Magneto-optical effects in second harmonic generation from W/Co/Pt nanofilms

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Abstract. Interfaces between ferromagnetic metals and nonmagnetic specimen attract much attention as they are very important for the formation of magnetic properties of nanostructures. Vice versa, specific magnetic ordering at such interfaces may provide new effects in their optical and nonlinear optical response. In this work we study the magnetization-induced effects in optical second harmonic generation (SHG) in W/Co/Pt-based thin films with the thicknesses of the Co layer of 2 – 10 nm. Besides common odd in magnetization effects in the SHG intensity, we observe additional one that is not expected for homogeneously magnetized ferromagnetic films, which consists in modulation of p-polarized SHG intensity under longitudinal magnetic field application. The phenomenological description of the observed effect is performed in terms of gradient and second-order in magnetization contributions to the SHG polarization, where gradient of magnetization along the normal to the structure plays the key role.

1. Introduction

Physics and technology of magnetic nanostructures is a hot research area known for a bunch of exciting effects originating from specific magnetic ordering at interfaces, which should appear as well in their optical and nonlinear optical response, first of all in surface-sensitive optical second harmonic generation (SHG) [1]. This is especially valid for the case of nanostructures composed of ferromagnetic and nonmagnetic materials, where such unique interface effects as perpendicular magnetic anisotropy, chiral magnetization states etc. have been demonstrated [2, 3, 4, 5]. Recently we studied magnetization-induced effects in SHG from bi- and trilayer films consisting of nanolayers of Co and nonmagnetic heavy metals (Pt, Ta). It was demonstrated that odd in magnetization intensity effect is observed for the p-polarized SHG in both reflection and transmission SHG schemes as the dc magnetic field was applied perpendicularly to the light, while it is symmetry-forbidden in dipole approximation for homogeneously magnetized structures. Moreover, it was found that the value of this "forbidden" magnetization-induced SHG effect depends on the symmetry of the nanostructures and is the largest in the case of asymmetric Pt/Co/W and Ta/Co/Pt films [6].

In order to investigate this effect in more detail, here we study the SHG in a nanostructure of the same nature, with the composition of W/Co/Pt, when applying dc magnetic field up to 1.5 kOe in longitudinal geometry.
2. SECOND HARMONIC GENERATION

In the electric dipole approximation, the SHG polarization is expressed as: \( P_{2\omega} = \hat{\chi}^{(2)D} E_\omega E_\omega \), where \( \hat{\chi}^{(2)D} \) is the electric dipole second-order susceptibility, \( E_\omega \) is the electric field of the fundamental light at the frequency \( \omega \). According to the Neumann’s principle, \( \hat{\chi}^{(2)D} \) is forbidden in the electric dipole approximation in the bulk of centrosymmetric materials, while it appears at the interfaces where the inversion symmetry is broken, thus giving rise to the interface SHG [7]. In the case of magnetic interfaces the magnetization-induced SHG (MSHG) in the most general form can be expressed as a series of the magnetization \( M \) [8]:

\[
\chi_{ijk}(M) = \chi_{ijk}^{(0)} + \chi_{ijklM}^{(1)} M_L + \chi_{ijklM}^{(2)} M_L M + \chi_{ijklM}^{(3)} \nabla_i M_M + \chi_{ijklM}^{(4)} M_L \nabla_m M_N \ldots ,
\]

where small letters in subindices correspond to polar vectors (\( E_\omega \) and \( \nabla \)), while the capital ones – to the axial vector \( M \). The first term \( \chi^{(1)} \) corresponds to nonmagnetic (crystallographic) dipole susceptibility, the second one is responsible for the common magnetooptical Kerr effect at the SHG wavelength in homogeneously magnetized media. The term \( \chi^{(2)} \) in Eq. (1) corresponds to the second-order (even) in \( M \) contribution to the nonlinear susceptibility, \( \chi^{(3)} \) and \( \chi^{(4)} \) describe the contributions induced by gradients of the magnetization; they are allowed in the bulk of centrosymmetric metals.

If the term \( \chi^{(1)} \) vanishes, which is realized for the case of p-polarized SHG and longitudinal or polar magnetic field application, odd in \( M \) MSHG effects are given by gradient terms described by \( \chi^{(3)} \) and \( \chi^{(4)} \); in what follows they are denoted as “forbidden” MSHG effect. It was explained in terms of “gradient” \( \chi^{(4)} \) contribution, particularly given by the components like \( \chi_{xxxXzZ}^{(4)} \), \( \chi_{xzzXzZ}^{(4)} \) etc, all of them containing gradients of \( M \) along the normal to the surface (along the \( (OZ) \) direction); the coordinate frame is shown in Fig. 1. Besides, one can consider the second-order in \( M \) interface contribution described by \( \chi^{(2)} \). Thus the SH intensity can be written as:

\[
I_{2\omega} \approx |P^{\text{even}} + P^{M\text{M}} + P^{\nabla}|^2 .
\]

On the other hand, the SHG intensity can be expressed as a sum of even and odd in \( M \) contributions as

\[
I_{2\omega} \approx |P^{\text{even}} + P^{M\text{odd}}(M)|^2 ;
\]

these notation are convenient for the analysis of odd in magnetization effects in the SHG intensity and are used below when describing the experimental results.

![Figure 1. MSHG experimental geometry with the coordinate frame; \( \theta \) is the angle of incidence.](image-url)

3. RESULTS AND DISCUSSION

In this work we studied isotropic magnetic trilayer films of the composition W(3)/Co(x)/Pt(3) with various Co thicknesses \( (x = 2, 3, 4, 10 \text{ nm}) \). The samples were grown by magnetron
sputtering on glass substrates using a AJA 2200 multichamber system at the basic pressure of $10^{-5}$ Pa, the layers are denoted starting from the substrate.

