Tuning of the transverse magneto-optical Kerr effect in magneto-plasmonic crystals

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Abstract. The spectral properties of the transverse magneto-optical Kerr effect (TMOKE) in periodic metal–dielectric hybrid structures are studied, in particular with respect to the achievable magnitude. It is shown that the TMOKE is sensitive to the magneto-optical activity of the bismuth-substituted rare-earth iron garnet, which is used as a dielectric material in the investigated structures. For samples with larger Bi substitution level and, consequently, larger gyration

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constant, the magnitude of the TMOKE increases and reaches 13% in the case of a Bi$_{1.8}$Lu$_{1.2}$Fe$_{3.6}$Al$_{1.4}$O$_{12}$ magnetic film. Further, it is demonstrated that the TMOKE vanishes at the high-symmetry points of the Brillouin zone (at the $\Gamma$ and X points). The main enhancement of the TMOKE takes place near the resonances of the surface plasmon polaritons (SPPs) at the metal/magnetic–dielectric interface. However, near the degenerate resonances of the SPPs at the air/metal and metal/magnetic–dielectric interfaces the TMOKE is increased by the air/metal SPPs as well. This phenomenon is explained in terms of a coupled oscillator model.

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1. Introduction

The amplitude, phase and polarization of light passing through a magnetized medium can be modulated by the medium's magnetization [1, 2]. On the other hand, it was demonstrated recently that one can manipulate the medium’s magnetization optically on sub-nanosecond time scales [3]. This makes magneto-optics quite promising as a tool for controlling light in modern optical devices.

Magneto-optical effects that provide modulation of light intensity are represented by the transverse magneto-optical Kerr effect (TMOKE). The TMOKE is observed when obliquely incident light illuminates a magnetic film magnetized in-plane and perpendicular to the incidence plane. The TMOKE is characterized by the parameter $\delta$, which represents the relative change in the intensity $I$ of the light reflected by the medium when its magnetization $M$ is reversed:

$$\delta = \frac{I(M) - I(-M)}{I(0)}.$$  (1)

The TMOKE allows one to investigate the magnetic properties of a medium and can be utilized in magneto-optical data storage [2, 4]. The relative change in the reflected light intensity due to the TMOKE in ferromagnets such as nickel or cobalt is of the order of $10^{-3}$, limiting TMOKE applicability [2]. Recently, several concepts were proposed to resonantly increase the TMOKE [5–10]. Specifically, these concepts involve surface plasmon polaritons (SPPs)—coupled oscillations of the electromagnetic field and the electron plasma in a metal,
which propagate along a metal/dielectric interface [11]. The combination of plasmonics and magneto-optics looks mutually beneficial. On the one hand, the magnetic field is an efficient tool to control SPPs [12, 13], and on the other hand SPPs can be utilized to enhance magneto-optical effects [14–30].

Early papers on the interplay between SPPs and magneto-optics addressed SPPs propagating along the smooth surface of a ferromagnetic film [31, 32] or along a smooth semiconductor surface in an external transverse magnetic field [33, 34]. In that case, the magnetic field modifies the SPP wave vector, but leaves the polarization of the SPP unchanged.

Periodic perforation of metal layers provides Fano resonances in transmission or reflection spectra due to SPP excitation [35], which are usually referred to as Wood anomalies [36]. The appearance of Fano resonances in the spectra of the perforated metallic ferromagnets gives a several times enhancement of the TMOKE [5, 21, 37]. Owing to a periodicity comparable to the SPP wavelength, such structures can be referred to as magneto-plasmonic crystals (MPCs). However, ferromagnetic metals introduce optical losses that are too large to be acceptable in practical applications. Reduction of optical losses was achieved in bimetallic systems of noble and ferromagnetic metals, either smooth or perforated [6, 7, 22, 23, 38–41]. The SPPs can propagate relatively long distances along the noble metal/air interface and they enhance the TMOKE, which takes place at the ferromagnetic surface. This concept provided further enhancement of the TMOKE by several times. In [13], a Mach–Zehnder interferometer was used to demonstrate that minute magneto-optical phase shifts of the SPPs in a gold/cobalt bilayer structure could be detected. Coexisting SPPs and localized surface plasmons were studied in Au/Co/Au trilayer films covered by Au nanodiscs with a SiO$_2$ spacer [7].

