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Formation of skyrmion states in ion-irradiated CoPt thin films

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The possibilities of controlled exposure to ion irradiation (He^+ with an energy of 20 keV and a fluence in the range from $3 \cdot 10^{14}$ to $3 \cdot 10^{15}\,cm^{-2}$) as a method for modifying the magnetic properties and domain structure of $Co_{0.35}Pt_{0.65}$ thin ferromagnetic films have been studied. It was found that the ion irradiation causes a change in the Dzyaloshinskii–Moriya interaction constant and a change in the skyrmion density that correlates with it. This result shows the possibility of homogeneous ion irradiation as a way to control the micromagnetic structure, namely the process of formation of skyrmion states.

Keywords: ferromagnetic thin films, ion irradiation, domain structure, magnetic force microscopy, Dzyaloshinskii-Moriya interaction, skyrmions.

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1. Introduction

Thin multi-layered films and alloys of the ferromagnet/heavy metal type are regarded as prospective carriers for magnetic recording [1–5]. The fundamental „bite“ of magnetic information in these layers is a specific magnetic nanoparticle — magnetic skyrmion. A skyrmion is a topologically protected spin configuration with dimensions ranging from a few nanometers to a tenth of the micron fraction. It is believed that the reason for the formation of skyrmions in thin films is the Dzyaloshinskii–Moriya magnetic interaction (DMI), which manifests itself in structures with strong crystalline or structural and hence magnetic anisotropy [1–7].

Some good examples of such anisotropic structures are thin CoPt films, which in recent years have been investigated by several groups [8–12]. These are highly coercive materials with large magnetocrystalline anisotropy constant. CoPt alloys represent a huge variety of crystalline structures with different types of symmetry, including structures $L1_1$ and $L1_0$ [8]. The existence and coexistence of different magnetic phases dictate different magnetic interactions, which in turn determine a variety of magnetic properties. This diversity is enriched by multi-layered CoPt structures, in which the exchange between Co layers is highly dependent on the thickness parameters of Co and Pt [9–12].

Generalizing the above, we can conclude that the magnetic and micromagnetic properties of multi-layers and CoPt alloys can be controlled in a wide range, varying composition and manufacturing conditions. In this work, ion irradiation was used as a technological reception for controlling the magnetic properties of CoPt films. We have previously demonstrated the influence of ion irradiation on the magnetic properties of $Co_{0.45}Pt_{0.55}$ alloy formed by electron-beam evaporation in high vacuum [13]. Magnetic-force microscopy showed that after irradiation with He^+ ions with 20 keV and $3 \cdot 10^{14}\,cm^{-2}$ fluence in a $Co_{0.45}Pt_{0.55}$ layer, there was an activation of isolated round domains (supposedly magnetic skyrmions), while for $Co_{0.35}Pt_{0.65}$ films irradiated with He^+ with a fluence of $4 \cdot 10^{14}\,cm^{-2}$, besides isolated circular domains, 360-degree domain walls (1D-skyrmions) are observed [14]. At the same time, the study of CoPt films by the Mandelstam–Brillouin spectroscopy showed an increase in the shift between the Stokes and anti-Stokes spectrum components and a significant increase in the Dzyaloshinskii–Moriya interaction in the irradiated samples [14].

This paper presents detailed results on the possibility of controlled radiation effects on the magnetic properties of $Co_{0.35}Pt_{0.65}$ ferromagnetic alloy films produced by electron-beam evaporation. In particular, the range of used helium ion fluence was significantly increased from $3 \cdot 10^{14}$ to

$3 \cdot 10^{15}\,\mathrm{cm}^{-2}$ (compared to work [14]). This allowed to obtain new data on the change of micromagnetic structure due to ion irradiation, see the transition from the formation of 360-degree domain walls to the predominant appearance of isolated round domains, show the correlation between the density of these domains and the Dzyaloshinskii–Moriya interaction constant calculated from the Mandelstam–Brillouin spectroscopy.

2. Experimental technique

Structures with thin CoPt ferromagnetic films were produced by electron beam evaporation of targets from high-purity materials in high vacuum. The formation process was carried out at a temperature of 200°C and included two steps: first, a dielectric layer Al_2O_3 of ~ 1 nm thickness was applied to the substrate of semi-insulating GaAs(100) layer to prevent cobalt diffusion in semiconductor [15], then the layers Pt and Co were alternating with a tenfold repetition: $[\mathrm{Co}(0.2\,\mathrm{nm})/\mathrm{Pt}(0.5\,\mathrm{nm})]_{\times 10}$. Thus CoPt(2/5) metal films with a total thickness of about 7 nm were formed.

