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Methods of modulation of micromagnetic characteristics of multilayer thin-film systems [Co/Pt]

© M.V. Dorokhin[1], P.V. Demina[1], A.V. Zdoroveishchev[1], D.A. Zdoroveishchev[1], A.V. Kudrin[1], I.L. Kalentyeva[1], V.N. Trushin[1], M.P. Timiryazeva[1,2], A.G. Temiryazev[1,2]

[1] Lobachevsky State University,
Nizhny Novgorod, Russia
[2] Fryazino Branch, Kotel'nikov Institute of Radio Engineering and Electronics, Russian Academy of Sciences,
Fryazino, Moscow oblast, Russia
E-mail: dorokhin@nifti.unn.ru

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Multilayer magnetic films were formed, representing 10 alternating layers of Co and Pt with a thickness of 3 and 5 Å, respectively ($[Co(3)Pt(5)]_{10}$). Two technological modifications of the multilayer Co/Pt structure are considered, one of which (doubling the thickness of the Co and Pt layers in one of the ten bilayers) allows for small adjustments, and the other (increasing the thickness of the Co layer by 3.5 times in one of the 10 bilayers) significantly changes the micromagnetic parameters: the density of skyrmions in a magnetized film in a zero magnetic field. The effect obtained is associated with modulations of the degree of mixing between the Co and Pt layers, which in the first case lead to some change in the composition of the Co_xPt_{1-x} solid solution in the center of the multilayer structure, and in the second case to the formation of an unmixed Co layer in the center of the structure; the boundaries between this layer and the Co_xPt_{1-x} solid solution provide a significant change in magnetic properties.

Keywords: magnetic anisotropy, thin films, ferromagnetism.

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1. Introduction

Thin-film multilayer structures and alloys based on ferromagnetic/heavy metal systems are of interest because of their unique magnetic and micromagnetic properties [1–4]. Such materials have magnetic anisotropy from the point of view of magnetic properties, the type of which is controlled by the technological parameters of film production. For instance, the studies in Ref. [3,4] showed the presence of perpendicular magnetic anisotropy in multilayer thin-film Co/Pt structures with a layer thickness of units of angstrom ensuring the position of the axis of light magnetization in the direction normal to the surface. Variations of the thicknesses of the ferromagnetic and heavy metal layers (independent or simultaneous), variations of the degree of mixing between the layers ensure a significant variation of the magnetic anisotropy constants and a corresponding variation of the position of the axis of light magnetization of the ferromagnetic film [5–7]. Thus, the technological parameters make it possible to control the magnetic properties of the films under consideration within a wide range. The possibility of formation of isolated magnetic domains with the possible size of hundreds or tens of nanometers has been shown from the point of view of the micromagnetic structure for ferromagnetic/heavy metal systems (in particular, Co/Pt). Such domains are a natural memory element in which a bit of information corresponds to the direction of magnetization [5,8]. The size, shape and density of micromagnetic elements depend on the structure and composition of the films and can also be controlled by varying the technological parameters of their growth [5–8].

This paper considers new methods of modification of technological parameters ensuring both smooth adjustment and significant restructuring of the magnetic properties and micromagnetic structure of thin Co/Pt films formed by electron-beam evaporation in the mode of alternating sputtering of Co and Pt targets with an electron beam. Earlier structural studies demonstrated that magnetic and micromagnetic properties are determined by the degree of diffusive blurring of the heterogeneities between the layers of the multilayer film and the composition of the solid solution of Co_xPt_{1-x}, which is formed in the system as a result of diffusive mixing of Co and Pt [9]. Local modulations were introduced in this study into a multilayer structure with strong diffusion blurring of layers in the form of an increase of the thickness of one Co/Pt bilayer out of ten or an increase of the thickness of the Co layer in one of ten bilayers. It is shown that such modulations provide additional tools for controlling the properties of ferromagnetic films by changing the uniformity and degree of mixing.

