05,13

Manipulating the Micromagnetic Structure of Multiphase CoPt Thin Films by Varying Layer Thicknesses

© M.V. Dorokhin[1], P.B. Demina[1], A.V. Zdoroveyshchev[1], D.A. Zdoroveyshchev[1], A.G. Temiryazev[1,2], M.P. Temiryazeva[2], I.L. Kalent'eva[1], V.N. Trushin[1]

[1] Research Institute for Physics and Technology, Lobachevsky State University of Nizhny Novgorod, 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, demina@phys.unn.ru

Received April 17, 2023
Revised April 17, 2023
Accepted May 11, 2023

The magnetic properties and micromagnetic structure of $[Co/Pt]_{10}$ multilayer magnetic films formed by independent variation of the Co and Pt layer thicknesses have been studied. The possibility of the film magnetization parameters manipulation was shown. It is found that the micromagnetic structure of the layers is significantly modified with a change in the thickness of the layers which correlates with the magnetization data. In particular, magnetic force microscopy revealed a system of magnetic skyrmions for a number of structures. The skyrmion density was found to be dependent on the growth conditions which in turn correlates with the shape of the magnetic hysteresis loop. Changing the thickness of the Co and Pt layers makes it possible to control the density of skyrmions in the range from 0.2 to 10.5 mkm^{-2}.

Keywords: micromagnetic structure, skyrmions, CoPt films, electron beam evaporation.

DOI: 10.21883/PSS.2023.06.56106.15H

1. Introduction

Multilayer films of ferromagnetic/heavy metal with an asymmetric layer alternation structure have been the subject of intensive research in recent years [1]. Interest in such materials is due to the complex of unique properties that allow the formation of a special micromagnetic structure with nanoscale magnetic domains — skyrmions. Skyrmions are considered to be a promising candidate for bit carrier: they are characterized by two stable states of magnetization and their small size enables ultra-high recording densities. Techniques for controlling the skyrmion size and density consist of varying the structure and composition of the magnetic film by changing the process growth parameters [1]. It is known from literature sources, including calculated phase diagrams, that the CoPt combination can form Co, CoPt, Co_3Pt, $CoPt_3$ and Pt. The structure of these phases may differ considerably: the phase may have different crystal system or be polymorphous altogether. The micromagnetic structure, however, is highly dependent on the specific phase composition and can change significantly even when it is slightly modulated [2–4].

The present paper is devoted to the study of methods for controlling the micromagnetic structure in Co/Pt multilayer films by precisely changing the thickness of Co and Pt layers. The ferromagnetic/heavy metal structures studied are distinguished by the method of their fabrication (electron-beam evaporation) which provides a high flexibility in controlling the parameters of the technological process and, consequently, the structure of the films formed. Change in the growth conditions not only allows the phase composition but also the homogeneity of the heterointerfaces between the individual layers to be altered. Films with blurred heterointerfaces are among the new objects in which the physics of magnetic interactions has not yet been studied.

2. Procedure

In this paper, multilayer films $[Co/Pt]_{10}$ formed by alternating sputtering of ferromagnetic/heavy metal layers on GaAs substrate were studied. The structures were obtained by electron beam evaporation in vacuum at 200°C. The film composition was set by the ratio of sputtering times of Co and Pt layers. Under these growth conditions, a multilayer film is formed with incomplete mixing between the layers. Two series of samples were considered: with varying thickness of Co in each bilayer from 1 to 6 Å with constant thickness of Pt (5 Å); with varying thickness of Pt in each bilayer from 3 to 7 Å with constant thickness of Co (4 Å). Each film was formed from 10 bilayers. The table shows the process parameters of the samples tested.