Another approach demonstrated recently by Belotelov et al [8, 10, 15] is to substitute the ferromagnetic metal by a noble metal layer with low optical absorption and to introduce a magnetic–dielectric layer into the MPC. It allows the SPPs to propagate with relatively low losses right at the magnetized interface without any spacing which makes the TMOKE even larger [8]. This enhancement is mediated by the SPPs at the metal/magnetic–dielectric interface leading to Fano resonances in the transmittance and reflectance spectra that are sensitive to a transverse magnetic field.

Here, we concentrate our studies on several features of the TMOKE in MPCs of similar structure consisting of a gold grating on top of a magnetic layer of bismuth-substituted rare-earth iron garnet on a gadolinium gallium garnet (GGG) substrate. In particular, we compare the TMOKE signal strength in the MPCs comprising magnetic layers of different magneto-optical activity. Further, we investigate the TMOKE dispersion in the vicinity of some specific points of the first Brillouin zone. These points include the center and the edges of the Brillouin zone and points where the dispersion curves of the SPPs propagating along two surfaces of the metal grating intersect. Finally, we compare TMOKE data measured in reflection and transmission geometries.

2. Experimental technique

Each MPC sample under study in this paper consists of a nanometer-sized gold grating on top of a bismuth iron garnet (BIG) magnetic film of few micrometers thickness deposited onto a GGG substrate. Apart from minor differences in the grating parameters and fabrication techniques, the major difference between the samples lies in the relative amount of bismuth ions (per stoichiometric formula unit) inside the magnetic films, which influences the magneto-optical
Table 1. Overview of the magneto-optical film compositions, fabrication techniques and magneto-optical properties along with gold-grating geometrical parameters of the investigated MPC samples. \( d \) is the gold grating period, \( r \) is the slit width of the gold grating, \( h_m \) is the thickness of the gold grating and \( h_d \) is the magnetic film thickness. Diagonal (\( \epsilon \)) and non-diagonal (gyration \( g \)) terms of the permittivity tensor are given at photon energy 1.55 eV.

| Sample | Chemical composition | Fabrication technique | \( \epsilon \) (nm) | Geometrical parameters (nm) |
|--------|----------------------|-----------------------|---------------------|----------------------------|
| 1      | Bi\(_{1.4}\)Lu\(_{1.2}\)Fe\(_{1.6}\)Al\(_{1.4}\)O\(_{12}\) | RF magnetron sputtering | \( \epsilon = 5.29 + 0.017i \) | \( d = 605 \) |
|        |                      |                       | \( r = 160 \) | \( h_m = 125 \) |
|        |                      |                       | \( h_d = 905 \) | \( h_m = 125 \) |
| 2      | (BiSmTm)\(_3\)(FeGa)\(_5\)O\(_{12}\) | Liquid phase epitaxy | \( g = 0.0051 \) | \( r = 85 \) |
|        |                      |                       | \( h_m = 100 \) | \( h_d = 5095 \) |
|        |                      |                       | \( d = 505 \) | \( h_m = 125 \) |
| 3      | Bi\(_{0.4}\)(YGdSmCa)\(_2\)d(FeGeSi)\(_5\)O\(_{12}\) | Liquid phase epitaxy | \( g = 0.0017 \) | \( r = 110 \) |
|        |                      |                       | \( h_m = 120 \) | \( h_d = 2500 \) |

Properties of the garnets [42]. The bismuth substitution level can be seen to be decreasing from sample 1 to 3 in table 1. A sketch of the principle design is given in figure 1(a).

For the magneto-optical measurements, we used a tungsten halogen lamp with stability better than 0.1%. The light was imaged onto a 100 \( \mu \)m pinhole in order to establish a point source of white light. The transmitted light was then collimated with an achromatic lens (focal distance of \( f = 150 \) or alternatively 300 mm) and focused onto the sample using a second achromatic lens with a focal distance of \( f_2 \leq f_1 \). A diaphragm has been used between the doublets in order to reduce the maximum angle of the incident light cone to below 1\(^\circ\). The light was focused on the sample into spots with diameters from 50 to 300 \( \mu \)m, depending on the dimensions of the grating patterns. To perform measurements at different angles of light incidence, the sample was mounted on a rotation stage. The zero-order transmission and reflection signals were spectrally dispersed with a single monochromator (linear dispersion 4.8 nm \( \text{mm}^{-1} \) at 900 nm) and detected with a charge-coupled device camera. The overall spectral resolution was about 0.5 nm. The p-plane linear polarization of the light was established by a polarizer in the excitation path, i.e. before the sample. Magnetic fields up to \( B = 180 \) mT were applied in transverse geometry using a water cooled electromagnet. During measurement the sample was kept at room temperature.

