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Structure and Magnetoresistive Properties of Three-layer Films $Co_{(1-x)}Cr_x$/Cu/Co

O.V. Bezdidko[1], S.A. Nepijko[1,2], Yu.O. Shkurdoda[1], Yu.M. Shabelnyk[1,*]

[1] *Sumy State University, 2, Rymsky-Korsakov St., 40007 Sumy, Ukraine*
[2] *Institute of Physics, Johannes Gutenberg University Mainz, 7, Staudingerweg, 55128 Mainz, Germany*

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The paper presents experimental results concerning the structural-phase state, diffusion processes and magnetoresistive properties of three-layer films $Co_{(1-x)}Cr_x$/Cu/Co/substrate (S) with Cr concentration in the upper layer up to 30 at. %. It is shown that the phase composition of both as-deposited and thermostabilized at the temperature of 700 K film samples with the layer thickness of 1÷30 nm corresponds to α(Co), f.c.c.(Cu) and $\alpha(Co_{1-x}Cr_x)$. Studies of diffusion processes have shown that in the systems $Co_{(1-x)}Cr_x$/Cu/Co/S the individuality of the layers is largely preserved both in the as-deposited state and after annealing at $T_a = 700$ K with the penetration of Cr atoms through the Cu layer. It was found that for as-deposited and annealed at temperatures of 400 and 550 K films $Co_{(1-x)}Cr_x$/Cu/Co/S with $c_{Cr} < 30$ at. % in the upper layer, $d_F = 20÷30$ nm and $d_N = 3÷15$ nm there is an atypical character of the behavior of the field dependences of the magnetoresistance is due to the different values of the spin asymmetry coefficients of the ferromagnetic layers α

Keywords: Diffusion, Anisotropic magnetoresistance, Giant magnetoresistance.

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1. INTRODUCTION

At the present stage of the development of materials science, the results of studies of the physical properties of magnetically inhomogeneous film materials are of great interest. Layered structures [1-3], or high-entropy alloys [4] in which spin-dependent electron scattering (SDES) can be realized deserve special attention. This is due to both the study of the features of SDES and the possibility of creating functional elements with improved operational characteristics [5-8].

A significant amount of experimental and theoretical results have been accumulated in the study of the phenomenon of giant magnetoresistance (GMR). It has been shown theoretically that inversion (sign reversal) of the GMR effect is possible provided that the spin asymmetry in electron scattering is opposite in adjacent ferromagnetic layers [9]. Therefore, the aim of this work was to study the features of the magnetoresistive properties of the asymmetric three-layer structures $Co_{1-x}Cr_x$/Cu/Co/substrate (S) with Cr concentration in the upper layer of $x \le 30$ at. %.

2. EXPERIMENTAL DETAILS

Three-layer films based on the $Co_{(1-x)}Cr_x$ ($x \le 30$ at. %), Co, and Cu were obtained in a vacuum chamber at a residual atmosphere gas pressure of 10^{-4} Pa. The Co-Cr film alloy was obtained by simultaneous evaporation of metals from independent sources (Cr – from a tungsten tape, Co – by an electron beam gun). Three-layer structures were formed by the method of layer-by-layer deposition. The films thickness was determined using a quartz resonator with a measurement error of ± 5 %.

The study of the crystal structure and phase composition of the films was performed by electron microscopic and electronographic methods (Transmission Electron Microscope ПЕМ-125К (SELMI, Ukraine).

The longitudinal and transverse magnetoresistance (magnetic field in the plane of the film) and the thermomagnetic treatment of the films were measured in a special setup under ultra-high 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 (1):

$$[R(B) - R(B_c)]/R(B_c), \quad (1)$$

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

3. RESULTS AND DISCUSSION

Let's first consider the structural and phase state of recently deposited and thermally stabilized three-layer films at a temperature of 700 K. Based on the electron microscopic and diffraction studies, it was found that the as-deposited films have a fine-crystalline structure (grain size less than 10 nm) (Fig. 1a). For these as-deposited samples, wide diffraction rings, obviously belonging to the f.c.c.(Cu), α(Co), h.c.p. α(Co-Cr) phases, are electronically detected (Fig. 1b, Table 1). Consequently, during the formation of the coating layer, a solid solution of chromium in cobalt Co (Cr) is formed, and this phase has a h.c.p. lattice. This statement is confirmed by the results of [10, 11] according to which the formation of solid solutions in the Co-Cr system fulfills the size criterion (the maximum difference in atomic radii is 2.4 %) and the proximity of electronegativity (1.7 for Co and 1.6 for Cr). In addition, according to the phase diagram of the Co-Cr system [12] (Fig. 2) at the concentration Cr $x \le 30$ at. %, a ferromagnetic solid solution based on h.c.p. lattice was formed.

