Ultrafast optical modulation of magneto-optical terahertz effects occurring in a graphene-loaded resonant metasurface

S. Zanotto[1*] , C. Lange[2], T. Maag[2], A. Pitanti[3], V. Miseikis[4], C. Coletti[4], R. Degl'Innocenti[5], L. Baldacci[6], R. Huber[2], and A. Tredicucci[7]

[1]Istituto Nazionale di Ottica – CNR, and LENS, Via Nello Carrara 1, 50019 Sesto Fiorentino (FI), Italy
[2]Department of Physics, University of Regensburg, 93040 Regensburg, Germany
[3]NEST, Istituto Nanoscienze – CNR and Scuola Normale Superiore, Piazza San Silvestro 12, 56127 Pisa (Italy)
[4]CNI@NEST, Istituto Italiano di Tecnologia, P.za S. Silvestro 12, 56127 Pisa, Italy
[5]Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom
[6]Scuola Superiore Sant'Anna, Institute of Life Sciences, P.za Martiri della Libertà 33, 56127 Pisa, Italy
[6]NEST, Istituto Nanoscienze-CNR and Dipartimento di Fisica "E. Fermi", Università di Pisa, L.go Pontecorvo 3, 56127 Pisa, Italy
*corresponding author: simone.zanotto@ino.it

ABSTRACT

In this paper we investigate the effect of a static magnetic field and of an optical pumping on the transmittance of a hybrid graphene-split ring resonator metasurface. Experimental evidence of a significant modulation of the transmitted spectra has been observed, both for what concerns the sole optical pumping, and for the combination of optical pumping and magnetostatic bias. The transmittance modulation traces retain peculiar spectral fingerprints which induces the interpretation of the phenomenon as a non-trivial interplay between the bare graphene response and the split ring resonance.

Keywords: terahertz, graphene, split-ring, magneto-optic, metasurface

1. INTRODUCTION

Terahertz science and technology underwent a flourishing development in the last decade. Strong advancements of sources, detectors, and optical elements have been reported, thus enabling several key applications [1,2]. Among the most promising optical materials for the terahertz spectral range is graphene, whose unique optical response, in conjunction with its tunability, enabled the realization of effective modulators, switches, absorbers, and detectors [3-17].

Nonetheless, there are still some issues connected with the manipulation of terahertz polarized light, and, more specifically, with the realization of non-reciprocal optical elements such as polarization rotators and optical isolators. The main reason is that the terahertz range lies in the intermediate region between optical and microwave spectral regions, and the materials currently employed in either of the above cited spectral regions are not suitable for operation in the terahertz. For instance, ferrite isolators are commonly employed up to 500 GHz, but above this frequency their insertion losses become prohibitive. Hence, there is much interest in exploring other materials for magneto-optic applications in the terahertz range. First-principle consideration revealed that graphene deserves a significant potential in this regards, and some implementations of actual devices have been reported recently [18-19].

Still, the devices reported so far operate in a static regime, where the electrons in the graphene lie in a stationary state, with a distribution dictated by the sole doping and, possibly, electrostatic biasing. Further degrees of freedom can be however accessed by controlling the graphene electrons through a pump optical beam. In this paper we report the magneto-optical response of a graphene layer coupled to a split-ring resonator metasurface upon an intense, ultrafast optical excitation. Effects complementary to these of the bare magnetic field have been observed.

2. MEASUREMENT TECHNIQUE AND DEVICE STRUCTURE

2.1 Magneto-optic pump-and-probe optical setup

The experimental setup devoted to the magneto-optical, pump-and-probe studies is sketched in Fig. 1. A 35 femtosecond, transform-limited pulse with central wavelength 800 nm is split in two beams. One of them, the pump beam, is sent on the sample through a hole in the parabolic mirror which serves for the focusing of the probe beam. An optical delay stage allows for tuning the arrival time of the pump pulse with respect to the probe pulse. The probe pulse is generated through a non-linear optical process taking place in a ZnTe crystal, subsequently focused on the sample, collected through a parabolic mirror and imaged on a second ZnTe crystal which serves for the electro-optic sampling technique.

The setup has been designed in order to allow the mounting of a helium-flow cryostat provided with THz optical access and a split-coil magnet. At the sample location a static magnetic biasing of up to 5 Tesla is achievable, while maintaining the sample at a temperature of 4 K.

