JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. **8** No 4(1), 04028(7pp) (2016)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том **8** № 4(1), 04028(7cc) (2016)

Physical Properties of Film Alloys Based on Ferromagnetic and Noble Metals (review). II. Film Materials Based on Co and Ag or Au

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(Received 10 September 2016; published online 29 November 2016)

The results of analysis of literature and own results on the phase composition, electrophysical and magnetoresistive properties of film based granular alloys Co and Ag or Au. As between film systems based on Fe and Ag or Au and Co and Ag or Au much in common, then this work can be seen as a continuation of [1], although we observed significant withdrawal of these two groups of film materials in structural-phase state, causing some change in electrophysical and magnetoresistive properties. The increase of the value of temperature coefficient resistance film Ag (Co) at the $c_{Co} \geq 26$ at.% and the strain coefficient at the $c_{Co} \cong 70$ at.%. It is concluded that the value of GMR completely determined by the temperature measurement and perfection of granulate state.

Keywords: Films Ag(Co) and Au(Co), Structural and phase state, Electrophysical and magnetoresistive properties, Temperature coefficient of resistance, Strain coefficient.

DOI: 10.21272/jnep.8(4(1)).04028 PACS numbers: 75.70.i, 75.47.Np

1. INTRODUCTION

In a previous paper [1] we analyzed the issue of phase composition and electrophysical and magnetoresistive properties of the film materials based on Fe and Ag or Au. Particular attention is drawn to the fact that so far no single system of symbolic notation of samples in the initial state in their formation methods simultaneously or layered condensation (allowed many authors incorrect designation: ($Me_1 - Me_2$), Me_1/Me_2 and $(Me_1)_x/(Me_2)_{1-x}$). Even in more inconsistency takes place in the marking of samples in the form of solid solutions with elements of granular state or not, which studies the electrophysical and magnetoresistive properties.

Orderly our notation more accurately describes the structure-phase state film samples to facilitate the understanding of processes in films as a solid solution (s.s.) or granular alloy film $Me_1(Me_2) + G$. It is also necessary to point out that, despite the many similarities films between systems based on Fe and Ag or Au and Co and Ag or Au, we realized clear their separation, as in the first case are formed granulated s.s. both on fcc Ag or Au, and based on the bcc Fe. In the case of films based on Co and Ag or Au, the formation s.s. based on Ag or Au. This it served the cause reason for us to consider these two groups of film materials separately. In the said withdrawal should also focus on the possibility of an orderly phase formation Ll_0 FeAu at the concentrations $c_{Fe} > 50$ at.% (see, references [47, 48] in work [1]), which was observed in the films based on Co and Ag or Au.

2. FILM MATERIALS BASED ON Co AND Ag

2.1 Structural and Phase State

Great interest in the physical properties of film systems based on Co and Ag stimulated considerable amount of research phase state as bilayer films and multilayers [2, 3-11]. In one of the first work [3], which were performed diffraction studies phase of multilayers Co/Ag prepared by molecular beam epitaxy, it was concluded that the formation of granules Co in the Ag matrix. The authors [3] called this granular system metastable alloy, although designated as intermetallic CoAg (that again is a terminological inconsistency, as discussed in work [1]). Interpretation of the metastable alloy as that found in other authors, it's just kind of extrapolation to bulk samples, although under the thermodynamics of small systems in the film samples may increase the equilibrium concentration of dissolved components. In other words, granulated film alloys should be interpreted as equilibrium thermodynamic system. The authors of [9] based on XRD studies concluded that the formation of films based on Co and Ag oversaturated (this interpretation in terms of bulk samples) s.s. Ag (Co) with fcc lattice решіткою (a = 0,4073-0,4083 nm). Note that the authors [9] as [3-6, 11], the observed X-ray lines separately from fcc s.s. Ag(Co) (although it focuses on this attention, and designate them as lines Ag) and hexagonal Co (obviously this is a reflection of granules Co). Summary of diffraction studies [10] indicate that the lattice parameter s.s. Ag (Co) very little different from the lattice parameter Ag films in a wide range of concentrations Co atoms (Fig. 1).

