Intense magnetooptical effect in magnetoplasmonic crystals

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Abstract. Significant increase of the intensity magnetooptical effect (transversal Kerr effect) is observed in transmission for a magnetoplasmonic crystal consisting of the perforated noble-metal film attached to a smooth magnetic dielectric layer. It is largely due to the magnetic field induced shift of the Fano resonances in transmission associated with the surface plasmon polaritons excited at the metal/magnetic-dielectric interface. It is shown that the quasi-guided modes of the magnetic layer also lead to the enhancement of the intensity magnetooptical effect. The considered magnetoplasmonic structures are of great importance for applications in telecommunication and molecular sensing as they also drastically enhance other magnetooptical effects.

1. Introduction

The magnetic field influence on light was discovered by M. Faraday in the mid of the 19th century [1]. Nowadays, magnetooptical effects are considered for nanophotonics applications requiring manipulation of light at gigahertz frequencies [2,3]. A prospective example is the intensity magnetooptical effect - transverse magnetooptical Kerr effect (TMOKE) providing an intensity change of light reflected from a ferromagnetic metal if an applied external magnetic field is reversed [4]. TMOKE allows one to investigate magnetic material properties and can be utilized in magnetooptical data storage [3,5]. For its observation the magnetic field has to be perpendicular to the plane of light incidence. The TMOKE is then characterized by the relative change of reflected light intensity \( I(\mathbf{M}) \) when a medium’s magnetization \( \mathbf{M} \) is reversed:

\[
\delta = \frac{I(\mathbf{M}) - I(-\mathbf{M})}{I(0)}. \tag{1}
\]

This change originates from the magnetic field induced change of boundary conditions at the magnetic layer surface, and attains maximum values for oblique incidence of p-polarized light. Usually, for smooth ferromagnetic metals \( \delta \) is on the order of \( 10^{-3} \) limiting its applicability [3,6]. The...
TMOKE’s counterpart in transmission may also occur, for which a necessary condition is a difference between the opposite magnetic-film boundaries. However, it is difficult to observe because of the small transmission through ferromagnetic metals in addition to its small magnitude [7].

In this paper we investigate both experimentally and theoretically a magnetic structure in which transmission is not vanishing and the TMOKE effect attains significantly enhanced values. It is achieved by using a special metal-dielectric structure – a magnetoplasmonic crystal. It consists of two thin films: a smooth ferromagnetic dielectric and a noble metal periodically perforated with subwavelength slits (figure 1). Surface plasmon polaritons (SPP) can be efficiently excited in this material. So the material combines gyrotropic, plasmonic and nanostructuring features. At the same time its constituents possess large value of the magnetooptical parameter (specific Faraday rotation) due to a ferromagnetic dielectric (bismuth rare-earth iron garnet) and on the other hand they have record small optical losses for the long wavelengths visible and near IR ranges.

Alliance between magnetooptics and plasmonics has been investigated since 70s of the previous century [8-25]. Early papers on the interplay between SPP and magnetooptics addressed SPPs propagating along the smooth surface of a ferromagnetic film [8-9] or along a smooth semiconductor surface in a transverse external magnetic field [10]. In that case, the magnetic field modifies the SPP wave vector but leaves its transverse magnetic polarization (TM) unchanged. An SPP assisted increase of the TMOKE at a ferromagnetic metal surface was reported [11-14] as well as an enhancement of the TMOKE in bimetallic systems consisting of a noble and a ferromagnetic metal [15-17]. Nanostructured plasmonic systems were investigated in [18-23], where a resonance increase of the TMOKE was reported for one-dimensional Co and Fe gratings [18,19], for noble metals in a high external magnetic field [20], and for noble-metal/ferromagnetic-metal multilayers with periodic perforations or protrusions [21,22]. The main disadvantage of most of the aforementioned approaches is that the optical losses associated with the presence of a ferromagnetic metal are relatively high. If the ferromagnetic metals were avoided as in case of pure semiconductor or noble metal systems, huge external magnetic fields exceeding several Tesla would be necessary to make the TMOKE at least comparable with the effect in ferromagnets. In this respect the case of the periodic structures with low loss magnetic dielectric and a noble metal studied in this work seem much more advantageous. This conclusion is supported by the previous theoretical studies [24,25].