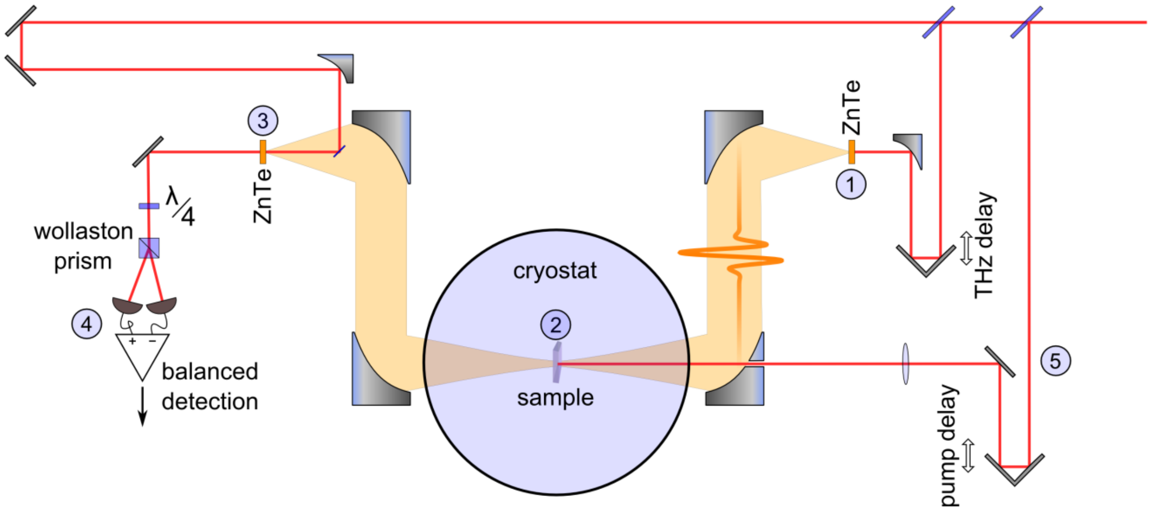

Figure 1. Schematics of the electro-optic terahertz pump-and-probe setup employed for the sample characterization The cryostat also includes a split-coil magnet.

2.2 Device structure

The device structure is schematized in Fig. 2 (a). On a double-polished, undoped GaAs wafer, several arrays of split ring resonators [20-21] have been patterned through ordinary optical lithography, metal deposition, and lift-off techniques (details in Fig. 2 (b-c) and in the figure caption). Subsequently, a thin (approx. 20 nm thick) silica layer has been deposited through RF sputtering. Finally, a graphene monolayer has been transferred on the sample surface. Graphene growth has been performed on a separate host copper foil by low-pressure chemical vapour deposition (CVD) using methane as a carbon precursor [22]. After chemically etching the copper growth substrate, the graphene was transferred to the sample using a supporting polymethyl-methacrylate film. This technique enabled to cover a wide area of the split-ring resonator patterned sample, as shown in Fig. 2 (d). This point is crucial since, in order to get a satisfactory signal-to-noise ratio from the optical measurement, a huge number of identical split-ring resonators have to be probed. Here, indeed, we were able to cover several groups of SRRs, thus enabling to perform experiments on patterns having different characteristics.

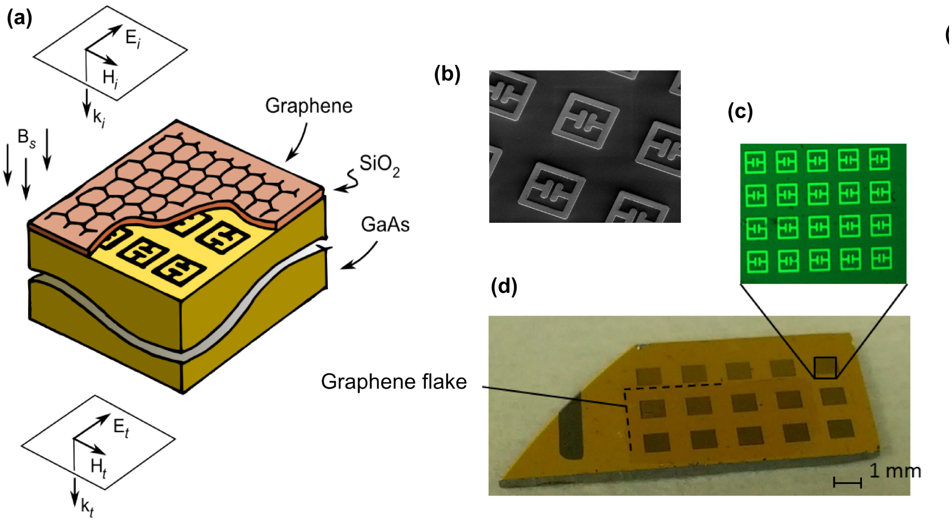

Figure 2. Sample schematics (a), electron microscope image of the split rings (b), optical microscope image of the split-rings (c), and overall view of the fabricated sample (d).

