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Abstract. Nonlinear magneto-optical Kerr is used to study the magnetic properties of thin bilayer films composed of cobalt and two different non-magnetic metals, platinum and tantal. Our experiments reveal different nonlinear-optical response of these two types of structures related to specific magnetic properties of Co/Ta and Co/Pt interfaces.

1. Introduction

Thin metallic ferromagnetic films and nanostructures are a subject of intensive studies for a long time [1]. High perspectives for applications of such structures in various types of magnetic, magneto-optical and spintronic devices are well recognized, while their development and successive applications is impossible without the full understanding of the underlying fundamental physical effects. One of the most hot topics here is the role of interfaces in operation of magnetic nanostructures, which requires the search of new effective diagnostic tools. Among others, the technique of optical second harmonic generation (SHG) is known due to its high sensitivity to the properties of nanostructures and interfaces [2, 3]. Moreover, this sensitivity is further improved in the case of magnetic interfaces as the typical values of magneto-optical effects are at least one order of magnitude larger as compared to their linear optical analogues [4, 5].

It was demonstrated that magneto-optical effects in SHG allow to study antiferromagnetic and even vortex magnetization in different types of nanostructures [6, 7], while in some cases second-order in magnetization effects are of principal importance for the formation of the SHG response [8]. In this work, we study the magnetic field induced effects in optical second harmonic generation that appear in planar structures composed of a heavy (Pt, Ta) and a ferromagnetic (Co) metals. The main idea is to visualize the effects of specific interface interactions induced

by the composition of the structures, including possible formation of chiral magnetization distribution at metallic interfaces of different composition.

2. Experimental details

The structures under study are bilayer films of the composition Co(20nm)/Pt(3nm) and Co(20nm)/Ta(3nm). The choice of the nonmagnetic metals is justified by their high and different values of spin-orbital constants. Polycrystalline metal films on silicon or glass substrates were fabricated using a high vacuum magnetron AJA 2200 multichamber system at the basic pressure of 10^{-5} Pa. Some of the structures were grown in a constant in-plane dc magnetic field in order to introduce magnetization easy axis. At the same time, the films reveal isotropic in-plane structure.

Magnetic properties of the samples were studied using the vibrational magnetometry and magneto-optical (MO) methods in the geometry of the longitudinal MO Kerr effect (MOKE). Both methods confirm the existence of the in-plane magnetization easy axis; the typical saturating magnetic field values being 60÷80 Oe.

Brillouin light scattering (BLS) technique was used for the determination of the value of the Dzyaloshinskii Moriya interaction (DMI) specific for each of the two samples described above. Radiation of the CW semiconductor laser at 532 nm wavelength was used as the probe radiation. We use BLS spectroscopy in backscattering configuration to investigate the nonreciprocal propagation of spin waves in the Damon-Eshbach geometry. The sample was in-plane magnetized and the wavevector of spin waves is perpendicular to the equilibrium magnetization direction. The shift between the peaks of Stokes and anti-Stokes components in BLS spectrum at a given value of wavelength is directly proportional to the DMI value. The corresponding procedure is described in detail elsewhere [9]. The performed measurements show that Co(20nm)/Pt(3nm) film reveals the higher value of the DMI constant as compared to Ta-containing one, in accordance with the results reported by other researchers [10].

For the nonlinear optical studies we used the Ti:sapphire laser as the source of the fundamental beam (820 nm wavelength, pulse duration of 80 fs, repetition frequency of 80 MHz, mean power of 50 mW). The laser radiation was focused on the sample into a spot of approximately 50 μm in diameter, the angle of incidence being 45°. The SHG radiation reflected from the samples was spectrally selected and detected by a photomultiplier operating in the photon counting mode. Nonlinear MOKE was studied as the longitudinal magnetic field was applied to the samples (see Fig. 1). In order to reveal the effect of magnetic anisotropy on the appearance of the nonlinear magneto-optical effects, the SHG experiments were carried out for various orientations of the magnetization easy axis with respect to the longitudinal magnetic field.

Atomic force microscopy was used for the studies of surface morphology of the studied structures, while magnetic microscopic images of the samples were obtained by magnetic force microscope (SmartSPM (AIST-NT)) over the area of 10μm$\times10\mu$m.

3. Results and discussion

Atomic force microscopy image of Co/Pt bilayer films shown in figure 1, a reveals that the roughness of the surface of the heavy metal is rather small and does not contain surface defects. On the contrary, magnetic force microscopy indicates inhomogeneous distribution of magnetization, which can not be associated with surface defects.

