Nonlinear TMOKE enhancement in 1D Au/Py magnetoplasmonic crystals

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Abstract. Resonant optical properties of the magnetoplasmonic crystals, which support propagation of surface plasmon polaritons (SPPs) accompanied by magnetooptical effects, have found success in magnetic field driven control of optical radiation. In this work we investigate the resonant magneto-optical effects in the second harmonic generation in the magnetoplasmonic crystal formed by gold/pemalloy bilayer covering dielectric grating. Strong transverse magnetooptical Kerr with the contrast up to 30% is revealed in the spectral vicinity of the SPP excitation.

1. Introduction
Magnetoplasmonic crystals (MPCs) are periodic metal-dielectric nanostructures, which bind together the excitation of surface plasmon polaritons (SPPs) and magnetooptical effects. Resonant optical properties of the SPP localized in close proximity to the interface of metal and dielectric make the optical response of the plasmonic crystals very sensitive to properties of media adjacent to the interface that was harnessed in sensing of molecules and gases [1]. Particularly, magnetization of media in MPCs can shift resonant frequencies of SPPs. For example, the interface of metal and magnetoactive dielectric semispaces supports propagation of SPPs, the wave vector magnitude of which is determined by both dielectric permittivities of neighboring media and the magnetization:

\[ k_{spp} = k_0 \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}} (1 + \alpha M), \]

where \( \varepsilon_m, \varepsilon_d \) are permittivities of metal and dielectric, respectively, \( M \) - magnetization perpendicular to the SPP propagation direction, \( \alpha = a (-\varepsilon_m \varepsilon_d)^{-1/2} (1 - \varepsilon_m^2 / \varepsilon_d^2)^{-1} \), \( k_0 = \omega / c \) is the vacuum wave number, \( a \) is the gyration constant [2]. This sensitivity of SPP in MPCs to the applied magnetic field gives rise to the resonant manifold increase of intrinsically weak magneto-optical effects, e.g., transverse magneto-optical Kerr effect (TMOKE) consisting of the variation of the reflection coefficient of light under the magnetization of medium perpendicularly to the plane of light incidence. It has paramount importance in control of light routing and remote optical sensing of magnetic field strength [3].

It should be noted that the excitation of SPPs requires special efforts caused by the wavevector mismatch between \( k_{spp} \) and the vacuum wave number. In plasmonic crystals it is cured by the light diffraction involving the reciprocal lattice vector \( G \) of the periodic structure that leads to the quasi-phase matching condition

\[ k_{spp} = k_{||} + mG, \]

where \( k_{||} \) is the component of the wavevector in the plane of incidence and \( m \) is an integer.
where $k_{\text{spp}}$ is the wavevector of SPP, $k_{||}$ is the tangential component of the wave vector of incident plane wave and $m$ is the diffraction order.

In the field of investigation of magneto-optical effects in MPCs special attention is paid to the structures composed of noble and ferromagnetic metals [4, 5]. In this case both high-quality plasmon resonances supported by noble metals, like gold or silver, exhibiting lowest optical losses in visible spectral range, and high magneto-optical activity are combined in one structure. It results in a pronounced enhancement of magneto-optical effects, for example, TMOKE [6].

Being odd to the magnetization, the magnitude of TMOKE is characterized by the magnetic contrast given by

$$\rho = \frac{I(+M) - I(-M)}{I(+M) + I(-M)}$$

that is a relative change of reflected light intensity for opposite directions of the medium magnetization $M$. The magnetic contrast $\rho$ has a typical value of about $10^{-3}$ that requires the intense efforts to detect it. In turn the nonlinear optical phenomena, particularly the second harmonic generation (SHG), can be harnessed to significantly intensify the effects of magnetic field in the optical response of MPCs [7]. Having nonlinear local field factors dependence, the resonant enhancement of the SHG in the spectral vicinity of the excitation of SPP at the fundamental frequency has been demonstrated [8, 9, 10]. As well being sensitive to the optical properties of the media interface, pronounced nonlinear TMOKE was achieved via SPP excitation in periodically perforated Au/Co films [11].

This work is aimed at the investigation of the SHG in the MPCs based on one-dimensional dielectric lattice covered by thin gold and ferromagnetic permalloy films. Special attention is paid to the nonlinear TMOKE under the excitation of SPPs. Several tens of percent of the magnetic contrast in the SHG in the spectral vicinity of SPP excitation is revealed, that makes such structures promising for the magnetic field sensing.

2. Samples, experimental results and discussion

![Figure 1](image1.png)

**Figure 1.** Experimental geometry. Light incidence plane XY, the sample is rotated around Z-axis, magnetic field is applied along Y-axis.

