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Influence of structure on the magnetoresistive properties of discontinuous multilayer structures based on $\mathrm{Ni_{80}Fe_{20}}$ and SiO

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This work presents the results of experimental studies on the structure and magnetoresistive properties of $[\mathrm{Ni_{80}Fe_{20}}(d_{\mathrm{Ni80Fe20}})/\mathrm{SiO}(5)]_5/\mathrm{S}$ discontinuous multilayer structures. It is shown that for both as-deposited and annealed structures at temperatures of 400, 500, 600, and 700 K, with an effective thickness of $\mathrm{Ni_{80}Fe_{20}}$ layer $d_{\mathrm{Ni80Fe20}} = 6\text{-}8$ nm, anisotropic magnetoresistance is observed. It was found that annealing at 500 K for samples with $d_{\mathrm{Ni80Fe20}} = 4\text{-}5$ nm leads to the emergence of isotropic magnetoresistance due to the formation of insulating interlayers between magnetic granules. After annealing at 600 K, anisotropic magnetoresistance reappears in structures with $d_{\mathrm{Ni80Fe20}} > 4$ nm due to the breakdown of the structural continuity of the insulating layers.

Ключові слова: discontinuous multilayer structures, ferromagnetic alloy, crystal structure, magnetoresistance, annealing.

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Introduction

The practical interest in nanocomposites is driven by their potential application as magnetosensitive elements in nanoelectronics, spintronics, and computing technologies [1-3]. Using such materials enhances the reliability of devices while significantly reducing their size, weight, power consumption, and cost [3-6]. Moreover, their relatively high specific electrical resistivity makes them suitable for high-frequency applications. Discontinuous multilayer structures based on ferromagnetic alloys and insulator layers are particularly promising among such materials. Despite substantial experimental and theoretical studies on granular structures [7-10], several questions remain unresolved. Specifically, the influence of the size and shape of magnetic granules, as well as their distribution within the insulator matrix, on the magnetic and magnetoresistive properties remains unclear. Meanwhile, less attention has been paid to studying layered structures based on ferromagnetic metals and insulator layers obtained by layer-by-layer deposition [11-15]. However, recent research has demonstrated that such structures offer several advantages over granular films. In particular, discontinuous multilayer structures can be considered composite materials with an amorphous insulating matrix that does not contain ferromagnetic metal atoms, has nearly uniform magnetic granule sizes, and allows for controlled modification of these sizes when necessary [15-19].

Given the above considerations, this study aimed to investigate the crustal structure and magnetoresistive properties of discontinuous multilayers $[\mathrm{Ni_{80}Fe_{20}}(d_{\mathrm{Ni80Fe20}})/\mathrm{SiO}(5)]_5/\mathrm{S}$ multilayers.

I. Experimental detail

The $[\mathrm{Ni_{80}Fe_{20}}(d_{\mathrm{Ni80Fe20}})/\mathrm{SiO}(5)]_5/\mathrm{S}$ discontinuous multilayers were prepared at room temperature in an HV chamber with a base pressure of 10^{-4} Pa. The method of layer-by-layer electron-beam evaporation was used for the deposition of samples. The thickness of the magnetic layers (d_{Ni80Fe20}) varied from 4 to 8 nm. The thickness of the layers was controlled by two independent in situ quartz resonators with an accuracy of 10%.

The magnetoresistive properties were measured using a software-hardware complex with current-in-plane geometries in an external magnetic field from 0 to 600 mT. All measurements were performed at room temperature. The measuring current was $I = 1$ mA. The value of longitudinal (magnetic field in the sample plane and parallel to current) and transverse (magnetic field in the sample plane and perpendicular to current) magnetoresistance have been calculated by equation $\mathrm{MR} = (R(B) - R(B_0))/R(B_0)$, where $R(B)$ is the current value of resistance in the magnetic field B; $R(B_0)$ is the resistance of the sample in the field of the B_0.

To investigate the annealing temperature effect on magnetoresistive properties, samples were annealed in step-increasing temperatures within the $T_{\mathrm{ann}} = 300$-700 K range, staying at each temperature for 20 min. The annealing was performed in a vacuum chamber with a 10^{-3} Pa pressure.