The micromagnetic structure of the formed CoPt films was studied by magnetic force microscopy (MFM) using a Smart SPM (AIST-NT) microscope using a low-magnetic-momentum [5] probe as a „standard" method. Domain structure changes were studied by MFM scanning with a PPP-LM-MFMR probe (Nanosensors), whose magnetic

Technological parameters of CoPt films

Number sample	Thickness Co in one layer, Å	Thickness Pt in one layer, Å	Thickness of one layer of CoPt film, Å	Marking samples
1	1	5	6	1/5
2	2	5	7	2/5
3	3	5	8	3/5
4	4	5	9	4/5
5	5	5	10	5/5
6	6	5	11	6/5
7	4	3	7	4/3
8	4	4	8	4/4
9	4	6	10	4/6
10	4	7	11	4/7

moment is sufficient to form skyrmions using the method described in papers [6,7]. To change the magnetic domain structure, samples „sub-magnetization“ was performed by bringing the MFM probe to a minimum distance from the surface and moving the probe over a certain area (in mode similar to MFM scanning). A separate area of the films was remagnetized from 3×3 to 5×5 mkm as a result of the probe's magnetic field being applied to the surface.

Measurements of the anomalous Hall effect acted as an estimate of the magnetization of the films as the Hall resistance $R_H(H)$ is a non-linear function of the external magnetic field and has both a component proportional to the magnetic field strength and a component proportional to the structure magnetization $R_s(M)$ [8,9].

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.

3. Results and discussion

According to the previously obtained results, the formed films are a disordered polycrystalline system with crystallite sizes less than 10 nm [10]. Fig. 1, *a* shows typical X-ray diffraction spectra of the structures (1, 2, 5, 6 and 10). For samples 1, 2, 5 and 10, the position of the main maximum does not correspond to the lines for Co and Pt or Co-Pt compounds. The analytical software interprets the indicated peaks as $CoPt(x)$ solid solution with a composition dependent on the thickness ratio. Note, that this spectrum is characteristic of most studied structures, the only difference being the precise position of the main diffraction maximum (DM). Fig. 1, *b* shows the dependence of the position of the main DM on the ratio of the Co thickness to the total thickness of the layers. A monotonic increase in the angular position of the DM with increasing relative thickness Pt can be seen. Note also, that for structures with small relative cobalt thickness, the DM peaks are shifted to Co (Fig. 1, *b*), which indicates incomplete dissolution of platinum; it is also confirmed by the presence of low-intensity peaks corresponding to DM from the undissolved Pt layer. An example of such a spectrum for sample 2 is shown in Fig. 1, *a* (curve 2/5). An exception to these patterns is the spectrum of a sample with a relatively thick Co (6 Å) layer, where broad lines are identified with maxima at angles corresponding to the DM positions from atomic cobalt and platinum (Fig. 1, *a*, curve 6/5).

Previous studies [10], have shown that CoPt film composed of 10 period Co and Pt with thicknesses of 3 and 5 Å, respectively, is heterogeneous, and its composition is modulated with a period close to the value of Co and Pt layer thicknesses. When varying the relative thickness of the layers, the degree of mixing of Co and Pt obviously depends on the specific values of d_{Co} and d_{Pt}. In structures with small Co thicknesses, it seems that only part of the platinum at the Co/Pt interface participates in cobalt diffusion and dissolution, so the film is probably a solid solution layer separated from each other by platinum layers. Co thickness increase provides a situation of complete Co and Pt dissolution, although the high peak widths and earlier results suggest heterogeneous mixing and preservation of the periodicity of the composition. With a relatively large Co thickness of 6 Å, the dissolution of cobalt by platinum is no longer ensured, which fundamentally changes both the structural and magnetic properties.