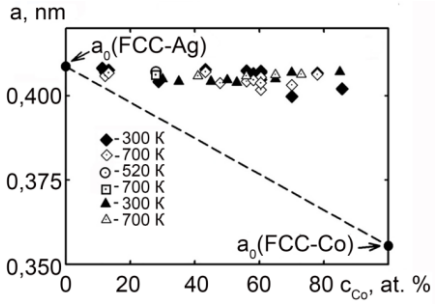

Fig. 1 – Dependence of parameter lattice s.s.Ag(Co) versus common concentration of Co atoms in film system. From work [10]

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This can be interpreted as stabilization of s.s. Ag(Co) substitution, which causes a slight decrease in the lattice parameter. Note that in work [2] presented a similar dependence a versus c_{Fe}. Based on the data [2, 3, 10] can be stated the fact that in the concentration range up to 80 at. % Co is no phase transition to the cubic or hexagonal Co, as is the case with films based on Fe and Ag or Au, where observed phase transition fcc s.s. Ag (Fe) or Au (Fe) → s.s. α-Fe(Ag) or α-Fe(Au) [14, 22, 23].

2.2 Electrophysical Properties

One of the first works, which studied the electrical properties (we are talking about TCR) granulated multilayers [Ag(4)/Co(0,2-1,0)]15/S, is the work [15]. The authors have established that at the $d_{Co} \le 0{,}5$ nm dependence of TCR versus d_{Co} abruptly changes its inclination and value (Fig. 2). This behavior TCR authors explain superparamagnetic as granules the $d_{Co} \le 0{,}5$ nm and an effective spin-dependent scattering of electrons (SDSE) on magnetic granules. This conclusion is partly confirmed by measurement of resistivity: $\rho \cong 3{,}4 \cdot 10^{-7}$ Ohm·m ($d_{Co} < 0{,}5$ nm) and $\rho \cong 2{,}8 \cdot 10^{-7}$ Ohm·m ($d_{Co} > 0{,}5$ nm), i.e. the low ohmic spin channel causes a general decrease of resistivity and in accordance with the ratio $\beta = (1/\rho)(\partial\rho/\partial T)$, – an increase of TCR.

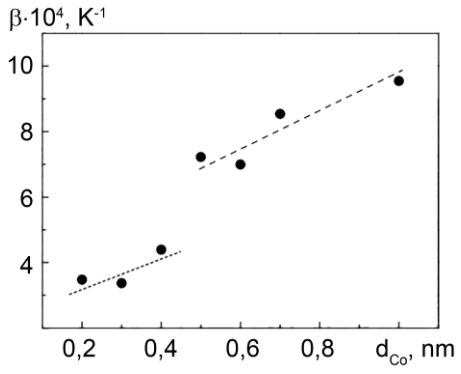

Fig. 2 – Temperature coefficient of resistivity versus Co thickness in granular multilayers $[Ag(2)/Co(d_{Co})]_{15}$ at RT. Decrease of TCR happens at the $c_{Co} \cong 26$ at. %. From work [15]

Systematic research of electrical properties held by us in work [16], whose purpose was to examine the impact of granular state film samples based on Co and Ag their electrical properties and compared the experimental values TCR with billing based on the phenomenological model for the temperature coefficient of resistance granular thin film alloys.

Single-layer and two-layer film systems ((Ag + Co)/S, Ag/Co/S) were obtained in vacuum chamber VUP-5M at a gas pressure of residual atmospheric 10^{-3}-10^{-4} Pa. When condensation Ag used resistive method of deposition and evaporation Co conducted as using electron beam gun, and resistive method. Film deposition was conducted as in one-layer from two sources and alternating condensation component.

In both cases there was stabilization of the granular film s.s. Ag(Co) (Fig. 3) in the process of condensation and the next annealing.