Taking into account the in-plane magnetic anisotropy of the composed bilayered films, the SHG studies were performed for various azimuthal orientations of the *dc* longitudinal magnetic field, **H**, with respect to it. As an example, Fig. 2a shows the dependence of the SHG intensity on magnetic field which reveals the hysteresis loop, the dc field and the easy axis of the Co/Pt bilayer film being perpendicular to each other; p-polarized fundamental beam was used and the
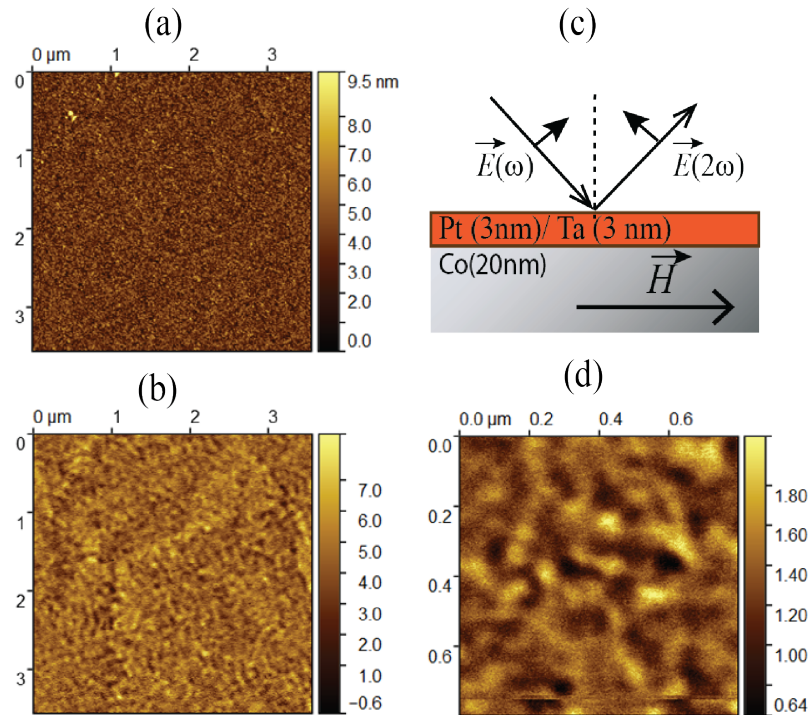
Figure 1. (a) atomic force microscopy image and (b,d) magnetic force microscopy images of Co/Pt bilayer film on different scales; (c) scheme of the second harmonic generation experiment with Co/(Pt,Ta) bilayer films; the *dc* magnetic field $\vec{H}$ is applied in the longitudinal geometry.

analyzer in the SHG registration channel was oriented in between p- and s-polarizations. Thus the SHG polarization plane rotation can be measured through the changes of the SHG intensity in such a scheme.

It can be seen from the figure 2,a that modulation of the SHG intensity under the reversal of the external magnetic field is about 35%, which corresponds to the maximal SHG polarization plane rotation to dozens of degrees counted out of the incident p-polarization of the fundamental beam. This exceeds the linear magneto-optical effect by nearly two orders of magnitude and is typical for the second-order nonlinear magneto-optical effects in ferromagnetic films. Moreover, the saturation of the SHG signal happens for relatively high values of the external magnetic field, H¿200 Oe. This exceeds similar values obtained by linear magneto-optical Kerr effect and magnetometry measurements, which characterize predominantly the magnetic properties of bulky cobalt film. This discrepancy should be attributed to different location of the second harmonic sources as compared to those producing the linear magneto-optical response, as SHG arises mostly at the interfaces between the ferromagnetic and heavy metals. Higher saturation of interfaces has been reported previously for a number of ferromagnetic metals [11].

In order to study chiral magnetization states at interfaces between a heavy (Pt,Ta) and a ferromagnetic metal (Co), we had to get rid of common magneto-optical Kerr effect, so we studied the magnetic-field induced effects in p-polarized SHG in the geometry of the longitudinal MOKE. According to symmetry arguments [5], the p-polarized SHG radiation is not affected by the longitudinal magnetic field, while may reveal chiral magnetic ordering at the interfaces, as was suggested in [12].