The magnetic properties of the structures were studied by measuring the magnetic field dependences of the Hall resistance. For the structures with different Co and Pt thicknesses, four different curve shapes were recorded, shown in Fig. 2. Structures with low Co content are characterized

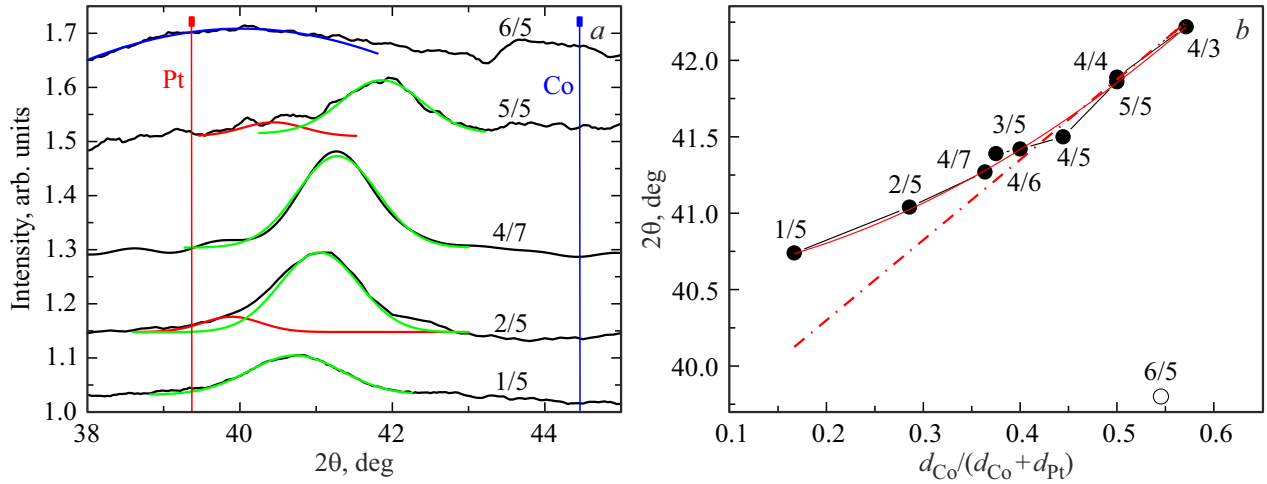

Figure 1. *a* — X-ray diffraction spectra measured in the area corresponding to the peak from the formed film. The numbers correspond to the sample labeling. *b* — Dependence of X-ray diffraction peak position on the ratio of Co and Pt thicknesses, the dashed line in the diagram shows the linear function of the solid solution (0 = Pt, 1 = Co).

by pronounced perpendicular magnetic anisotropy with the axis of easy magnetization lying in the growth direction — perpendicular to the surface (Fig. 2, *a*). The hysteresis loop width increases with increasing Co content. For structures with a Co layer thickness from 3 to 4 Å the shape of the hysteresis loop changes, there are additional breaks in the dependence which are typical of structures containing two interacting magnetic phases (Fig. 2, *b*) [11]. The hysteresis loop width (coercive field) in this thickness range decreases with increasing thickness.

For structures with nominally identical Co and Pt thicknesses (4/4 and 5/5 Å), the easy magnetization axis shifts in direction to the layer plane, which manifests itself in the „slope“ of the hysteresis loop (Fig. 2, *c*). The value of the residual resistance for these structures becomes lower than the magnetization saturation resistance. In the case of maximum thicknesses of cobalt or platinum, the axis of easy magnetization lies in the layer plane, which manifests itself in the absence of residual magnetization in the dependence $R_H(H)$ and a significant increase in the magnetic saturation field (Fig. 2, *d*).

The above trends can most clearly be seen in the relative thickness Co dependence of the two hysteresis loop parameters: the saturation magnetic field and the ratio of the residual resistance (in zero magnetic field) to the saturation magnetization resistance (R_O/R_S). This ratio is proportional to the ratio of residual magnetization to saturation magnetization (M_r/M_s). The above dependencies are shown in Fig. 3, *a, b*, respectively. It can be clearly seen that as the relative thickness of Co increases, there is a rotation of the easy magnetization axis in the plane of the films, which manifests itself in an increase in the magnetic saturation field and a reduction of the residual Hall resistance to zero.