The crystalline quality of the as-grown samples of the manufactured structures was investigated by the X-ray diffractometry method at the Bruker D8 Discover installation (equipment of IFM RAS Collective Research Center).

The resulting CoPt(2/5) structures were divided into samples with size 6×6 mm and were uniformly irradiated on the ILU-3 accelerator with He^+ ions with 20 keV energy, with the fluence of ions (F) from $3 \cdot 10^{14}$ to $3 \cdot 10^{15}\,\mathrm{cm}^{-2}$. The choice of energy and the helium ion fluence range was made taking into account the calculation of the distribution of defects by the depth of structures carried out using SRIM software [16] and a calculator to estimate the material sputtering coefficient at ion irradiation [17]. In addition, based on earlier calculations using SRIM software, it has been noted that the ion action with selected parameters may cause significant penetration of cobalt and platinum atoms into the Al_2O_3 dielectric layer, at the same time, oxygen and aluminum do not fall into the CoPt film [14]. In addition, these calculations showed that the Al_2O_3 layer with a thickness of 1 nm at selected ion irradiation conditions effectively prevents the penetration of Co and Pt atoms into the semiconductor part of the structure.

The Faraday magnetooptic effect (the external magnetic field is applied perpendicular to the film surface) at the laser radiation wavelength 980 nm and the Hall effect were studied on the as-grown and irradiated samples (ordinary — when the external magnetic field is perpendicular to the film plane, and planar — when the external magnetic field is in the film plane). Finally, the magnetic relationships of the Faraday angle (Q_F) in the field 0 to ± 4500 Oe, ordinary (R_H) and planar (PHE) Hall resistance in the range ± 2.5 T were analyzed.

The domain structure of the as-grown and irradiated samples was studied by magnetic force microscopy (MFM) with the Smart SPM (AIST-NT) microscope „standard“ technique using a low magnetic moment probe [18]. The study of the domain structure changes leading to the formation of skyrmions was carried out by the PPP-LM-MFMR scan probe (Nanosensors) with the high magnetic moment described in work [19].

The samples were investigated by the Mandelstam–Brillouin spectroscopy (MBS) method, which allows to study the dispersion characteristics of spin waves in magnetics and to evaluate spatially irregular precession of magnetic moments (spin excitations with different wave numbers) in ferro- and ferrimagnetic materials [20] at frequencies in the range from 1 to 1000 GHz. Analysis of shifts of the Stokes and anti-Stokes Mandelstam–Brillouin scattering frequencies allows estimating the energy of the Dzyaloshinskii–Moriya interaction — the main parameter used in the interpretation of the skyrmion states in the magnetic medium [21].

All tests were carried out at room temperature.

3. Results and discussion

The X-ray diffraction studies of CoPt(2/5) layer structures were carried out using two methods. The first is a measurement in the symmetric Bragg–Brentano geometry with a linear positional-sensitive LynxEye detector. The second method — measurement in sliding incidence geometry with a parabolic Goebel mirror on the primary beam at an incidence angle on the sample of 2 degrees and a Soller slit in combination with a point detector to increase the sensitivity to the crystalline phases of thin surface films. It should be noted that in both cases, X-ray diffraction shows the most intense peak (111) of CoPt phase with a face-centric cubic (FCC) lattice near 41 degrees at a diffraction angle of 2Θ. Fig. 1 shows the spectrum of CoPt(2/5) film structure obtained in the symmetrical Bragg–Brentano geometry. The size of the CoPt phase coherence scattering regions by the width of the diffraction peak is 7 nm, which corresponds to the thickness of the film and indicates good mixing of Co and Pt. Bar lines indicate table positions of peaks for different stoichiometry of CoPt alloy, Pt and Co metal layers. The green line shows the position of the peak corresponding to the FCC lattice Pt phase, the gray line — to Co, the red line — to alloy $Co_{25}Pt_{75}$, and the blue line — to alloy $Co_{50}Pt_{50}$. The above data suggest that in the thin CoPt(2/5) layer produced by electron-beam evaporation, the subnanometer layers of Co and Pt are sufficiently well mixed (as evidenced by the absence of peaks in the area of 2Θ for Co and Pt). In addition, a linear approximation of the stoichiometry showed that the CoPt(2/5) films tested corresponded to $Co_{35}Pt_{65}$ alloy.