Figure 1(b) represents a typical transmission spectrum, which exhibits a characteristic Fano feature corresponding to an SPP resonance. The transmitted or reflected light intensities \( I(E) \) near a Fano resonance are given by the sum of a resonant \( I_r \) and a non-resonant \( I_{nr} \) contribution:

\[
I(E) = I_r(E) + I_{nr},
\]

where \( E = \hbar \omega \) is the photon energy. The resonant channel is due to plasmon excitation, while the non-resonant one represents direct transmission or reflection. The resonantly scattered wave
Figure 1. (a) The magneto-plasmonic heterostructures consist of a gold grating on top of a planar ferromagnetic–dielectric (bismuth iron garnet film) grown on a non-magnetic substrate (gadolinium gallium garnet). The parameters of the gold film for the three samples are listed in table 1. The sketch shows the TMOKE geometry, i.e. $\mathbf{M}$ is perpendicular to the plane of incidence and is in the sample plane. Incident light is p-polarized, i.e. its electric field $\mathbf{E}$ lies in the plane of incidence. (b) An exemplary Fano feature in the transmission spectrum (circles) of sample 1 for $\theta = 0^\circ$ incidence. The data has been fitted with equation (3) (solid curve), which gives the resonance position and width of $\hbar \omega_i = 1.64$ eV and $\Gamma_i = 0.06$ eV, respectively (cf figure 2(a)).

experiences a phase shift of $\pi$ across the resonance energy, resulting in constructive and destructive interference with the non-resonantly scattered light and thus the characteristic asymmetric lineshape of the features in the transmission and reflection spectra (see figure 1(b)) [35].

In order to determine the position of the SPP resonances in the transmission and reflection spectra, they have been approximated by a sum of up to three Fano shapes located at different energies:

$$I = \sum_{i=1}^{3} A_i (1 + E_i Q_i)^2 / (1 + E_i^2) + D$$

(3)

with $E_i = (\omega - \omega_i) / (\Gamma_i / 2)$ and $D = D_0 + D_{\text{lin}} \omega$. Here, $A_i$, $\omega_i$, and $\Gamma_i$ are the amplitude, position and width of resonance $i$, while $Q_i$ is a parameter describing its degree of asymmetry, which depends on the ratio of the intensities of the two interfering resonant and non-resonant channels. All other contributions with weak spectral dependence are taken into account by $D$. The number of Fano resonances in equation (3) corresponds to the maximum of three branches of SPP dispersions that are simultaneously present in the energy range of the transmission/reflection spectra (see figure 4(a)).

For TMOKE measurements the magnetic field was applied along the MPC slits and perpendicular to the incidence plane (see figure 1(a)). The spectral dependence of $\delta$ has been evaluated using equation (1) where the transmission or reflection spectra measured for different directions of the magnetic field $B$, i.e. of the magnetization $M$, have been used.
3. Dependence of the transverse magneto-optical Kerr effect (TMOKE) magnitude on the bismuth substitution level

Figure 2(a) shows the transmission spectrum of sample 1 for incidence angles between $\theta = 0^\circ$ and $12^\circ$. The transmitted intensity normalized to the spectral distribution of the illuminating white light source is color coded and given in per cent. The position of the resonances corresponding to SPPs excited on the gold/magnetic–dielectric interface by the $\pm 2$nd diffraction orders are indicated by open circles. These data points were obtained by fitting the individual transmission features by the Fano formula (equation (3)). The lines serve as guides to the eye to illustrate the energy dispersion of the two modes.

