Femtosecond magneto-optics in magnetoplasmonic nanostructures

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Abstract

We disclose ultrafast manifestations of magnetoplasmonics by observing nontrivial evolution of transverse magneto-optical Kerr effect within 45-fs pulses reflected from an iron-based magnetoplasmonic crystal. The effect takes place for resonant SPP excitation, has opposite signs of time derivative for different slopes of the resonance, and is addressed to magnetization-dependent dispersion relation of SPPs.

1. Introduction

Since its establishment by Michael Faraday in 1845, magneto-optics found numerous applications in science and technology, including, to name a few, methods such as magnetic circular dichroism and magneto-optical microscopy, and devices such as magneto-optical isolators and memory. However, in the overwhelming majority of experiments, magneto-optical effects are measured with continuous-wave light sources. It was only in 1996 that magneto-optics resorted to subpicosecond scale [1]. As femtosecond pulse sources were developed, immense possibilities in magnetic information recording and readout were attained [2]. A big downside of this approach to induce time-dependent magneto-optical response lies in the requirement of intrinsic changes to be brought into the medium by a high-power laser source.

In order to pursue shorter time scales of light-matter interaction one may consider short-living solid state excitations, such as polaritons. Surface plasmon polariton (SPP), namely, an electromagnetic wave bound to free-electron plasma of metal, is a short-living excitation spanning for up to several hundreds of femtoseconds. Interaction of a femtosecond laser pulse with plasmonic nanostructures recently emerged as a topic for research bringing about, to name a few, laser pulse amplitude [3] and polarization shaping [4] with plasmonic media. On the other hand, it was shown that external quasistatic magnetic field can be used to control dispersion of SPP in magnetic media [5]; despite immense number of works on magnetoplasmonics emerged during the last three decades [6, 7, 8], magnetoplasmonic effects have been considered static so far.

In this work, we experimentally demonstrate manifestations of time-dependent transverse magneto-optical Kerr effect (TMOKE) within 45-fs laser pulses reflected from a one-dimensional iron-based magnetoplasmonic crystal. We show that exciting SPPs with magnetization-dependent dispersion law allows one to control the shape of the reflected pulse. TMOKE evolution is shown to have either positive or negative time derivative depending on the spectral position of the incident pulse's carrier wavelength λ_c with respect to the SPP resonance wavelength. Proper grounds are given for this effect within the Lorentzian spectral line shape approach.

2. Idea, sample and methods

The idea of creating the appropriate conditions for observing the intra-pulse time-dependent TMOKE is illustrated in Fig. 1. The femtosecond pulse excites an SPP in a one-dimensional iron grating—the so-called magnetoplasmonic crystal. SPP has a magnetization-dependent dispersion relation:

$$k_{\text{SPP}}(M) = \frac{\omega}{c}\sqrt{\frac{\varepsilon}{\varepsilon+1}}\left[1+\alpha g(M)\right], \qquad (1)$$

where ε is the dielectric permittivity of iron, $\alpha = [\sqrt{-\varepsilon}\times(1-\varepsilon^2)]^{-1}$, and g is the absolute value of the gyration vector of iron. Lifetime of SPP is limited by radiative and dissipative losses down to the values of no greater than 1 ps. Therefore, the resultant reflected pulse is perturbed with respect to the initial shape and contains information about SPP towards the end of the pulse, that is depicted with the elongated tail of the reflected pulse. The end of the pulse, therefore, is more sensitive to enhanced magnetoplasmonic effects; hence, one should expect rising of TMOKE towards the end of the pulse.

We briefly describe an experimental method to point out the nontrivial evolution of transverse magneto-optical Kerr

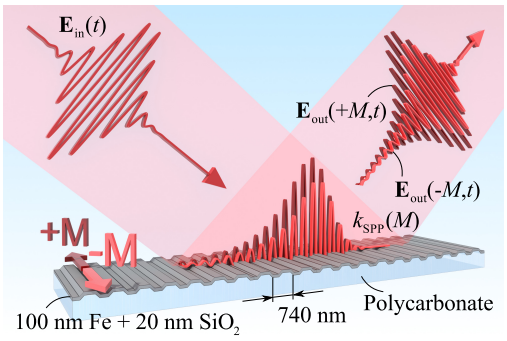

Figure 1: Illustration of the ultrafast time-dependent transverse magneto-optical Kerr effect.

