Pump-probe spectroscopy in gold-garnet magnetoplasmonic metasurfaces

T V Dolgova¹, M A Kiryanov¹, P K Nurgalieva¹, G S Ostanin¹, A I Musorin¹, H Uchida², M Inoue², A A Fedyanin¹

¹Faculty of Physics, Lomonosov Moscow State University, 119991 Moscow, Russia
²Department of Electrical and Electronic Information Engineering, Toyohashi University of Technology, 1-1 Tempaku-cho, Toyohashi, Aichi 441-8580, Japan
dolgova@nanolab.phys.msu.ru

Abstract. Ultrafast modulation of optical response is realized in a multi-resonant magnetoplasmonic metasurface under resonant femtosecond pump. A saturation of resonant probe transient transmission is shown at dielectric quazi-waveguide mode.

1. Introduction
Active plasmonics and magnetoplasmonics rely on the resonance modes for optical response control by dielectric function tuning via laser-induced hot-carrier generation and external magnetic field [1-4]. Multi-resonance systems open a new possibility to handle the ultrafast response using resonances of different nature and their coupling. Thus, both plasmonic and high-quality-factor polaritonic magnetooptical resonances tunable by the azimuthal angle were realized in 2D hybrid metal-dielectric magnetoplasmonic metasurfaces [5]. Resonant femtosecond probe is sensitive to the ultrafast heating by non-resonant pump. The ultrafast modulation of surface-plasmon resonant transverse magnetooptical Kerr effect was shown in one-dimensional nickel magnetoplasmonic crystals [6].

In this paper a saturation of a resonant probe transient transmission is shown in a magnetoplasmonic metasurface under resonant femtosecond pump. The effect is observed within the quasi-waveguide dielectric mode.

2. Experimental
The metasurface is an array of gold nanospheres with a diameter of 100 nanometers and a period of 600 nanometers on a quartz substrate. The array is covered with a 100-nanometer layer of bismuth-substituted yttrium-iron garnet. Gold nanospheres as meta-atoms provide electromagnetic field concentration at electric dipole plasmonic resonance at 810 nm for normal incidence (pump region), and that at 735 nm with quadrupole resonance at 620 nm for the angle of incidence of 20° (probe region).

The peculiarity of such structures is the possibility of excitation of the quasi-waveguide mode at 530 nm. The detailed sample description and scanning electron microscopy image can be found in [5]. The ultrafast optical responses of the magnetoplasmonic metasurface are measured using the pump-probe technique. A regeneratively amplified Ti:sapphire laser with 800 nm central wavelength, 50 fs pulse duration, and 1 kHz repetition rate is a source of laser radiation. The pulse is divided into a pump and a probe pulses with controllable delay. The 0.6 mJ/cm² pump pulse is directed normally to the studied sample. The probe line generates supercontinuum pulse covering the range from 450 to 750 nanometers.
with a total power density not more than 1 nJ/cm$^2$. The angle of probe pulse incidence to the sample is of 18 degrees. The spot sizes of the pump and probe pulses on the sample surface are of 800 and 400 μm, respectively.

To determine the ultrafast optical response, a differential transmittance coefficient $\Delta T/T$ was introduced, that is the normalized difference between the transmittance in the presence and in the absence of a pump pulse. Fig. 1 shows the spectrum of the differential transmittance of the supercontinuum for the 500-femtosecond delay of the probe.

![Fig 1. $\Delta T/T$ spectrum for 500 fs time delay and 0.6 mJ/cm$^2$ pump fluence](image)

Maximum of differential reflectance is achieved at 730 nm associated with the excitation of surface lattice resonance. This resonance appears due to the spectral overlap of dipole surface plasmon and the Rayleigh anomaly for (-2, +1) diffraction order in the Bi:YIG layer. Maxima of differential reflectance at 615 nm and 670 nm wavelength are related to the overlap of quadrupole surface plasmon and the fading of diffraction orders in quartz (+1,0) and Bi:YIG (+2,0). The peaks at 530 nm and 555 nm wavelength are caused by waveguide propagation of (0,+-2) diffraction orders in the Bi:YIG layer. The reason of the dip at the wavelength of 500 nm is the maximum of the increase of imaginary part of the dielectric constant due to the interband transition in gold. The ultrafast modulation of the optical response of a magnetoplasmonic metasurface is associated with the interaction of the pump pulse resonant to the dipole mode with the electron and phonon systems of the sample. The presence of the resonances associated with the electronic systems of gold nanospheres and yttrium-iron garnet allows us to separate the modulation of the optical response of metal nanospheres and the dielectric layer.

The dependence of the maximum amplitude at the wavelength of 527 nanometers on the intensity of the pump pulse is not linear starting from 0.3 mJ/cm$^2$ (Fig.2). Saturation of the effect is observed for higher fluencies. The differential transmittance is linear with pump fluence at the other resonant features associated with the gold nanospheres. The saturation is a consequence of the nonlinear interaction of a femtosecond pulse with an yttrium-iron garnet. A saturation effect reduces during first picoseconds and disappears after electron-phonon relaxation.
3. Conclusions
The effect of transient transmission saturation is probed by white-light supercontinuum at the dielectric quazi-waveguide mode of an yttrium-iron garnet magnetoplasmonic metasurface. The saturation effect fades out with electron-phonon relaxation. The differential transmittance for the localized nanosphere plasmon modes shows linear behavior for the same pump fluence range.

4. Acknowledgements
T.V.D. thanks Russian Foundation for Basic Research (grant №20-02-00758, experiment). M.A.I. thanks Russian Science Foundation (grant №19-72-00168, calculations). K.M.A. thanks MSU Quantum Technology Centre for the support. This research was performed according to the Development program of the Interdisciplinary Scientific and Educational School of Lomonosov Moscow State University “Photonic and Quantum technologies. Digital medicine”.

References
[1] Bossini, D. Belotelov, V. I. Zvezdin, A. K.; Kalish, A. N.; Kimel, A. V. Magnetoplasmonics and Femtosecond Optomagnetism at the Nanoscale. ACS Photonics 2016, 3, 1385.
[2] Pohl, M., Belotelov, V. I., Akimov, I. A., Kasture, S., Vengurlekar, A. S., Gopal, A. V., Zvezdin, A. K., Yakovlev, D. R., Bayer, M. Plasmonic crystals for ultrafast nanophotonics: Optical switching of surface plasmon polaritons. Phys. Rev. B: Condens. Matter Mater. Phys. 2012, 85, No. 081401.
[3] N. Rotenberg, J. Caspers, H. M. van Driel, Tunable ultrafast control of plasmonic coupling to gold films, Phys. Rev. B 80 (2009).
[4] V.V. Temnov, Ultrafast acousto-magneto-plasmonics, Nat. Photon. 6, 728 (2012).
[5] Musorin, A. I.; Chetvertukhin, A. V.; Dolgova, T. V.; Uchida, H.; Inoue, M.; Luk’yanchuk, B. S.; Fedyanin, A. A. Tunable multimodal magnetoplasmonic metasurfaces. Appl. Phys. Lett. 2019, 115, 151102.
[6] I.A. Novikov, M.A. Kiryanov, P.K. Nurgalieva, A.Yu. Frolov, V.V. Popov, T.V. Dolgova, and A.A. Fedyanin. Ultrafast Magneto-Optics in Nickel Magnetoplasmonic Crystals. Nano Lett., 2020, 20, 12, 2606.
[7] G.V. Hartland, Optical studies of dynamics in noble metal nanostructures, Chem. Rev. 111, 3858 (2011).