

Modification of the Interfacial Dzyaloshinskii–Moriya Interaction in Cobalt/Heavy Metal Films Irradiated with Helium Ions

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Abstract—The effect of irradiation with helium ions of thin cobalt films on a layer of heavy metals (Pt and W) on the interfacial Dzyaloshinskii–Moriya interaction (iDMI) is studied experimentally. The iDMI value is measured via Brillouin light scattering (BLS). The hysteresis loops of the samples are measured via optical magnetometry. The iDMI value varies nonmonotonically in the range of 0.1–0.6 J m⁻² at fluences of 10¹⁴–10¹⁶ cm⁻².

Keywords: ferromagnetic films, interfacial Dzyaloshinskii–Moriya interaction, perpendicular anisotropy

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INTRODUCTION

In fact, topological soliton solutions for the magnetization distribution have been known since the late 1970s [1, 2]. However, scientists became seriously interested in them only after they discovered similar states in chiral magnetic materials without an inversion center [3–5]. Since that time, they have been called magnetic skyrmions [6]. The disadvantage of chiral magnets is that the interfacial Dzyaloshinskii–Moriya interaction (iDMI), which stabilizes magnetic skyrmions, is usually weak in them. Therefore, skyrmions can be stable only in a narrow range of magnetic fields and low temperatures [7]. The symmetry of spatial inversion in ultrathin multilayer films of ferromagnetic and heavy metals is violated at the interface between the layers, which leads to a strong iDMI [8], which stabilizes magnetic skyrmions at room temperature. To date, chiral magnetic structures caused by iDMI have been experimentally observed in Mn/W films [9], Fe/Ir films [10], and Co/Pt films [11]. Despite the fact that the stability of magnetic skyrmions may be due to iDMI, it is necessary to find out the ways of their nucleation in a system that is initially in a homogeneous state. One of the ways is to pass current through geometrically inhomogeneous bridges [12]. Another way is the magnetization reversal of a nanostructured magnetic film in a uniform magnetic field [13]. In particular, such nanostructuring can be pro-

vided by local irradiation of the film with a beam of helium ions [14, 15], which can lead to the stabilization of the skyrmion in the region with a reduced magnetic anisotropy even when there is no iDMI [16].

It is well known that the anisotropy coefficient of magnetic multilayer Co/Pt films can be changed by irradiation with helium ions [17, 18]. When the fluences are small, the magnitude of the perpendicular anisotropy decreases; when the fluences are large, the film can acquire planar anisotropy. This change in anisotropy is due to the mixing of atoms at the interface of two metals. The value of iDMI is due to the same boundary effects at the interface between a ferromagnetic metal and a heavy metal with a strong spin-orbit interaction, as is the magnitude of the perpendicular magnetic anisotropy. Accordingly, it should also change when the films are irradiated with helium ions. The effect of boundary irregularities on the iDMI value was obtained both theoretically (based on ab initio calculations) [19] and experimentally for three-layer Ta/CoFeB/MgO structures [20] and CoPt alloy films [21] irradiated with helium ions. Another alternative way of changing the mutual arrangement of atoms at the interface between magnetic and heavy metals is mechanical deformation (compression or tension) of the sample. Recently, it was demonstrated that such deformations lead to a significant change in the value of iDMI [22].