II. Results

To correctly interpret the results of the magnetoresistive properties studies of $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers, we first consider the features of their phase state and crystal structure (Figs. 1, 2). The results of electron microscopy studies on films with relatively large effective thicknesses of $Ni_{80}Fe_{20}$ layer ($d_{Ni80Fe20} = 6$-10 nm) indicate that these as-deposited films are nanocrystalline, with an average nanocrystal size of about 8-12 nm (Fig. 2a). The phase state of such samples corresponds to the *fcc*-Ni_3Fe with a lattice parameter of $a = 0.355$–0.358 nm. For as-deposited samples with ultrathin magnetic layers ($d_{Ni80Fe20} = 2$-5 nm), a reduction in the average nanocrystal size to 4-7 nm is observed (Fig. 2d). A detailed analysis of electron diffraction patterns for such samples is complicated due to significantly broadened diffraction maxima (Fig. 1). It can only be noted that the structure of such films is close to amorphous, with the broadening of maxima possibly related to the small crystallite size.

The size of magnetic granules significantly depends

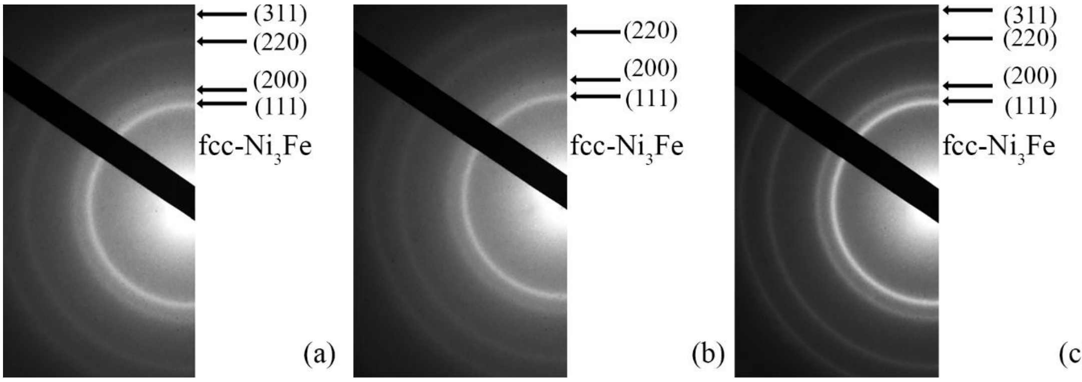

Fig. 1. Diffraction patterns for $[Ni_{80}Fe_{20}(4)/SiO(5)]_5/S$ discontinuous multilayers after deposition (a) and annealing at 500 K (b) and 700 K (c).