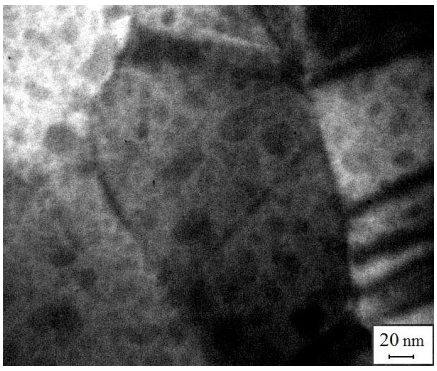

Fig. 3 – Microstructure of granular alloy, obtain at the annealing of film [Ag(17) + Co(26)]/S to 800 K. S – substrate

The calculation of the concentration Co atoms is based on the ratio:

$$c_{Co} = \frac{D_{Co} d'_{Co} \mu_{Co}^{-1}}{D_{Co} d'_{Co} \mu_{Co}^{-1} + D_{Co} d_{Ag} \mu_{Ag}^{-1}}$$

where D – density, μ – atom mass, d'_{Co} – effective thickness of Co film, with atoms formed of s.s. and granules, which calculated based on ratio:

$$d'_{Co} = d_{Co} - \frac{4}{3}\pi r_0^3 \cdot \frac{d_{Co} + d_{Me}}{\left(2r_0 + \Delta l_{gr}\right)^3} \qquad (2)$$

Control estimated concentration of c_{Co} was carried out by X-ray microanalysis.

Typical temperature dependence of the resistivity for the system illustrated in Fig. 4. When heated in the first cycle in the temperature range 300-600 K is a slight increase of resistivity, and a decrease in the range of 600-850 K, which may be associated with healing of defect structure and recrystallization processes which cause reduction of grain-boundary contributions carrier scattering in the total resistance of the system. Upon cooling, the resistivity decreases monotonically, due to the completion of relaxation processes at the first cycle of annealing.

The experimental dependence of the effective concentration of the Co in film systems obtained by sequential (points 1 and 3) or both (points 2 and 4) condensation film systems with a total thickness of 40-50 nm show on Fig. 5. The results of up to 10 % consistent with the calculation based on the ratio of (1) specified in work [1].

We have also studied the strain properties of granular films based on Co and Ag (in Section 3.2 [1] presented similar results for films based on Au and Fe). Typical depending $\Delta R/R(0)$, R and $(\gamma_l)_{dif} = \frac{1}{R_i} \cdot \frac{\partial R_i}{\partial \varepsilon_{li}}$ versus ε_l shown in Fig. 6. The resulting average values $(\gamma_l)_{dif}$ correlate well with those values for single-layer films Ag(38)/S (i.e. $c_{Co} = 0$ at.%) and Co(70)/S (i.e. $c_{Ag} = 0$ at.%), which is illustrated in Fig. 7.

Note that the cause anomalous low values (Fig. 6b, c; Fig. 7) can be explained by the strain dependence of electrocarried parameters (specular reflection from external surfaces and the transmission parameter at

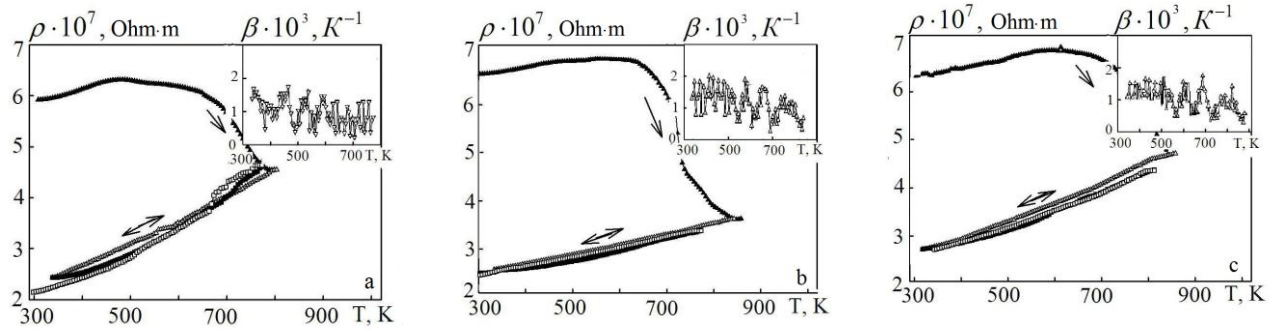

Fig. 4 – Dependence of resistivity and temperature coefficient of resistivity (in the insets) versus temperature for films Ag(34)/Co(15)/S (a), Ag(21)/Co(18)/S (b) and [Ag(22) + Co(15)]/S (c). Effective concentration of atoms Co, at.%: 32 (a), 52 (b) and 45 (c)

