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Magnetization-induced anisotropy of second harmonic generation in thin cobalt films

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ABSTRACT

Magnetization-induced effects in the symmetry properties of optical second harmonic generation (SHG) are studied in thin cobalt films. We demonstrate that the application of an external magnetic field leads to the appearance of a strong SHG azimuthal anisotropy from the isotropic Au/Co structure. Symmetry analysis of the SHG dependencies, supported by the SHG interferometry measurements, indicates that the effective magnetic and crystallographic second-order susceptibilities are both of the same order of magnitude.

1. Introduction

Nonlinear optics provides unique possibilities for the investigation of the electronic properties of a matter [1]. Among various nonlinear optical methods, second harmonic generation (SHG) is known as a highly sensitive probe of surfaces and interfaces of centrosymmetric materials. In that case, the second-order polarization is localized in a thin subnanometer thick surface layer. A good correlation between the surface crystallographic symmetry and of the SHG anisotropy has been demonstrated for a number of structures [2,3]. In particular, it allowed to visualise the structural phase transition in upper monolayers of Si monocrystals using the SHG probe [4].

For the case of magnetics, additional symmetry operations appear that describe the modification of the nonlinear-optical response under the reversal of the magnetization, **M** [5,6]. In that case the components of the second-order susceptibility tensor, $\hat{\chi}^{(2)}$, can be divided into two parts, odd and even in **M**. The former one is proportional to *M* and changes its sign under the reversal of the magnetization, $\hat{\chi}^{(2)odd}(\mathbf{M}) = \hat{\chi}^{(2)odd}(-\mathbf{M})$, while the latter is independent on the direction of **M**. The corresponding expression for the SHG field can be written as: $\mathbf{E}(\mathbf{M}) = \mathbf{E}^{even} \pm \mathbf{E}^{odd}(M)$ and thus describes the nonlinear magnetooptical effects [7,8]. It is worth noting that the magnetization-induced susceptibility appears only in the regions with broken central symmetry. This brings about a high sensitivity of the SHG probe to the magnetization of surfaces and internal interfaces of centrosymmetric media [6,9,10].

The symmetry analysis of the second-order susceptibility of magnetized surfaces was performed in [6]. It was shown that the symmetry of $\hat{\chi}^{(2)odd}$ and $\hat{\chi}^{(2)even}$ components is different; $\hat{\chi}^{(2)odd}$ describes new operations that vanish for the case $\mathbf{M}=0$, while $\hat{\chi}^{(2)even}$ components reveal the same symmetry as the crystallographic (nonmagnetic) susceptibility. This should find confirmation in the SHG azimuthal anisotropic dependencies from a magnetized and nonmagnetized medium. At the same time, to the best of our knowledge, there has been no direct experimental confirmation of this effect. In this paper we study the appearance of a magnetization-induced azimuthal anisotropy of the optical second harmonic reflected from a planar isotropic Co/Au film.

2. Samples and experimental set-up

The samples under investigation are thin cobalt films deposited on Si(1 0 0) substrate, coated by a gold layer with a thickness of ~ 0.9 nm. The Au/Co bilayer films were fabricated by ion beam sputtering in a single deposition cycle as described in detail elsewhere [11]. Two samples were studied, with the thickness of the Co layer d_{Co} = 0.5 and 10.0 nm; a schematic of the structure is shown in Fig. 1(a). The morphology of the Au/Co surface was studied by means of the atomic force microscopy (Femtoscan 0 0 1) over an area of 5 μm × 5 μm. It was shown that the Au surface is smooth and homogeneous, the average roughness being less than 1 nm [11]. Magnetization measurements have shown that the studied planar layered structures are ferromagnetic and isotropic within the film plane.

For the nonlinear-optical experiments an Nd:YAG^{3+} laser was used with the wavelength of 1064 nm, repetition rate of 25 Hz and pulse duration of 15 ns. The angle of incidence was 40°.

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The SHG radiation reflected from the samples was spectrally selected by appropriate color and interference filters, passed through an analyzer and then detected by a PMT and gated electronics. The sample was placed on a holder in a constant transversal magnetic field of about 3.1 kOe created by permanent NdFeB magnets. In this way simultaneous azimuthal rotations of the sample and of the magnet could be performed (Fig. 1(b)).

3. Experimental results

3.1. Nonmagnetic SHG

Azimuthal angular dependencies of the SHG intensity were studied first in nonmagnetized samples. The intensity of the *p*-polarized SHG component turned out to be isotropic and by two orders of magnitude larger as compared with the intensity of the *s*-polarized one. The SHG scattering indicatrices, i.e. the dependencies of the SHG intensity on the polar angle of scattering, demonstrate that the second harmonic is generated in the direction of the specular reflection. Such properties of the second-order nonlinear response correspond to the fulfillment of the so-called *s*-prohibition rule [12,13] and indicate that the Au/Co structure is isotropic and smooth. Thus the nonlinear optical response of nonmagnetized Au/Co/Si planar structures should be described by a second-order susceptibility tensor corresponding to an isotropic surface.