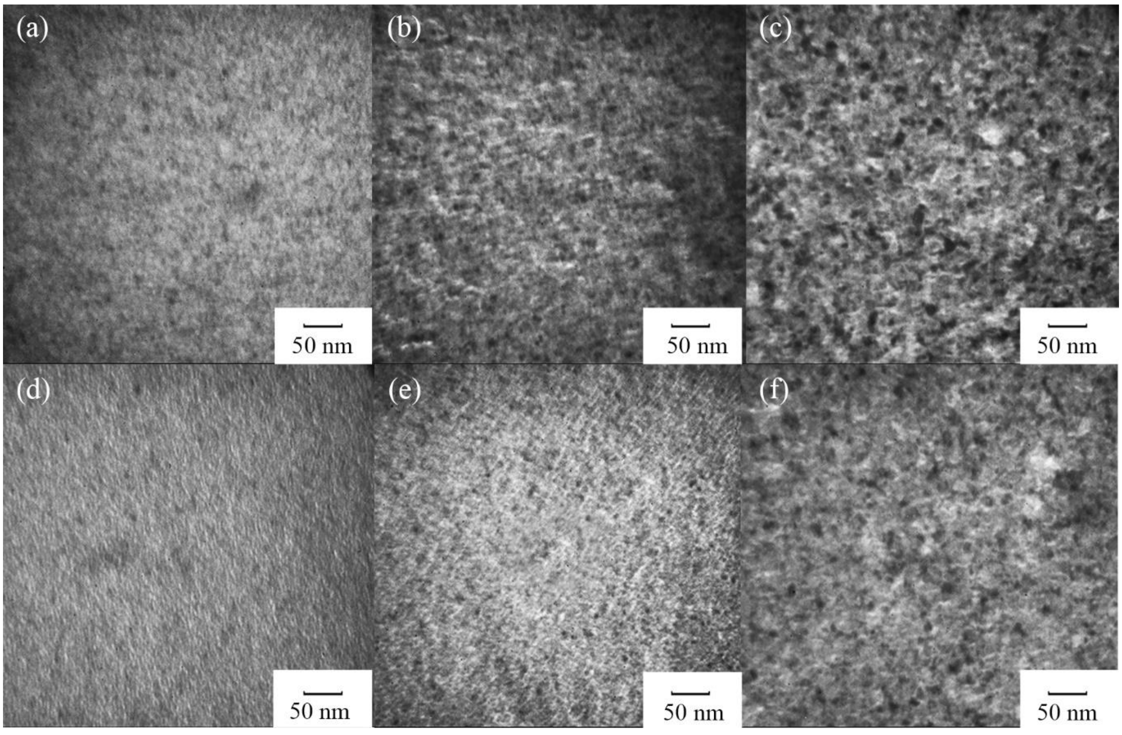

Fig. 2. TEM images for $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers with $d_{Ni80Fe20} = 8$ nm (a-c) and 4 nm (d-f) after deposition (a, d) and annealing to 500 K (b, e) and 700 K (c, f).

on the effective thickness of the magnetic layers. Fig. 3a shows the dependence of the average crystallite size on the effective thickness of the $Ni_{80}Fe_{20}$ layer. For $d_{Ni80Fe20}$ = 4 nm, the size of magnetic granules is approximately 5 nm. At these thicknesses, the layers may not be structurally continuous. In this case, an infinite ferromagnetic cluster forms, and the layer becomes "electrically continuous". As the effective thickness of the $Ni_{80}Fe_{20}$ layers increases to 6 nm, an increase in crystallite size to 8 nm is observed. At these thicknesses, structurally continuous metallic layers form.

Annealing at 500 K does not significantly change the structure and phase composition (Fig. 1b). However, even a slight (1.1-1.25 times) increase in crystallite size in structures with a thickness of $Ni_{80}Fe_{20}$ layer of 4-5 nm leads to the formation of isolated islands with insulatщк channels between them, 1-2 nm in width (Fig. 1d). This is confirmed by the magnetoresistive studies presented below. Additionally, based on Ref. [20], the formation of oxides around the crystallites is possible, although in this case, oxides were not detected via electron diffraction.

Significant structural changes are observed only after annealing at 700 K (Fig. 1c). For all samples, the average crystallite size of 1.5-2 times is growth (Fig. 2c, 2f). The phase composition of all samples annealed at 700 K corresponds to *fcc*-Ni_3Fe. The formation of oxide phases after annealing at 700 K was also not detected via electron diffraction.

Consider the results of magnetoresistive effect studies in as-deposited and annealed at 400, 500, 600, and 700 K structures with $d_{Ni80Fe20}$ = 4-8 nm. Figs. 4a and 4b illustrate the field dependence of magnetoresistance for as-deposited discontinuous multilayers with different thicknesses of the $Ni_{80}Fe_{20}$ layer. All as-deposited samples exhibit anisotropic magnetoresistance (AMR), characteristic of structurally continuous single-layer films of $Ni_{80}Fe_{20}$ alloy.

According to electron microscopic studies, in the process of deposition of such structures, "infinite" ferromagnetic clusters are formed, and, as a result, the structures are "electrically continuous". The formation of extended ferromagnetic clusters causes the realization of anisotropic magnetoresistance [21-23]. The value of the AMR at room temperature for these samples ranges from 0.05% to 0.15%, depending on the thickness of the $Ni_{80}Fe_{20}$ layers. Relatively small values of AMR can be attributed to a decrease in saturation magnetization resulting from the high defectiveness of as-deposited $Ni_{80}Fe_{20}$ layers and their structural inconsistencies.