After annealing at a temperature of 700 K, in the $Co_{(1-x)}Cr_x$/Cu/Co/S films the same phases are present as

* y.shabelnyk@aph.sumdu.edu.ua

in the just deposited samples: α(Co), f.c.c.(Cu), α(Co-Cr) (Fig. 1d). The width of the diffraction maxima after annealing decreases significantly, but the lines of the α(Co) phases and solid solution α(Co-Cr) as a result of close interplanar distances, the electron diffraction patterns are not separated.

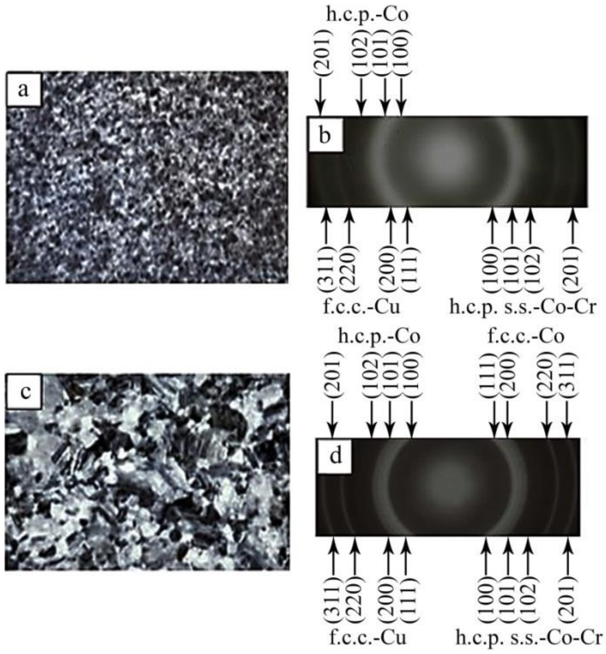

Fig. 1 – Crystal structure and electrograms of as-deposited (a, b) and annealed at a temperature of 700 K (c, d) $Co_{(1-x)}Cr_x$/Cu/Co film system (x = 20 at. %, $d_{Co\text{-}Cr}$ = 25 nm, d_{Cu} = 10 nm, d_{Co} = 25 nm)

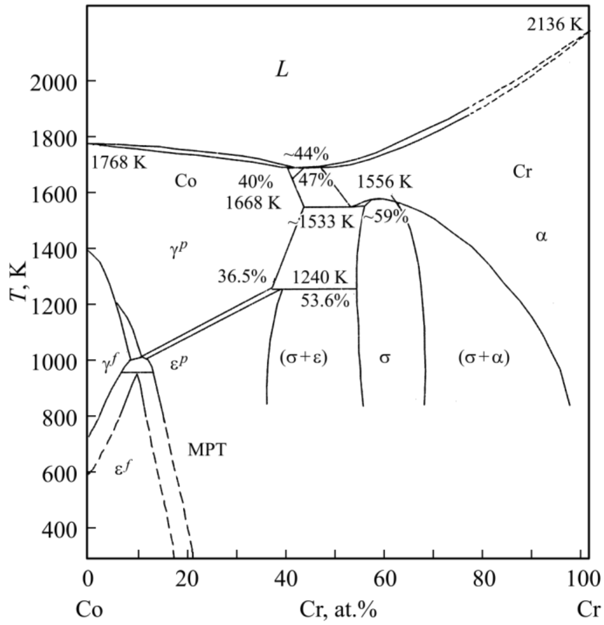

Fig. 2 – State diagram of the Co-Cr system [10] ε^f and ε^p are solid solutions based on h.c.p.(Co) with ferromagnetic and paramagnetic properties, respectively; γ^f and γ^p are solid solutions based on f.c.c.(Co) with ferromagnetic and paramagnetic properties, respectively