3. PUMP-AND-PROBE EXPERIMENTAL RESULTS

3.1 Response at a fixed delay time

In a previous work [23] we have showed that the graphene-split ring resonator hybrid metasurface exhibits a pronounced magneto-optic effect, i.e., the transmittance of the metasurface is modulated upon an application of a magnetic field in correspondence to the SRR resonance. Here we show that a further modulation is obtained when a pump pulse excites the sample prior to the arrival of the probe pulse. Fig. 3 (a) shows the spectrum of the transmitted pulse collected at zero magnetic field and without the pump excitation, which clearly shows that at a well definite resonance exists at the frequency of about 0.8 THz. Fig. 3 (b) instead recalls the effect discussed in Ref. [23], i.e., the transmittance modulation which occurs when a static magnetic field is applied to the sample. When pumping the sample with a pulse having an average intensity of 600 μW/cm^2, the curves reported in Fig. 3 (c) are instead obtained. The pump pulse precedes the probe pulse by 1.3 picoseconds, thus we expect that the charge excited from the valence band to the conduction band of the graphene has not underwent recombination.

While the general trend for the pumped case is similar to that observed in the unpumped one, a smoother resonant lineshape can be noticed. We interperet this result as a consequence of the injected carrier concentration, which increases the effective Fermi energy and the graphene conductivity. Indeed, it has been shown previously [23] that an increase in the Fermi energy is responsible for a less pronounced feature in the relative transmittance spectra. In general, the overall behaviour of the system can be ascribed to a semiclassical response of the graphene carriers, essentially due to the relatively low mobility of transferred graphene. In other words, the absence of a clear signature of the Landau levels in the optical spectra is consistent with previous reports in the literature [24, 25].

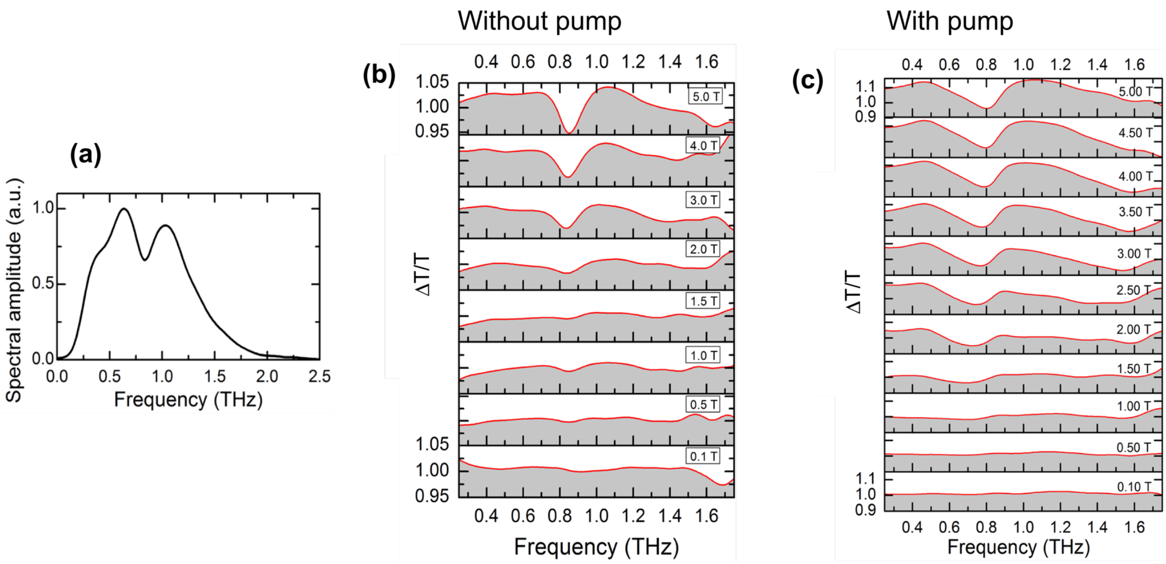

Figure 3. Panel (a): spectral content of the probe pulse after interaction with the sample. The dip at 0.8 THz is ascribed to the excitation of the split-ring resonance. Panels (b) and (c): transmittance modulation as a function of the magnetic field applied to the sample, either in absence or in presence of a pump beam.