Consider the effect of the annealing temperature on the nature of the field dependencies and the magnitude of the magnetoresistance of the discontinuous multilayers. Annealing at 400 K does not change the nature of the field dependencies of the magnetoresistance and its value. Significant changes in the magnetoresistive properties of such structures are observed only after annealing at a temperature of 500 K (Fig. 4c, d). The dependences of the magnetoresistance on the induction of the applied external magnetic field in the transverse and longitudinal geometries of measurement for discontinuous multilayers with $d_{Ni80Fe20}$ = 4 nm and 5 nm are isotropic (Fig. 4d). In most cases, the field dependences obtained in different geometries coincided with the experimental error, and no magnetoresistive hysteresis was observed in the field dependences of magnetoresistance. This fact suggests that the influence of magnetoresistance anisotropy is negligible. The maximum value of isotropic magnetoresistance at room temperature is approximately 0.1%. Such peculiarities of the magnetoresistance behavior can be explained by the structural features described in detail above. According to electron microscopic studies, after annealing the samples at a temperature of 500 K, the formation of granules with insulator channels between them with a width of 1-2 nm is observed. The insulator layers prevent direct ferromagnetic exchange between adjacent islets within the same layer but allow interisland tunneling [24-27]. Also, the structural continuity of the insulator layer is likely to be preserved, which excludes direct ferromagnetic exchange between magnetic islands of neighboring layers. Thus, a structure is formed in which an insulator barrier separates the magnetic granules, so the transfer of an electron from one granule to another will be carried out only due to the tunneling effect.

For structures with $d_{Ni80Fe20}$ = 6-8 nm, in which an infinite ferromagnetic cluster is formed in the initial state, annealing at a temperature of 500 K does not lead to its fragmentation (formation of a granular structure). As a result, the character of the magnetoresistance remains anisotropic Fig. 4e.

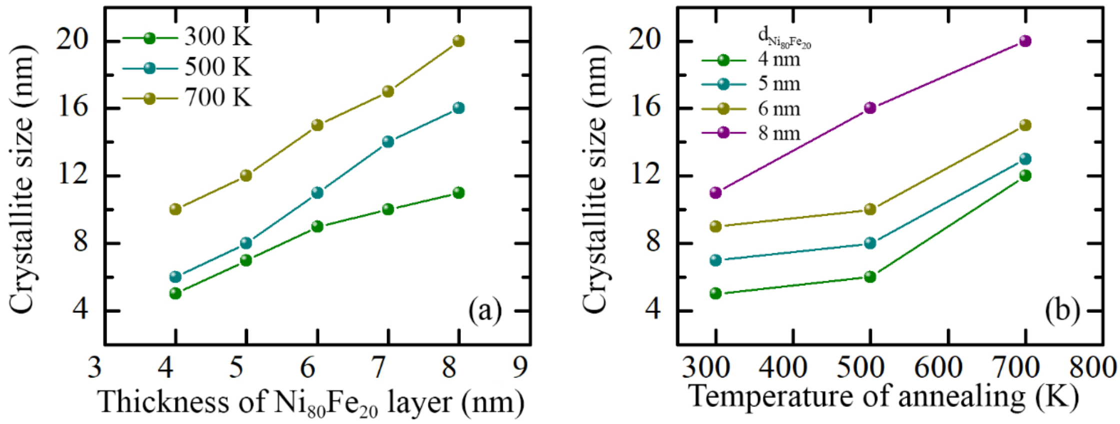

Fig. 3. The average size of crystallites as a function of the effective thickness of $Ni_{80}Fe_{20}$ layers (a) and annealing temperature (b) for $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers.