The TMOKE signal of this sample is depicted in figure 2(b) for both positive and negative incidence angles $\theta$. A comparison with figure 2(a) shows that the positions of the TMOKE resonances follow closely the positions of the plasmonic resonances. This is because the observed change in transmission upon application of an external magnetic field, given by the parameter $\delta$, is induced by a shift of the plasmonic resonance. The magnetization $M$ of the magnetic film leads to a change of the wave number $k$ of the plasmonic mode. In the
Figure 3. Magnetic field dependence of the TMOKE in transmission measured under oblique incidence for three different samples with varying bismuth content $c_{\text{Bi}}$. The amplitude of a particularly strong TMOKE peak for each sample is shown. The maximum attainable value of the effect rises with increasing bismuth content. The solid lines are used to guide the eye.

In order to illustrate the origin of the higher TMOKE values, we compare its magnetic field dependence in samples with different bismuth substitution levels. Figure 3 shows the dependence of the maximum achievable TMOKE amplitude in dependence of the applied external magnetic field $B$ for samples 1–3 (see table 1).

All three curves show a progression characteristic for the magnetization curve in ferromagnetic BIG materials: for low-field strengths, there is a linear dependence of the effect on the applied magnetic field since the magnetic domains, originally oriented along the easy axis, are turned along the external field. Further increase of the magnetic field leads to a saturation of the magnetization $M$ and thus of the observed TMOKE. The saturation values for the effect grow with the bismuth substitution level. Sample 3 has a small amount of bismuth and therefore its magneto-optical activity is weak. As a result the TMOKE is limited to a maximum value of 1.5% in this sample. For sample 2, whose bismuth substitution level is $c_{\text{Bi}} \approx 1$, the magnetic...
field dependence is given by the red dots. For this sample the saturation value of the TMOKE amplitude lies around 7%. The largest TMOKE amplitude is measured for sample 1 amounting to 13% under saturated magnetization.

The experimental data clearly demonstrate that the magnitude of the TMOKE induced by a magnetic medium is dependent on its gyration \( g \), i.e. on its magneto-optical activity, which can be tailored by optimal choice of chemical composition. The other important aspect is the value of the saturating magnetic field. It is closely related to the magnetic anisotropy of the magnetic film and depends on the composition as well as on the thickness. The magnetic films of samples 2 and 3 have a uniaxial magnetic anisotropy perpendicular to their surfaces, while the magnetic film of sample 1 has a uniaxial magnetic anisotropy that forms a small angle with the sample surface. Because of the differences in their composition their saturating magnetic fields vary. Sample 2 has the smallest one of about 60 mT.

4. TMOKE related to surface plasmon polaritons from different interfaces: spectral tunability

While the achieved TMOKE magnitude values are already on the scale that would be manageable in practical device applications, further potential of MPCs lies in the interaction of both of its SPP modes, which are provided owing to the two interfaces of the noble metal. Here, it is interesting to investigate the effect of possible coupling between SPPs from different interfaces on the TMOKE. This coupling should take place in the regions of intersecting SPP dispersions. Here, we concentrate on sample 2 with moderate bismuth substitution level whose plasmon dispersions show the desired intersections within the first Brillouin zone (1.BZ). Transmission and reflection spectra have been measured across a wide range of incidence angles and are summarized in figure 4. The fitting results obtained using equation (3) are shown in figure 4(a) as full (reflection) or open (transmission) circles for the Au/BIG SPPs (red) and the Au/air SPPs (black) for different angles of the 1.BZ. The results are in good agreement with a Drude model calculation of the SPP dispersions in the empty lattice approximation, which is shown in figure 4(b). Exemplary spectra as well as the corresponding Fano fits can be found in figures 4(c)–(e). However, a closer look reveals a deviation from the model at photon energies around 1.4 eV, where the experiment reveals an anti-crossing and a subsequent splitting of the branches of the SPP dispersions at the intersection, which is characteristic of strong coupling.

Corresponding TMOKE measurements are shown in figures 5(a) and (b) for angles close to the center and the edge of the 1.BZ.

