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TEMPERATURE EFFECT ON MAGNETORESISTIVE PROPERTIES OF Fe AND Co ISLAND FILMS

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Magnetoresistive properties of ultrathin Fe and Co films with effective thicknesses $d = 3 \div 30$ nm are studied in a temperature interval of (150–700) K. For films with $d = 3 \div 10$ nm, the electrical conductivity is found to be thermally activated and the tunnel magnetoresistance effect is revealed. A decrease of the temperature down to 150 K in measurements on as-condensed films is found to result in the appearance of the magnetoresistive hysteresis and a reduction of the tunnel magnetoresistance (TMR) in a thickness interval of 10–15 nm. The maximum TMR value in the fields up to 0.7 T amounted to 1% for the as-condensed Fe films with an effective thickness of 17 nm and the Fe films with an effective thickness of 8 nm annealed at a temperature of 520 K.

Keywords: island film, tunnel magnetoresistance, spin-dependent tunneling, thermally induced conductivity.

1. Introduction

Interest in ultrathin metal films is related to the facts that, first, they are an initial phase of condensation of film objects (the initial stage of film growth) and, second, they are objects demonstrating specific electrophysical properties, if this growth stage is stabilized. As in the case of granulated metal-insulator composites [1–3], the spin-dependent tunneling of electrons is also possible in island films, which can be of interest for applications (magnetic devices for increasing the memory density, sensors of physical parameters, and other elements of nanoelectronics and spintronics [4, 5]).

The specific features of magnetoresistance in ultrathin films follow from their island structure. Vacuum gaps interfere the direct ferromagnetic exchange between neighbor islands, but allow the electron tunneling between them. The tunneling probability depends on the magnetization vector orientation in neighbor islands and is sensitive to the applied magnetic field. It is maximum, when the magnetic moments in adjacent granules are oriented in parallel, i.e. the spin-dependent conductivity is realized, which results in the appearance of the tunnel magnetoresistance (TMR) effect [6, 7]. The magnitude of tunnel magnetoresistance substantially depends on the properties of dielectric barriers between ferromagnetic particles, including the properties of the ferromagnet-insulator interface. Since the TMR in granular composites is determined only by the spin-preserving charge tunneling, the processes of tunneling through barriers, when the spins of charge carriers are not preserved, do not contribute to the magnetoresistance.

The issue concerning the influence of the temperature on the TMR magnitude is also challenging. The authors of works [8–10] assumed that the spin polarization is proportional to the magnetization of a sur-

face layer in ferromagnetic particles, and it diminishes under the influence of spin-wave excitations as the temperature increases. As a result, the TMR magnitude decreases, as the temperature grows. Another factor that governs a change of the magnetoresistance at the temperature growth is the presence of parallel channels for electrons to pass the inter-island gaps, in which the spin is not preserved. The role of processes, in which the gaps are passed without spin preservation, grows with the temperature elevation, which also gives its contribution to the magnetoresistance reduction.

The authors of work [11] showed that the emergence of a percolation cluster and the change of the magnetic state in cermet films result in the appearance of temperature dependences of the magnetoresistance of various types. For instance, for films with the content of a magnetic material less than the percolation threshold, the tunnel magnetoresistance increases, as the temperature falls down. For films, in which the volume fraction of the ferromagnetic material exceeds the percolation threshold, a different type of the temperature dependence of the magnetoresistance was revealed.

Proceeding from the aforesaid, the aim of this research was to determine how the temperature and the effective thickness affect the tunnel magnetoresistance in iron and cobalt island films.

2. Experimental Technique

Ultrathin one-layer Co and Fe films from 1 to 30 nm in thickness were obtained in a vacuum chamber at a pressure of residual atmospheric gases of 10^{-4} Pa [12]. The films were condensed by evaporating the metals with a purity not lower than 99.98% with the use of an electron-beam gun. The film condensation was carried out onto a substrate at room temperature at the rate $\omega = (1 \div 2)$ nm/s, depending on the evaporation regime. When studying the electro- and magnetoresistive properties, polished glass plates with preliminarily rendered contacts were used as substrates.

The magnetoresistance was measured, and the thermomagnetic treatment of the films was carried out in a special installation under conditions of ultrahigh oil-free vacuum (10^{-6}–10^{-7} Pa) in a dc magnetic field with the strength up to $H = 150$ kA/m and in a temperature interval from 150 to 700 K.

