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Observation of magneto-optical second-harmonic generation with surface plasmon excitation in ultrathin Au/Co/Au films

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Magnetization-induced second-harmonic generation with surface plasmon excitation in an ultrathin Au/Co/Au multilayer structure has been investigated. The resonant coupling of surface plasmons with the fundamental light results in drastic changes of the second-harmonic intensity and a sign reversal of nonlinear magneto-optical effects. Model analysis of the observed phenomena is given on the basis of the multiple interference of interface nonlinear contributions calculated using the Green's functions formalism. © *1999 American Institute of Physics.* [S0003-6951(99)01328-5]

Surface collective electron oscillations, also known as surface plasmons (SP), can be excited in noble metals below the plasma frequency and may give rise to a variety of linear and nonlinear phenomena.[1] The coupling of the electric field at optical frequencies with SP in metallic multilayer films results in an increase of the linear magneto-optical effects.[2] It has been shown experimentally and theoretically that the optical second-harmonic generation (SHG) is strongly enhanced due to the SP excitation.[1,3] Since the observation of magnetization-induced SHG,[4] it has been established that SHG is a powerful method to study magnetic surfaces and interfaces. This method was intensively used for investigations of new nonlinear magneto-optical phenomena in different film compositions,[5,6] showing that magnetization-induced SHG is very sensitive to the crystallographic, magnetic, and electronic structure of thin films.

In this letter we present the experimental observation of nonlinear magneto-optical phenomena related to the surface plasmon excitation in an ultrathin Au/Co/Au multilayer structure. The measurements have been done using the attenuated total reflection technique (ATR) in the Kretschmann geometry.[1] The coupling of the SP with light at the fundamental frequency gives rise to drastic changes of second-harmonic (SH) intensity. SH magnetic contrast exhibits a sign reversal when the SP are excited. We developed a model on the basis of the Green's functions (GF) approach, which allows us to describe different contributions of the film interfaces to the SHG. Model calculations were found to be in good agreement with the observed phenomena.

In crystallographically centrosymmetric layers the SHG is only allowed, in the electric-dipole approximation, at surfaces and interfaces where the space-inversion symmetry is broken. In the presence of a magnetization $\mathbf{M}$, the nonlinear polarization $\mathbf{P}^{2\omega}$ can be written as

$$P_i^{2\omega}=\epsilon_0\chi_{ijk}^{N}E_jE_k\pm\epsilon_0\chi_{ijk}^{M}(\pm\mathbf{M})E_jE_k, \tag{1}$$

where E_j and E_k are components of the optical electric field at the fundamental frequency ω. The nonlinear susceptibility tensors χ_{ijk}^{N} and χ_{ijk}^{M} describe nonmagnetic and magnetic contributions to the nonlinear polarization $\mathbf{P}^{2\omega}$, respectively. Neglecting dissipation in the medium, χ_{ijk}^{N} is a real tensor element and χ_{ijk}^{M} is a pure imaginary.[7] In the presence of absorption, both tensors are complex, thus allowing an interference between the two contributions. This interference gives rise to new nonlinear phenomena such as nonlinear magneto-optical rotation[5] and nonlinear circular dichroism.

The ultrathin Au/Co/Au films were deposited on a 1-mm-thick float glass substrate in a high vacuum chamber, following the procedure described in Ref. 8. In the Au(3 nm)/Co(3 nm)/Au(25 nm)/glass structure, the easy magnetization axis was located in the film plane. Pulsed coils producing an in-plane magnetic field up to 1 kOe were specially designed to magnetize this sample at saturation. The glass substrate was optically coupled to a half-cylindrical glass lens using a refractive index adaptation liquid.