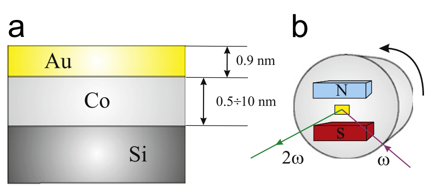

Fig. 1. The schemes of (a) Au/Co/Si structure and (b) of the magnetization-induced anisotropy measurements.

3.2. Magnetization-induced anisotropy

Magnetization-induced anisotropy of the SHG intensity reflected from an isotropic Au/Co/Si structure was studied when rotating the sample together with the permanent magnetic field around an axis normal to the plane of the sample. Fig. 2 shows the dependencies of the SHG intensity on the azimuthal angle of the sample, ψ, measured for the film with d_{Co} = 10.0 nm for different combinations of polarizations of the fundamental and SHG waves. In this experiment the angle ψ defines also the azimuthal orientation of the in-plane magnetic field. The values $\psi = 0^\circ$ and 180° correspond to the magnetic field oriented perpendicularly to the plane of incidence (transversal geometry). It can be seen that a pronounced anisotropy of the SHG intensity appears for all combinations of polarizations, contrary to the case of a nonmagnetized sample. In particular, anisotropic *s*-polarized SHG appears that is absent in the case of a nonmagnetized structure (Fig. 2(b),(d)). The symmetry analysis of these dependencies is presented below.

In the case of structurally isotropic magnetic films, magnetization-induced changes of the nonlinear-optical response can be described in terms of the SHG magnetic contrast defined as $\rho_{2\omega} = (I_{2\omega}(\psi = 0^\circ) - I_{2\omega}(\psi = 180^\circ))/(I_{2\omega}(\psi = 0^\circ) + I_{2\omega}(\psi = 180^\circ))$ [6]. The appearance of the SHG magnetic contrast is evident from the dependencies shown in Fig. 2. It is caused by a modification of the SHG intensity as the transversal component of the dc magnetic field is being changed. The maximal values of the transversal magnetic field correspond to the azimuthal angles $\psi = 0, 180^\circ$. For the $p-\omega$, $p-2\omega$ and $s-\omega$, $p-2\omega$ combinations of polarizations the SHG magnetic contrast is about $\rho_{2\omega} = 83$ and 70%, respectively.

Magnetization-induced anisotropy of the SHG intensity in the $p-\omega$, $p-2\omega$ combination of polarizations was studied for Au/Co structures with the cobalt layer thickness of d_{Co}=10.0 and 0.5 nm; the corresponding experimental curves are shown in Fig. 3. It can be seen that these dependencies are very similar; the SHG magnetic contrast of about 73% is observed for the structure with a thinner cobalt layer. Slightly different shape of $I_{2\omega}(\psi)$ curves is probably due to different relative values of the

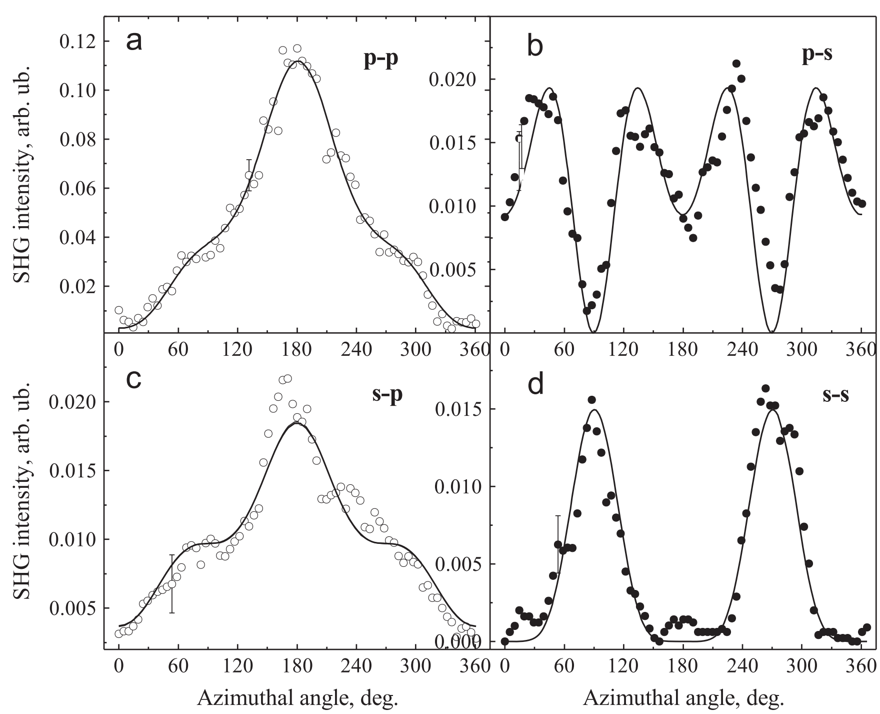

Fig. 2. Anisotropic dependencies of the SHG intensity measured for (a) $p-\omega$, $p-2\omega$; (b) $p-\omega$, $s-2\omega$; (c) $s-\omega$, $p-2\omega$ and (d) $s-\omega$, $s-2\omega$ combinations of polarizations of the fundamental and SHG radiation for Au/Co/Si film, d_{Co}=10 nm.