Intensity-dependent reflectance modulation of femtosecond laser pulses in GaAs nanocylinders with magnetic resonances

Varvara V Zubyuk, Polina P Vabishchevich, Maxim R Shcherbakov, Alexander S Shorokhov, Anna A Fedotova, Sheng Liu, Gordon A Keeler, Tatyana V Dolgova, Isabelle Staude, Igal Brener and Andrey A Fedyanin

1 Quantum Electronics Department and Quantum Technology Center, Faculty of Physics, Lomonosov Moscow State University, Moscow 119991, Russia
2 Sandia National Laboratories, Albuquerque, New Mexico 87185, United States
3 Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA
4 School of Applied and Engineering Physics, Cornell University, Ithaca, NY 14853, USA
5 Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-University Jena, 07743 Jena, Germany

E-mail: zubyuk@nanolab.phys.msu.ru, fedyanin@nanolab.phys.msu.ru

Abstract. We experimentally demonstrate modulation of reflectance in periodic arrays of subwavelength gallium arsenide nanocylinders with Mie-type resonances due to absorption saturation and changes in the refractive index of the semiconductor material of metasurface. The intensity-dependent reflectance modulation of up to 30% in the vicinity of the magnetic dipole resonance at a low laser fluence below 200 μJ/cm² is shown by I-scan measurements.

1. Introduction

Phenomenon of saturable absorption of semiconductors is widely used in optical devices such as semiconductor saturable absorber mirror (SESAM) [1]. These devices have become a key component of ultrafast passively mode-locked and Q-switched laser sources [2, 3]. The saturation of optical absorption was demonstrated in different materials and systems such as plasmonic nanocomposites [4, 5]. Recently, semiconductor nanoparticles and metasurfaces with Mie-type resonance have proven themselves as potentially useful for nanophotonic applications [6, 7]. This happened since they possess completely new properties in comparison with bulk materials [8, 9] and reveal enhanced interaction with electromagnetic radiation [10, 11]. The dielectric metasurfaces also demonstrate low absorption losses as compared to plasmonic structures. In this work, we experimentally demonstrate the intensity-dependent reflectance modulation of femtosecond laser pulses in GaAs nanocylinders samples that exhibit the Mie-type resonances.
Figure 1. (a) A scanning electron microscope image of the GaAs metasurface. (b) Reflection spectra of the GaAs nanocylinders sample with a magnetic dipole (MD) resonant peak at $\lambda \sim 880$ nm for p- and s-polarization of the incident laser radiation at $\theta = 12^\circ$.

2. Results and discussion

2.1. Metasurface sample

The samples are periodic arrays of GaAs nanocylinders with silica caps on top, nanocylinders situated on AlGaO nanocylinders that are situated on a bulk GaAs substrate (Fig.1(a)). The metasurface was fabricated using a previously published procedure [11, 12]. The diameter and height of the nanocylinders are 200 nm and 300 nm, respectively, the diameter-to-pitch ratio is 0.5, and the height of the AlGaO nanocylinders is also approximately 300 nm. Measured reflectance spectra of GaAs nanocylinders arrays at $\theta = 12^\circ$ are close to the normal angle of incidence and exhibit peaks both for p- and s-polarized light that correspond to the magnetic dipole (MD) resonance at the same wavelength in the near infrared (Fig.1(b)). The MD resonance for p-polarized light shifts to the long-wavelength range as the angle of incidence is increased.