In this paper we demonstrate experimentally the SPP assisted significant enhancement of the TMOKE in the magnetoplasmonic crystals and investigate the influence of the quasi-guiding modes of the magnetic film on the TMOKE.

2. Eigenmodes of the magnetoplasmonic crystal
Optical properties of the considered magnetoplasmonic crystal are strongly dependent on the eigenmodes excited in the system by the incident light. These are SPPs propagating along the metal/air and metal/magnetic-dielectric interfaces, cavity modes in the metal grating slits and guiding modes of the magnetic film. Let us consider the influence of these modes on the magnetooptical response and on the TMOKE effect, in particular. At this we characterize the magnetic medium by the permittivity tensor \( \varepsilon^{(m)} \) having the following non-zero components: \( \varepsilon_{11} = \varepsilon_{22} = \varepsilon_{33} = \varepsilon_m, \ varepsilon_{13} = ig, \ varepsilon_{31} = -ig \), where \( g \) is the value of medium’s gyration [3]. At this we assume that the magnetization of the layer is along y-axis (figure 1). The typical values for \( g \) are about 0.001, so further we neglect effects that are nonlinear on magnetization.
Figure 1. Magnetoplasmonic crystal. $k^{(i)}$ is the wave vector of incident light.

Let us start from the SPPs at the horizontal interfaces of the metallic grating. It is well known that excitation of the SPPs in the metal-dielectric structures causes so called Wood anomalies [26]. In the case of sufficiently high grating ($h>50\text{nm}$) their dispersion is well approximated by

$$\kappa = k_o \sqrt{\frac{\varepsilon_{\text{me}}}{1 + \varepsilon_{\text{me}}}}, \quad (2)$$

where $k_o$ is the vacuum wave number and $\varepsilon_{\text{me}}$ is the dielectric constant of the metal. There is no gyrotropic input here so one can expect rather weak influence of the magnetization on the metal/air SPP and consequently there are no any pronounced TMOKE features related to these SPPs excitation.

In this respect, the SPPs at the metal/magnetic-dielectric interface and guiding modes in the magnetic dielectric are more promising since they are localized in the vicinity of the gyrotropic part of the magnetoplasmonic crystal. Considering the magnetic layer placed between the metal without perforation and a substrate the following equations for their dispersion relations can be obtained from Maxwell’s equations with the corresponding boundary conditions:

$$\frac{\gamma_m}{\varepsilon_m} \left( \frac{\gamma_m + \gamma_s}{\varepsilon_m + \varepsilon_s} \right) + \frac{\gamma_m}{\varepsilon_m} \frac{\gamma_s}{\varepsilon_s} + \left( \frac{\gamma_m}{\varepsilon_m} \right)^2 + \frac{g \kappa}{\varepsilon_m^2} \left( \frac{\gamma_m}{\varepsilon_m} - \frac{\gamma_s}{\varepsilon_s} \right) \tan(\gamma_m h_m) = 0 \quad (3a)$$

for the SPPs and

$$\frac{k_{zm}}{\varepsilon_m} \left( \frac{\gamma_m + \gamma_s}{\varepsilon_m + \varepsilon_s} \right) + \frac{\gamma_m}{\varepsilon_m} \frac{\gamma_s}{\varepsilon_s} + \left( \frac{k_{zm}}{\varepsilon_m} \right)^2 - \frac{g \kappa}{\varepsilon_m^2} \left( \frac{\gamma_m}{\varepsilon_m} - \frac{\gamma_s}{\varepsilon_s} \right) \tan(k_{zm} h_m) = 0 \quad (3b)$$

for guiding modes, where $h_m$ is the magnetic layer height, $\gamma_i = (\kappa^2 - \varepsilon_i k_o^2)^{1/2}$, $i = m, \text{me}, s$, $k_{zm} = (\varepsilon_m k_o^2 - \kappa^2)^{1/2}$, $k_o = \omega \varepsilon_m^{-1}$, index ‘s’ refers to the substrate. It follows from (3) that the propagation constant $\kappa$ of both types of eigenmodes is dependent on magnetization.