![Figure 2](image2.png)

**Figure 2.** SHG (dotted lines) and nonlinear TMOKE (bubbles) azimuth dependence for pump wavelengths of 830 nm (yellow) and 860 nm (red).

The experimental studies were carried out for the MPC structure consisting of gold film with the thickness of 80 nm and ferromagnetic permalloy (Py) $\text{Ni}_{80}\text{Fe}_{20}$ film with the thickness of 10 nm, consistently sputtered on a one-dimensional polymer diffraction grating with a period of 600 nm located on the glass substrate.
The studies of the SHG and nonlinear TMOKE in MPCs were performed using femtosecond titanium: sapphire laser tunable in the spectral range of 720 nm – 900 nm with the repetition rate of about 80 MHz, power of about 15 mW and the pulse duration of about 60 fs. The p-polarized light beam was incident at the angle of \( \theta = 22.5^\circ \) and focused by lens with \( f = 5 \) cm on the MPC; the reflected light was collected by another lens and second harmonic (SH) signal selected by appropriate color filters was detected by a photomultiplier tube. The experimental studies were carried out under the azimuth rotation of the structure around its normal, while keeping the angle of incidence \( \theta \) constant [Fig. 1]. The structure was located between two permanent neodymium magnets with the field strength of about 2 kOe, leading to the saturation magnetization of the permalloy layer in the MPC. The magnetic field was oriented along the y-axis that is perpendicular to the plane of the light incidence (xz), as shown in Fig. 1.

We revealed that considered structure supports the propagation of the SPPs, which appear as dips in the linear reflection spectrum of light that in our case takes place for both s- and p-polarizations of the incident light. We are focused on the SPP resonances located in the spectral vicinity of 860 nm, which correspond to the surface optical waves localized at the interface of Au/Py/Air and for the diffraction order \( m = -1 \) [see Eq. 2]. We demonstrated that the magnetic contrast of linear TMOKE is the most pronounced for these resonances compared to the SPPs localized at the interface SiO\(_2\)/Au. We revealed that the excitation of SPPs at the fundamental frequency in the considered structure is accompanied by the resonant Fano-type dependence of the SH intensity in the reflected light as the function the azimuth angle \( \phi \). It is shown by dashed curves in Fig. 2 for fundamental wavelengths of \( \lambda = 830 \) nm and \( \lambda = 860 \) nm. In this figure the SPP at the fundamental frequency corresponds to diffraction order \( m = -1 \) [see Eq. 2] and it is excited at the azimuth angle of about \( \phi = 30^\circ \) for \( \lambda = 830 \) nm and \( \phi = 20^\circ \) when \( \lambda = 860 \) nm. We suppose that specific form of the azimuth dependence of the SH intensity stems from the interplay of the local field enhancement of the fundamental light and the interference of SH light waves emitted by various parts of the MPC, where phase of nonlinear light sources changes at the variation of \( \phi \) through the resonant value.

We revealed that the TMOKE in the SH intensity is resonantly enhanced in the close vicinity of the minimum of the SHG, that is close to the azimuth angle of the SPP excitation. This is illustrated by dots in Fig. 2 for both wavelengths. As seen at the variation of the azimuth angle, \( \rho \) abruptly changes with sign reversal. We achieved 20% TMOKE in SHG for fundamental wavelength of \( \lambda = 830 \) nm and 30% for \( \lambda = 860 \) nm, while nonresonant nonlinear TMOKE is about 2 – 5%. Obtained resonant values of TMOKE are significant for the structures where magneto-optical properties are introduced by the magnetooptical material film with thickness of only 10 nm. We emphasize that observed nonlinear magnetic contrast \( \rho \) is by 4 orders of the magnitude larger than the linear magnetic contrast that is about \( 3 \cdot 10^{-3} \) for considered structure. Obtained large values of magnetic contrast confirm superior possibilities of such type of MPC for the magnetic field sensing.

3. Conclusion
In this work we studied the SHG and nonlinear TMOKE in the MPC formed by a one-dimensional dielectric diffraction grating covered by continuous gold and ferromagnetic permalloy films that binds together diffraction-assisted excitation of the SPPs with high figure of merit and the magnetooptics. We revealed resonant effects of the excitation of the SPPs at fundamental frequency in the SHG, which appear as Fano-like shape of the angular dependence of the SH power. We observed that it is accompanied by manifold increase of the TMOKE in the SHG, which reaches the contrast of 30%, giving the opportunity of the application of considered structures for the magnetic field sensing.

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