The electric resistance was measured, by using the two-point measurement scheme with the help of a universal digital voltmeter V7-46/1. Since the specimen resistance rather than the resistivity is measured when the magnetoresistance is researched, the error of those measurements did not exceed 0.02%.

3. Experimental Results and Their Discussion

In order to determine the magnetoresistance anisotropy and reveal the positive anisotropic magnetoresistance in the Fe and Co island films, the measurements were carried out with the "current in the film plane" configuration, by using two orientations of a magnetic field with respect to the current (the longitudinal and transverse magnetoresistances). From the isotropic field dependences of the longitudinal and transverse magnetoresistances obtained at room temperature, it follows that they coincide within the experimental error limits. Therefore, the field, temperature, and size dependences will be presented below only for the longitudinal magnetoresistance.

It should be noted that the isotropy of field dependences means that the influence of the magnetoresistance anisotropy is not essential, and the observed dominating effect is a result of the spin-dependent tunneling of electrons between ferromagnetic islands, because the mutual orientation of the current and the field has no matter for this mechanism [13]. Therefore, in what follows, only the results of researches concerning the tunnel magnetoresistance will be reported.

From the field dependences of the magnetoresistance obtained for the as-condensed Co (Fig. 1, *a*) and Fe (Fig. 1, *b*) films, it follows that if the temperature is lowered down to 150 K, there appears a magnetoresistive hysteresis. Similarly to the case of cermat films, such features in the field dependences can be explained as follows [14].

When cooling down without an external magnetic field, the dipole fields of large clusters and the percolation cluster stimulate the magnetic moments of islands to be ferromagnetically ordered. As a result, the initial tunnel resistance is lower than that in the system with non-correlated magnetic moments. Due to the application of an external magnetic field, this ordering is destroyed. The magnetic moments of separate particles are oriented randomly along the easy axes of each island. The orientation of easy axes is determined by the shape anisotropy (for elongated clusters or island chains) or the magnetocrystalline

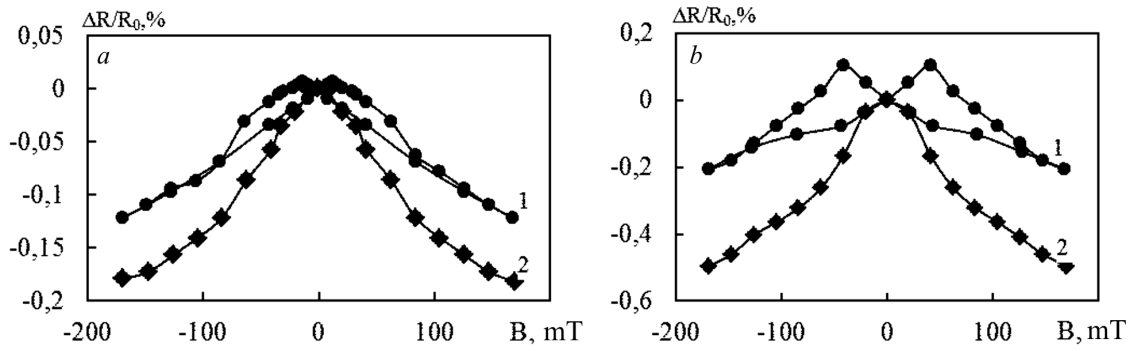

Fig. 1. Field dependences of the longitudinal magnetoresistance for as-condensed Co (d = 15 nm) (*a*) and Fe (d = 17 nm) (*b*) films obtained at various temperatures: 150 (*1*) and 300 K (*2*)