If the magnetic layer is rather thin the SPPs dispersion law is close to one for the interface between the metal and the substrate: $\kappa = k_o \left[ \varepsilon_s \varepsilon_{\text{me}} (\varepsilon_s + \varepsilon_{\text{me}})^{-1} \right]^{1/2}$, so it is almost magnetization independent. With the increase of $h_m$ the influence of magnetization on the SPP dispersion gets more pronounced according to (3b) and also the optical spectra acquire additional features related to the excitation of guiding modes that are also magnetic dependent as seen from (3a). If $h_m$ exceeds the
value \[ [-\left(\epsilon_{me} + \epsilon_m\right)]^{1/2}(k_0\epsilon_m)^{-1} \] that is about hundred nanometers the SPPs dispersion becomes independent of the layer height and the dispersion law (3a) takes the following form:

\[
\kappa = k_0\epsilon_m\epsilon_{me}^{-1}\left(1 + g\left(-\epsilon_m\epsilon_{me}^{-1}\right)^{1/2}\left(1 - \epsilon_m^2/\epsilon_{me}^2\right)^{-1}\right). \tag{4}
\]

The essential point here is the effect of magnetooptical nonreciprocity. It implies that the dependence of the SPP and guiding modes propagation constants on magnetization is odd and hence for fixed magnetization the two waves propagating in opposite direction have different phase velocities.

If an eigenmode is excited by an incident light it leads to appearance of the resonant features in optical reflection and transmission spectra. For that the projection of the incident wave number on the sample surface \( k_x = k_0\sin\theta \), where \( \theta \) is the incident angle, should be equal to the eigenmode’s propagation constant \( \kappa \). As the latter is now dependent on magnetization the resonant wavelengths are shifted with respect to the non-magnetic case. Moreover, according to magnetooptical nonreciprocity the shifts are of opposite signs for opposite directions of magnetization. That leads to emergence of the TMOKE described by (1). Its value is dependent not only on the shift value but also on the shape of the resonance curve. It can be shown that the TMOKE value is proportional to the derivative of reflection/transmission on wavelength, so the spectral shape of the TMOKE resonance has negative minimum followed by positive maximum or visa versa. The TMOKE value can be rather high in comparison with that in case of a smooth magnetic film.

For the case of the plasmonic crystal depicted in figure 1 the excitation condition for eigenmodes is:

\[
\kappa = k_0\sin\theta + mG, \tag{5}
\]

where \( G = 2\pi d^{-1} \) is the reciprocal lattice vector, \( d \) is the period of the plasmonic crystal and \( m \) is an integer. In this case the eigenmodes are leaky and they also contribute to the transmitted wave in the far field. That is why, it is more correct to call the guiding waves the quasi-guiding ones. The correspondent resonant features in transmission and reflection spectra have the typical shape consisting of minimums followed by maximums or visa versa. The shapes of this kind are called Fano resonances [27]. They appear in cases when a resonant process is accompanied by a non-resonant one. In our case the former is the eigenmode excitation via one of the diffraction orders and the latter is the wave propagating through the structure.

The dispersion laws for the quasi-guiding modes and the SPPs are influenced by periodicity and the slits in the metal grating. However, if the slits are rather narrow being compared to the eigenmode wavelength the empty lattice approximation can be applied, according to which \( \kappa \) can be estimated from (2), (3) or (4). Near the centre and the edges of the Brillouin zone (\( \kappa = mG/2 \)) this approximation is not applicable as the wave propagation is strongly influenced by periodicity. The excitation of the quasi-guiding modes and the SPPs at metal/magnetic interface is accompanied by resonant enhancement of TMOKE as described above. Due to the periodicity of the structure nonreciprocal propagation is prohibited at the centre and the edges of the Brillouin zone, so TMOKE near these points of the Brillouin zone vanishes.

Besides there are eigenmodes of another kind that are Fabry-Perot cavity modes. Since they are localized mainly inside the slits they are almost magnetization independent and don’t significantly contribute to TMOKE.