Typical images of the micromagnetic structure of the samples are shown in Fig. 4. In the center of the MFM image, there is an area of film remagnetized by the probe in the mode discussed in the methodical part of the paper. The area size ranges from 3×3 to 5×5 mkm^2, as discussed above. At the image periphery, a domain structure characteristic of non-magnetized CoPt film is observed. The exception is sample 6/5 (Fig. 4, *d*), not showing any magnetic contrast in the MFM image. This is due to the location of the easy magnetization axis in the film plane for this sample, in this configuration, the magnetic domains cannot be resolved by the magnetic probe used. The magnetized areas of the MFM images show features in the form of dark contrasting spots interpreted as skyrmions [10]. Previous studies of similar structures [10] by Lorentzian transmission electron microscopy (LTEM) have shown that similar dark contrast spots in MFM images of CoPt films can be matched with a characteristic contrast feature in the LTEM image, which is unambiguously interpreted as a Néel skyrmions. The characteristic blurring of the circular skyrmion shape into an oval is probably due to insufficient resolution of the probe, which does not allow the separation of closely spaced adjacent skyrmions.

The size and concentration of the skyrmions (N_{sk}) depends on the Co layer thickness. The term „skyrmion concentration“ refers to the number per unit area. Note, that the N_{sk} parameter correlates with the type of magnetoelectric Hall resistance dependence. In structures with the smallest relative Co layer thickness characterized by the magnetic properties shown in Fig. 2, *a*, a homogeneous magnetic contrast without features is recorded (Fig. 4, *a*). As the Co (2/5) thickness increases, the type of magnetic field dependence R_H changes (Fig. 2, *b*), with low-density skyrmion being recorded. Skyrmion density was calculated by recalculating the skyrmion number in the acquired images relative to the magnetized area, with oval features estimated as two skyrmions (assuming that closely spaced skyrmions give a homogeneous contrast). This calculation is an assessment, and the resulting N_{sk} value is highly