The experimental setup and the geometry of measurements in the ATR configuration are shown in Fig. 1. The mode-locked Ti:sapphire laser pumped by an Ar laser was used as a source of 100 fs light pulses with a repetition frequency of 86 MHz at the wavelength of $\lambda = 800$ nm. The beam with an average power of 30 mW was focused on the sample in a spot of about 100 μm in diameter. In front of the sample a RG715 filter was used in order to eliminate a possible parasitic SHG generated at the surfaces of the optical

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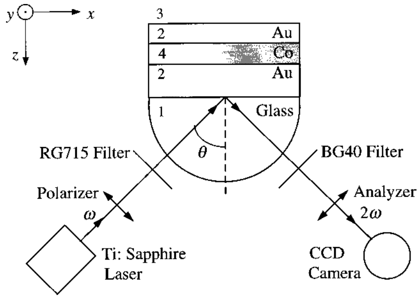

FIG. 1. Schematics of the experimental setup.

elements. After the sample, a BG40 filter was used to block the fundamental light. The SH intensity generated by the Au/Co/Au film was measured using a high-sensitivity cooled charge-coupled device camera. We used p-polarized fundamental light (TM mode), which excites SP near an incidence angle θ_p with a wave vector k_p defined by the SP dispersion equation[1]

$$k_p=\frac{\omega}{c}\sqrt{\frac{\epsilon_1\epsilon_2}{\epsilon_1+\epsilon_2}},\tag{2}$$

where ϵ_1 and ϵ_2 are the dielectric constants of the two media at the interface where SP are excited. SP may be coupled with evanescent optical waves from a half-cylindrical glass lens at the angle

$$\theta_p=\arcsin\left(\frac{k_p c}{\omega\sqrt{\epsilon}}\right),\tag{3}$$

where ϵ is the dielectric constant of the glass. For interfaces between optically isotropic media with p-polarized excitation, when there are only x and z components in the fundamental electric field (see Fig. 1), the nonlinear polarization $\mathbf{P}^{2\omega}$ can be described for the longitudinal geometry ($\mathbf{M}\|x$) by the matrix expression

$$\mathbf{P}^{2\omega}=\epsilon_0\begin{pmatrix} 0 & \chi^N_{xxz} & 0 \\ \chi^M_{yxx} & 0 & \chi^M_{yzz} \\ \chi^N_{zxx} & 0 & \chi^N_{zzz}\end{pmatrix}\begin{pmatrix} E_xE_x \\ 2E_xE_z \\ E_zE_z\end{pmatrix}.\tag{4}$$

For the transversal geometry, when $\mathbf{M}\|y$, $\mathbf{P}^{2\omega}$ can be written as

$$\mathbf{P}^{2\omega}=\epsilon_0\begin{pmatrix} \chi^M_{xxx} & \chi^N_{xxz} & \chi^M_{xzz} \\ 0 & 0 & 0 \\ \chi^N_{zxx} & \chi^M_{zxz} & \chi^N_{zzz}\end{pmatrix}\begin{pmatrix} E_xE_x \\ 2E_xE_z \\ E_zE_z\end{pmatrix}.\tag{5}$$

The nonzero tensor components are determined on the basis of symmetry considerations. One must note that, due to the symmetry, $\chi^M_{yzz}=-\chi^M_{xzz}$ in Eqs. (4) and (5).

The variations of the reflectivity of the Au/Co/Au film at the fundamental light frequency versus the angle of incidence θ are shown in Fig. 2(a). The signal increases up to the total internal reflection angle $\theta_t\simeq 43.5°$ and then strongly decreases down to the angle $\theta_p\simeq 45°$. The sharp minimum in the intensity of the reflected light indicates that SP are excited near this angle. A similar behavior of the reflected light at $\lambda=632.8$ nm in Au/Co/Au film has already been observed by Safarov *et al.*[2]. A theoretical modelization of the reflection coefficients and magneto-optical effects in Au/Co/Au films in the ATR configuration on the basis of the GF technique has been achieved by Kosobukin.[9] It was established that in the Au/Co/Au/glass structure SP are excited at the air/Au interface. The coupling of the light with SP gives rise to a strong enhancement of the magneto-optical linear Kerr effect.[2]