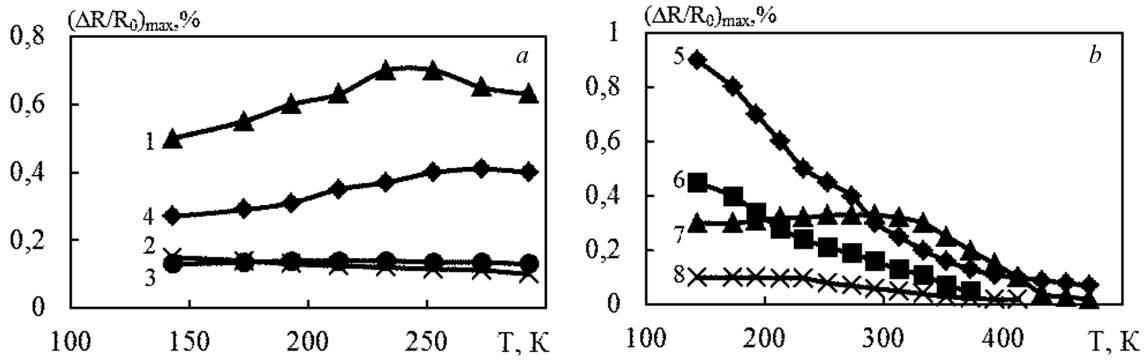

Fig. 2. Temperature dependences of TMR for unannealed (*a*) and annealed (*b*) Fe (*1, 2, 5, 6*) and Co (*3, 4, 7, 8*) films. d_{Fe} = 17 (*1*), 10 (*2*), 8 (*5*), and 10 nm (*6*); d_{Co} = 10 (*3*), 15 (*4*), 8 (*7*), and 9 nm (*8*). The annealing temperature equals 520 K for Fe films and 700 K for Co films

anisotropy for separate islands. As a whole, this new magnetic state will be characterized by larger resistances in comparison with the state obtained after the cooling without external magnetic field, because the latter state is a metastable low-energy one [14]. For larger applied magnetic fields, the resistance decreases owing to the alignment of particles' magnetic moments along the field direction. As a result, there arises a maximum in the initial dependence of the magnetoresistance on the magnetic field induction. If the induction of the applied magnetic field vanishes, the system does not return into the state similar to that obtained at the cooling in the zero field, and the film resistance is higher than that obtained after the first cooling [15].

From the dependences of the tunnel magnetoresistance on the temperature presented for the ultrathin Fe and Co films that were not annealed or annealed at various temperatures (Fig. 2), it follows that they can have various characters depending on the effective film thickness. In an effective-thickness interval of 20–25 nm for the as-condensed Co films and 25–30 nm for the Fe films, the reduction of the measurement temperature to 150 K gives rise to an insignificant increase of the magnetoresistance (Fig. 2, *a*, curves *2* and *3*). For narrower effective thicknesses ($d_{Fe} < 25$ nm and $d_{Co} < 15$ nm), the tunnel magnetoresistance decreases by a factor of 1.3 to 5 (Fig. 2, *a*, curves *1* and *4*), when the temperature decreases to 150 K. For the Fe films with effective thicknesses of 5–10 nm annealed at a temperature of 520 K, the tunnel magnetoresistance increases by 2–5 times, as the temperature decreases (Fig. 2, *b*, curves *5* and *6*).

Let us consider the factors giving rise to a change of the magnetoresistance at the variation of the temperature in more details. A change of the $(\Delta R/R_0)_{max}$ ratio due to the variation of the temperature can result from the variation of both the difference $\Delta R_{max} = R_0 - R_S$ (R_0 is the electrical resistance of a specimen in the absence of a magnetic field,