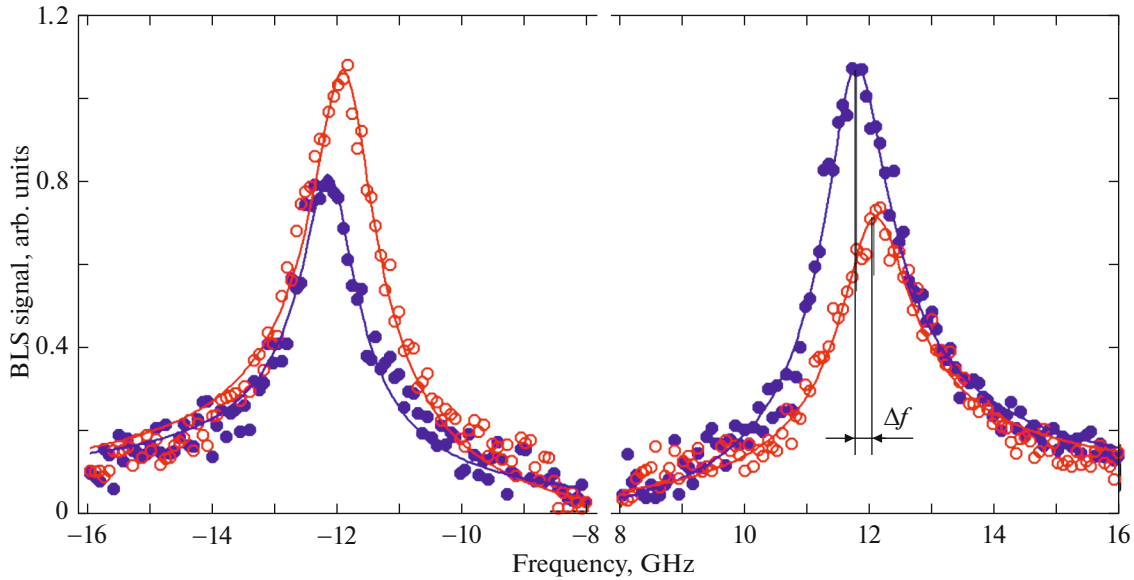


Fig. 1. BLS spectra of a cobalt film on a heavy metal substrate (the data correspond to an unirradiated Co/Pt film) at H of 1 T (red circles) and H of -1 T (blue circles). Solid lines correspond to the Lorentz approximation of the experimental data. Δf is frequency shift between the Stokes and anti-Stokes peaks (for the color picture, see electronic version of the journal).

In this work, we study the effect of irradiation with helium ions of cobalt films on the substrate of heavy metals (Pt and W), which demonstrate a strong spin-orbit interaction, on the magnetization, anisotropy, coercive properties, and iDMI value. It is shown that the value of iDMI changes nonmonotonically when the fluence is 10^{14} – 10^{16} cm^{-2} , while the value of magnetization remains unchanged. In addition, Co/Pt films exhibit a well-known change in the anisotropy from the easy-axis anisotropy to the planar anisotropy at fluences of $\sim 3 \times 10^{15}$ cm^{-2} .

EXPERIMENTAL

Magnetic films were grown by magnetron sputtering in argon (pressure is 4×10^{-3} Torr) on a 22×22 mm^2 glass substrate with a tantalum buffer layer (10 nm). Then, a layer of heavy metal (Pt or W) with a thickness of 10 nm and a cobalt layer with a thickness of 1 nm were deposited. The thickness of the cobalt film for the preparation of the samples was chosen as thin as possible, since iDMI appears on the surface and its effect on the magnetic properties of the sample decreases with an increase in the thickness of the magnetic film. The layer growth rate is 0.125 nm s^{-1} for Co and 0.25 nm s^{-1} for Pt. The magnetic layer was covered with a 5 nm thick layer of light metal (Al) to prevent oxidation. Subsequently, each sample was divided into eight parts with a size of $\sim 5 \times 10$ mm^2 , and these parts were uniformly irradiated with He^+ ions. Irradiation with He^+ ions was carried out using an ILU-3 ion-beam accelerator with an energy of 30 keV at ion fluence values of 10^{14} , 3×10^{14} , 6×10^{14} , 10^{15} , 3×10^{14} ,

and 10^{16} cm^{-2} . This sample preparation technique ensures that samples with different fluences will have the same initial structure. The unirradiated portion of the sample used as a reference sample. The energy of helium ions of 30 keV was chosen due to the fact that this energy level is standardly the maximum energy level of helium ions in commercially available FIB lithography devices (focused ion beams) and allows achieving the best focusing, and hence, lithographic resolution [14, 15, 23]. The penetration depth is hundreds of microns. Consequently, helium ions are not implanted into thin-film metal structures, the ions accumulate deeply in the substrate. The ILU-3 ion-beam accelerator was developed by the NRC “Kurchatov Institute” in the 1960s; this is an accelerator of medium-mass ions with magnetic separation. After leaving the magnetic field, a vertical ribbon-type ion beam hits the collector device and is scanned horizontally with a high voltage at a frequency of 80 Hz with an amplitude that prevents medium-mass ions from entering the rectangular hole in the diaphragm. The magnitude of the fluence is regulated both by the density of the ion beam and by the exposure time, which was on the order of tens of minutes.