2.2. I-scan measurements

We measure the intensity of femtosecond laser pulses that are reflected from the GaAs metasurface using I-scan measurements in reflection geometry. The wavelength $\lambda = 830$ nm is set not far from the bandgap of GaAs ($\lambda = 870$ nm) to maximize the effect of absorption saturation. This laser radiation generates electron-hole pairs due to the single photon absorption processes. The free carrier plasma affects the complex dielectric permittivity; the intensity-dependent reflectance is detected at the normal angle of incidence. The GaAs sample shows the enhanced modulation order of magnitude larger in comparison with the GaAs substrate (Fig.2(a)). The reflectance coefficient as a function of the incident laser intensity for the GaAs nanocylinders sample is indicated by black dots and for the substrate by blue dots. We demonstrate intensity-dependent laser pulse modulation of up to 30% in the vicinity of the magnetic dipolar resonance at a low pump fluence below 200 $\mu$J/cm$^2$. We found excellent agreement of the experimental results with fullwave simulations that use a model with changes in the complex dielectric permittivity induced by free carriers.
Figure 2. (a) Reflectance coefficient as a function of the incident laser intensity for the GaAs nanocylinders sample (black dots) and for substrate (blue dots). (b) Different contribution to the changes in extinction coefficient as a function of the incident laser intensity for the GaAs material induced by free carriers: BF – bandfilling contribution, BS – bandgap shrinkage contribution, Dt – Drude term and Total contribution (Total=BF+BS+Dt).

2.3. Reflectance modulation mechanism
The laser radiation generates electron-hole pairs due to the single photon absorption processes and this leads to the changes in the complex dielectric permittivity. We provided the full-wave simulations with different changes in complex refractive index and find out that reflectance modulation occurs mainly due to changes in extinction coefficient. These changes are determined by three contributions [13] and total change become negative:

$$\Delta \kappa = \Delta \kappa_{BF} + \Delta \kappa_{BS} + \Delta \kappa_{Dt} < 0,$$

where the indexes of $\Delta \kappa_{BF,BS,Dt}$ correspond to BF – bandfilling contribution, BS – bandgap shrinkage contribution, Dt – Drude term and $\Delta \kappa$ is the total changes: Total=BF+BS+Dt. The main contribution to the total change is made by the bandfilling. The reflectance spectrum of the GaAs metasurface is strongly modified under intense laser radiation due to the high sensitivity of Mie-type resonance to material parameters.

3. Conclusions
In conclusion, we have experimentally demonstrated intensity-dependent reflectance modulation of the laser pulse in subwavelength GaAs nanocylinders exhibiting localized Mie-type resonances. The modulation of reflectance coefficient is up to 30% and occurs under free carriers generation at a low laser fluence below $200 \mu J/cm^2$. The reflectance modulation mechanism is complex and is determined primarily by absorption changes due to bandfilling effect.

Acknowledgments
This work was partly supported by financial support from Ministry of Education and Science of the Russian Federation (grant N14.W03.31.0008, calculation of optical spectra). Experimental part was performed with the support of the grant of the President of the Russian Federation for leading scientific schools (grant No. NSh-6862.2018.2 — optical spectra), supported in part by Russian National Technology Initiative, Russian Foundation for Basic Research (grant No. 16-29-11811 — I-scan measurements), and by the German Research Foundation through the Emmy Noether Programme (STA 1426/2-1). Parts of this work were supported by the
U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering and performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

References
[1] Keller U et al. 1996 IEEE J. Sel. Top. Quantum Electron. 2 435–451
[2] Keller U 2003 Nature 424 831
[3] Saraceno C et al. 2012 IEEE J. Sel. Top. Quantum Electron. 18 29–41
[4] Plaksin O et al. 2008 J. Appl. Phys. 103 114302
[5] Kumar M et al. 2014 Plasmonics 9 129–136
[6] Shcherbakov M et al. 2015 Nano Lett. 15 6985–6990
[7] Shcherbakov M et al. 2017 Nat. Commun. 8 17
[8] Evlyukhin A B et al. 2012 Nano Lett. 12 3749–3755
[9] Decker M et al. 2015 Adv. Opt. Mat. 3 813–820
[10] Shorokhov A et al. 2016 Nano Lett. 16 4857–4861
[11] Liu S et al. 2016 Nano Lett. 16 5426–5432
[12] Liu S et al. 2016 Adv. Opt. Mat. 10 1457–1462
[13] Bennett B et al. 1990 IEEE J. Quantum Electron. 26 113–122