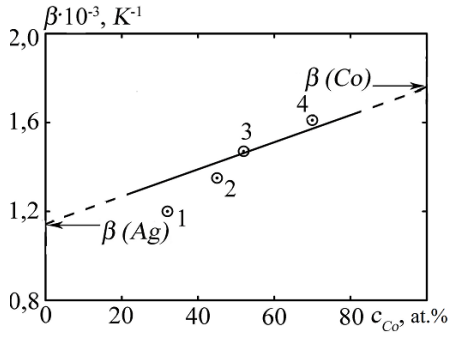

Fig. 5 – Experimental dependence β of granular s.s. Ag(Co) versus effective concentration of Co atoms. The values β (Ag) and β (Co) correspond film thickness of 40 nm

the grain boundary of electrons, SDSE on granules and the mean free path of electrons λ_0) [12]. Obviously, these strain dependences can specify both an increase and decrease $(\gamma_l)_{dif}$ and $(\gamma_l)_{\text{int}}$, as observed in this case. According to modern concepts increase γ_l fully determined by the efficiency of grain boundary scattering of electrons and strain dependence λ_0, while decrease γ_l connected with efficiency of surface electrons scattering. As follows, at the transition to film systems based on amorphous and polycrystalline films there are relatively large value (to 30 units) $(\gamma_l)_{dif}$ and $(\gamma_l)_{\text{int}}$ (see, for example, results [13], for film system based on Fe and a-Gd). This can be explained by an effective grain boundary scattering of electrons in the film Fe and the strong of strain dependence λ_0 in the film a-Gd.

2.3 Magnetoresistive Properties

When analyzing the MR properties known work, we conditionally divide into two groups: a) work, which presents the results for multilayers where exceptionally important role played by the interface electron scattering and possibly missing elements of granular state [15, 17-21], and b) work, which samples represent granulated alloys where exceptional role SDSE on magnetic granules Co [2-6, 9, 12, 23-29].

Summary of results of the first group of works presented in Table 1.

The results [15, 17, 20, 21] may need to be clarified in terms of their structural state because under the same experimental conditions the authors [4] observed in multilayers Co/Ag formation of Co granules with size 1.5 nm. Perhaps, SDSE on the interfaces and granules causes great value GMR ≅ 22 % as in work [4] and in works [17, 20, 21].

Table 2 presents compiled our and the results by different authors who interpret the structural state of the samples as granular alloy.

Comparing the GMR value for two conventional groups that we proposed (Tables 3 and 4), probably unnecessary, since in pure multilayers not realizated [4]. The GMR value is primarily determined by the temperature of measuring and total (effective) concentration of magnetic components based on formed by system of granules.

3. FILM MATERIALS BASED ON Co AND Au

3.1 Structural and Phase State

In early works (see, for example, [31, 32]) question about phase state superlattices Co/Au analyzed and no attention was paid to the influence of the thickness of Co layers on the value GMR (see, Subsection 2.3). Most likely work [33] was the first, which focuses attention on granular state s.s. Au (Co) in films with a thickness 50 nm. Granules size ranged from 1.5-3 to 10 nm, which substantially affect the value of GMR. Conclusion regarding granular state was later confirmed in works [10, 34].