The hysteresis loops of the films were studied by measuring the magneto-optic Kerr effect (MOKE) in polar and meridional geometry using the method of crossed polarizers. A highly stabilized He–Ne laser ($\lambda = 632$ nm) was used as a radiation source. The light incident on the sample was p polarized. Unfortunately, the small thickness of the cobalt film and, accordingly,

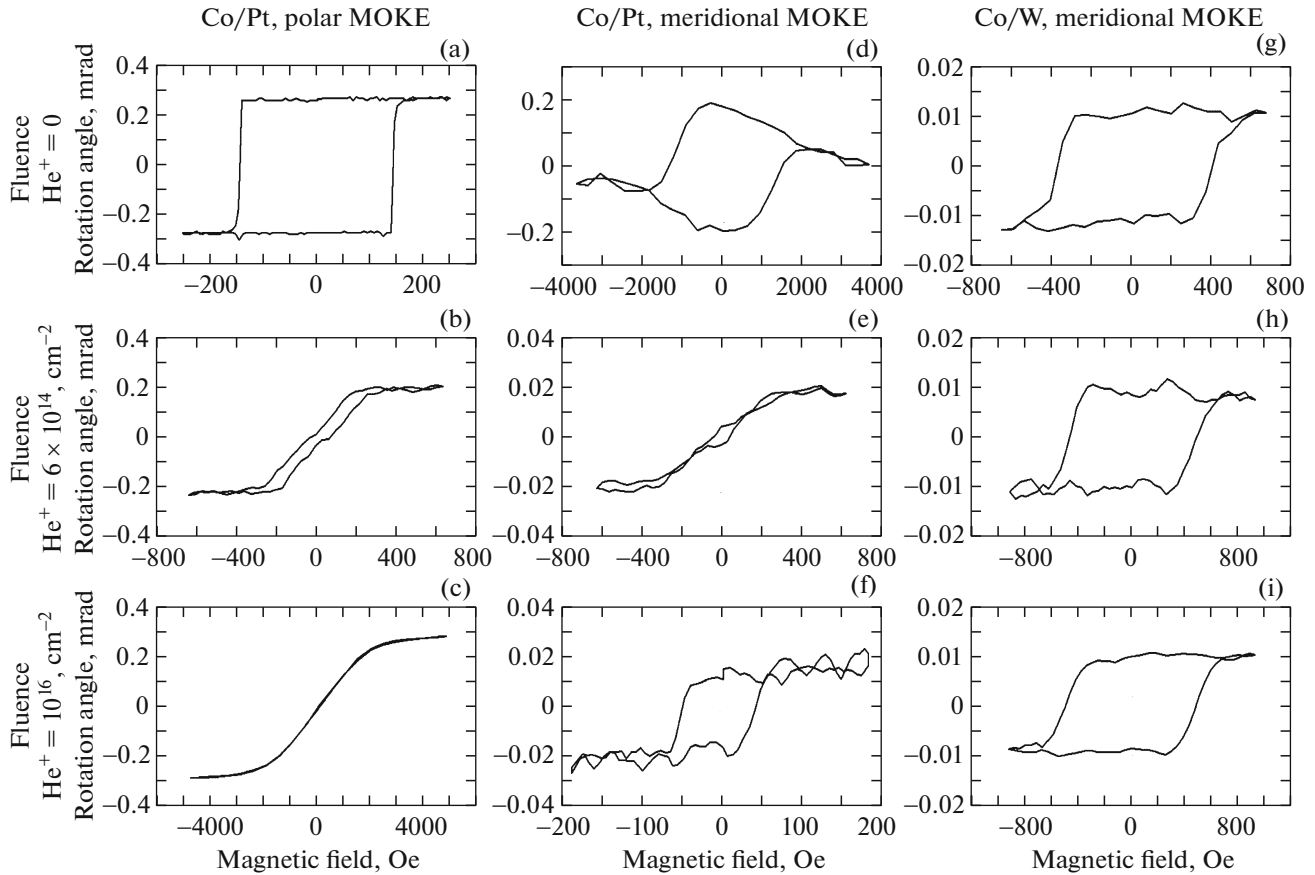


Fig. 2. Typical form of MOKE hysteresis for the samples. (a–c) Polar MOKE in Co/Pt; (d–f) meridional MOKE in Co/Pt; and (g, h, i) meridional MOKE in Co/W. (a, d, g) Data for unirradiated samples; (b, e, h) data for the samples with fluence of $6 \times 10^{14} \text{ cm}^{-2}$; and (c, f, i) data for the samples with fluence of 10^{16} cm^{-2} . The sign of the anisotropy of the Co/Pt film changes with increasing fluence. When magneto-optical rotation is measured in meridional form for a magnetic film with perpendicular anisotropy (unirradiated Co/Pt film), both the meridional MOKE and polar MOKE contribute to the measured effect.

the weak leakage fields did not allow measurements of the domain structures formed during the magnetization reversal via magnetic force microscopy, although such attempts were made by us.