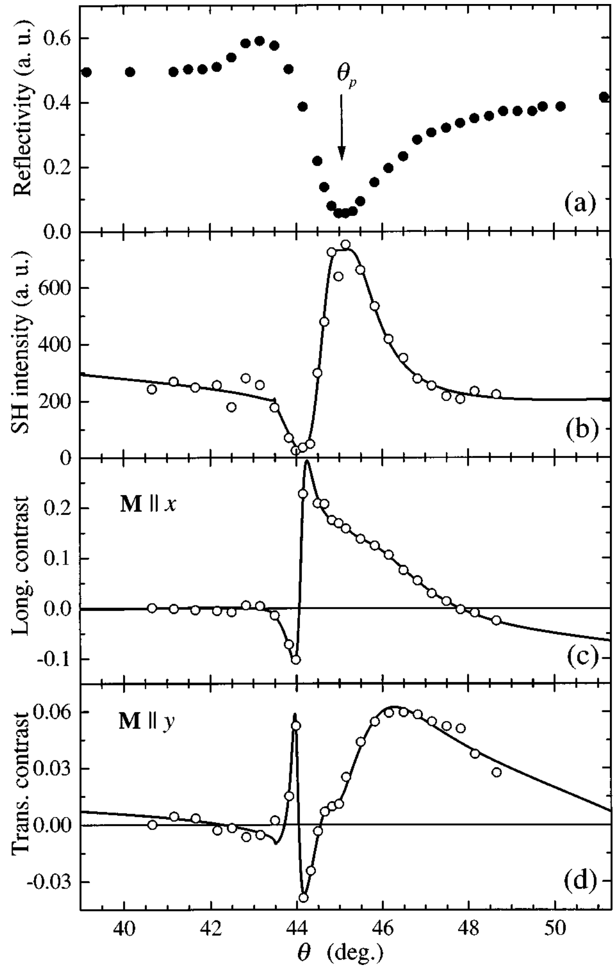

FIG. 2. Variations with the incidence angle θ of the reflectivity at the fundamental frequency (a), the nonmagnetic SHG intensity (b), and the SH magnetic contrast in longitudinal (c) and transversal (d) geometries.

The total SH intensity measured in longitudinal geometry ($\mathbf{M}\|x$) with p-polarized input and p-polarized output is shown by dots in Fig. 2(b). According to Eq. (4) there is no magnetic contribution to the nonlinear polarization in this configuration. The SH signal has a strong minimum at the angle $\theta\simeq 44°$. It reaches a maximum near θ_p, decreases, and is constant for angles $\theta>47°$.

The dots in Fig. 2(c) show the experimental longitudinal ($\mathbf{M}\|$) SH magnetic contrast ρ, defined as

$$\rho=\frac{I^{2\omega}(+\mathbf{M})-I^{2\omega}(-\mathbf{M})}{I^{2\omega}(+\mathbf{M})+I^{2\omega}(-\mathbf{M})}=\frac{2|\chi^N||\chi^M|\cos\varphi}{|\chi^N|^2+|\chi^M|^2},\tag{6}$$

versus the angle θ (φ is the optical phase difference between the even and odd tensor elements). We chose the position of the analyzer $\alpha=80°$ in order to obtain a good signal-to-noise ratio ($\alpha=0$ corresponds to the p polarization). The contrast has a sharp jump at $\theta\simeq 44°$. Near the angle of SP excitation θ_p, ρ decreases drastically and then changes sign at $\theta\simeq 48°$. The same type of measurements were done in transversal configuration ($\mathbf{M}\|y$) for p-input and p-output polarizations and are shown in Fig. 2(d). The sign of the transversal SH contrast changes three times in a narrow angle interval $\theta_t < \theta < 45°$. Then, ρ increases strongly and has a broad maximum at $\theta \simeq 46.5°$.