3.2 Electrophysical Properties

These electrophysical properties as resistivity, temperature coefficient of resistance or strain coefficient regarding of granular film alloys s.s. Au (Co) currently remain poorly understood and, in general, are not in sight researchers. In this connection, we emphasize the work [35], which is devoted to the study of transport properties model peculiar film system, which of Co_n clusters (diameter of 1-3 nm) embedded in a Au matrix. The authors showed that in this case the temperature dependence ρ is consistent with the model of Bloch-Gruneisen and value λ_0 the mean free path of electrons are found in the ratio:

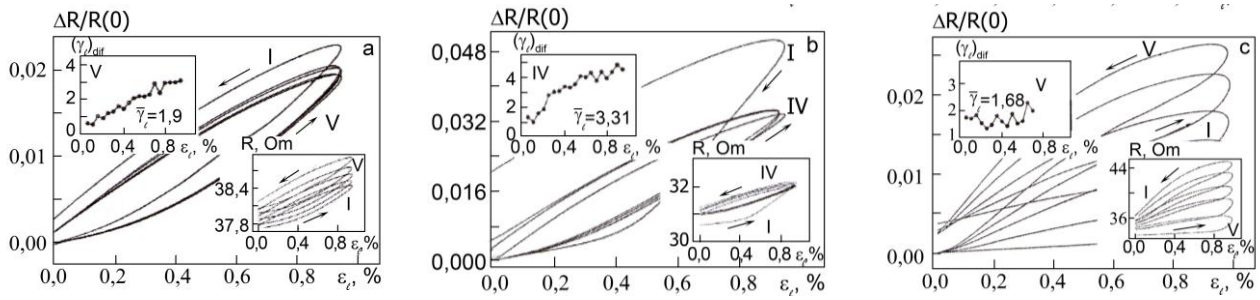

Fig. 6 – The strain dependences $\Delta R/R(0)$, R and $(\gamma)_{dif}$ for granular films s.s.Ag (Co) with different common concentration c_{Co}, at.%: 85 (a), 73 (b) and 52 (c). I – V – numbers of strain cicles

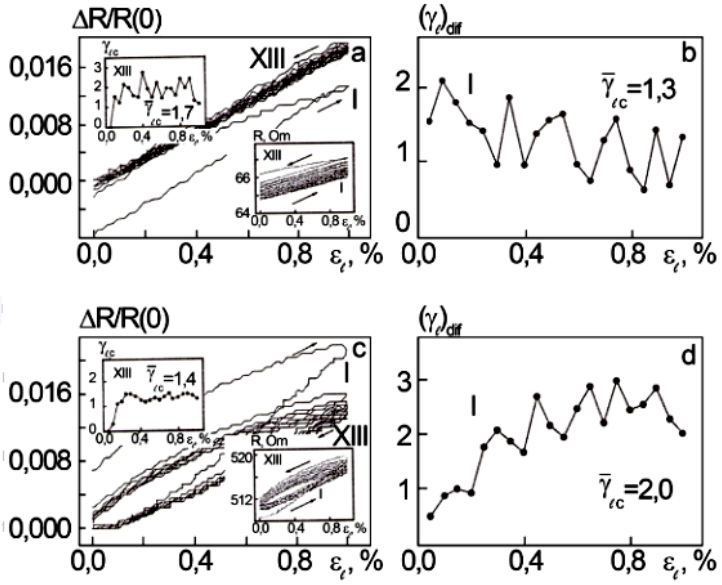

Fig. 7 – Dependence $\Delta R/R(0)$, R and $(\gamma)_{dif}$ versus ε_l for film Ag(38)/S (a, b) and Co(70)/S (c, d) [36]

Table 1 – GMR in multilayers based on Ag and Co

No	Film system (in the notation authors)	Experimental technique	T_a^{**}, K	GMR, %	Literary source
1	Co/Au/Co	T_s = 240-300 K	–	There was great MR	[30]
2	Ag(0,2-6)/Co(0,6-1,5); Common thickness to 1000 nm	T_m^* = 4,2K CIP-MR and CPP-MR	4,2	50 (CPP-MR); $\pi = \frac{CPP-MR}{CIP-MR} = 10$ (at the $d_{Ag} \cong$ 6 nm)	[18, 19]
3	Multilayers $[Ag(1\text{-}4)/Co(0,2\text{-}2)]_{15}$	Ion sputtering T_s = 300 K	–	0,12 % ($d_{Ag} \cong$ 0,2); 1,2 %($d_{Ag} \cong$ 4 nm)	[15]
4	$[Co(0,5\text{-}3)/ Ag(1,5)]_{20}$ $[Co(1,5)/Ag(1,5)]_{50}$	Electrodeposition Ion sputtering T_s = 300 K	– –	9,2 9,1	[20] [21]