To measure the iDMI value, we used the Brillouin light scattering (BLS) in the Damon–Eshbach geometry [24]. A typical BLS spectrum is shown in Fig. 1. Solid lines correspond to the Lorentz approximation; the shift of the Stokes peak and anti-Stokes peak is denoted as Δf . According to the standard approach, the iDMI constant is calculated as [22, 24]

$$D = 2M_s \Delta f / (\pi \gamma k), \quad (1)$$

where M_s is saturation magnetization; k is a wave-number; and $\gamma = 176 \text{ GHz T}^{-1}$ is a gyromagnetic ratio. The M_s value that we used in our calculations is $1.1 \times 10^6 \text{ A m}^{-1}$, which is a typical value for cobalt thin films [25, 26]. A typical BLS spectrum is shown in Fig. 1 (it corresponds to the original Co/Pt structure).

RESULTS AND DISCUSSION

The hysteresis loops obtained by magneto-optical magnetometry is shown in Fig. 2. The original Co/Pt films exhibit easy-axis anisotropy. This can be seen from the characteristic hysteresis loop in a perpendicular field with remanent magnetization equal to the saturation magnetization (Fig. 2a). As the fluence increases, the coercive field gradually decreases, while the remanent magnetization remains equal to the saturation magnetization up to a fluence of $3 \times 10^{14} \text{ cm}^{-2}$ (Fig. 3). In this case, the hysteresis loop in the longitudinal field has a characteristic complex form when the residual signal exceeds the signal in saturation (Fig. 2d). This waveform is explained as follows. Measurements in the meridional geometry are carried out at an incidence angle of 45° . In this case, the wave vector of the electromagnetic wave has two components (both a component lying in the plane of the sample and a component perpendicular to it). Accordingly, both the polar effect (since there is remanent magnetization perpendicular to the surface) and meridional

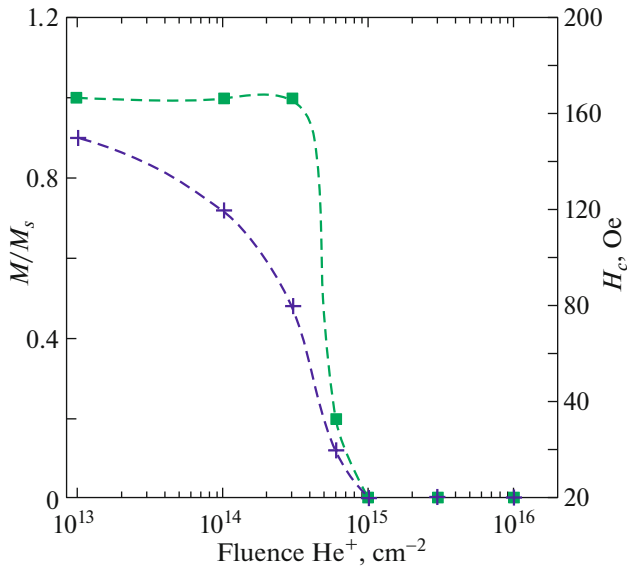


Fig. 3. The dependence of the remanent magnetization (green squares) and the coercive electric field (blue crosses) on the He^+ ion fluence when the Co/Pt film is magnetized in a magnetic field perpendicular to the sample surface (for the color picture, see electronic version of the journal).

effect (since the saturation magnetization is in the plane of the sample) contribute to the measured effect. Thus, the curve is the sum of the magnetization curve in the perpendicular direction and in the longitudinal direction. At $6 \times 10^{14} \text{ cm}^{-2}$ (Fig. 2b), the shape of the magnetization curve changes: the remanent magnetization M_z decreases to 20%, while the remanent magnetization upon magnetization in the plane (Fig. 2e) vanishes. This behavior indicates that although the easy-axis anisotropy remains, its value decreases so much that the uniformly magnetized state in zero field becomes unstable and the system breaks up into domains. As fluence increases (10^{15} – 10^{16} cm^{-2}), the remanent magnetization M_z vanishes (Figs. 2c and 3), and there is a characteristic hysteresis with the remanent magnetization equal to the saturation magnetization upon magnetization in the plane. This behavior clearly indicates that the easy-axis anisotropy transforms into the planar anisotropy.