To approve the above qualitative analysis we need to conduct a comprehensive numerical modelling of the electromagnetic properties of the structure. It is accomplished on the basis of the rigorous coupled waves analysis (RCWA) extended to the instance of gyrotropic materials [28-29]. At this, special factorization rules are used for the improvement of the algorithm’s convergence.
3. Magnetoplasmonic sample and experimental set up

Magnetic part of the magnetoplasmonic structure is a 2.5 micron thick bismuth-substituted rare-earth iron garnet film of composition Bi$_{0.4}$(Y,Gd,Sm,Ca)$_{2.6}$(Fe,Ge,Sn)$_5$O$_{12}$ grown by liquid phase epitaxy from Bi$_2$O$_3$: PbO : B$_2$O$_3$ melt on the Gd$_3$Ga$_5$O$_{12}$ substrate with orientation (111). The film possesses uniaxial magnetic anisotropy in the direction perpendicular to the film plane. The specific Faraday rotation is 0.46 deg/micron at wavelength 633 nm. The magnetoplasmonic sample of structure shown in figure 1 was fabricated by the thermal deposition of the gold layer on the bismuth-substituted rare-earth iron garnet film and subsequent electron beam lithography combined with the reactive ion etching in Ar plasma. The sample was characterized by AFM and SEM imaging. The grating parameters obtained are gold layer height $h$ is 150 nm, period $d$ is 595 nm and the air groove width $r$ is 110 nm.

For magnetooptical measurements we used a halogen lamp as a source of white light. The collimated light was focused at the sample into a spot with diameter of about 300-500 µm and aperture angle below 1°. In order to perform the measurements at different angles of light incidence the sample was mounted on rotation stage. Zero order transmission signal was spectrally detected with a single monochromator (linear dispersion 6.28 nm/mm) equipped with charged coupled device detector. The overall spectral resolution was below 0.3 nm. The polarization direction of the transmitted light was selected with a polarizer in the detection path. The magnetic fields up to 4 kOe were applied in transverse geometry using water cooled electromagnet.

4. Extraordinary TMOKE related to SPPs

Results of the experimental observation of TMOKE in zero order transmitted light are shown in figure 2 for different angles of light incidence.

![Figure 2](image-url)

Figure 2. Experimentally measured transmission (thick black curves) and TMOKE (thin red curves) for three different incidence angles: (a) $\theta =0.8^\circ$; (b) $\theta =5^\circ$; (c) $\theta =10^\circ$. Light is p-polarized and is incident perpendicularly to the slits.
Since height of the magnetic film in the investigated sample is rather large ($h_m=2.5 \, \mu m$) influence of the guided modes is not noticeable and main features of the spectra are related to SPPs and cavity modes. Indeed, it follows from (2) and (4) that the Fano resonance (A) is due to the SPP of the zero order at the air/gold interface, while the Fano resonances (B) and (C) are related to the SPPs of the second order at the gold/magnetic-film interface. The prominent transmission peak (D) is attributed to the collective Fabry-Perot cavity mode inside the slits.

To ensure that sample magnetization is nearly in-plane relatively large external magnetic field of 2000 Oe was applied. It can be seen that away from resonances $\delta$ is very small. Actually, $\delta$ cannot be measured experimentally which means that it does not exceed $10^{-3}$. On the contrary, at the resonances $\delta$ reaches $1.5 \cdot 10^{-2}$ demonstrating an increase by at least one order of magnitude. Electromagnetic modeling for the nonresonant case gives $\delta \approx 10^{-4}$. If compared to the case of uncovered bare iron garnet film the enhancement factor is even larger, namely it is up to $10^3$. So we can announce that a giant TMOKE in transmission is observed. The effect can be made larger if a magnetic film with higher specific Faraday rotation is used [30].

It is apparent that no noticeable TMOKE increase is observed for other resonant regions. It goes exactly in line with the discussion above and highlights the TMOKE’s sensitivity to excitation of different eigenmodes. For normal incidence the TMOKE is zero because of the full symmetry of the incident light with respect to the structure. As soon as the incidence is not normal the symmetry is broken and two SPP propagating into opposite directions are excited at slightly different frequencies and the TMOKE appears. The value of $\delta$ is almost $10^{-2}$ even if the incidence angle is as small as 0.8° (figure 2(a)). At the transition from normal to slightly oblique incidence transmission spectrum does not change noticeably while TMOKE spectrum does. It demonstrates TMOKE sensitivity to slight changes of the illumination conditions. If the incidence angle is larger (e.g. it is 5°, figure 2(b)) two eigenfrequencies of the SPP propagating in opposite directions differs significantly and give birth to two Fano resonances in transmission. The TMOKE accompanies both resonances, but $\delta$ has opposite signs at them reflecting the fact that these resonances are due to the SPP propagating in opposite directions. So the sign of the TMOKE at the magnetoplasmonic crystals is also sensitive to the direction of the SPP propagation. Further increase of the incidence angle shifts the plasmonic resonances further apart, but the value of the TMOKE remains almost steady (figure 2(c)).