Characterization of magnetic properties of CoPt (2/5) films and effects of ionic irradiation on them in a sufficiently wide range of fluence of ions — from $3 \cdot 10^{14}$ to $3 \cdot 10^{15}\,\mathrm{cm}^{-2}$ was based on the results of the study

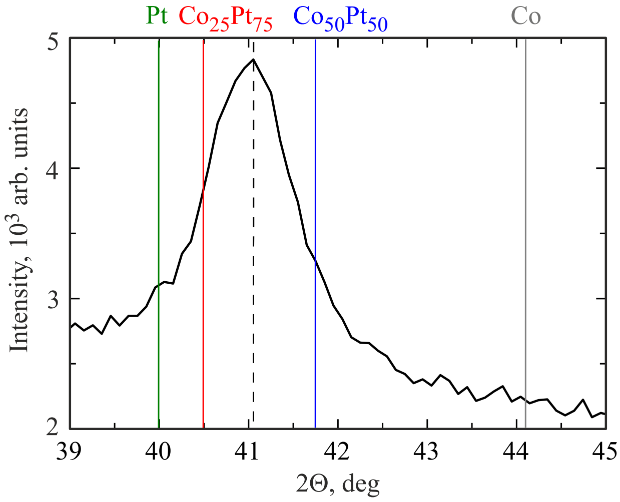

Figure 1. The X-ray diffraction spectrum of the structure with CoPt(2/5) film obtained in the symmetric Bragg–Brentano geometry. Bar lines indicate table positions of peaks for different stoichiometry of CoPt alloy, Pt and Co metal layers. The green line shows the position of the peak corresponding to Pt, the gray line — to Co, the red line — to alloy $Co_{25}Pt_{75}$, and the blue line — to alloy $Co_{50}Pt_{50}$.

of the Faraday magnetooptic effect and the Hall effect. The results of measuring the magneto-field dependencies of the Hall ordinary resistance $R_H(H)$ and planar Hall effect PHE(H), determined by magnetization behavior depending on magnetic field, are given in Fig. 2. The $R_H(H)$ dependency of the as-grown sample contains a hysteresis loop with a coercive field of ~ 700 Oe, with the resistance value in the zero magnetic field being equal to the value in the saturation field, and this type is maintained up to a fluence value of $5 \cdot 10^{14}$ cm^{-2}. At higher values F there is a narrowing of the hysteresis loop and a decrease in the value of R_H in the zero magnetic field, and at maximum helium ion fluence, the nonlinear dependence of $R_H(H))$ goes to saturation in the magnetic field above 7500 Oe (inset in Fig. 2, *a*). At the same time, the measurement of the Hall planar effect shows a monotonic reduction of the saturation field value from 8000 Oe for the as-grown sample to 1500 Oe for the structure after irradiation with $F = 3 \cdot 10^{15}$ cm^{-2} (Fig. 2, *b* and *c*). The observed effects indicate that the residual magnetization decreases with increasing He^+ fluence from $3 \cdot 10^{14}$ to $1 \cdot 10^{15}$ cm^{-2} due to the chosen radiation exposure conditions for CoPt(2/5) films. When ion fluence exceeds $1 \cdot 10^{15}$ cm^{-2} the lateral component of the CoPt alloy light magnetization axis is significantly increased, as measured by the planar Hall effect.

The behavior of the magnetofield dependences of the Faraday angle of the as-grown and helium-irradiated samples is shown in Fig. 3. Here we present the dependences of the coercive field and the ratio of the Faraday angle in the zero magnetic field to the value of the Faraday angle corresponding to the saturation magnetic field on the helium ion fluence. In general, the behavior of these dependences also characterizes a significant decrease in the perpendicular anisotropy of the magnetization vector when irradiation with a fluence greater than $5 \cdot 10^{14}$ cm^{-2} is applied.