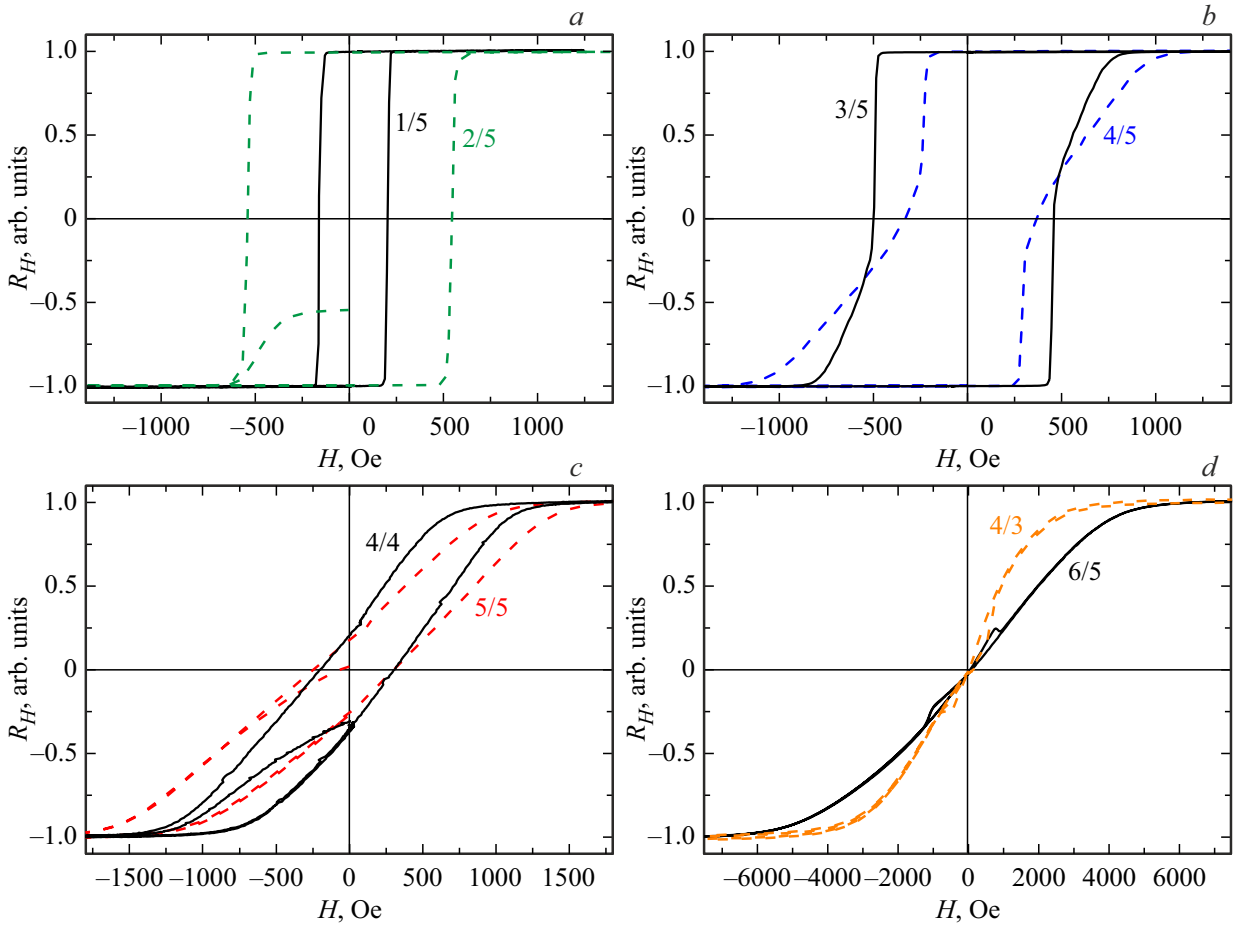

Figure 2. Magnetofield dependence of Hall resistance. The numbers in the diagrams correspond to the sample markings.

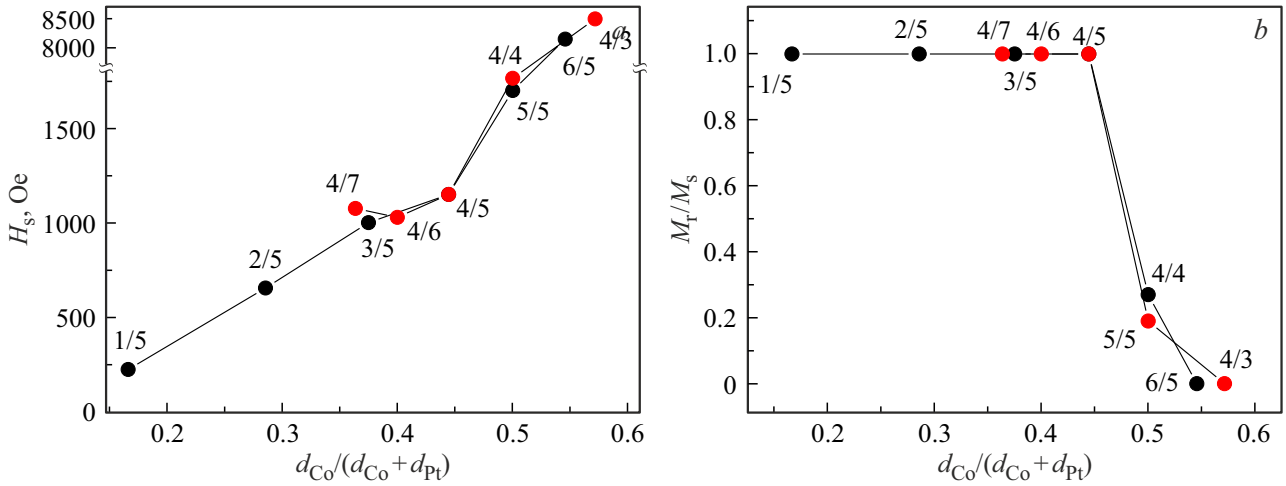

Figure 3. Dependence of saturation magnetic field (a) and ratio of residual resistance to saturation magnetization resistance (b) on relative thickness of Co.