2. Experiment

Thin-film Co/Pt structures were formed on Si/Al_2O_3 substrates by electron beam evaporation in high vacuum [9,10] at a temperature of 200°C. The growth was carried out by successive application of ten 3 and 5 Å thick layers of Co and Pt, respectively. Thus, the basic type of structures was a multilayer film $[\mathrm{Co}(3)\mathrm{Pt}(5)]_{10}$. The properties of such structures depending on the thicknesses of Co and Pt were considered earlier in [9,10]. The film structure was intentionally modified at the micro level in this study by technological modulation during the sputtering process without changing the overall thickness of the film. The modification comprised the following:

– doubling of the thickness of one bilayer in the center of a multilayer film (and reduction of the total number of bilayers to 9) — structure A. This provided the structure of the form $[\mathrm{Co}(3)/\mathrm{Pt}(5)]_4[\mathrm{Co}(6)/\mathrm{Pt}(10)]_1[\mathrm{Co}(3)/\mathrm{Pt}(5)]_4$;

– an increase of the thickness of the cobalt layer in one of the periods by 3.5 times (and a decrease of the total number of bilayers to 9) — Structure B. This provided the structure of the form $[\mathrm{Co}(3)/\mathrm{Pt}(5)]_4[\mathrm{Co}(11)/\mathrm{Pt}(5)]_1[\mathrm{Co}(3)/\mathrm{Pt}(5)]_4$;

– the structure B — $[\mathrm{Co}(3)/\mathrm{Pt}(5)]_{10}$ formed under similar conditions and equivalent to the structures considered in [9] was used as a control. This structure was formed to compare the results.

The phase composition and crystal structure of the formed films were studied by X-ray diffraction using a Bruker D8 Discover X-ray diffractometer. The crystal structure of the GaAs substrates on which the multilayer metal film was formed corresponds to a single-crystal and is not considered in this paper.

The magnetic properties were evaluated by measuring the anomalous Hall effect at room temperature. The Hall resistance $R_{\mathrm{H}}(H)$ is a nonlinear function of the external magnetic field and has both a component proportional to the magnetic field strength (R_0) and a component proportional to the magnetization of the structure $R_s(M)$ [11,12]. The value of $R_0 \ll R_s$ in the saturation region of magnetization in the studied structures. Therefore, R_0 is assumed to be zero, and the dependence $R_H(H)$ is thus similar to the magnetic field dependence of magnetization. The comparison of curves $R_H(H)$ for different structures allows drawing conclusions about the modulation of magnetization with varying technological parameters.

The micromagnetic structure of the formed Co/Pt films was studied by magnetic force microscopy (MFM) using microscope Smart SPM (AIST-NT) in the „two-pass“ mode using a probe with a low magnetic moment [13]. The changes of the domain structure were studied by MFM scanning using PPP-LM-MFMR probe (Nanosensors) the magnetic moment of which is sufficient for the formation of skyrmions by the method described in Ref. [14,15]. The samples were magnetized for changing the magnetic domain structure which consisted in placement of the MFM probe at a minimum distance to the surface and its movement over a certain area (in a mode similar to MFM scanning). A separate region of films with sizes from 3×3 to $5 \times 5\,\mu\mathrm{m}^2$ was magnetized as a result of exposure of the surface to the magnetic field of the probe.

3. Results and discussion

A series of structural studies of samples similar to those formed in this paper were performed in Ref. [9,10]. The results showed that the individual layers of Co and Pt in films of the type $[\mathrm{Co}(3)/\mathrm{Pt}(5)]_{10}$ are strongly mixed with each other and represent a solid solution $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ with periodically varying composition x [9.10]. The obtained results of the study of structures confirm the mixing of Co and Pt layers and the formation of a solid solution $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ of variable composition x like in Ref. [9,10]. The X-ray diffraction spectra of the obtained films are shown in Figure 1, *a* and *b*. The only peak recorded on the spectra (apart from peaks from the substrate) is the line corresponding to the solid solution $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ although layers with a large thickness value were embedded in the film structure. Presumably, uniform diffusion mixing does not take place in the studied structures, but the individual layers are so strongly mixed with each other that only a peak corresponding to a solid solution with some averaged composition is recorded on the X-ray diffraction spectra. Figure 2 shows the magnetic field dependences of the Hall resistance, reflecting changes of the magnetization. The dependence $R_H(H)$ of the control structure B is a closed hysteresis loop with a singularity of the form „wasp waist“. A similar singularity is recorded in a number of cases in two-phase magnetic films [9]. Figure 3 shows MFM images