It should be noted that the authors of [13, 14] showed the metastable (s.s.) h.c.p.(Co-Cr) solid solution with a lattice parameter a = 0.355-0.362 nm in Co/Cu films after annealing at $T_a \geq 600$ K. This Co-Cu solid solution partially decomposes upon cooling to 300 K with the release of α(Co) particles. In the case of the $Co_{(1-x)}Cr_x$/Cu/Co/S system, the formation of Co-Cu solid solution was not detected in the electron diffraction patterns, possibly due to the relatively large thickness of the Co layer (d_{Co} = 20-30 nm) and the small thickness of the copper layer (d_{Co} = 2-10 nm). The average grain size in annealed films, compared to as-deposited, increases by about 5-10 times, depending on the thickness of the layers (Fig. 1a, c).

Table 1 – Deciphering of electrograms from as-deposited and annealed in high vacuum to T_a = 700 K $Co_{(1-x)}Cr_x$/Cu/Co (x = 20 at. %, $d_{Co\text{-}Cr}$ = 25 nm, d_{Cu} = 10 nm, d_{Co} = 25 nm) film system

As-deposited				Annealed to 700 K			
No	d, nm	*hkl*	Phase	No	d, nm	*hkl*	Phase
1	0.216	100	h.c.p-Co, h.c.p-s.s. Co-Cr	1	0.217	100	h.c.p-Co, h.c.p-s.s. Co-Cr
2	0.207	111	f.c.c.-Cu	2	0.205	111	f.c.c.-Cu
3	0.191	101	h.c.p-Co, h.c.p-s.s. Co-Cr	3	0.190	101	h.c.p-Co, h.c.p-s.s. Co-Cr
4	0.182	200	f.c.c.-Cu	4	0.177	200	f.c.c.-Cu
5	0.148	102	h.c.p-Co, h.c.p-s.s. Co-Cr	5	0.146	102	h.c.p-Co, h.c.p-s.s. Co-Cr
6	0.126	110 220	h.c.p-Co, h.c.p-s.s. Co-Cr, f.c.c.-Cu	6	0.127	220	f.c.c.-Cu
7	0.107	311 201	f.c.c.-Cu, h.c.p-Co, h.c.p-s.s. Co-Cr	7	0.125	110	h.c.p-Co, h.c.p-s.s. Co-Cr
8				8	0.107	311 201	f.c.c. - Cu h.c.p - Co, h.c.p - s.s. Co-Cr

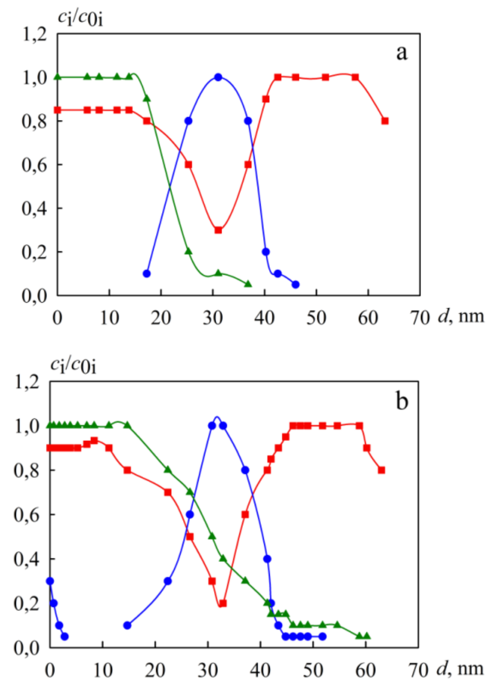

Fig. 3 – Diffusion profiles for three-layer films of $Co_{(1-x)}Cr_x$/Cu/Co (x = 20 at. %, $d_{Co\text{-}Cr}$ = 25 nm, d_{Cu} = 10 nm, d_{Co} = 25 nm) in as-deposited (a) and annealed to 700 K (b) state (♦–Co, ●–Cu, ▲–Cr)

As a result of a layer-by-layer analysis of the components of these systems (Fig. 3a), it was found that the just deposited samples obtained at room temperature, irrespective of the thickness of the layers, have a small region of mutual diffusion. The formation of this region can be associated with condensation-stimulated diffusion and diffusion along the intergranular borders (see, for example, [15, 16]).

