

# Magnetization Induced Optical Second Harmonic Generation from Interfaces between Ferromagnetic and Heavy Metals

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**Abstract:** Magnetization induced effects in optical second harmonic generation from interfaces between heavy and ferromagnetic metals reveal an important role played by magnetic anisotropy and second-order in magnetization effects in their nonlinear response. © 2019 The Author(s)

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## 1. Introduction

Optical second harmonic generation is a well-known surface sensitive probe providing unique information about the most important parameters of surfaces and interfaces [1]. It appears due to the symmetry breaking due to the structural discontinuity. Moreover, as it is accompanied by the time-inversion symmetry breaking specific to hyrotropic media, this results in high efficiency in diagnostics of interface magnetism demonstrated in a large number of papers [2, 3]. At the same time, specific role played by the Dzyaloshinskiy-Moria interaction (DMI) at the interfaces between a ferromagnetic and a heavy metals on the nonlinear optical response of a single or multiple interfaces, has not been studied in much detail. Here we study the magnetic field induced effects in optical second harmonic generation that appear in planar structures composed of heavy (Pt, Ta) and ferromagnetic (Co) metals, with a special emphasize on the role of magnetic anisotropy.

## 2. Methods

Magnetic characterization of the samples was performed by vibrational magnetometry and magneto-optical Kerr effect (MOKE). For the nonlinear optical studies, p-polarized radiation of the 80 femtosecond pulsed Titanium sapphire was used at the wavelength of 780 nm and the mean power of about 100 mW. SHG radiation reflected from the samples in the direction of the specular reflection was detected by a photomultiplier (PMT) operating in the photon counting mode. Nonlinear MOKE was studied as the longitudinal magnetic field of the strength up to 1.5 kOe was applied to the samples.

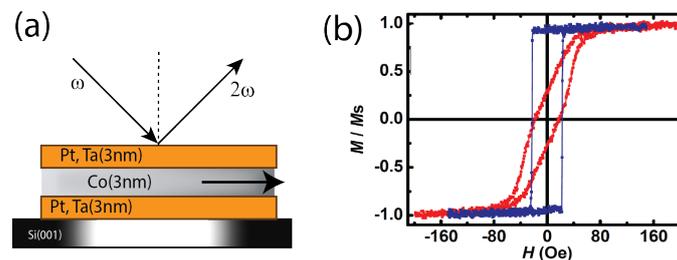


Fig. 1. a) Scheme of a trilayer magnetic structure composed of a 3 nm thick Co layer surrounded by 3 nm thick layers of heavy metals; b) MOKE loops for the Ta/Co/Pt structure using the vibrational magnetometer, blue and red curves correspond to orientation of the easy and heavy magnetization axes parallel to the applied field.

We used the two experimental schemes for the visualization of the nonlinear magneto-optical interface effects. First, the analyzer oriented at  $45^\circ$  with respect to the p-polarization was introduced prior to the PMT, so that the relevant nonlinear magneto-optical effect of the SHG polarization plane rotation appeared as the magnetic hysteresis of the SHG intensity. This scheme is a well-known experimental approach. Second, p-polarized SHG

intensity was measured, which excluded the registration of the SHG polarization plane rotation, while allowing to study less trivial nonlinear optical contributions.

The samples were made by high vacuum magnetron sputtering as an in-plane dc magnetic field of 1 kOe was applied. Three-layer planar structures of the composition  $HM/Co/HM$ , all the layers being 3nm thick (Fig. 1,a),  $HM$  being a heavy metal like Pt, Ta etc. with different constants of the Dzyaloshinskiy-Moria interaction that influence the magnetic properties of interfaces.

### 3. Results and Discussions

Magnetometry and MOKE measurements showed that all the structures possess a in-plane magnetic anisotropy, so the following nonlinear magneto-optical studies were performed for various orientations of the magnetic field with respect to the magnetization easy axis (Fig. 1,b). The most pronounced nonlinear MOKE is attained as the in-plane orientation of the easy axis is  $45^\circ$  relatively to the applied magnetic field. Figure 2 shows the SHG magnetic hysteresis loops associated with the SHG polarization plane rotation (Fig. 2,a) and for the p-polarized SHG, which reveals only the SHG intensity changes (Fig. 2,b).

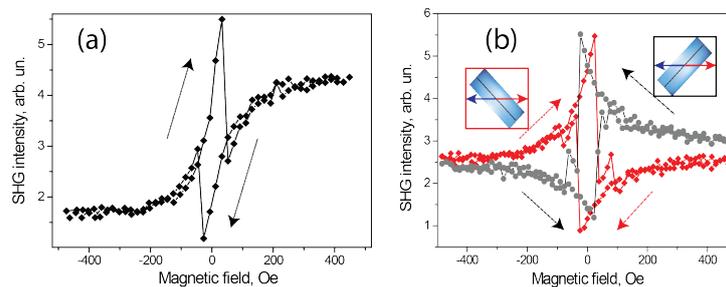


Fig. 2. SHG magnetic hysteresis loop measured for Ta/Co/Pt structure a) in the geometry of longitudinal MOKE as discussed in the body of the text, and b) for the p-polarized second harmonic, for  $\pm 45^\circ$  angle between the magnetization easy axis and the applied magnetic field (red and grey symbols, respectively, as shown in the insets); arrows denote the directions of the magnetic field changing.

A strong modulation of the SHG intensity close to zero magnetic field is observed in both experimental geometries for Ta/Co/Pt 3-layer film, while the relevant minimum and maximum in Fig. 2a depend on the azimuthal orientation of the structure. This corresponds to the in-plane magnetization rotation and appearance of the transversal Kerr effect, as well as to the second-order in magnetization effects, which stems from the performed modelling. Similar effect takes place for the p-polarized SHG (Fig. 2,b), where the difference of the SHG hysteresis shapes for different samples' orientation is quite distinct.

Summing up, we used the SHG probe for the studies of magnetic properties of interfaces between ferromagnetic (Co) and heavy metals (Pt,Ta). We proved by nonlinear magneto-optical Kerr effect that inversion of the longitudinal magnetic field leads to the in-plane magnetization rotation, leading to a sharp SHG peaks associated with the transversal M component. The observed strong azimuthal anisotropy of the shapes and values of the SHG magnetic hysteresis loops can be due to the formation of the unidirectional magnetic anisotropy, specific for the samples with definite compositions. An important role of the second-order in magnetization effects in the formation of their nonlinear response is shown. We also suggest a phenomenological description of the observed nonlinear optical effects.

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