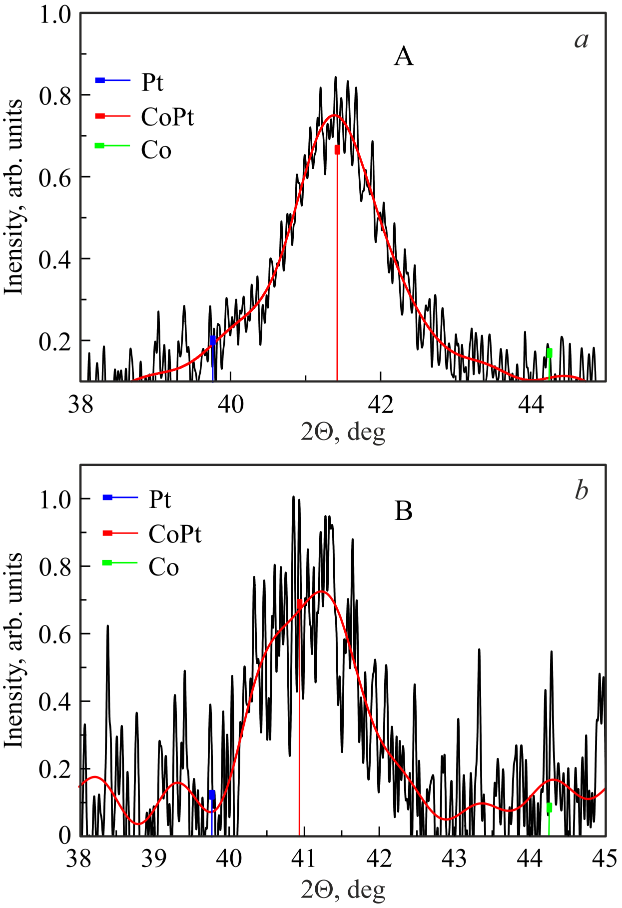

Figure 1. Measured X-ray diffraction spectra of Co/Pt samples Structure A (*a*) and Structure B (*b*). The spectral region in which peaks from the Co/Pt film were observed is shown.

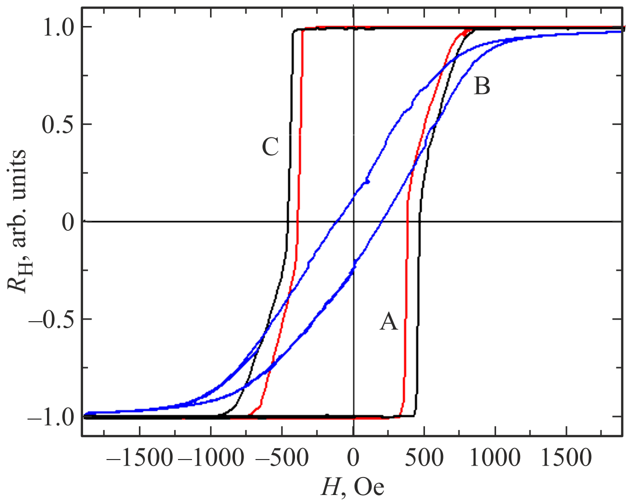

Figure 2. Magnetic field dependences of the Hall resistance measured for samples A (curve A) and B (curve C) formed with a deviation from the growth pattern and the control sample B (curve C).

of the micromagnetic structure of the formed samples. Dark regions of $\sim 0.1\,\mu$m in size previously interpreted as skyrmions [9] based on Lorentz transmission microscopy data are observed in the magnetized region in the MFM image of the control structure (Figure 3,*c*). The density of skyrmions, defined as the number of skyrmions per unit area, is $\sim 1\,\mu\text{m}^{-2}$, which is relatively small. It should be noted that for the control structure, both the magnetic properties and the micromagnetic structure (including the density of skyrmions) are consistent with the previously obtained results in Ref. jcite9. Technological modulations of the layers result as will be shown below, to significant changes in the magnetic characteristics and micromagnetic structure compared with the control structure. An increase of the thickness of one Co/Pt bilayer by two times in one of the 9 bilayers (structure A) results in a decrease of the width of the hysteresis loop from 475 Oe to 355 Oe (Figure 2, curves B and A, respectively). The micromagnetic pattern experiences some transformation: the density of skyrmions calculated according to Figure 3,*a* is $\sim 1.2\,\mu\text{m}^{-2}$, round micromagnetic singularities identified in the MFM image are transformed into extended micromagnetic elements (Figure 3,*a*). Presumably, the extended elements correspond to skyrmions located close to each other, the distance between which is lower than the resolution of a magnetic force microscope.