* T_m – measurement temperature; ** Ta – annealing temperature

Table 2 – GMR in granular multilayers and film systems based on Ag and Co

№	Film system (in the notation authors)	Experimental technique	Size of Co granules, nm	T_a, K	GMR, %	Literary source
1	2	3	4	5	6	7
1	Granular system Co-Ag as a metastable alloy	Magnetron sputtering, T_s = 77 K, $d \cong$ 2000 nm	2 13	470 875	50; 61; 67; 84	[22]
2	Epitaxial Co/Ag granular film alloy	T_s = 300, 570, 720 K	–	–	10 (32 at.%Co); 13 (45 at.%Co);	[3]

1	2	3	4	5	6	7
3	Single-layer films $Co_x Ag_{1-x}$, where $8 \le x \le 46$ at. %	Simultaneous ion sputtering components, T_s = 400 K	–	300 500	3 (7,5 at.%Co), 6 (17 at.%Co); 2,2 (7,5 at.%Co), 9 (20 at.%Co); 13,8(22 at.%Co)	[23]
4	Granular multilayer $Co_{21}Ag_{79}$(50)	Simultaneous condensation, T_s = 520 K	1,5	295	21,5	[4]
5	Granular films Co_xAg_{1-x}, where $0 \le x \le 100$ at. %	Simultaneous condensation, T_s = 300, 400 K	the average size 10 nm	300 500	9,5 13,0	[5]
6	Granular alloy Ag(20)/Co(d)/Ag(20); Co(0,1-1)/Ag(1); Co(0,25)/Ag(0,5-2)	Simultaneous condensation	–		– 12 12	[30]
7	Magnetron sputtering alloy Co- Ag; different meaning Co_xAg_{1-x}	Simultaneous condensation, thickness 220 nm	2,2 (26 at.%), 5,1 (40 at. %)	–	34 (10K; 34 at.%Co); 10,1 (300K; 34 at.%Co);	[24]
8	Nanogranular system Co/Ag; different meaning Ag-Co with segregation of Co atoms	Magnetron sputtering of alloy, T_s = 300 K	–	500	90,0 (T = 5 K); 39,8 (T = 570 K)	[9]
9	Granular film alloy Ag-Co	Co-sputtering, d = 300 nm	–	–	15 (31 at.%Co)	[26]
10	Granular films Ag-Co with c_{Co} = 5-40 at.%; different meaning Co_xAg_{1-x}	Electrodeposition	3,5-9	–	At the 10 K: 3 (2 at.%Co), 15 (14 at.%Co), 13,5 (40 at.%Co); At the 300 K: 0,3 (2 at.% Co), 3,5 (14 at.% Co), 3,4 (40 at.% Co),	[27]
11	Granular films Co_xAg_{1-x} (x = 33-52 at. %)	Sputtering of alloy, d = 310 nm	9-18	570-720	1,5-4,5	[6]
12	Granular films Co-Ag	Electrodeposition	–	–	To 6 (20-30 at.%Co)	[28]
13	Nanocrystalline films Co-Ag; different meaning CoAg	Laser vaporization, T_s = 300 K, d = 70 nm	Grain sizes, nm: < 5 (as-deposition); 10 (on film/S/ interface)		16 (at the 10 K); 12 (at the 77 K); 3,8 (at the 300 K);	[29]
14	Ferromagnetic alloys Co-Ag	Electron-beam evaporation and others. technology	–	–	To 6	[2]

$$\frac{1}{\lambda_0(\mathrm{Au}+\mathrm{Co}_n)} = \frac{c_{\mathrm{Au}}}{\lambda_0(\mathrm{Au})} + \frac{c_{\mathrm{Co}}}{\lambda_0(\mathrm{Co})},$$

allowing indirectly contribute to develop the opinion on clusters of transport properties (n – the number of atoms in cluster).