The strength of the externally applied magnetic field was held constant at \( B = 170 \text{ mT} \). Remarkably, the TMOKE amplitude vanishes at both symmetry points, i.e. at \( \theta = 0^\circ \) and \( 61^\circ \). At these points, two SPPs are excited simultaneously having opposite wavenumbers \( k \). Consequently, the TMOKE for each of the SPPs differs in sign and cancels to zero (see also [8]). The shift of the signals at angles larger than zero, indicated by the red dashed lines, follows the Au/BIG SPP dispersion shown in figure 4(a). Additionally, at the intersection of both SPPs, figure 5(b) clearly shows TMOKE features that shift according to the Au/air SPP (black dashed line), whose resonances are expected to be independent of the magnetization of the dielectric.

The presence of the effect at the air/metal SPP for photon energies in the vicinity of the metal/magnetic–dielectric SPP excitation can be understood in terms of a coupled oscillator model. In the framework of this model, the SPPs at the two interfaces of the metallic grating are considered as two oscillators with their coupling coming from the finiteness of the grating.
Figure 4. Dispersion relations of the SPPs at the Au/BIG (red) and Au/air (black) interfaces of sample 2 within the first Brillouin zone (a) as obtained by fitting equation (3) to the SPP features in the transmission (see figures 4(c) and (e)) and reflection (see figure 4(d)) spectra and (b) by means of a Drude model calculation for a grating period $d$ and the dielectric constant of the magnetic film $\varepsilon_d = 4.5$. The intersection of the SPP dispersion curves leads to a splitting, i.e. anti-crossing around an incidence angle of $40^\circ$ in figure 4(a). Selected transmittance and reflectance spectra are shown in figures 4(c) and (e), as well as figure 4(d), which also give the results of the Fano fits (red curves). The spectra are shifted for clarity. The dashed lines correspond to zero level.

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thickness and from the slits. The frequencies of the coupled modes are given by

$$\omega_\pm^2 = \frac{1}{2} (\omega_d^2 + \omega_a^2 \pm \sqrt{(\omega_d^2 - \omega_a^2)^2 + 4 \eta_1 \eta_2}),$$

(5)

where $\omega_d$ and $\omega_a$ are the eigenfrequencies of the oscillators, i.e. the frequencies of the metal/magnetic–dielectric and air/metal SPPs, respectively, and $\eta_{1,2}$ are the coupling coefficients depending on the grating parameters. If $\omega_d$ and $\omega_a$ differ significantly so that $|\omega_d^2 - \omega_a^2| \gg \sqrt{\eta_1 \eta_2}$, then the SPPs at the two interfaces exist almost independently of each other and $\omega_\pm \approx \omega_d, \omega_a$. As $\omega_d$ and $\omega_a$ approach each other, the coupling between the SPP modes increases and their frequencies $\omega_\pm$ become dependent on both $\omega_d$ and $\omega_a$. The resonance frequency $\omega_d$ depends on the magnetization according to equation (4), leading to a dependence of the frequencies of both modes defined by equation (5) on the magnetization. That is why, in the vicinity of the crossing of the dispersion curves for the air/metal and metal/magnetic–dielectric SPPs, the TMOKE is enhanced at either of the modes. As will be discussed further, this
Figure 5. The TMOKE measured in transmission with sample 2 at incidence angles (a) close to \( k = 0 \) (from 0\(^\circ\) to 10\(^\circ\)) and (b) from 41\(^\circ\) to 61\(^\circ\), i.e. close to the edge of the first Brillouin zone. The dashed lines are guides to the eye representing the shift of the TMOKE features. Surprisingly, close to the intersection of the SPP dispersions, the curves show features whose shift can be attributed not only to the Au/BIG magneto-plasmon (red dashed lines) but also to the Au/air SPP (black dashed line, compare figures 4(a) and (b)). At both \( k = 0 \) as well as at the 1.BZ edge, the TMOKE amplitude is zero due to symmetry considerations. The contour plot in (c) shows the calculated near field spatial distributions of the square of the absolute value of the magnetic field \( |H|^2 \) at \( \theta = 49^\circ \) and photon energy 1.38 eV.

Enhancement is the result of the magnetization induced shift of the plasmonic resonance rather than of vanishing transmittance.

The maximum amplitude of the TMOKE in this sample is obtained at 1.4 eV and 47\(^\circ\), i.e. in the vicinity of the SPP anti-crossing, reaching 8.5\%. Additionally, in the same range of angles, enhanced optical transmission is observed, which exceeds 25\%. This value, which is considerably higher than would be expected from direct transmission through the grating slits, involves tunneling of the air/metal SPP to the metal/magnetic–dielectric interface due to the finite grating depths and subsequent radiative scattering into the forward direction [43].