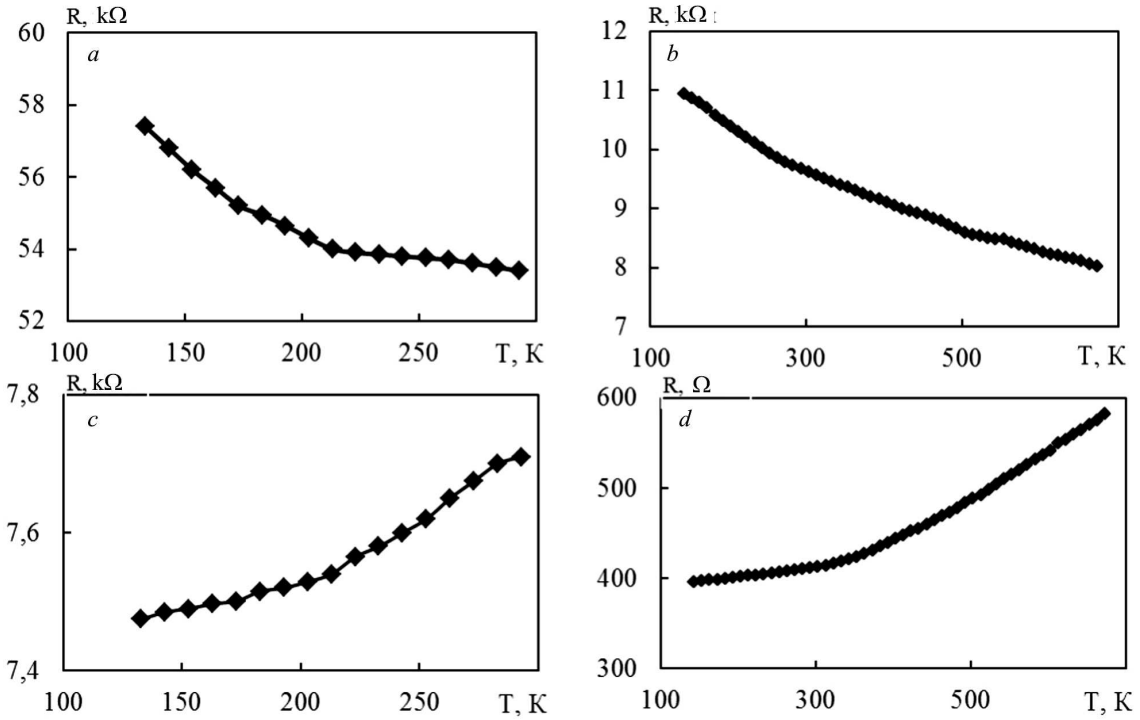

Fig. 3. Temperature dependences of the resistance for as-condensed Co films (*a, c*) and Co films annealed at a temperature of 700 K (*b, d*). $d = 12$ (*a*), 8 (*b*), and 25 nm (*c, d*)

and R_S is this resistance measured in the maximum field) and R_0. Really, both those quantities (R_0 and R_S) are temperature-dependent. But they can change with the temperature in such a way that the ratio $(\Delta R/R_0)_{\max}$ can increase, as well as can decrease. Therefore, in order to understand better the influence of the temperature on the magnetoresistance, we have studied the dependences of the resistance and the quantity $\Delta R_{\max}$ on this parameter.

Let us first consider the features in the temperature dependences of the electrical resistance obtained for the ultrathin Fe and Co films. For the as-condensed Fe ($d_{\mathrm{Fe}} < 25$ nm) and Co ($d_{\mathrm{Co}} < 15$ nm) films, the so-called dielectric regime of conductivity is realized in a temperature interval of 150–300 K (Fig. 3, *a*), which is evidenced by the negative temperature coefficient of resistance (TCR) typical of the nonmetallic character of conductivity (like the conductivity in semiconductor or amorphous materials). The conductivity is governed by the thermally activated tunneling of electrons between the metal islands separated by vacuum gaps. The electric resistance of a film in the dielectric mode increases, as the temperature decreases. Most models for the conductivity of island films are based on two mechanisms allowing electrons to overcome the potential barrier between the islands: the classical one, when an electron penetrates through the barrier owing to its energy that exceeds the barrier height, and the quantum-mechanical one, when an electron penetrates through the barrier by means of the quantum-mechanical tunneling effect. Leaving a thorough theoretical analysis of both those mechanisms aside (see, e.g., work [16]), we have to note that, unfortunately, none of them describes the experimental dependences of the electrical resistance on the effective film thickness (the island size).

At present, the concepts of the thermally induced tunneling of electrons from one island to another one dominate. In this case, the tunneling activation energy is mainly determined by the dimensions of islands and the gaps between them, and does not depend on the work function of electrons from metal. Then the specific resistance of a film can be written in the form [17]