Annealing of samples with layer thickness d_{Co} = 20-30 nm, $d_{Co\text{-}Cr}$ = 20-30 nm and d_{Cu} = 5-15 nm at temperature T_a = 700 K leads only to insignificant further interpenetration of Co and Cu atoms (Fig. 3b). Although it should be noted that the diffusion of Cr atoms during annealing is much more intense. For annealed films, Cr atoms penetrate through the Cu layer and subsequent interaction of Co and Cr atoms in the Co base layer occurs.

Consider the results of a study of the magnetoresistive properties of three-layer films. For $Co_{(1-x)}Cr_x$/Cu/Co/S as-deposited (Fig. 4a) and annealed films at temperatures of 550 K (Fig. 4b) and 700 K (Fig. 4c) with $x < 30$ at. %, d_F = 20-30 nm, d_N = 3-10 nm, atypical behavior of the field dependences of the magnetoresistance is observed (for anisotropic and giant magnetoresistance) (Fig. 4a, b, d) [17, 18]. As the induction of the external magnetic field decreases from the saturation field B_S to 0, the value of the electrical resistance first increases slightly and then sharply decreases. Then, after changing the direction of the external magnetic field, the resistance reaches its minimum value and increases sharply, reaching its maximum value with a further increase in the magnetic field induction. A further increase in the magnetic field induction leads to a sharp decrease in the magnetoresistance. Note that it is impossible to explain the course of the field dependences of the magnetoresistance by the superposition of spin-dependent electron scattering and spin-orbit interaction.

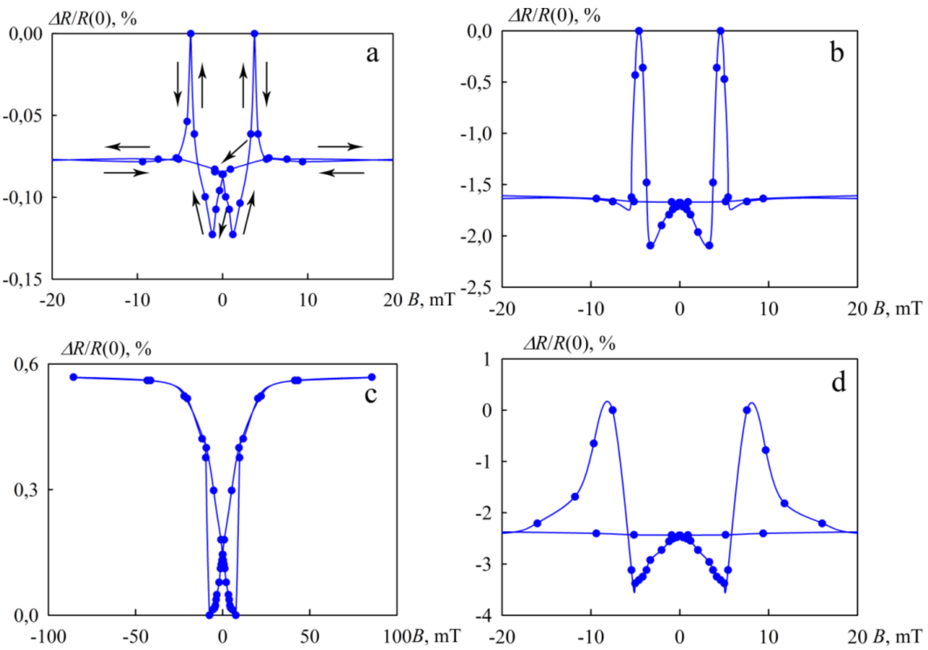

Fig. 4 – Dependence of the longitudinal MR on the induction of an external magnetic field for the as-deposited (a) and annealed T_a = 550 K (b, d) and 700 K (c) $Co_{(1-x)}Cr_x$/Cu/Co (x = 20 at. %, $d_{Co\text{-}Cr}$ = 20 nm, d_{Cu} = 5 nm, d_{Co} = 20 nm) film system (f, b, c – temperature measurement is 293 K; d – 120 K)

Normally, the influence of anisotropic magnetoresistance in structures with spin-dependent scattering of conduction electrons manifests itself in different values of magnetoresistance in fields large beyond the saturation field for transverse and longitudinal measurement geometries. The reason for the appearance of additional minima in the field dependences of magnetoresistance, in our opinion, may be different values of the spin asymmetry coefficients of the ferromagnetic layers α ($\alpha < 1$ for Co-Cr alloy and $\alpha > 1$ for Co film). The value of the magnetoresistance at room temperature is 0.3-0.5 % for just deposited samples, and 1-2 % for annealed samples at a temperature of 550 K. With a decrease in the measurement temperature to 120 K (Fig. 4d), the nature of the field dependences of the magnetoresistance does not change significantly.