5. Transmission and TMOKE versus height of the magnetic film
The results of the numerical simulations showing the transmission and TMOKE dependence on the magnetic layer height are presented in figure 3.

![Figure 3](image-url)
Figure 4. The dispersion curves for guiding modes of magnetic layer calculated from (3b) and (5) for \( m = \pm 2 \).

The principal features on the TMOKE spectrum (figure 3(b)) are series of resonances that are dependent on the magnetic layer height and they are accompanied by transmission dips. These are the contribution of the magnetic layer quasi-guiding modes excited by ±2nd diffraction orders. Their excitation conditions estimated from (3b) and (5) are shown in figure 4 and there is rather nice correspondence with transmission and TMOKE resonances. The difference can be explained by the only approximate applicability of the empty lattice approximation.

According to (3b) and (5) each quasi-guiding mode in the plasmonic crystal is characterized by two integers \( m \) and \( n \), the former denoting the diffraction order via which the mode is excited and the latter coming from solution of (3b) for fixed \( m \). Within the spectral range shown in figures 3 and 4 the quasi-guiding modes only with \( m = \pm 2 \) are excited. It should be noted that opposite signs of \( m \) correspond to opposite directions of modes propagation and hence according to the discussion above the signs of TMOKE are opposite for \( m = 2 \) and \( m = -2 \), that is clearly seen in figure 3(b). For the case of normal incidence the difference in dispersion between modes with the same values of \( n \) but opposite values of \( m \) vanishes and so does TMOKE since this situation corresponds to the quasi-momentum \( \kappa = 0 \) from the first Brillouin zone (see Section 2).

As we have already discussed in Section 4 the other features in transmission spectrum (figure 3(a)) don’t demonstrate strong magnetooptical response. One of them is increase in transmission at wavelengths about 680 nm that is independent of the magnetic layer thickness. It is Fabry-Perot resonance inside the slits of metallic grating. The others are transmission dips at wavelengths about 610 and 650 nm. They are related to the SPPs excitation at the metal/air interface and therefore they don’t contribute much to TMOKE.

It should be noted that for rather large values of magnetic layer height the excitation of modes is less effective. Moreover, if the structure is illuminated with light with small coherence length the modes are not excited at all, so in our experiments they were not observed.

6. Conclusion

We demonstrate experimentally that the magnetooptical effect can be drastically increased via excitation of the SPPs in the magnetoplasmonic crystals – periodic structures consisting of metal and magnetic dielectric perforated layers. The increase of the TMOKE in transmission is measured to be up to tree orders of magnitude if compared with a smooth magnetic dielectric film. The main reason for this enhancement is the magnetization induced shift of the SPP associated Fano resonances in transmission spectra. This shift makes more significant input in the TMOKE signal for larger gradients of the Fano resonances. However, noticeable influence of the SPPs on the TMOKE is observed only...
for the SPP propagating along the metal/magnetic-dielectric interface. Plasmonic excitations at the other interface hardly contribute into the TMOKE enhancement because they are not affected much by the gyrotropic properties of the underlying magnetic layer.

The TMOKE increase takes also place if quasi-guided TM modes of the magnetic layer are excited. Similarly to the SPPs their resonance frequency is linearly dependent on the magnetic layer transverse magnetization.

So TMOKE in magnetoplasmonic crystals is sensitive to the type of the eigenmodes of the structure. Moreover, it changes sign for SPPs or quasi-guiding modes traveling in opposite directions. Thus, TMOKE can become an important tool for complete characterization of plasmonic nanostructures.

Even though here we paid here only attention to TMOKE, our previous study [31] allows us to claim that the proposed class of magnetoplasmonic heterostructures can be used to enhance also other magnetooptical effects such as the Faraday effect or the longitudinal Kerr effect. With these parameters the structures are very promising for applications in ultra high sensitivity devices and optical data processing.

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