A 3.5-fold increase of the thickness of the Co layer to 11 Å in one of the 9 bilayers (structure B) results in a much more significant transformation of properties (Figure 2, curves C and B). The magnetic hysteresis loop undergoes significant changes: the magnitude of the coercive field decreases from 475 to $\approx$ 100 Oe, the ratio of Hall resistances in the zero and maximum magnetic fields R_o/R_s decreases (in structures A and In it was equal to 1). The so-called „slope“ of the hysteresis loop ensures a change of the micromagnetic structure: the period of the domain structure is significantly reduced. A more detailed scanning of the remagnetization result shows that the probe splits the domains into round skyrmions, but the density of skyrmions

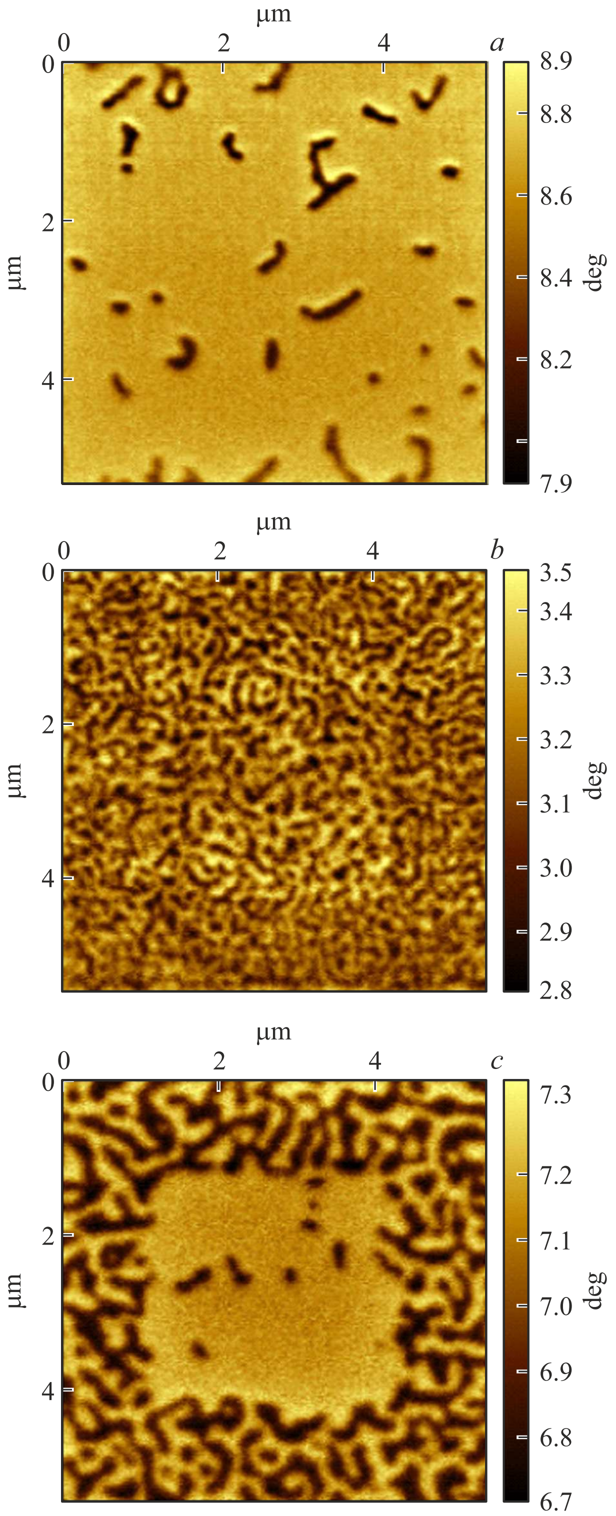

Figure 3. MFM images of samples A (*a*), B (*b*) and C (*c*). The scan area of structure A was $5\,\mu\text{m}^2$, the scan area of structures B and C was $3\,\mu\text{m}^2$.

increases by more than an order of magnitude compared to the original structure. A part of the skyrmions located close to each other is resolved as an elongated micromagnetic element (Figure 3, *b*).