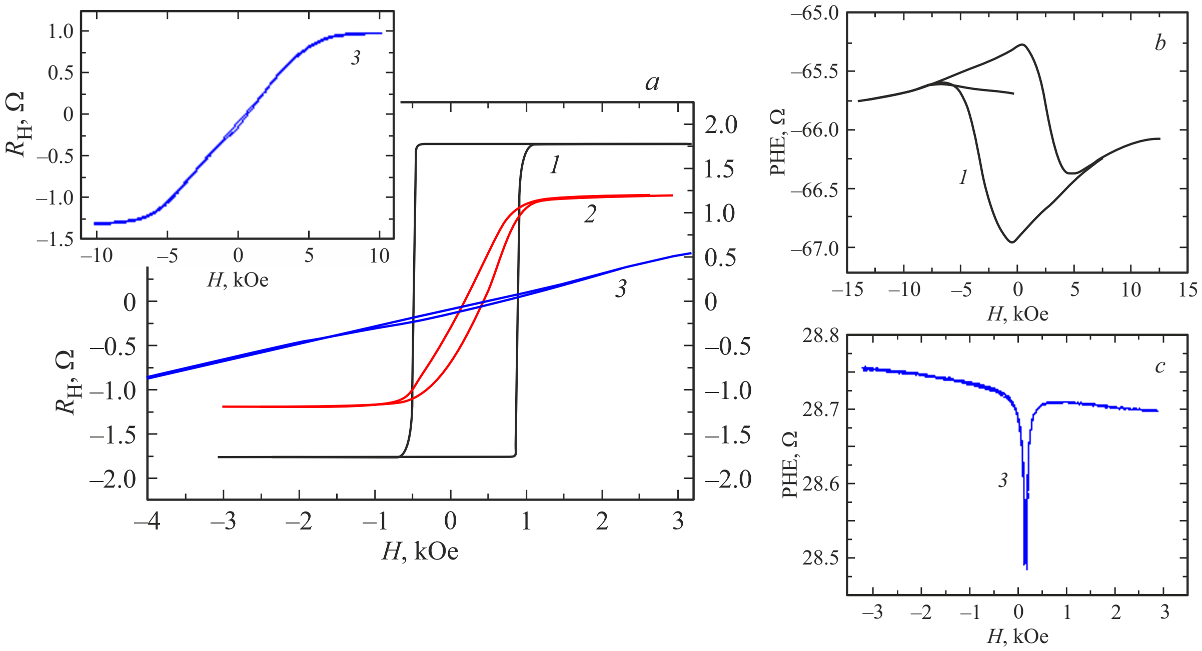

Figure 2. Magnetic field dependences of the Hall resistance: *a* — ordinary; *b* and *c* — planar for the as-grown and ion-irradiated helium with different fluence of CoPt(2/5) film samples. The dependence *1* corresponds to the as-grown sample, the dependence *2* — to the sample irradiated with $F = 7 \cdot 10^{14}$ cm^{-2}, dependence *3* is a sample irradiated with $F = 3 \cdot 10^{15}$ cm^{-2} (on the inset to fragment *a* this dependence is shown in large magnetic fields up to 11000 Oe).

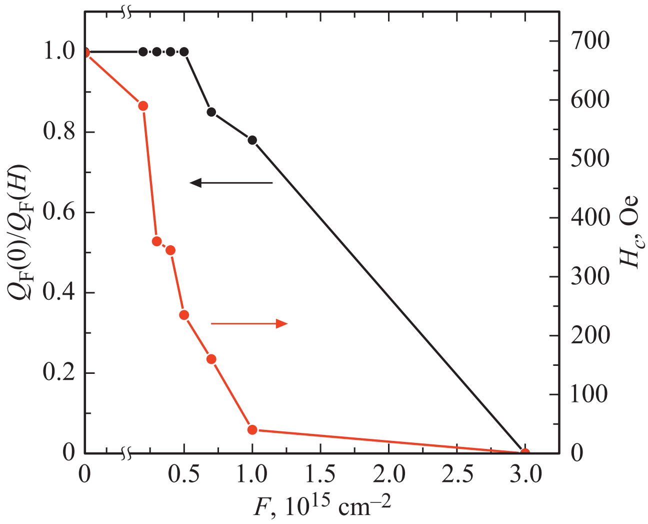

Figure 3. The dependencies of the coercive field H_c and the relative values of the Faraday angle $Q_F(0)/Q_F(H)$ on the helium ion fluence for the CoPt(2/5) structure.