inaccurate. However, because of strong differences in the skyrmion number for different structures, the N_{sk} value seems to be a good quantitative indicator to compare MFM images with each other. So for structure 2/5, the image registers 10 characteristic dark contrast spots on the remagnetized probe area $3.5 \times 3.5\,\text{mkm}^2$, which corresponds to ≈ 0.81 skyrmion at $1\,\text{mkm}^2$ (i. e. $N_{sk} = 0.81\,\text{mkm}^{-2}$).

Increasing the thickness of the Co layer leads to an increase in N_{sk}. The highest skyrmion densities were recorded for structures with relatively large thicknesses

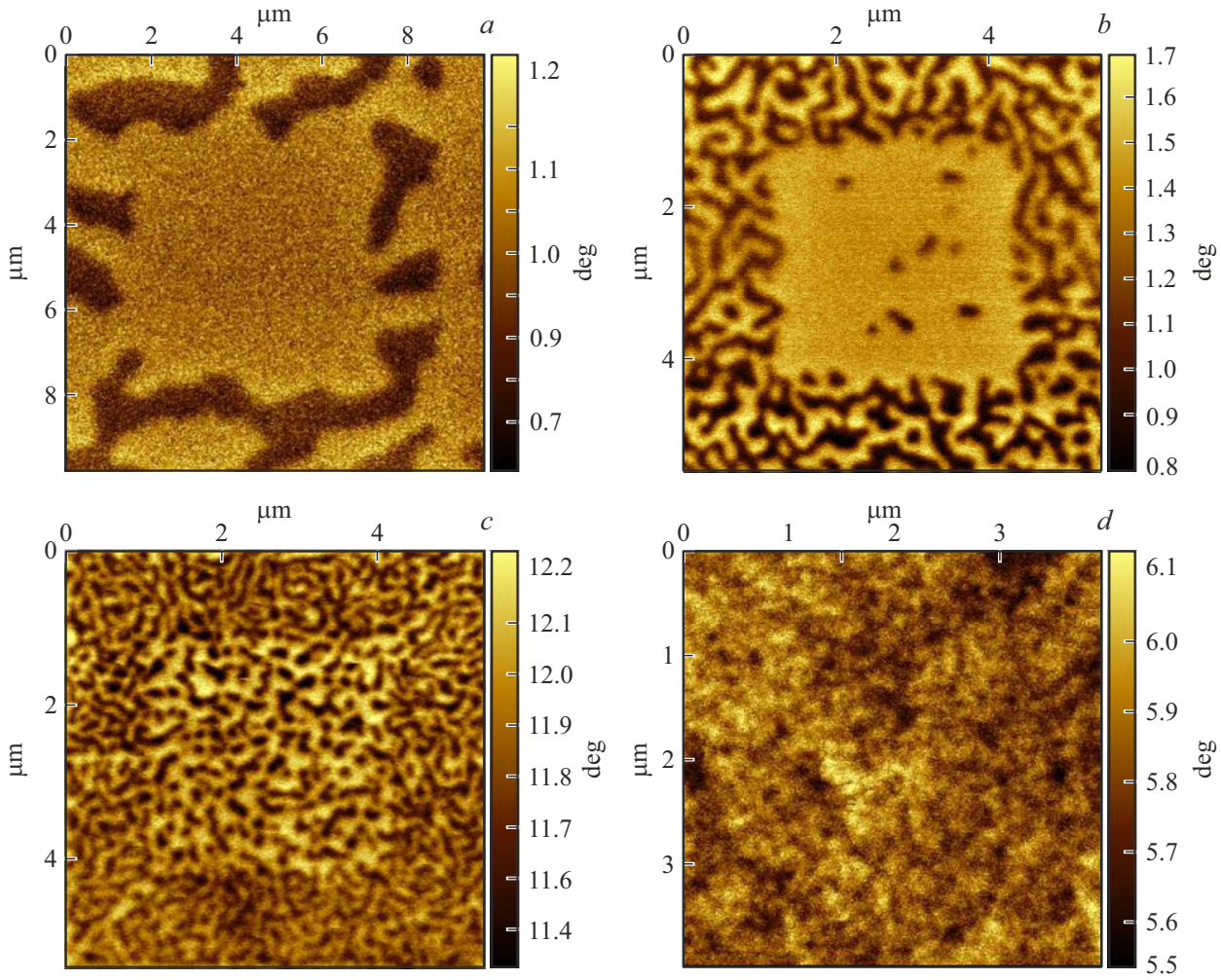

Figure 4. MFM images of the surface of films pre-magnetized by a magnetic force microscope probe. The thicknesses of the Co/Pt layers in each bilayer in Å: *a* — 1/5, *b* — 3/5, *c* — 4/4, *d* — 6/5.

of 4/4 (Fig. 4, *c*) and 4/5 — 128 skyrmions per remagnetized area $3.5 \times 3.5\,\text{mkm}^2$ ($N_{\text{sk}} = 10.5\,\text{mkm}^{-2}$) and 43 skyrmions per remagnetized area $3.5 \times 3.5\,\text{mkm}^2$ ($N_{\text{sk}} = 3.5\,\text{mkm}^{-2}$), respectively.

The structures with 4/4 composition are characterized by a decrease in residual magnetization relative to the saturation magnetization, with the highest observed skyrmion density being recorded in the MFM image. Increasing the layer thickness to 5/5 results in a labyrinth domain structure with no isolated micromagnetic features after probe exposure, i. e. the micromagnetic structure cannot be formed using the magnetization method described in the methodology section. A further increase in thickness of Co leads to a significant transformation of the micromagnetic structure: due to the reversal of the easy magnetization axis in the direction lying in the film plane, a homogeneous magnetic contrast is visible in the image (Fig. 4, *d*).

Let us proceed to discuss the results obtained. Magnetic force microscopy, Hall effect and X-ray analyses have shown the possibility of significant modification of the magnetic properties and micromagnetic structure in multilayer [Co/Pt] thin films. In particularly, the possibility of controlling the skyrmion density within wide limits has been shown. The maximum skyrmion number in these films is given by the composition Co/Pt = 4/4. These films are characterized by a magnetic field dependence of magnetization in the form of a sloping hysteresis loop.