The change in the type of anisotropy of Co/Pt films irradiated with ions was well known even earlier, but we would like to draw attention to the following important experimental fact. Regardless of the fluence, the magnitude of the magneto-optical rotation at saturation remains unchanged and is 0.2 rad in polar geometry and 0.02 rad in meridional geometry. This indicates that the magnetization of the films does not change in the fluence range of 10^{14} – 10^{16} cm^{-2} . This is important for the subsequent calculation of the iDMI value.

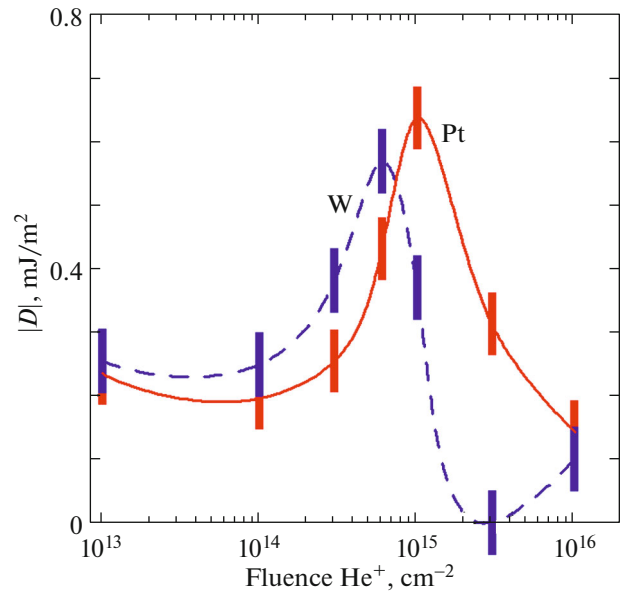


Fig. 4. The dependence of the absolute value of the iDMI coefficient of Co films on a sublayer of heavy metals Pt (solid red line) and W (blue dashed line) on the fluence of helium ions (for the color picture, see electronic version of the journal).

As for the Co/W structures, they initially exhibit a planar anisotropy, and the shape of the hysteresis loop and the remanent magnetization almost independent of the He^+ fluence in the range under study (Figs. 2g–2i).

The calculated iDMI values of Co/Pt films and Co/W films are shown in Fig. 4. It should be noted that if the iDMI constant of Co/Pt film is positive, then the iDMI constant of Co/W film is negative. Nevertheless, absolute values of the constants depending on the fluence are shown in Fig. 4 for comparison. The iDMI value is calculated by formula (1). Since the magnetization does not depend on the fluence, the change in the shifts of the Stokes peak and anti-Stokes peak (Δf) observed in the BLS is due precisely to the change in the iDMI constant. The nonmonotonic dependence of D on the fluence in Co/Pt films exhibits a maximum at fluences of 10^{15} cm^{-2} , while the iDMI constant increases by a factor of three compared to unirradiated films. A further increase in fluence leads to a rapid decrease in iDMI value. A similar effect is observed for Co/W films with the only difference that the peak of the iDMI value corresponds to slightly lower fluences ($6 \times 10^{14} \text{ cm}^{-2}$).

The change in the iDMI value is apparently associated with a change in the structure of the interfilm interface, but this issue requires additional study. Note that a similar nonmonotonic change in the iDMI constant of the Ta/CoFeB/MgO structure irradiated with ions was reported in [20]. In this case, the maximum iDMI value corresponded to a fluence of $1.2 \times 10^{15} \text{ cm}^{-2}$, which is in good agreement with our results.

At the same time, the maximum iDMI value increased threefold in comparison with unirradiated control samples, which also quantitatively corresponds to our results.

CONCLUSIONS

The possibility of modifying the interfacial Dzyaloshinskii–Moriya interaction in cobalt films on the substrate of a heavy metal (Pt and W) under irradiation with 30 keV helium ions is demonstrated. It is found that the iDMI constant increases at fluences of $\sim 6 \times 10^{14} - 10^{15} \text{ cm}^{-2}$, which should lead to a decrease in the energy of the domain wall and facilitate the formation of magnetic skyrmions. Local modification of magnetic films with ion beams can serve as a tool for the formation of magnetic nanostructures for skyrmion spintronics.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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