (a) (0 – 1)% and (b) (0 – 2%). S – substrate

As a conclusion, film alloys of FePd and FePt ($L1_0$ phase) and Fe_3Pd and Fe_3Pt ($L1_1$ phase) can be effective as sensitive elements of flexible sensor electronics, since the $L1_0$ and $L1_1$ phases are thermally stable up to 1570 K [12], low sensitive to deformations, and the GMR or AMR effects occurs in them with weak magnetic fields (up to 1 T).

4. CONCLUSIONS

1. In film alloys FePd, FePt, Fe_3Pd and Fe_3Pt, abnormally small (from 1.9 to 2.2 units) values of the coefficient of strain sensitivity are observed at deformations up to 2%.
2. The dependence of magnetoresistance on the induction of an external magnetic field in films based on Fe and Pd or Pt in parallel, perpendicular, and transverse measurement geometries has an isotropic character, and its value, as the value of the resistance, monotonically decreases with increasing of induction.
3. In FePd and FePt film alloys with a thickness of 20 to 50 nm, signs of GMR are observed, and the MR increases a bit due to ordering processes and recrystallization processes; the MR value is (0.1-0.4) %.
4. Film alloys based on Fe and Pd or Pt can be used as sensitive elements of flexible magnetic field sensors, since FePd and FePt ($L1_0$ phase) and Fe_3Pd and Fe_3Pt ($L1_1$ phase) are thermally stable in a wide range of temperatures.

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Деформаційні та магніторезистивні властивості низькоентропійних функціональних матеріалів на основі Fe і Pd або Pt

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У статті наведені результати досліджень магніторезистивних властивостей низькоентропійних нанорозмірних плівкових матеріалів на основі Fe та Pd або Pt як складових високоентропійних плівкових сплавів. Зразки формувалися методом пошарової конденсації з наступною термооброкою, що дозволило отримати системи з атомним упорядкуванням, в яких реалізуються ефекти анізотропного (АМР) і гігантського (ГМО) магнітоопору. Дослідження проводилися в трьох геометріях вимірювання. Товщину окремих шарів плівкових матеріалів підбирали таким чином, щоб відповідно до діаграм стану Fe-Pd та Fe-Pt для масивних зразків в плівкових системах стабілізувались стійкі в широкому інтервалі температур фази FePd або FePt і $FePd_3$ або $FePt_3$, що підтверджено електронографічними дослідженнями. У таких сплавах спостерігалися аномально малі (від 1,9 до 2,2 одиниць) значення коефіцієнта тензочутливості при деформації до 2 % та ознаки ГМО з малою амплітудою від 0,1 до 0,4 %. Установлено, що плівкові сплави на основі Fe і Pd або Pt можуть бути використані як чутливі елементи гнучких датчиків магнітного поля з широким робочим діапазоном температур.

Ключові слова: Низькоентропійні матеріали, Плівкові сплави на основі Fe та Pd або Pt, Процеси впорядкування атомів, Аномально низький коефіцієнт тензочутливості, Магнітоопір.