$$\rho = \rho_0 (1 + \beta T) + C \exp (E_a/kT), \qquad (1)$$

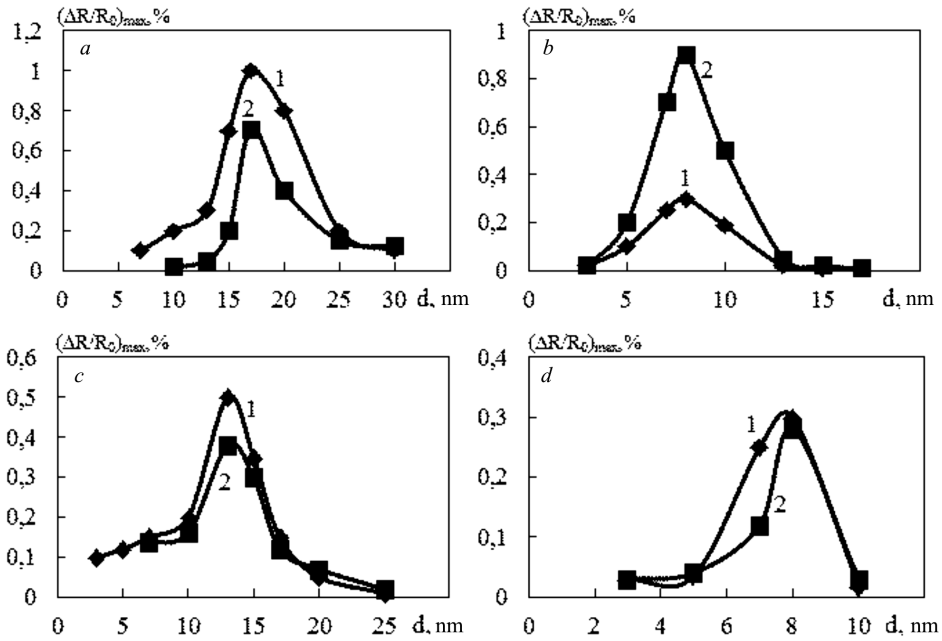

Fig. 4. Dependence of TMR on the effective thickness for as-condensed (*a, c*) and annealed (*b, d*) Fe (*a,b*) and Co (*c, d*) films. The measurement temperatures are 300 (*1*) and 150 K(*2*). The annealing temperature is 520 K for Fe films and 700 K for Co ones

where the first term describes the ordinary (metallic) dependence of the resistance, and the second one is associated with a certain thermally activated process.

If the effective thickness of as-condensed Fe and Co films increases to 20–25 nm, the TCR absolute value (the TCR itself is negative) decreases and approaches zero. Evidently, this behavior corresponds to approximately identical contributions of the terms in expression (1). If the effective film thickness exceeds 25–30 nm, only the dependences $R(T)$ with positive TCR values are observed (Fig. 3, *c*), which is typical of metals and corresponds to the dominant role of the first term [18]. The films that have effective thicknesses less than 10 nm and are annealed at a temperature of 700 K demonstrate also the exponential temperature dependences of their resistance (Fig. 3, *b*). If their effective thicknesses grow, a transformation to the metallic dependence $R(T)$ takes place (Fig. 3, *d*).

Using the data obtained for the films with the exponential temperature dependence of their resistance, we determined the activation energy of conductivity E_a. The calculation gave values within the interval 10^{-2}–10^{-3} eV, which are typical of pure metals [19]. With the growth of the effective film thickness, the activation energy of conductivity E_a decreases (the distances between the islands become shorter). The maximum value of the activation energy of conductivity equals 0.1 eV. Hence, the activation energy is determined by the size of islands and the distances between them, rather than the film composition.

Now, let us consider the variation of $\Delta R_{\max}$ with the temperature for the Fe and Co films. In the case of annealed films, the variation of $\Delta R_{\max}$ depends on the effective thickness. For instance, for the Co films with an effective thickness of 5–7 nm that are annealed at a temperature of 700 K, the value of $\Delta R_{\max}$ does not change, as the temperature decreases from room temperature to 150 K. For the Co films in the interval of effective thicknesses from 8 to 10 nm, the value of $\Delta R_{\max}$ increases by a factor of 1.2–1.5. For the Fe films with $d_{\mathrm{Fe}} = 5 \div 10$ nm that are annealed at a temperature of 520 K, the value of $\Delta R_{\max}$ increases by 10 to 20 times. The described variations in the resistance and the value of $\Delta R_{\max}$ result in the existence of various types of of the temperature dependence for the magnetoresistance.

Let us analyze in detail why the quantity $\Delta R_{\max}$ is changed. The situation is obviously similar to the case of granular composites [14]. When analyzing the tem-

perature dependence of $\Delta R_{\max}$, one should consider the competitive influences from the percolation cluster and separate superparamagnetic particles in the presence of the dipole-dipole interaction. In this case, there are plenty of narrow tunnel barriers with a high conductance. The temperature dependence of $\Delta R_{\max}$ will be driven by the competition between two following processes: the magnetic blocking of separate clusters and the formation of a magnetically ordered system of clusters. At low temperatures because of blocking processes, the larger number of clusters that do not interact with one another contribute to the tunnel magnetoresistance. This process leads to an increase of $\Delta R_{\max}$. If the temperature decreases further, the processes of magnetic ordering stimulate the ordering of the moments of separate clusters, i.e. the formation of a magnetically ordered system over the whole specimen, and a reduction of the magnetoresistance. As a result of this competition, there emerges a maximum in the temperature dependence of the magnetoresistance.