The scheme of the MSHG experiment is presented in Fig. 1. As a pump we used the p-polarized radiation of the pulse Ti:sapphire laser with the following parameters: wavelength of 800 nm, pulse duration of approximately 20 fs, repetition frequency of 80 MHz. The pump was focused to the surface of the sample in a spot of 30 μm in diameter; the transmitted SHG light passed through the color BG39 filters, an analyser (Glan-Tailor prism) and was detected by a PMT operating in the photon counting mode. The analyser was set at (i) 45° between p- and s-polarizations (the so called mixed polarization, ”allowed” geometry), or (ii) p-polarization of the SHG radiation (”forbidden” geometry). The samples were placed in the magnetic fields formed by an electromagnet ($H < 1.5$ kOe).

![Figure 2](image)

**Figure 2.** (A) Longitudinal MOKE hysteresis loop (triangular symbols) and analogous ”allowed” SHG magnetic field dependence (open square symbols) measured for W(3)/Co(3)/Pt(3) film. (B) ”Forbidden” SHG effect for structures W/Co/Pt with various Co thicknesses of $\theta = 30$°. (C) Estimated odd terms of the second-order susceptibility as a function of angle of incidence, $|\chi_{eff}^{odd}|(\theta)$, for W/Co/Pt films with various Co thicknesses.

Figure 2(A) shows MOKE and MSHG magnetic hysteresis loops in the ”allowed” geometry. Longitudinal magnetooptical Kerr effect (MOKE) was used for the characterization of the magnetic properties of the films, when using the probe radiation of He-Ne laser. As an example, MOKE hysteresis loop for W(3)/Co(3)/Pt(3) film is shown in Fig. 2(A) (black symbols). One can see that the width of the MOKE hysteresis is roughly twice as large as in the case of the MSHG. This may be related to different localization of the MOKE and MSHG sources, in the bulk of the cobalt film and at its interfaces, are their different magnetic properties. Using Eq. (1), one can estimate the ratio of $|P^{odd}|$ and $|P^{even}|$ of $|P^{odd}|/|P^{even}| \sim 0.56$.

Figure 2(B) shows magnetic hysteresis loops measured in the geometry of the ”forbidden” MSHG effect for the angle of incidence $\theta = 30$° for the samples with Co thicknesses $x = 2, 3, 4, 10$ nm; the magnetic field was applied in the longitudinal geometry, i.e. in plane of the
samples and in the plane of incidence, (XOZ). Firstly, pronounced peaks in the SHG intensity for the magnetic fields lower than the saturation fields are seen, which can be explained as remagnetization of the structure by rotation of the average magnetization \( M \) in the plane of the sample through the transversal MOKE geometry. Secondly, one can observe the asymmetry of the SHG intensity in saturating magnetic fields (of up to 600 Oe) of the opposite directions. It can be characterized by the MSHG magnetic contrast \( \rho_{2\omega} = \frac{I^+_{2\omega} - I^-_{2\omega}}{I^+_{2\omega} + I^-_{2\omega}} \), where \( I^+_{2\omega} \) and \( I^-_{2\omega} \) denote the SHG intensity for positive and negative magnetic fields, respectively. The existence of the MSHG contrast indicates the presence of odd with respect to \( M \) terms in the SHG polarization that appear in Eq. (2). As the common MOKE in SHG is forbidden under these conditions, we propose that this effect can be explained in terms of contribution of \( P_{MM} \) and \( P_{\nabla} \) from (1).

Measuring the "forbidden" effect for all W/Co/Pt samples for the incidence angles \( \theta = 5^\circ, 10^\circ, \ldots, 45^\circ \) revealed an increase of mean SHG intensity \( \langle I_{\text{mean}} \rangle = 0.5(I^+_{2\omega} + I^-_{2\omega}) \) with increasing \( \theta \). Similar phenomenon was observed for Pt/Co/W thin films (opposite order of layers) [6]. Such dependence represents the primary influence of \( z \)-component of nonlinear polarization on magneto-induced SHG. By calculating the mean SHG intensity and magnetic contrast, we estimated odd contribution as \( |P_{\text{odd}}| \sim |\hat{\chi}_{\text{eff}}^{\text{odd}}| \sim 0.5|\rho_{2\omega}|\sqrt{\langle I_{\text{mean}} \rangle} \). Figure 2(C) demonstrates the dependence of this quantity on the angle of incidence \( |\hat{\chi}_{\text{eff}}^{\text{odd}}| (\theta) \), which reveals quite similar values \( |\hat{\chi}_{\text{eff}}^{\text{odd}}| \) for all \( x \) values at angles of incidence \( \theta = 15^\circ \ldots 30^\circ \). A rise at \( \theta \geq 30^\circ \) can be explained as an increase of relative contribution of \( P_{MM} \) and \( P_{\nabla} \) with polar \( z \) indices.

4. Conclusion

Summing up, we studied experimentally magnetic field induced effects in optical second harmonic generation from W/Co/Pt nanolayered films. We demonstrate that when excluding the detection of common magneto-optical effects at the SHG wavelength in the longitudinal magnetic field, that is expected for homogeneous magnetic films, odd in magnetization changes of the intensity of the p-polarized SHG are observed. These studies confirm that gradient in magnetization terms of the second-order susceptibility should be necessarily taken into account when considering the MSHG effects in thin magnetic films.

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