Figure 2,b,c present the dependencies of the SHG intensity on the longitudinal magnetic field for the p-in (ω), p-out (2ω) polarization combination for the Co(20nm)/Pt(3nm) film oriented
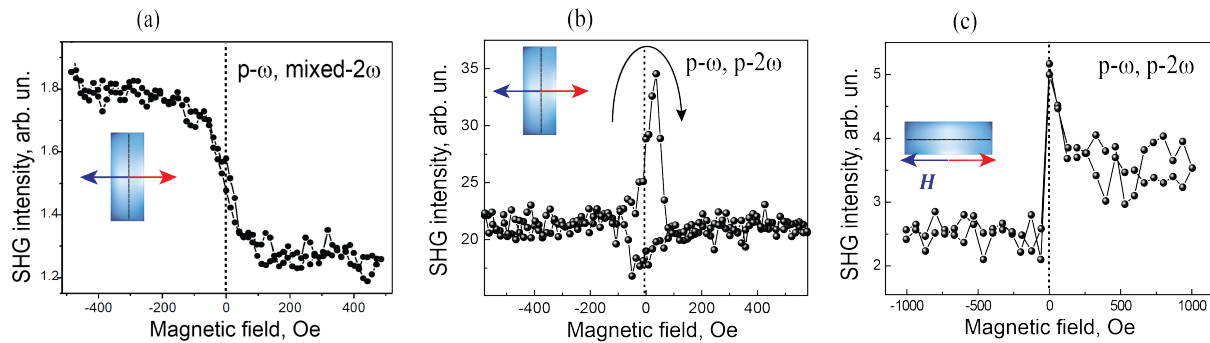
Figure 2. (a) Nonlinear magneto-optical longitudinal Kerr effect at the SHG wavelength for the Co/Pt film, the easy axis is perpendicular to the magnetic field; dependencies of the p-polarized SHG intensity on the longitudinal magnetic field in Co/Pt film with the easy axis (b) perpendicular and (c) parallel to the longitudinal dc magnetic field.

perpendicular (panel b) and parallel (panel c) with respect to the external magnetic field. The following main features of these dependencies should be mentioned:

(i) First, for both easy axis orientations, dramatic variation of the SHG intensity around H=0 are observed, namely, one can see a sharp asymmetric peak in the vicinity of zero magnetic field, where the second harmonic intensity changes nearly twice. We attribute this effect to the re-magnetization process. It can be either in-plane re-orientation of magnetization accompanied by the appearance of the transversal magnetization-induced SHG component, which should contribute to the p-polarized second harmonic signal, so the SHG response is determined by the vector sum of the non-magnetic (crystallographic) and transversal magnetization-induced SHG terms, which may be linear or quadratic in magnetization. Or it can be a strongly inhomogeneous distribution of magnetization. In the latter case, the averaged square of the relevant SHG component can also produce additional SHG source at close to zero magnetic field. Thus under the magnetization reversal, first- and second-order in magnetization effects in the polarization at the second harmonic wavelength are valid, which lead to the changes of the shape of the SHG magnetic hysteresis loops and the contrast of the SHG intensity.

It is also worth noting that in the SHG magnetic hysteresis loops, the magnetization reversal reveals a pronounced asymmetry with respect to the projection of the external dc magnetic field on the magnetization easy axis. This was analyzed when measuring the corresponding dependencies of the SHG intensity for different orientations of the magnetization easy axis. In all cases, a strong SHG peak was registered at zero H values, while the changes of the SHG intensity for the positive and negative magnetic field can be even of the opposite signs. Quantitatively this can be characterized by the SHG magnetic contrast introduced in a common way, $\rho_{2\omega} = [I_{2\omega}(H) - I_{2\omega}(-H)]/[I_{2\omega}(H) + I_{2\omega}(-H)]$, where $I_{2\omega}(\pm H)$ is the SHG intensity for the positive and negative magnetic field. So depending on the tilt angle of the magnetic easy axis relatively the applied dc magnetic field, the $\rho_{2\omega}$ values can differ in sign. This effect is the most pronounced in the case of the presence of Pt/Co interfaces and needs further investigation, while it is clear now that the magnetization reversal is determined to much extend by the in-plane magnetic anisotropy of the interfaces in bilayer films.