effect within femtosecond laser pulses. Time-dependent TMOKE $\delta(t)$ is defined as:

$$\delta(M,t) = \frac{I(M,t) - I(-M,t)}{I(0,t)}, \quad (2)$$

where $I(t)$ is the envelope function of the pulse. In order to obtain the information about $\delta(t)$, a correlation function (CF) measurement setup was used. A train of 45-fs laser pulses from a Coherent Micra Ti:sapphire oscillator was precompressed at a chirped-mirror assembly in order to account for dispersive optics in the setup. The pulses were split into two beams, one being the signal beam with the sample in it, and the other one being the gate beam with a 3 fs-step delay line. Two electrically serial electromagnets placed around the sample were used to apply quasistatic magnetic field in TMOKE configuration, i.e., in the plane of the sample perpendicularly to the plane of incidence. The magnets were driven by an AC current source at the frequency of $f_M = 117$ Hz and provided about 30 mT magnetic flux density that is enough to saturate magnetization of iron in the sample. Placing the non-magnetic sample holder and the magnets into an iron housing was noticed to prevent other optomechanical components from mechanical oscillations at the frequency of the external magnetic field. The pulses in both beams were brought together by a parabolic mirror in a 100 μm-thick beta-barium borate crystal producing sum-frequency radiation towards the core of a multi-mode optical fiber. The sum-frequency signal represents the second-order intensity correlation function of the intensity profiles of the two beams:

$$I_{\text{CF}}(M,\tau) = \int_{-\infty}^{\infty} \left[I_{\text{sig}}(t)(1+\delta(M,t)/2)\right] I_{\text{gate}}(t-\tau)dt, \quad (3)$$

and is therefore inherently dependent on the $\delta(M,t)$ function of the signal pulse. The 2 m-long fiber transferred the signal to a distant Si photodiode coupled to two lock-in amplifiers. Two signal values were measured: the first one was the photocurrent amplitude I_{CF,f_C} at the frequency of the optical chopper $f_C = 420$ Hz that in the absence of the external magnetic field gives the value of:

$$I_{\text{CF},f_C}(\tau) \propto \int_{-\infty}^{\infty} I_{\text{sig}}(t) I_{\text{gate}}(t-\tau)dt. \quad (4)$$

The second one was the photocurrent amplitude I_{CF,f_M} at the frequency of the external magnetic field that gives the value of:

$$I_{\text{CF},f_M}(M,\tau) \propto \int_{-\infty}^{\infty} \delta(M,t) I_{\text{sig}}(t) I_{\text{gate}}(t-\tau)dt. \quad (5)$$

Finally, the ratio of the latter two gives the value:

$$\Delta(M,\tau) = \frac{I_{\text{CF},f_M}}{I_{\text{CF},f_C}} = \frac{I_{\text{CF}}(M,\tau) - I_{\text{CF}}(-M,\tau)}{I_{\text{CF}}(\tau,0)}, \quad (6)$$

that is a characteristic value of the time-dependent TMOKE signal. Although $\Delta(M,\tau)$ is a value that refers to $\delta(M,t)$ in an indirect fashion, its nontrivial dependence on τ indicates nontrivial dynamics of TMOKE—note that if $\delta(M,t)$ is otherwise time-independent, Δ exactly equals the static TMOKE value.

Measurements of $\Delta(M,\tau)$ were performed for a sample of one-dimensional magnetoplasmonic crystal based on a commercially available digital versatile disk polycarbonate template having periodic corrugation with the depth of approximately 50 nm and the period of 750±10 nm. The dielectric template was covered by a 100 nm layer of polycrystalline iron deposited by magnetron sputtering and protected by a 20 nm-thick silica layer from the top. Angular-dependent reflection coefficient spectroscopy indicates two branches of SPP modes forming a plasmonic band gap at the near-normal incidence. Based on the spectroscopy results, the angle of incidence of laser radiation onto sample was chosen to be $\theta = 5^\circ$. The particular angle of incidence was chosen intentionally (a) for the reflectance maximum within one of the SPP branches to be situated in the spectral vicinity of the laser radiation spectrum, and (b) to eliminate the TMOKE arising from the iron surface itself, that scales with $\sin^2 2\theta$. Prior to time-dependent measurements, static TMOKE spectroscopy was performed. Firstly, significant enhancement of TMOKE is observed as compared to the value measured for a plain iron film $\delta_{\text{Fe}} \approx 10^{-3}$ at the same angle of incidence. Strong wavelength dependence is observed in the TMOKE spectrum in the vicinity of the SPP resonance. This feature is connected to the magnetization-dependent dispersion relation of SPP shown in Eq.(1). We will see below that Eq.(1) also explains the time evolution of the TMOKE within the pulse.