The domain structure of the as-grown and irradiated samples was investigated by the MFM method in the „standard“ two-pass scanning mode of the films. It should be noted that the investigated structures were always in the demagnetized state before the beginning of the MFM measurements. In this case, a probe with extremely low magnetic moment was used, which does not change the magnetic structure. The collected data are presented in Fig. 4. As the result of irradiation with F from $1 \cdot 10^{14}$ to $5 \cdot 10^{14}$ cm^{-2}, no noticeable changes in the labyrinth domain structure are observed. At high fluence values, $7 \cdot 10^{14}$ cm^{-2} and higher, the size of the domains begins to decrease, indicating a decrease in the domain wall energy, which may also be due to a decrease in the orthogonal anisotropy of the magnetization vector due to ion irradiation.

Scanning the films with the Nanosensor probe, which has a high value of magnetic moment, leads to changes in the domain structure of the original and irradiated samples

Parameters of the as-grown and irradiated CoPt(2/5) films (shift between the Stokes and anti-Stokes spectrum components, Δ, and the DMI constant, D) obtained by the Mandelstam–Brillouin spectroscopy

F, 10^{14} cm^{-2}	0	3	4	5	7	10
Δ, MHz	214	220	431	527	527	60
D, mJ/m^2	0.278	0.285	0.560	0.685	0.681	0.078

and the formation of skyrmion states (Fig. 5). First, the $5 \times 5\,\mu$m^2 region is scanned in two passes, and the film is partially remagnetized by the probe in the scanning region. Then, in one pass at a distance of about 100 nm above the surface, a larger region of $10 \times 10\,\mu$m^2 is scanned. For the as-grown CoPt(2/5) film, 1D-skyrmions (360-degree domain walls with widths ~ 150 nm) are observed in the MFM image. For irradiated CoPt(2/5) films, a transition to circular skyrmions (small cylindrical isolated domains with „reverse“ magnetization — dark objects) is observed, and at $F = 3 \cdot 10^{14}$ cm^{-2} and more an increase in their number is recorded. At a fluence of $7 \cdot 10^{14}$ cm^{-2} a very dense lattice of individual circular skyrmions is observed (a more detailed image is also shown in Fig. 5). In the case of a helium ion fluence of $1 \cdot 10^{15}$ cm^{-2} or higher, skyrmions are not observed (fragments f and g of Fig. 5). To quantitatively interpret the micromagnetic structure, the parameter n_{sk} — skyrmion densities was introduced. This parameter is understood as the number of skyrmions per unit area ($1\,\mu$m^2) and is calculated by dividing the total number of skyrmions on the MFM scan by the area of this scan.

It is suggested that the mechanism of formation of 1D-skyrmions and small circular isolated domains may be related to the interaction between the probe-moved domain wall and the as-grown domain structure of CoPt films [22]. Studies by the Mandelstam–Brillouin spectroscopy revealed the presence of a shift between the Stokes and anti-Stokes spectral components (Δ) in the as-grown and irradiated samples of CoPt(2/5) films (table).

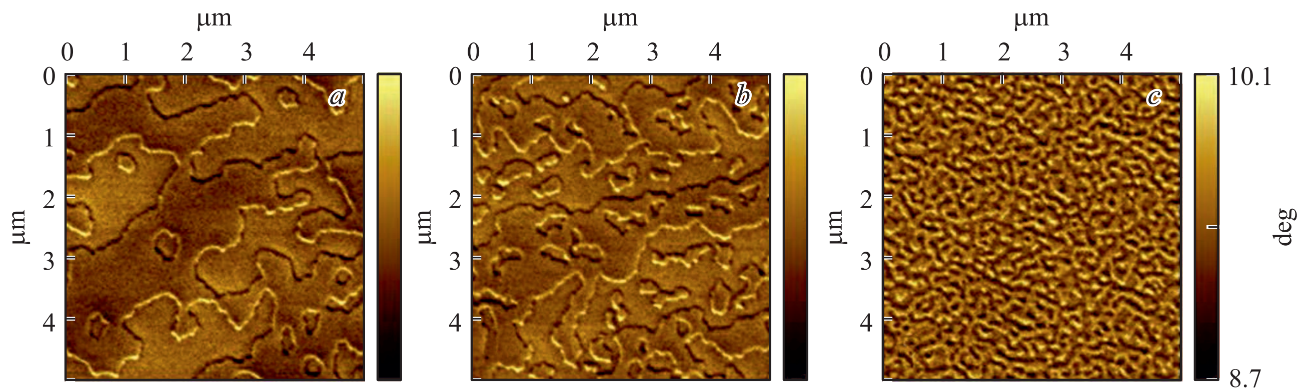

Figure 4. MFM images of the as-grown and irradiated with helium ions with different fluences, cm^{-2}: a — 0, b — $7 \cdot 10^{14}$, c — $1 \cdot 10^{15}$ CoPt(2/5) films obtained by low magnetic moment probe scanning.