Similar measurements were performed for the case of Co(20 nm)/Ta(3 nm) bilayer film, the corresponding results are shown in Fig. 3. The averaged SHG intensity in this structure is much less than for the Co/Pt one, which is attributed to different surface nonlinear susceptibility at Co/Ta interface. Again, one can see a pronounced longitudinal magneto-optical Kerr effect at the second harmonic wavelength, for the used experimental geometry the SHG polarization
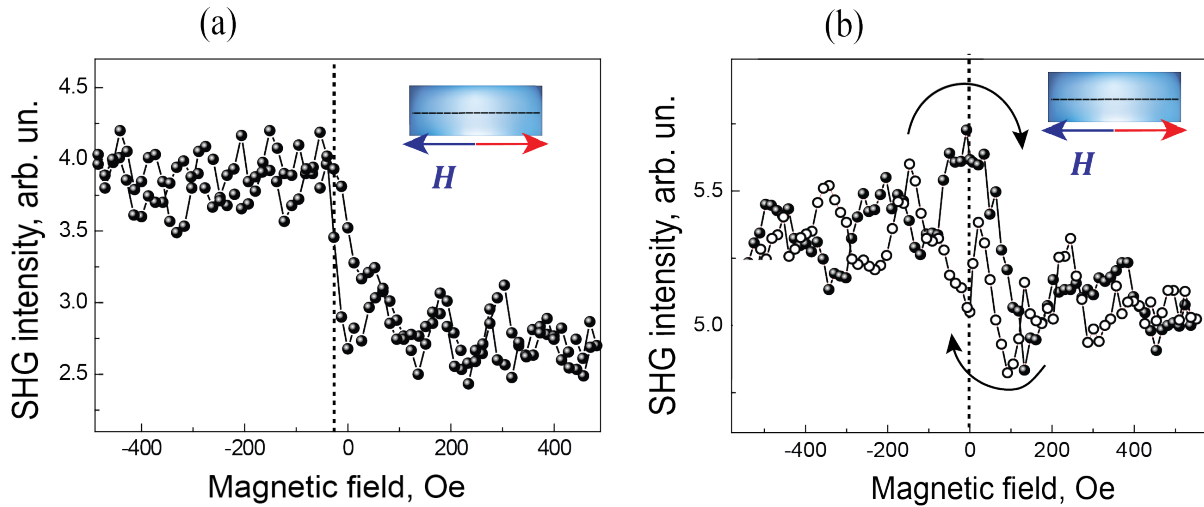
Figure 3. (a) Nonlinear magneto-optical longitudinal Kerr effect at the SHG wavelength and (b) dependence of the p-polarized SHG intensity on the longitudinal magnetic field measured for the Co/Ta film. In both cases the longitudinal *dc* magnetic field is applied parallel to the magnetization easy axis

plane rotation is about 10 degrees. The dependence of the p-polarized SHG on the longitudinal magnetic field also shows a modulation, while rather noisy, due to small SHG intensity and low signal to noise ratio. Nevertheless, the averaged SHG intensity for the negative values of the *dc* magnetic field is larger than that for the positive fields, as opposed to the results of Co/Pt film.

(ii) A surprising finding is a nonzero contrast of the SHG intensity for the case of the Co/Pt and Co/Ta structures observed for the applied *dc* magnetic fields far beyond the remagnetization region of bulky cobalt. Really, one can see in Fig. 2,c and Fig. 3,b that even for the magnetic field much higher than the saturating one, the SHG contrast is $\rho_{2\omega} > (15 \pm 3)\%$ for Co/Pt films and is $\rho_{2\omega} > (3 \pm 1.5)\%$ for Ta-containing bilayer. As was mentioned above, the sign and the value of this contrast of the SHG intensity depends on the mutual orientation of the magnetization easy axis and the external magnetic field.

Taking into account the interface sensitivity of the magnetization-induced SHG effect, this should be attributed to different magnetization distribution at Co/Ta and Co/Pt ones. Namely, the formation of inhomogeneous and chiral magnetic states can work here, which can be expected for FM/NM interfaces. This assumption is supported by the magnetic force microscopy (see figure 1), as well as by the BLS studies. The latter confirm the existence of the DMI at cobalt/platinum interface, while the effect is much less pronounced due to large thickness of cobalt.

4. Summary
Summing up, we performed the second harmonic generation studies of magnetization-induced effects in two types of bilayer structures, Co(20nm)/Pt(3nm) and Co(20nm)/Ta(3nm). Strong nonlinear magnetooptical effect of the SHG polarization plane rotation is observed in the geometry of the longitudinal Kerr effect, which is typical for the nonlinear magneto-optics. We demonstrate the existence of a strong magnetic field induced modulation of the SHG intensity for the geometry that excludes the observation of linear in magnetization SHG effect. We attribute the underlying mechanism to the magnetic chirality-driven SHG contribution at the considered interfaces.

Acknowledgements

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