As the temperature falls down, the number of thermally activated carriers decreases exponentially, and they are located only in large islands, for which the electrostatic energy is low. In other words, the tunneling order drastically grows, as the temperature decreases. Hence, the distribution of island dimensions in the film plays a crucial role for the magnetoresistance. The abnormal growth of the magnetoresistance at low temperatures is associated with the involvement of higher-order processes of tunneling, i.e. the spin-dependent tunneling of electrons between large islands involving small islands located between them in the presence of the strong Coulomb blockade (under definite conditions, the tunneling through a tunnel barrier is decreased because of the Coulomb repulsion between the electrons and the charged island) [20, 21].

From the dimensional dependences obtained for the Fe and Co films at various temperatures (Fig. 4), it follows that the TMR increases with a decrease of the effective thickness (the increase of the vacuum gap width) up to a certain limit. The TMR maximum is observed at an effective thickness of 17 nm for the as-condensed Fe films and 13 nm for the Co films.

After the annealing, the TMR maximum shifts toward smaller effective thicknesses, which is associated with the growth of islands by means of their coalescence. At small enough effective thicknesses (3–5 nm), the vacuum gap width substantially increases (to 5–10 nm), and the island film transforms into a system of completely isolated islands, for which the tunneling and, accordingly, tunnel magnetoresistance are absent at all. It should be noted that the positions of maxima in the dimensional dependences are not changed, if the temperature decreases to 150 K. In the case of as-condensed Fe (Fig. 4, *a*) and Co (Fig. 4, *c*) films and Co films annealed at a temperature of 700 K (Fig. 4, *d*), the TMR values decrease together with the temperature. The Fe films annealed at a temperature of 520 K (Fig. 4, *b*) are the exception. For them, in an interval of effective thicknesses of 5–10 nm, the temperature decrease to 150 K results in a substantial growth of TMR.

4. Conclusions

In this work, it is found that as-condensed Fe films with $d_{\mathrm{Fe}} = 10 \div 25$ nm and as-condensed Co films with $d_{\mathrm{Co}} = 5 \div 17$ nm demonstrate the isotropic field dependences associated with the spin-dependent electron tunneling between ferromagnetic islands. The maximum magnitude of tunnel magnetoresistance in a magnetic field lower than 0.7 T amounts to 1% for the as-condensed Fe films with an effective thickness of 17 nm and the Fe films with an effective thickness of 8 nm annealed at a temperature of 520 K. The competition between two processes, the magnetic blocking of separate clusters and the formation of a magnetically ordered system of clusters results in the appearance of the temperature dependences of the tunnel magnetoresistance of various types. The dependences of the tunnel magnetoresistance on the effective thickness have a maximum associated with the formation of vacuum gaps with an optimum width of (2–3 nm).

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1. S. Mitani, H. Fujimori, S. Ohnuma. Spin-dependent tunneling phenomena in insulating granular systems. *J. Magn. Magn. Mater.* **165**, 141 (1997).
2. O.V. Stognei, A.V. Sitnikov, Yu.E. Kalinin, S.F. Avdeev, M.N. Kopytin. Isotropic positive magnetoresistance of nanogranular composite materials Co–Al_2O_n. *Fiz. Tverd. Tela* **49**, 158 (2007) (in Russian).
3. A.K. Fedotov, Yu.A. Fedotova, Yu.V. Kasyuk, A.V. Mazanik, M.A. Andreev, A.N. Suvorov, A.P. Ivanov, M. Marszalek. Electric conductivity of film composite