The abnormal behavior of the field dependences disappears for films with d_{Cu} = 5-15 nm after annealing at a temperature of 550 and 700 K, depending on the Cr content. So, at the concentration of Cr x = 5-10 at. % transition to typical field dependences occurs at a temperature of 700 K (Fig. 4c). Moreover, at d_{Cu} = 5-10 nm, typical anisotropic character of magnetoresistance is recorded. At d_{Cu} = 10-15 nm, isotropic field dependences are observed which are typical for GMR. For samples with the concentration of Cr x = 15-30 at. % in the covering layer, the transition to typical isotropic field dependences occurs at a temperature of 550 K. The dependence of the annealing temperature after which the anomalous behavior of the field dependences of the magnetoresistance on the Cr content disappears, in our opinion, is due to the different intensity of diffusion of Cr atoms in the base Co layer and the formation in it solid solution Co-Cr. The cause of the anisotropic character of magnetoresistance is the destruction of the structural integrity of the copper interlayer.

4. CONCLUSIONS

As-deposited films $Co_{(1-x)}Cr_x$/Cu/Co with the concentration of Cr in the upper layer x = 5-30 at. % consist of h.c.p.(Co), h.c.p.(Co-Cr) and f.c.c.(Cu). After annealing at a temperature of 700 K, the change in phase composition is not electronographically recorded.

The results of the study of diffusion processes showed that in systems $Co_{(1-x)}Cr_x$/Cu/Co/S the individuality of the layers is largely preserved both in the as-deposited state and after annealing at T_a = 700 K. For annealed films, Cr atoms penetrate through the Cu layer and subsequent mutual diffusion of Co and Cr atoms in the base layer.

For just deposited and annealed to 400 and 550 K films $Co_{(1-x)}Cr_x$/Cu/Co/S with $x < 30$ at. %, d_F = 20-30 nm, d_N = 3-10 nm the atypical character of behavior of field dependences of magnetoresistance caused by various values of coefficients of spin asymmetry of ferromagnetic layers is observed α ($\alpha < 1$ for Co-Cr alloy and $\alpha > 1$ for Co film).

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Структурні і магніторезистивні властивості тришарових плівкових структур $Co_{(1-x)}Cr_x$/Cu/Co

О.В. Бездідько[1], С.О. Непийко[1,2], Ю.О. Шкурдода[1], Ю.М. Шабельник[1]

[1] *Сумський державний університет, вул. Римського-Корсакова, 2, 40007 Суми, Україна*
[2] *Institute of Physics, Johannes Gutenberg University Mainz, 7, Staudingerweg, 55128 Mainz, Germany*

В роботі представлені експериментальні результати стосовно структурно-фазового стану, дифузійних процесів та магніторезистивних властивостей тришарових плівок $Co_{(1-x)}Cr_x$/Cu/Co/підкладка (П) з концентрацією Cr у верхньому шарі до 30 ат. %. Показано, що фазовий склад як свіжосконденсованих так і термостабілізованих при температурі 700 К плівкових зразків з товщиною шарів (1÷30) нм відповідає α(Co), ГЦК (Cu) та $\alpha(Co_{1-x}Cr_x)$. Дослідження дифузійних процесів показали, що у системах $Co_{(1-x)}Cr_x$/Cu/Co/П в значній мірі зберігається індивідуальність шарів як в невідпаленому стані, так і після відпалювання при $T_{відп}$ = 700 К з проникненням атомів Cr через прошарок Cu. Встановлено, що для свіжосконденсованих та відпалених за температур 400 та 550 К плівок $Co_{(1-x)}Cr_x$/Cu/Co/П з $c_{Cr} < 30$ ат. % у верхньому шарі, d_F = 20-30 нм та d_N = 3-15 нм спостерігається нетиповий характер поведінки польових залежностей магнітоопору обумовлений різними значення коефіцієнтів спінової асиметрії феромагнітних шарів α.

Ключові слова: Дифузія, Анізотропний магнітоопір, Гігантський магнітоопір.