Table 3 – Summary data for granulated film alloys

Film system in the initial state	c_{Co}, at.%	$\rho \cdot 10^7$, Ohm m, T_m = 300 K	$\beta \cdot 10^3$, K^{-1} averaging in the range of 300-550K
Au(4)/Co(14)/S	84	4,52	1,56
Au(8)/Co(14)/S	73	3,70	1,49
Au(29)/Co(14)/S	42	1,90	1,37
Au(48)/Co(10)/S	23	1,00	1,27

We have studied the temperature dependences ρ and TCR of film systems Au(4-48)/Co(10-14)/S. Fig. 8 illustrates the phase composition and crystal structure granular s.s. Au(Co) at different effective concentrations of atoms Co, formed in the layer of condensation of Co and Ag. The temperature dependence of ρ and TCR shown in Fig. 9.

Summary results of the study of the electrical properties are shown in Table 3. At first glance there appears a clear concentration dependence ρ and TCR (and this is to a large extent is), but because the concentration is given thickness of the individual layers, in dependence $\rho(c_{Co})$ and $\beta(c_{Co})$ a contribution and provides dimensional effect.

3.3 Magnetoresistive Properties

An analogy with the analysis of the survey results

Table 4 – GMR in granular multilayers and film systems

№	Film system (in the notation authors)	Experimental technique	Size of granules Co, nm	T_a, K	GMR, %	Literary source
1	Superlattices Co-Au; different meaning Co/Au; $[Co(0,5)/Au(1,6)]_{60}$ $[Co(3)/Au(1,6)]_{30}$	Molecular beam epitaxy	–	–	15 (T = 4,2 K) 2 (T = 4,2 K)	[32]
2	Layered Co/Au structures Co(10)/Au(4)/Co(10)	–	–	–	1,8 (T = 300 K) 2,7 (T = 4,5 K)	[37]
3	Sputtered Au-Co thin films	Conventional RF sputtering; simultaneous condensation, d = 15-100 nm	>10	370-670	13,5 (T = 300K, 23 at.%Co; 3 (T = 300 K, 9 at.%Co)	[33]
4	Multilayers Au/Co; different meaning Co-Au Au(36)/$[Co(2,4)$-$Au(1,1)]_{30}$/Au(5)/Si	Condensation on patterned subsfrates of Si by electron-beam evaporation	–	–	0,42	[40]
5	Multilayers films Co/Au; Co(0,5-2)/Au(0,3-2,5) Co(0,5-2)/Au(0,5-20)	Electrochemical deposition	–	–	1-5	[41]
6	Granulare alloy Co-Au; different meaning Co/Au multilayers	Electrodeposition; electron-beam deposition	–	573	4,7(38 at.%Co)	[34, 42]