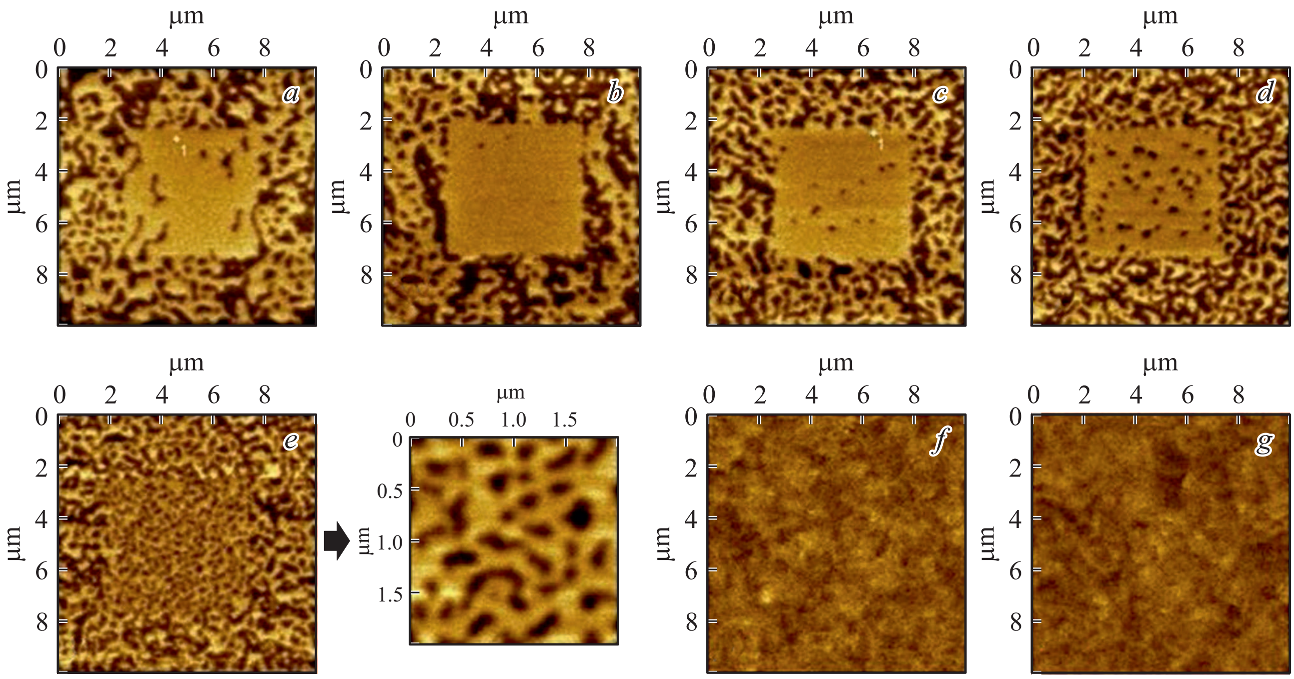

Figure 5. MFM images of the as-grown and irradiated with different fluence, cm^{-2}: *a* — 0, *b* — $3 \cdot 10^{14}$, *c* — $4 \cdot 10^{14}$, *d* — $5 \cdot 10^{14}$, *e* — $7 \cdot 10^{14}$ (the inset shows a more detailed image of the central region), *f* — $1 \cdot 10^{15}$, *g* — $3 \cdot 10^{15}$ CoPt(2/5) films obtained by high magnetic moment Nanosensor probe scanning (domain structure MFM creation).