Figure 5(c) gives an example of the optical magnetic field distribution \( |H|^2 \) near the grating (gray box) in the vicinity of the SPP anti-crossing (\( \theta = 49^\circ \) and photon energy 1.38 eV). It supports the above reasoning by showing simultaneous excitation of the air/metal and metal/magnetic–dielectric SPPs.
5. Transmission versus reflection

Although the enhancing mechanism of the TMOKE by SPPs has already been elucidated (see section 3 and [8]), the observed phenomena can be unambiguously identified as surface effects through measurements of the reflectivity from the gold side of the MPCs. This way, contributions from the bulk BIG can be excluded.

Figure 6 gives a comparison of the TMOKE in transmission (figure 6(a)) and in reflection from the top gold grating (figure 6(b)) at incidence angles of 31° and 33°. In full accord and with our expectations, the effect is observed in both reflection and transmission geometries, resembling each other in shape and position. The magnitude is reduced in reflection by a factor of 3, which can be explained as follows. The value of TMOKE is in first order proportional to the magnetization, i.e.

\[ \delta(E, M) = a(\partial I(E)/\partial E)M/I(E) \]

with a proportionality constant \( a \) (see e.g. [8]). In the vicinity of a Fano resonance, \( I(E) \) is given by equation (2), where \( I_r(E) \) can be expressed by a normalized function \( S(E) \) and an amplitude \( I_0 \) in the following way

\[ I_r(E) = I_0 S(E). \]

This leads to

\[ \delta(E, M) = \frac{a I_0 M(\partial S(E)/\partial E)}{I_0 + I_0 S(E)}. \]  

(6)
In transmission spectra, \( I_{nr} \) is usually much smaller than \( I_0 \), which makes \( \delta_T(E, M) \) almost independent of the transmitted light intensity: \( \delta_T(E, M) = a M (\partial S(E)/\partial E)/S(E) \). This proves that the enhancement of the TMOKE is due to the magnetization induced shift of the Fano resonance rather than due to low values of the transmitted light intensity. On the other hand, the contribution of \( I_{nr} \) in reflection is more pronounced and since it enters in equation (6) in the denominator it diminishes the TMOKE value.

Figure 6(b) also reveals a decreased linewidth as compared to the transmission signals, which is due to the different Fano resonance shapes of transmittance and reflectance (see figures 4(c) and (d)). The curves are superimposed by high-frequency oscillations with a period of about 50 meV originating from Fabry–Perot interferences inside the magnetic film of thickness \( h_d = 5.095 \mu m \). These oscillations are weaker in reflection with an amplitude at least one order of magnitude smaller than the TMOKE peaks. Consequently, the part of the reflected light that penetrates the bulk film as well as a possible contribution to the signal can be neglected.

6. Conclusions

The results illustrate the importance of MPCs for applications involving the TMOKE in either transmission or reflection geometries. It has been shown that the magnitude of the effect can reach up to 10% and even higher depending on the bismuth substitution level of the magneto-optically active layer. At the same time, these magnitudes can be achieved at low magnetic fields and in nanometer scale volumes due to the small saturation magnetic field (\( \leq 100 \text{ mT} \), see figure 3) and the high confinement of the SPPs to the film surfaces. Since the described phenomena are localized to few hundreds of nanometers in the direction perpendicular to the interfaces, corresponding to the skin depth of the SPPs in the adjacent materials, rather than being bulk phenomena, material costs can be largely reduced leading the way to miniaturized magneto-optical applications, such as all-optical switches and modulators. Furthermore, the results demonstrate that the effect can be arbitrarily tuned to energies, which are easily accessible by a broad range of commercially available laser systems, by a simple rotation of the sample and/or by appropriate design of the grating parameters. Despite the presumably advantageous transmission geometry for future applications, the reflection geometry provides one additional degree of freedom. Further potential is implied by the optical switching of the magnetization by means of the inverse Faraday effect instead of using external fields, which promises to accelerate switching times into the ultrafast regime.

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