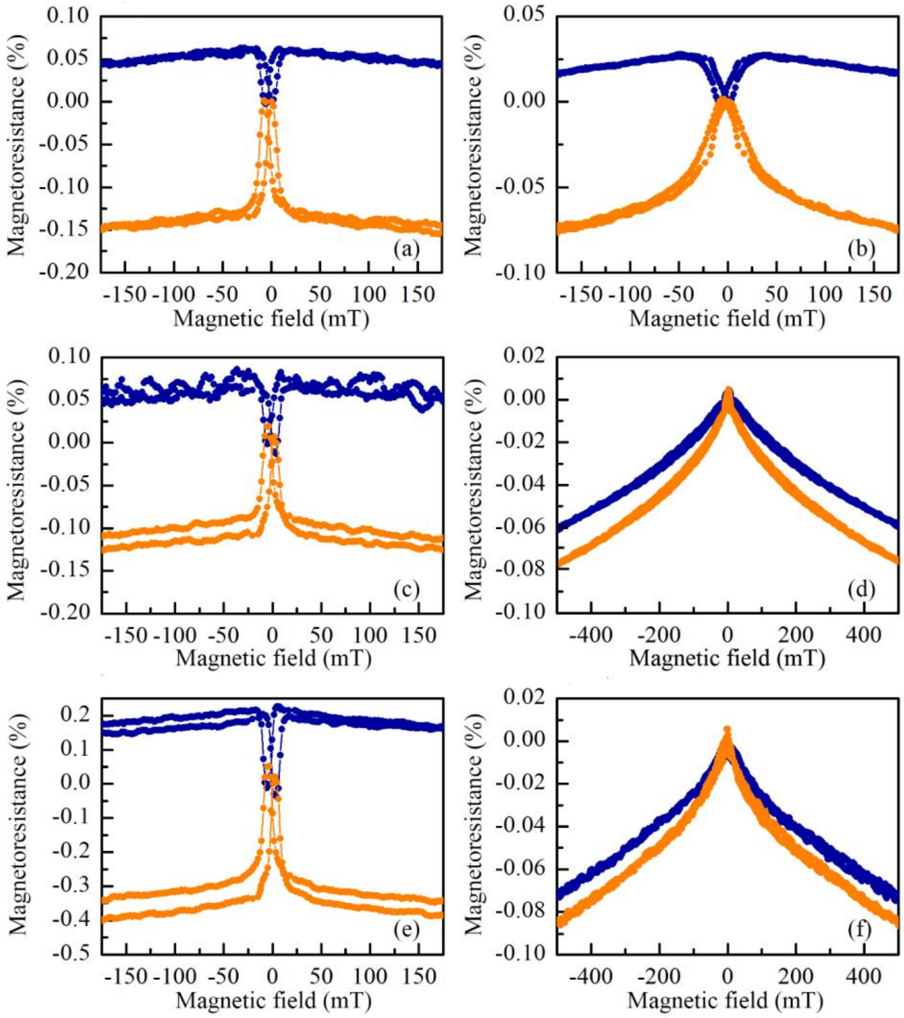

Fig. 4. Field dependencies of the magnetoresistance for $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers with $d_{Ni80Fe20}$ =8 nm (a, c, e) and 4 nm (b, d, f) after deposition (a, b) and annealing to 500 K (c, d) and 700 K (e, f).

Consider the peculiarities of the magnetoresistive properties of the discontinuous multilayers after annealing at 600 K. In this case, for structures with $d_{Ni80Fe20}$ = 6-10 nm, no change in the nature of the field dependencies was observed. Only a slight increase in both the transverse and longitudinal magnetoresistance values can be noted due to the increase in the size of metal granules and the healing of crystal structure defects.

The magnetoresistance behavior after annealing at 600 K of the discontinuous multilayers with $d_{Ni80Fe20}$ = 4 nm is presented in Fig. 4f. A transition to the anisotropic character of the magnetoresistance is observed after annealing. This magnetoresistance behavior can be explained by violating the structural continuity of the insulator layers. As a result, a direct ferromagnetic exchange appears between the magnetic granules of neighboring layers. This leads to the formation of a ferromagnetic cluster throughout the entire structure volume, resulting in the realization of anisotropic magnetoresistance.