The registered shift in absolute value did not change when the polarity of the magnetic field was changed. The obtained data make it possible to estimate the Dzyaloshinskii–Moriya interaction constant (D) in the initial and irradiated CoPt(2/5) films (table) and to analyze the effect of ion irradiation on its value and the number of formed skyrmions (Fig. 6). In the calculations, we used the value of the CoPt saturation magnetization, which was estimated earlier on a calibrated magnetometer and was about $1.1 \cdot 10^6$ A/m. For irradiated CoPt(2/5) layers, the DMI constant increases for a range of changes in He^+ ion fluence from 0 to $(5{-}7) \cdot 10^{14}\,cm^{-2}$ and reaches a maximum value of about $0.685\,mJ/m^2$. At higher ion fluence, the value of D drops dramatically. The dependence of the skyrmion density on the ion fluence behaves similarly (curve *2* in Fig. 6). Thus, Fig. 6 shows a perfectly clear correlation between the skyrmion density and the value of the DMI constant calculated from the measurements. Such a correlation testifies to the determining role of the Dzyaloshinskii–Moriya interaction in the formation of 360-degree domain walls and small cylindrical isolated domains with „reverse“ magnetization, which can be interpreted as skyrmions. Apparently, the Dzyaloshinskii–Moriya interaction is present in the as-grown samples of CoPt(2/5) films and is enhanced by ion irradiation.

As was shown earlier [14] on the basis of model calculations by means of SRIM software, the applied ion irradiation promotes asymmetric mixing of Co and Pt atoms in CoPt films, and this may underlie the mechanism of the observed effects of ion irradiation on their magnetic properties and domain structure. In particular, the perpendicular anisotropy decreases and the formation of skyrmion states increases. The formation of skyrmions depends on the domain wall energy, which, in turn, is influenced by the Dzyaloshinskii–Moriya interaction constant [23]. As the DMI constant increases, the domain wall energy decreases, and optimal conditions for the formation of skyrmion-like

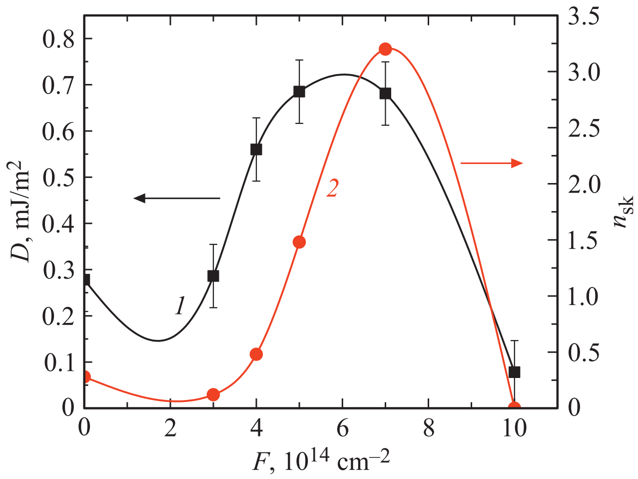

Figure 6. Dependences of the Dzyaloshinskii–Moriya interaction constant calculated from the results of studies of the as-grown and irradiated films by the Mandelstam–Brillouin spectroscopy (*1*), and the skyrmion density (*2*) on the ion fluence for CoPt(2/5) films.

domain structures under the influence of the local magnetic probe field arise.

4. Conclusion

This work demonstrates the use of uniform irradiation by He^+ ions with an energy of 20 keV in a wide range of fluence changes (from $3 \cdot 10^{14}$ to $3 \cdot 10^{15}\,cm^{-2}$) as a way to modify the magnetic properties and micromagnetic structure of CoPt(2/5) films formed by electron-beam evaporation in high vacuum. Comprehensive studies of the structural and magnetic properties of the as-grown and irradiated samples were carried out and their domain structure was investigated. A decrease in perpendicular anisotropy and activation of the correlated process of skyrmion state formation due to the observed strengthening of the Dzyaloshinskii–Moriya interaction in the irradiated samples were shown. This effect can be associated with a decrease in the domain wall energy due to anisotropic mixing of Co and Pt atoms under the action of ion irradiation, which leads to an increase in the Dzyaloshinskii–Moriya interaction constant and the appearance of optimal conditions for the formation of skyrmion-like domain structures under the influence of the local magnetic probe field.

We emphasize that ion irradiation is thus a new tool for controlling the type of skyrmion states and their density in thin-film magnetic structures.

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Measurements of the magnetization of CoPt films on a calibrated magnetometer were made by A.I. Gumarov at Kazan Federal University.

Conflict of interest

The authors declare that they have no conflict of interest.

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