3. Results and discussions

We prove the influence of SPPs on the shape of the pulses by measuring full width at half maximum of the correlation functions depending on the carrier wavelength as presented in Fig. 2(a). One can see the wavelength-dependent CF width of the pulse reflected from the sample; moreover, the function of CF width correlates well with the SPP resonance as provided in the reflection spectrum given in gray. The open dots denote the width of the intensity autocorrelation function as measured with an autocorrelator placed before the sample. The dashed line illustrates the Fourier-transform limit for sech^2-pulses.

The key results of the work are provided in Fig. 2(b-h). Time-dependent magneto-optical effect is demonstrated with the measured $\Delta(\tau)$ function for different λ_c with red dots superimposed with corresponding CFs. Zero time delay is each time defined at the CF maximum. In these experiments, the spectral bandwidth of the pulses was kept at 14 nm of intensity FWHM for each λ_c. The zero dispersion was set for $\lambda_c = 800$ nm, and Fourier-limited pulses were acquired for all of the λ_c values in use providing average time-bandwidth product of 0.31 ± 0.1 as measured with an autocorrelator just before the sample. The effect

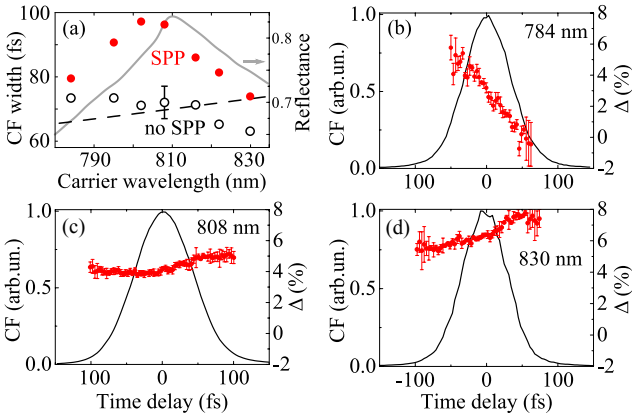

Figure 2: (a) Intensity correlation function width measured with the correlation setup (filled dots) and with an autocorrelator placed before the sample (open dots) as a function of the carrier wavelength. Black dashed curve denotes the Fourier-transform limit. Gray curve shows the reflection coefficient spectrum. (b-d) Correlation functions (curves) and time-dependent TMOKE represented by the $\Delta(\tau)$ function as denoted in Eq.(6) (dots with error bars) for the pulses with the carrier wavelength values of 784 nm, 808 nm, and 830 nm, respectively.

is seen to strongly depend on the part of the SPP resonance that is excited. Gradual increase of $\Delta(\tau)$ is observed for $\lambda_c > 802\,\text{nm}$. The reversed picture is observed for $\lambda_c < 802\,\text{nm}$ where $\Delta(\tau)$ is found to be decreasing either monotonically or on the average. Finally, TMOKE remains almost constant for $\lambda_c = 802\,\text{nm}$. Such a spectrally-selective behavior contributes to establishing the major role of SPPs in forming $\Delta(\tau)$.

The effect was studied in other configurations of the experiment. Illuminating the sample with s-polarized light should have provided no signal due to TMOKE properties. This is supported by no detectable signal at the frequency of the external magnetic field under CW illumination. However, a spurious TMOKE signal of about 0.4% was obtained in the case of CF measurement. In this case, the $\Delta(\tau)$ dependence looks like an interference pattern and could be resultant from the mechanical oscillations in the setup disturbing the far-field interference pattern formed by two beams incident onto the BBO crystal. Nevertheless, the magnitude of this effect is approximately one order of magnitude lower than that found in p-polarized light and thus cannot question our main result. The same procedure was also conducted for the sample azimuthally rotated by 90°. In this case, p-polarized static TMOKE yielded $(0.2 \pm 0.02)\%$ for all the wavelength values of interest, and no pronounced $\Delta(\tau)$ dependence was observed.

4. Conclusions

In conclusion, manifestations of time-dependent transverse magneto-optical Kerr effect are experimentally demonstrated in 45-fs laser pulses reflected from a one-dimensional iron-based magnetoplasmonic crystal. The effect is attributed to exciting SPPs with magnetization-dependent dispersion. Kerr effect evolution is shown to have either positive or negative time derivative depending on the position of the incident pulse's carrier wavelength λ_c with respect to the SPP resonance. Proper grounds are given for this effect within the Lorentzian spectral line shape approach. Being a subwavelength-thickness tailorable nanostructure, the iron-based plasmonic crystal under study is a promising tool for manipulating femtosecond laser pulses with the external magnetic field that is to find applications in the novel active plasmon-based telecom devices.

Acknowledgement

Authors thank A. A. Grunin and V. V. Rodionova for sample fabrication, and Chang-Won Lee for fruitful discussions. Support of Russian Foundation for Basic Research and Ministry of Education and Science of the Russian Federation is acknowledged.

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