Results of X-ray diffraction analysis suggest that mixing of Co and Pt atoms plays an essential role in multilayered [Co/Pt] magnetic films formed by electron-beam evaporation. Because of the absence of peaks corresponding to atomic cobalt in the X-ray spectrum, we assume that at Co layer thicknesses up to and including 5 Å it is the cobalt that dissolves completely, forming a complex system which may contain both an $CoPt_x$ solid solution and individual CoPt compound phases (for example, Co_3Pt). The exact phase composition cannot be detected due to the strong disorder of the film and the large width of the X-ray diffraction peaks. At large Co (6 Å) thicknesses, platinum does not diffuse to the full depth of Co during growth

and a system of atomic Co and Pt layers with a blurred heterogeneous boundary is formed. The easy magnetization axis of such a system probably lies in the film plane. Similar magnetic properties have been recorded for the structure Co/Pt = 4/3. In this case, the platinum thickness (3 Å) is not sufficient to dissolve all the Co, so a CoPt solid solution with periodically changing composition is formed, in such a system, presumably, there is no perpendicular magnetic anisotropy. No similar dramatic change in properties was observed when the Pt thickness was increased to a value of 7 Å moreover, peaks related to atomic Pt are recorded in a number of X-ray spectra. This leads to the conclusion that the main factor affecting the micromagnetic structure is the complete dissolution of the cobalt by the platinum, while the platinum itself may remain partially undissolved. The platinum is limited to forming the phase composition of $CoPt_x$ solution, and it is phase composition $CoPt_x$ determining the magnetic characteristics of the multilayer structure. The effect of undissolved platinum interlayers on the magnetic characteristics was not detected in the present experiments.