In order to explain our results, we developed a model involving multiple interference of the different interface contributions to SHG, which can be calculated using the GF technique. The GF approach was used for general description of SHG from surfaces and interfaces by Guyot-Sionnest, Chen, and Shen[10] and for analysis of the SHG dispersion at simple metal surfaces by Liebsch and Schaich.[11] The GF formalism is a convenient way for the consideration of the light propagation problem in multilayer structures, when the layer thickness is much less than the light wavelength. In our model we took into account the coupling of SP with the fundamental light, using a theory developed for the SP excitation in trilayer metallic systems.[9] The electric field $E_i^{2\omega}$ of the outcoming SH wave in the glass cylindrical lens can be represented as a sum of contributions from the interfaces of the film:

$$\begin{aligned} E_i^{2\omega} \propto\, & T^{24}T^{42}T^{21}\chi^{32}_{ijk}F^{32}_j F^{32}_k e^{2i(K_2 d'_2 + K_4 d_4 + K_2 d_2)} \\ & + T^{42}T^{21}\chi^{24}_{ijk}F^{24}_j F^{24}_k e^{2i(K_4 d_4 + K_2 d_2)} \\ & - T^{21}\chi^{24}_{ijk}F^{42}_j F^{42}_k e^{2iK_2 d_2} + \chi^{21}_{ijk}F^{21}_j F^{21}_k, \qquad (7) \end{aligned}$$

where T^{ab} are transmission coefficients for the SH light at the ab interface, and F_j^{ab} and F_k^{ab} are continuous local integral functions of the electric fields at the fundamental frequency. The numbering of interfaces is given in Fig. 1: 32 is the air/Au, 24 is the Au/Co interface, etc. K_2 and K_4 are the z components of the SH wave vectors in Au and Co. d_2, d'_2, and d_4 are the thicknesses of the Au buffer and cover layers and of the Co layer, respectively. One can suppose that $\chi^{42}_{ijk} = -\chi^{24}_{ijk}$, because the Au/Co and the Co/Au interfaces are perfectly symmetrical. The quantities T^{ab} and F_i^{ab} were calculated using, respectively, Eqs. (A7) and (3.16) in Ref. 9. The complex refractive indices of Au and Co were taken from Ref. 12. The SH intensity was calculated as $I^{2\omega} \propto |E_x^{2\omega}\cos\theta + E_z^{2\omega}\sin\theta|^2$ for the p-polarized SH light and as $I^{2\omega} \propto |E_y^{2\omega}|^2$ for the s polarization. We developed a program for the fitting procedure of the experimental data. Unknown components of the complex nonlinear tensors χ^{ab}_{ijk} were used as fit parameters. Nonmagnetic components of χ^{ab}_{ijk} were found fitting the data shown in Fig. 2(b). Then, nonmagnetic components of χ^{ab}_{ijk} were fixed and the fitting procedure was continued for the SH magnetic contrast using Eq. (6). The calculated curves are shown in Figs. 2(b)–2(d) by the solid lines. One can see a good agreement of the modelization with the experiment.

We demonstrated experimentally the magnetization-induced second-harmonic generation with the surface plasmon excitation in ultrathin Au/Co/Au structures. In the electric-dipole approximation the second-harmonic generation is allowed at magnetic and nonmagnetic interfaces in these films. At the angle θ_p, due to the coupling of SP with the fundamental light, the fundamental electric field is strongly enhanced at the nonmagnetic air/Au interface and redistributed to the magnetic Au/Co and Co/Au interfaces. This results in an enhancement of both the magnetization-induced and the nonmagnetic SHG. Drastic changes in the distribution of the fundamental field at the magnetic interfaces near θ_p result in a sign reversal of the SH magnetic contrast in the longitudinal and transversal geometries. Model calculations based on the Green's functions technique are shown to be in good agreement with the experimental results.

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