A further increase in the annealing temperature to 700 K, regardless of the thickness of the ferromagnetic layers, does not change the nature of the magnetoresistance. Only a 2-fold increase (up to 0.1%) in the value of isotropic magnetoresistance and a 2-3-fold increase (up to 0.3-0.5%) in the anisotropic magnetoresistance are observed (Fig. 5). This increase in magnetoresistance is due to a significant increase in the size of the granules (up to 12-20 nm), which is confirmed by the results of electron microscopic studies.

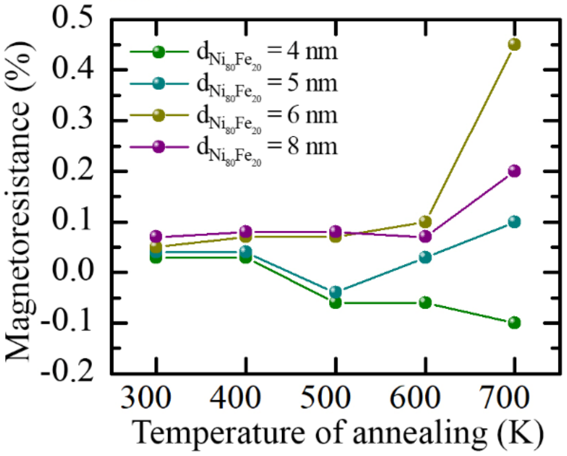

Fig. 5. Dependence of longitudinal magnetoresistance on annealing temperature for the $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers with different effective thicknesses of ferromagnetic layers. The negative value of the magnetoresistance shows

that the electrical resistance of the magnetized sample decreases when it is placed in a magnetic field.

Conclusions

All as-deposited and annealed (400, 500, 600, and 700 K) $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5/S$ discontinuous multilayers with effective thicknesses of $Ni_{80}Fe_{20}$ layer $d_{Ni80Fe20} = 6\text{-}10$ nm are nanocrystalline, with an average nanocrystal size of about 8-20 nm, depending on layer thickness and annealing temperature. The phase composition of these samples corresponds to *fcc*-Ni_3Fe. For as-deposited samples with ultrathin magnetic layers $d_{Ni80Fe20} = 4\text{-}5$ nm, the average nanocrystal size decreases to 4-7 nm. These structures form a continuous magnetic cluster, resulting in anisotropic magnetoresistance. An anisotropic magnetoresistance effect is observed for all as-deposited samples. After annealing at 500 K for structures with $d_{Ni80Fe20} = 4\text{-}5$ nm, a transition to isotropic magnetoresistance is observed. This is due to the formation of insulator channels between magnetic granules. Further annealing at 600 and 700 K significantly increases the crystallite size, disrupting the structural continuity of the insulator layers and forming metallic clusters throughout the structure. For such samples, anisotropic magnetoresistance reappears. Except for structures with $d_{Ni80Fe20} > 4$ nm, where the formation of an infinite metallic cluster is not observed due to the low density of metallic granules.

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Вплив структури на магніторезистивні властивості багатошарових структур на основі $Ni_{80}Fe_{20}$ та SiO

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У роботі представлені результати експериментальних досліджень структури та магніторезистивних властивостей шаруватих структур $[Ni_{80}Fe_{20}(d_{Ni80Fe20})/SiO(5)]_5$П. Показано, що для невідпалених та відпалених при температурі 400, 500, 600 та 700 К шаруватих структур з ефективною товщиною шарів $d_{Ni80Fe20} = 6$-8 нм спостерігається анізотропний характер магнітоопору. Встановлено, що відпалювання зразків при температурі 500 К з $d_{Ni80Fe20} = 4$-5 нм приводить до появи ізотропного магнітоопору внаслідок формування діелектричних прошарків між магнітними гранулами. Після відпалювання при температурі 600 К спостерігається знову поява анізотропного магнітоопору у структурах із $d_{Ni80Fe20} > 4$ нм внаслідок руйнування структурної суцільності діелектричних прошарків.

Ключові слова: багатошарові структури, феромагнітний сплав, кристалічна структура, магнітоопір, відпалювання.