The influence of phase composition $CoPt_x$ on the micromagnetic structure has been studied in paper [12]. The cited article demonstrates a significant influence of the phase composition on the magnetic properties and domain structure in layers $CoPt_x$. For the studied structures, similar mechanisms of magnetic interactions can be assumed, but the high heterogeneity of the composition, which includes interlayers of atomic platinum, should be taken into account. In particularly for certain phase compositions, a micromagnetic structure involving magnetic skyrmions is implemented.

Thus, element distribution profiles, the presence of dissolved and non-dissolved Pt layers and their thickness determine the nature of magnetic interactions in the films and the resulting micromagnetic structure. The practical result, however, is to establish a process method for controlling the micromagnetic structure over a wide range. In particular, it is shown that specifying the thickness of the layers allows the skyrmion density to vary within almost two orders of magnitude: from 0.22 mkm^{-2} to 10.5 mkm^{-2}.

Funding

This study was financially supported by the Russian Science Foundation (project No. 21-79-20186).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] L. Wang, C. Liu, N. Mehmood, G. Han, Y. Wang, X. Xu, C. Feng, Z. Hou, Y. Peng, X. Gao, G. Yu. ACS Appl. Mater. Interfaces **11**, 12098 (2019).
[2] P. Vlaic, E. Burzo. J. Opt. Adv. Mater. **12**, *5*, 1114 (2010).
[3] J.B. Newkirk, R. Smoluchowski, A.H. Geisler, D.L. Martin. J. Appl. Phys. **22**, 290 (1951).
[4] A.W. Rushforth, P.C. Main, B.L. Gallagher, C.H. Marrows, B.J. Hickey, E.D. Dahlberg, P. Eames. J. Appl. Phys. **89**, 7534 (2001).
[5] A.V. Zdoroveyshchev, O.V. Vikhrova, P.B. Demina, M.V. Dorokhin, A.V. Kudrin, A.G. Temiryazev, M.P. Temiryazeva. Int. J. Nanosci. **18**, *3–4*, 1940019 (2019).
[6] A.G. Temiryazev, M.P. Temiryazeva, A.V. Zdoroveishchev, O.V. Vikhrova, M.V. Dorokhin, P.B. Demina, A.V. Kudrin. FTT **60**, *11*, 2158 (2018). (in Russian).
[7] A.G. Temiryazev, A.V. Zdoroveischev, M.P. Temiryazeva. Izv. RAN, Ser. fiz. **87**, *3*, 362 (2023). (In Russian).
[8] J.C. Woolley, J.H. Phillips, J.A. Clark. J. Less-Com. Met. **6**, 461 (1964).
[9] N. Nagaosa, J. Sinova, S. Onoda, A.H. MacDonald, N.P. Ong. Rev. Mod. Phys. **82**, 1539 (2010).
[10] M.V. Dorokhin, A.V. Zdoroveyshchev, M.P. Temiryazeva, A.G. Temiryazev, P.B. Demina, O.V. Vikhrova, A.V. Kudrin, I.L. Kalentyeva, M.V. Ved, A.N. Orlova, V.N. Trushin, A.V. Sadovnikov, D.A. Tatarskiy. J. All. Comp. **926**, 166956 (2022).
[11] S.T. Ruggiero, A. Williams, C.E. Tanner, S. Potashnik, J. Moreland, W.H. Rippard. Appl. Phys. Lett. **82**, 4599 (2003).
[12] Y. Yang, J.S. Chen, G.M. Chow. J. Appl. Phys. **109**, 07B744 (2011).

Translated by Ego Translating