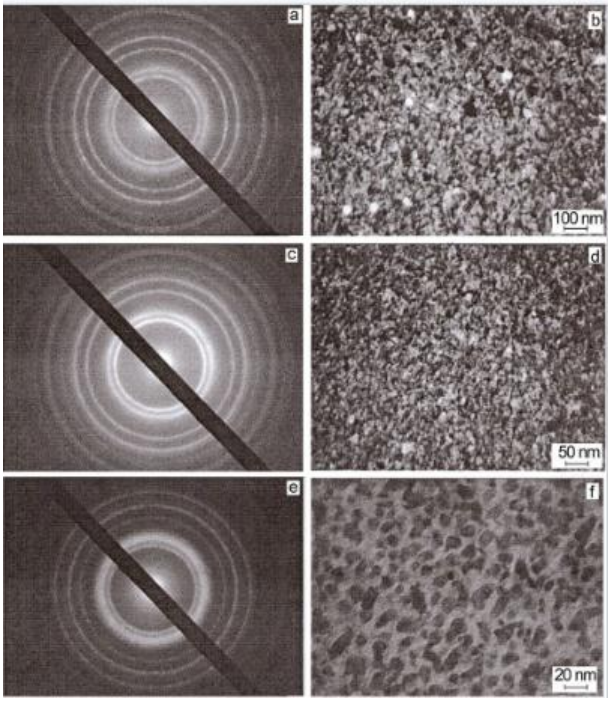

Fig. 8 – Phase composition and microstructure of granular films s.s. Au(Co). In the initial state: Au(48)/Co(10)/S, T_a = 600 K, c_{Co} = 23 at.% (a, b); Au(8)/Co(14)/S, T_a = 300 K, c_{Co} = 73 at.% (c, d) and Au(4)/Co(14)/S, T_a = 700 K, c_{Co} = 84 at.% (e, f)

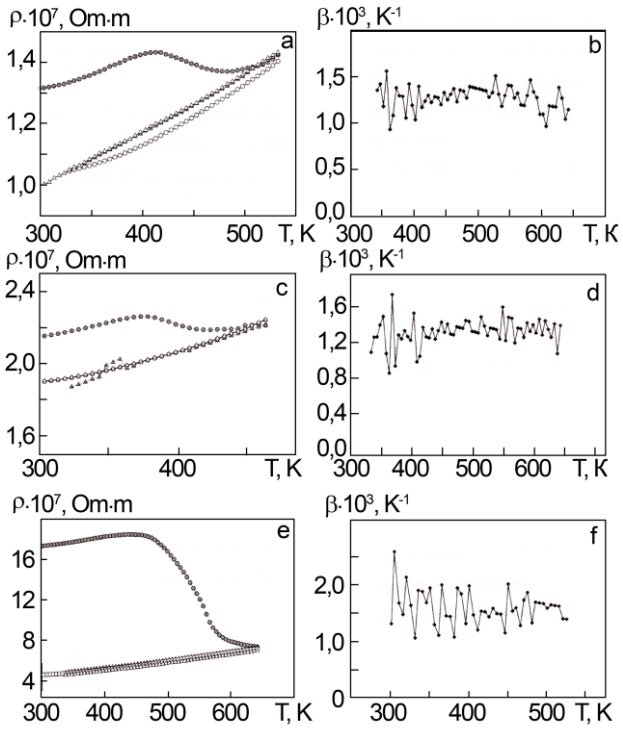

Fig. 9 – Temperature dependence ρ and TCR of granular films s.s. Au(Co). In the initial state: Au(48)/Co(10)/S, c_{Co} = 23 at.% (a, b); Au(29)/Co(14)/S, c_{Co} = 42 at.% (c, d) and Au(4)/Co(14)/S, c_{Co} = 84 at.% (e, f)

magnetoresistive properties of film systems based on Ag and Co (Tables 1 and 2) we realized the same generalization to film systems based on Au and Co (Table 4) based studies [32-34, 37-42]. The results is uniquely indicated that GMR value not significantly dependent on the structural state – multilayer or granulated film based s.s. Significant role of the concentration of magnetic components, which in turn causes a certain critical size granules, in which sold most effective SDSE.

4. CONCLUSIONS

The generalization own and literature data on structural and phase state and correlating it with electrophysical and magnetoresistive properties leads

to the following conclusions:

– in film systems based on Co and Ag or Au, obtained by different methods or the simultaneous deposition layer of condensation formed separate component disordered fcc s.s. Ag (Co) or Au (Co) in most of his cases with elements of granular state;

– optimum concentration of the Co granular system defines perfection and, consequently, the effectiveness SDSE;

– electrophysical properties (resistivity, TCR and SC) alloys granular s.s. Ag(Co) + G or s.s. Au(Co) + G defined conductivity of solid solutions and not systems granules;

– the value of GMR is not significantly dependent on the method of obtaining granular s.s., and fully determined by the temperature measurement and optimum concentration of cobalt atoms.

We note also that has great potential (see, for example, [43, 44]) use granular film alloys in sensor technology (thermistors, sensors of temperature, strain and magnetic field).

Work performed under the state budget project No 0115U000689 (2015-2017 years) and No 0115U000664 (2016-2018 years).

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