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Influence of the surface morphology on the magnetoresistance of ultrathin films of ferromagnetic metals and their alloys

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Abstract. The microstructure, morphology and magnetoresistive properties of as-deposited and annealed at 700 K discontinuous films of ferromagnetic metals and their alloys are investigated. It is established that the isotropic field dependences stemming from the spin-dependent electron tunneling between ferromagnetic islands are observed for as-deposited Co films with an effective thickness (d) of $d_{\mathrm{Co}} = 5$–25 nm as well as for Fe films ($d_{\mathrm{Fe}} = 10$–30 nm). It is shown that the maximum values of the isotropic magnetoresistance take place in the case when the size of the islands is equal to 3–5 nm and the width of the dielectric barrier between them is 1–3 nm. The maximum TMR values for the films annealed at 700 K are obtained when the dielectric barrier is 2–5 nm wide.

1 Introduction

The study of the tunnel metal-dielectric structures is a promising direction in the development of solid-state physics. This is related to their large-scale practical application in various devices of modern solid-state electronics and spintronics (magnetic devices intended to increase memory density, sensors of physical quantities and other elements of nanoelectronics and spintronics). In these structures, the dielectric layers impede the direct ferromagnetic exchange between the adjacent granules while admitting electron tunneling between them. Tunneling probability depends on the relative orientation of the magnetization vector in the adjacent granules and is sensitive to the applied magnetic field [1–3]. It is maximum when the moments in the adjacent granules are oriented in parallel, *i.e.* the spin-dependent tunneling conductance is implemented that leads to the tunnel magnetoresistance effect (TMR).

High TMR values obtained in magnetic tunnel junctions (MTJ) with a MgO dielectric layer enable to consider them as functional elements of a magnetoresistive memory cell [4]. However, relatively high tunnel switching currents (J) (10^6–10^8 A/cm^2) result in their significant heating, which is a critical condition for the creation of new-generation memory devices. In this regard, it is necessary to consider the alternative technological and design solutions for a detailed analysis of the spin-polarized transport mechanisms.

One of such solutions consists in the utilization of MTJ with the so-called vacuum barrier (vacuum tunnel structures) [5]. A great number of the latest unique experiments on switching the magnetic nanoisland Fe/W structures through the vacuum gap encourages experimental and theoretical studies in this direction.

The vacuum tunnel structures allow considering the features of spin transport if there is no exposure to the collateral effects on the layer interfaces. In addition, the use of a vacuum gap instead of a dielectric layer makes it possible to investigate the features of the spin-tunneling magnetic phenomena in strong field emission mode when voltages are higher than the dielectric breakdown threshold.

Thus, the objective of the study was to determine the general regularities of the morphology, crystal structure and phase composition influence on the magnetoresistive properties of island films of ferromagnetic metals and their alloys.

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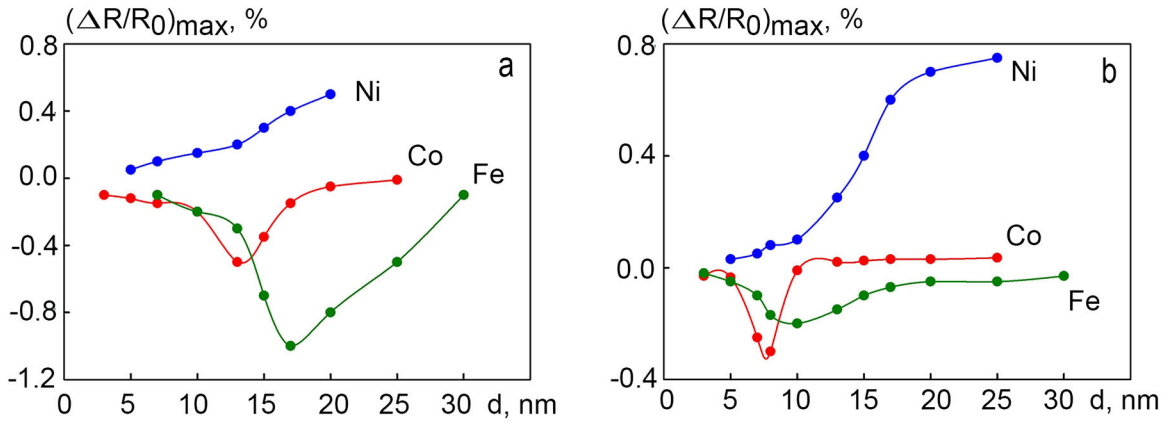

Fig. 1. Dependence of the magnetoresistance on the effective thickness for as-deposited (a) and annealed to 700 K (b) Ni, Co and Fe films.