Let's move on to discussing the results obtained. The dependence of the magnetic properties and the micromagnetic structure of the layers $[\mathrm{Co(x)/Pt(y)}]_{10}$ on the thickness of Co and Pt in the multilayer structure was obtained in the previous studies [9,10]. It was found in this study that the structures are strongly mixed films $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ with variable contents of Co and Pt unlike most of the known studies. The strong mixing of layers Co and Pt is evidenced by X-ray diffraction measurements of the samples, as well as TEM images of the cross-section, considered in Ref. [9]. A comparison of the TEM and X-ray diffraction data allows making the following conclusions: layers of solid solution of $\mathrm{Co}_z\mathrm{Pt}_{1-z}$ with an increased content of Co are formed in the multilayer structure instead of Co layers, and layers of $\mathrm{Co}_y\mathrm{Pt}_{1-y}$ with an increased content of Pt are formed instead of platinum. The composition $\mathrm{Co}_x\mathrm{Pt}_{1-x}$, revealed by X-ray diffraction studies (Figure 1) is the average value of the composition of the solid solution.

The presence of skyrmions in Co−Pt films is associated with the Dzyaloshinskii–Moriya Interaction (DMI) resulting in a special type of magnetic ordering [1,6–9]. The energy of the Dzyaloshinskii−Moriya Interaction is believed to include two components: volumetric and superficial. In the case of the structures studied in this paper for the surface component of the DMI, the interaction constant depends on the degree of blurring of the heteroboundaries of $\mathrm{Co}_z\mathrm{Pt}_{1-z}/\mathrm{Co}_y\mathrm{Pt}_{1-y}$, whereas the volume component of the DMI is associated with the presence of L_{10} phase in the system, the value of the volume constant usually increases with an increase of the total thickness of the film [16]. The total thickness of the film does not change in this study, the increase of the thickness of a separate layer in the center is attributable to a decrease of the total number of bilayers.

A Co/Pt bilayer with the doubled technological thickness is formed in the structure in accordance with the technology described above. The amount of diffusive blurring of the boundaries does not change, but the degree of mixing of Co and Pt in this particular bilayer slightly decreases, and the number of heteroboundaries decreases by one (because 9 bilayers are formed to preserve the total thickness of the film). Such a structural transformation results in a slight change of both the magnetic properties and the micromagnetic structure of the magnetized film.

It was previously shown that Co/Pt layers are mixed with each other in a multilayer structure up to 6 Å thick [9,10]. We assume that the formation of a layer of Co in a multilayer structure with a significantly larger thickness (structure B) in comparison with [9,10] prevents its complete mixing, as a result, a thin layer of pure Co with a strongly blurred diffusion heteroboundary is formed in the center of the film. This introduces two new boundaries to the system: $\mathrm{Co/Co}_x\mathrm{Pt}_{1-x}$ and $\mathrm{Co}_x\mathrm{Pt}_{1-x}/\mathrm{Co}$, which may be asymmetric because of different growth times and exposure to high temperature. The average composition of the films does not significantly change, which should not significantly affect the value of the volume constant of the DMI. At the same time, the significantly structurally different boundary of $\mathrm{Co/Co}_x\mathrm{Pt}_{1-x}$ most likely introduces a significant change in surface effects: both in the value of the magnetic anisotropy constant and the value of the constant of Dzyaloshinskii−Moriya interaction. This causes significant changes of the type of the magnetic hysteresis loop (Figure 2) and the micromagnetic structure of the film (an increase of the density of skyrmions in a magnetized film in a zero magnetic field), respectively.

4. Conclusion

Therefore, the study demonstrates two technological modifications of the multilayer Co/Pt structure without any change of its thickness, one of which (an increase of the thickness of one Co/Pt bilayer by two times in one of 9 bilayers) allows for small adjustments of micromagnetic parameters, and the other (an increase by 3.5 times of the thickness of the Co layer up to 11 Å in one of the 9 bilayers) allows for a significant changing of the micromagnetic parameters: the density of skyrmions in a magnetized film in a zero magnetic field, the magnitude of the coercive field. The effect obtained is associated with modulations of the degree of mixing between the Co and Pt layers, which in the first case lead to some change in the composition of the solid solution $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ in the center of the multilayer structure, and in the second — to the formation of an unmixed layer in the center of the structure Co; the boundaries between this layer and the solid solution $\mathrm{Co}_x\mathrm{Pt}_{1-x}$ provide a significant change in magnetic properties.

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Conflict of interest

The authors declare that they have no conflict of interest.

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