NiCoCr–MoS_2. *Nanosyst. Nanomater. Nanotekhnol.* **9**, 457 (2011) (in Russian).
4. O.I. Tovstolytkin, M.O. Borovyi, V.V. Kurylyuk, Yu.A. Kunyts'kyi. *Physical Basics of Spintronics* (Nilan, 2014) (in Ukrainian).
5. G. Herzog, S. Krause, R. Wiesendanger. Heat assisted spin torque switching of quasistable nanomagnets across a vacuum gap. *Appl. Phys. Lett.* **96**, 102505 (2010).
6. V. Soti. Tunneling magnetoresistance of FeCo/MgO/FeCo magnetic tunnel junctions. *Adv. Stud. Theor. Phys.* **5**, 775 (2011).
7. M. Ziese. Extrinsic magnetotransport phenomena in ferromagnetic oxides. *Rep. Progr. Phys.* **65**, 143 (2002).
8. C.H. Shang, J. Nowak, R. Jansen, J.S. Moodera. Temperature dependence of magnetoresistance and surface magnetization in ferromagnetic tunnel junctions. *Phys. Rev. B* **58**, R2917 (1998).
9. W.G. Wang, C. Ni, T. Moriyama, J. Wan, E. Nowak, J.Q. Xiao. Spin-polarized transport in hybrid (Zn, Cr)Te/Al_2O_3/Co magnetic tunnel junctions. *Appl. Phys. Lett.* **88**, 202501 (2006).
10. N.V. Dalakova, B.I. Belevtsev, E.Yu. Belyaev, A.N. Bludov, V.N. Pashchenko, M.G. Osmolovskii, O.M. Osmolovskaya. Resistive and magnetoresistive properties of pressed CrO_2 powders with various types of intergranular dielectric layers. *Fiz. Nizk. Temp.* **38**, 1422 (2012) (in Russian).
11. A.Ya. Vovk, V.O. Golub, L. Malkinski, A.F. Kravets, A. Pogoriliy, O. Shipil. Tunneling magnetoresistance in granular cermet films with particle size distribution. *J. Magn. Magn. Mater.* **272–276**, E1403 (2004).
12. V.B. Loboda, Yu.A. Shkurdoda, V.A. Kravchenko, S.N. Chursenko, V.N. Kolomiets. Structure and magnetoresistive properties of polycrystalline Co/Cu/Co films. *Metallofiz. Noveish. Tekhnol.* **33**, 161 (2011) (in Russian).
13. A. Milner, A. Gerber, B. Groisman. Spin-dependent electronic transport in granular ferromagnets. *Phys. Rev. Lett.* **76**, 475 (1996).
14. S. Sankar, A.E. Berkowitz, D. Smith. Spin-dependent transport of Co–SiO_2 granular films approaching percolation. *Phys. Rev.* B **62**, 14273 (2000).
15. A.Ya. Vovk, V.O. Golub, A.M. Pogoriliy, O.V. Shypil, A.F. Kravets. Temperature dependence of magnetotransport in CoFe–AlV_2DOV_3D granular films in the vicinity of the percolation threshold. *Metallofiz. Noveish. Tekhnol.* **24**, 1277 (2002).
16. L. Trusov, B. Kholmyanskii. *Metal Island Films* (Metallurgiya, 1973) (in Russian).
17. *The Technology of Thin Films: A Handbook.* Edited by L. Maissel, R. Glang (McGraw-Hill, 1970), Vol. 2.
18. Yu.O. Shkurdoda, A.M. Chornous, V.O. Kravchenko, V.B. Loboda. Magnetoresistive properties of nanocrystalline cobalt films. *Nanosyst. Nanomater. Nanotekhnol.* **14**, 293 (2016) (in Ukrainian).
19. P.G. Borzyak, Yu.A. Kulyupin. *Electronic Processes in Metal Island Films* (Naukova Dumka, 1980) (in Russian).
20. Yu.A. Kruglyak. *Nanoelectronics "from Bottom to Top"* (TES, 2015) (in Russian).
21. S. Mitani, S. Takahashi, K. Takanashi, K. Yakushuiji, S. Maekawa, H. Fujimori. Enhanced magnetoresistance in insulating granular systems: Evidence for higher-order tunneling. *Phys. Rev. Lett.* **81**, 2799 (1998).

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ВПЛИВ ТЕМПЕРАТУРИ
НА МАГНІТОРЕЗИСТИВНІ ВЛАСТИВОСТІ
СТРУКТУРНО НЕСУЦІЛЬНИХ ПЛІВОК Fe ТА Co

Р е з ю м е

Проведені дослідження магніторезистивних властивостей надтонких плівок Fe та Co з ефективними товщинами $d =$ $=$ 3–30 нм в інтервалі температур 150–700 K. Для плівок з $d =$ 3–10 нм електропровідність є термічно активованою і реалізується тунельний магнітоопір. Показано, що для свіжосконденсованих плівок зниження температури вимірювання до 150 K приводить до появи магніторезистивного гістерезису і зменшення величини тунельного магнітоопору в інтервалі ефективних товщин 10–15 нм. Максимальна величина тунельного магнітоопору в магнітному полі до 0,7 Тл становить 1 % для свіжосконденсованих та відпалених за температури 520 K плівок Fe ефективною товщиною 17 та 8 нм відповідно.