2 Experimental details

The films of Co, Fe, Ni as well as Co_xNi_{1-x} and Fe_xNi_{1-x} alloys of (1–30) nm effective thickness were obtained in a vacuum chamber at residual atmosphere gas pressure of 10^{-4} Pa [6]. Condensation of the films was carried out at room temperature of the substrate with the rate of $\omega = (0.5–1)$ nm/s depending on the modes of evaporation. In order to investigate the magnetoresistive properties, polished glass plates with the pre-applied contact areas were taken as substrates. For structural-phase research, KBr crystals and carbon films were used as substrates.

Thermal stabilization of the samples as well as the study of their electrical resistance dependences on the temperature were carried out according to the "heating-cooling" scheme with the constant velocity of (2–3) K/min in the temperature range of (300–700) K.

Measurement of the longitudinal and transverse magnetoresistance as well as thermomagnetic film processing were performed in a special facility in an oil-free ultrahigh vacuum environment of (10^{-6}–10^{-7}) Pa.

Structural-phase studies of the samples were carried out using a transmission electron microscope TEM-125K and an electron diffraction apparatus.

3 Results and discussion

In order to determine the magnetoresistance anisotropy and identify positive anisotropic magnetoresistance in structurally discontinuous films, the measurement was performed in configuration "current in the film plane" and for two magnetic field geometries with respect to the current (longitudinal and transverse magnetoresistance). From the obtained at room temperature isotropic field dependences of the longitudinal and transverse magnetoresistance it follows that they coincide within the experimental margin of error.

Figure 1 illustrates the longitudinal MR dependences on the effective thickness of newly condensed (fig. 1(a)) and annealed at 700 K (fig. 1(b)) Ni, Co and Fe films. These dependences have a non-monotonic behavior. A negative isotropic magnetoresistance is observed for as-deposited samples in the range of effective thicknesses of $d_{Co} = 5–25$ nm and $d_{Fe} = 10–30$ nm. This fact indicates that the magnetoresistance anisotropy impact is insignificant, while the observed dominant effect is not anisotropic and is conditioned by spin-dependent electron tunneling between the ferromagnetic islands, since the mutual orientation of current and field is not important for such a mechanism [7,8]. It should be noted that condensation was performed on a neutral non-oriented substrate (glass, carbon film) at room temperature ($T_s \approx 300\,\text{K} < 2T_m/3$) (where/here T_m is the melting temperature); therefore the diffusion mobility of atoms on the substrate was complicated.

The electron microscopic findings indicate the formation during deposition of many small crystallization centers and their simultaneous growth. As a result, practically the same structure (fig. 2(a), (c) and fig. 3(a)) was observed in unannealed films regardless of their composition and thickness. Irregularly shaped islands are of 2–5 nm in size, and the gaps between them take the form of channels of approximately identical 1–2 nm width [9]. Thus, the Volmer-Weber growth mechanism is implemented for the films. Thickness growth of Co and Fe films is structurally manifested only in increasing island density. The similarity in the morphology of the as-deposited Co and Fe films in a rather wide thickness range leads to the implementation of spin-dependent electron tunneling. The exception is Ni films whose tunneling magnetoresistance has not been detected. The maximum TMR value for the as-deposited films in a magnetic field up to 0.7 T makes 0.5% for Co and 1% for Fe.

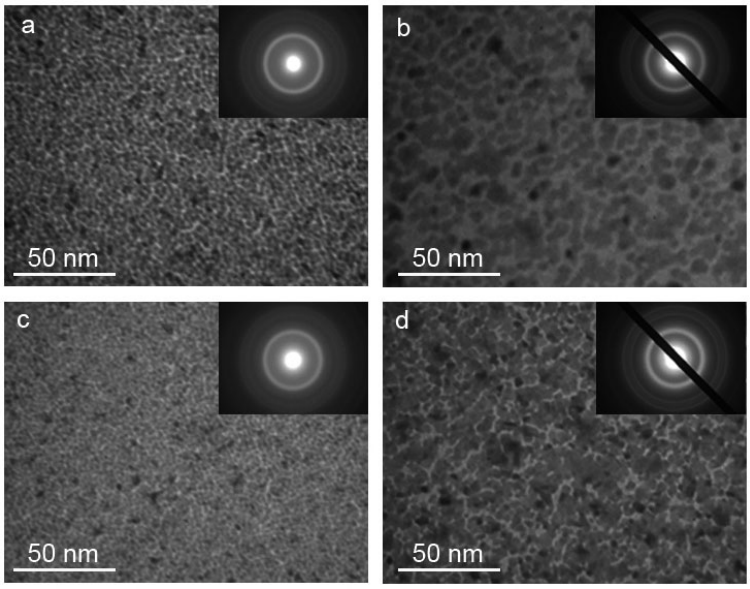

Fig. 2. Crystalline structure of the as-deposited ((a), (c)) and annealed to 700 K ((b), (d)) Co films at the thickness of $d_{\text{eff}} = 5$ nm ((a), (b)), 15 nm ((c), (d)).

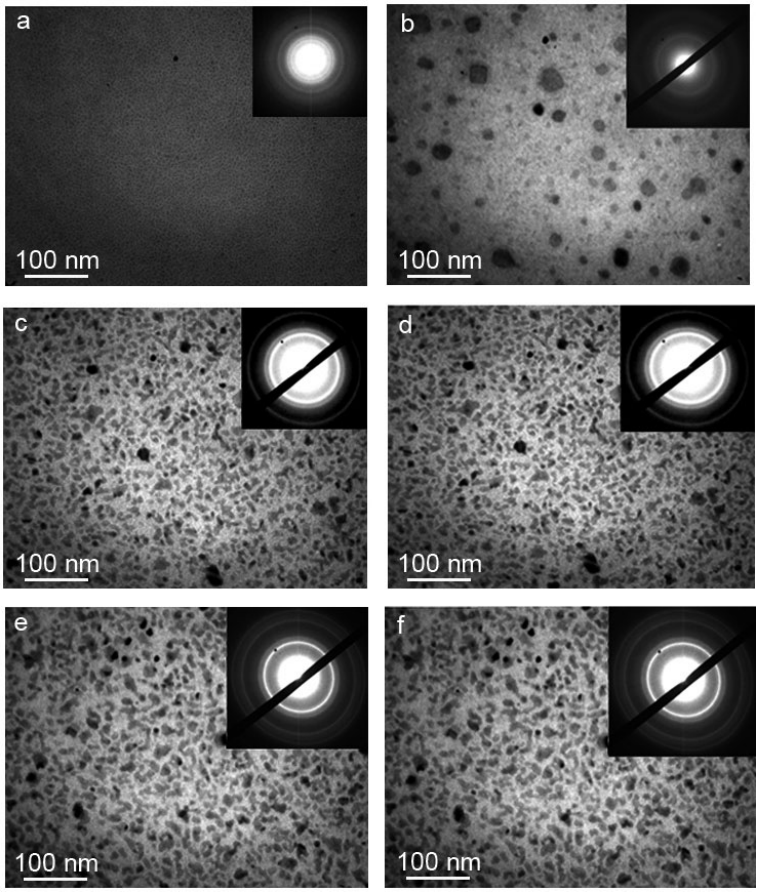

Fig. 3. The structure of ultrathin films of CoNi alloy. (a) as-deposited film; (b)–(f) after annealing to 700 K. The effective thickness of the films: (a) 20 nm ($c_{\text{Co}} = 20\%$); (b) 9 nm ($c_{\text{Co}} = 80\%$); (c) 9 nm ($c_{\text{Co}} = 10\%$); (d) 10 nm ($c_{\text{Co}} = 30\%$); (e) 15 nm ($c_{\text{Co}} = 70\%$); (f) 20 nm ($c_{\text{Co}} = 50\%$).

Annealing at 700 K entails significant differences in the morphological state of the film samples depending on their thickness. Let us analyze in more detail the impact of the film morphology on the magnetoresistive properties on an example of Co films. It should be noted that the size dependence of the tunneling magnetoresistance for island films is similar to the concentration dependence of the negative TMR for metal-dielectric composites [10]. When effective thickness is small ($d \sim$ 3–5 nm), simple island coarsening to 10–20 nm occurs due to the coalescence (fig. 2(b)), and the total number of islands 3–5 times decreases. This causes the increase in channel width (up to 2–7 nm) and TMR reduction as well as its weak dependence on the effective thickness (the so-called pre-percolation area). Such a pattern, but with a higher density of islands and narrower channels (2–5 nm), is observed when $d_{\mathrm{Co}} = 6$–7 nm. These morphological changes in the structure result in a sharp increase in the TMR value. The maximum magnetoresistance values ($\Delta R/R_{\max} = 0.35\%$) occur near the percolation threshold from the side of the dielectric area. The further increase in the effective thickness ($d_{\mathrm{Co}} =$ 8–10 nm) leads to the formation of a "bridge" structure (fig. 2(d)), and, consequently, to dramatic TMR reduction and appearance of the magnetoresistance anisotropy.

It should be noted that the maximum magnetoresistance values are implemented in the case when the dielectric barrier width between the adjacent granules is minimum (tunneling probability is maximum), but the effective exchange interaction between the electron spins belonging to different granules is still absent. Moreover, the films of both pure ferromagnetic metals and their alloys are in the superparamagnetic state. It is assumed in the work [11] that appearance of superparamagnetism is typical for island magnetic films. These data are in a good agreement with the data presented in [12], according to which Fe films of 3.9 nm thickness are island ones and are in the superparamagnetic state. When increasing the film effective thickness and coarsening the granules, there occurs interaction between their magnetic moments which leads to the magnetic medium ordering and, consequently, decline in the magnetoresistance effect. This process starts before the formation of an infinite cluster of metal granules that are in contact. Therefore, for the studied films, the TMR maximum is shifted relative to the percolation threshold towards smaller effective thicknesses. It should be also noted that TMR value, apart from the island size and the vacuum channel width, is affected by a number of factors: the presence of impurities and roughness at the interface between the islands and the vacuum barrier.

A negative isotropic magnetoresistance is also observed for the ultrafilms of $\mathrm{Co}_x\mathrm{Ni}_{1-x}$ and $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ ferromagnetic alloys throughout the whole range of their concentrations. The causes of the appearance of TMR for ultrathin films of ferromagnetic alloys are the same as in the case of island (discontinuous) films of pure metals. The results of the electron-microscopic investigations confirm the island structure of ultrathin films of $\mathrm{Co}_x\mathrm{Ni}_{1-x}$ and $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys. Figure 3 illustrates the structure of as-deposited and annealed at 700 K ultrathin films of $\mathrm{Co}_x\mathrm{Ni}_{1-x}$ alloy varying in effective thickness and concentration.

4 Conclusions

The as-deposited films of Fe and Co as well as of $\mathrm{Co}_x\mathrm{Ni}_{1-x}$ and $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys, whose effective thickness is below 20 nm, have an island structure with the islands (nanocrystals) measuring 2–5 nm and the channels of 1–3 nm wide. As a result of annealing at 700 K, due to coalescence the island sizes increase up to 15–25 nm, the channel width up to 2–7 nm depending on the film thickness.

It is determined that isotropic field dependences, which are caused by spin-dependent electron tunneling between the ferromagnetic islands, take place for as-deposited Co and Fe discontinuous films unlike Ni films. The maximum tunneling magnetoresistance observed for as-deposited Fe films ($d_{\mathrm{Fe}} =$ 15–20 nm) makes 1%. The maximum TMR values can occur when the island size is equal to 3–5 nm, and the width of the dielectric barrier between them is 1–3 nm. With increasing effective film thickness we observe the anisotropic character of the magnetoresistance.

The anisotropic magnetoresistance of up to 0.02% appears after annealing of Co samples in the thickness range of $d_{\mathrm{Co}} =$ 10–25 nm. Annealing does not result in the anisotropic magnetoresistance for Fe films of such thicknesses. For the annealed films of $\mathrm{Co}_x\mathrm{Ni}_{1-x}$ and $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys with an effective thickness of 5–10 nm we can observe the isotropic magnetoresistance of up to 0.2%.

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