3.2 Response in function of the delay time

To better assess the dynamics of the photoexcited carriers in graphene, we performed a series of measurements where the relative time between the pump and the probe pulse was varied. No magnetic field is applied here. In this experiment we focused on a different group of split-ring resonators, characterized by slightly different dimensions, which resulted in a transmittance spectrum featuring a double-dip (Fig. 4 (a)).

Here, we employed a pump fluence of 1.5 $\mu J/cm^2$, and swept the pump-probe relative time from -1 ps to +2 ps. As expected, for negative or zero relative time the transmittance does not get modulated, i.e., $\Delta T/T = 0$. When instead the pump-probe relative time assumes positive values, a double-peaked structure emerges from the noise background, gaining contrast as the relative time increases. Curiously it turned out that in this case $\Delta T/T$ assumes a negative value in the low-energy section of the spectrum, except for the resonance regions where $\Delta T/T$ is positive. Nevertheless, the effect is clear, and it could be harnessed to functional applications like ultrafast terahertz optical modulators.

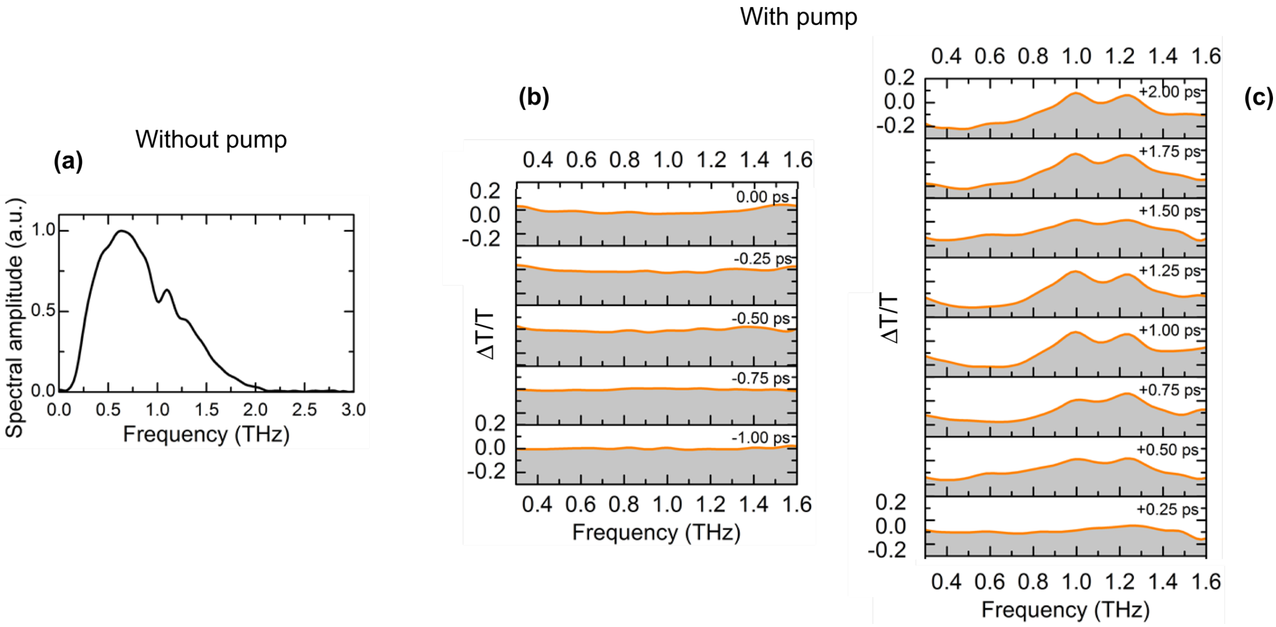

Figure 4. Panel (a): spectral content of the probe pulse after interaction with the sample. Here, a sample different from that considered in Figure 3 is under investigation. Panels (b) and (c): transmittance modulation as a function of the time delay between pump and probe pulses. Negative times means that the probe pulse is preceding the arrival of the pump pulse.

4. CONCLUSIONS

In summary, we reported of transmittance modulation upon ultrafast optical excitation and magnetostatic biasing of a hybrid metasurface comprising a graphene monolayer closely spaced from an array of split ring resonators. Positive and negative values have been observed on the transmittance modulation spectra, and interpreted as an effect of the Fermi level of the charge carriers in the conduction band. These results open interesting perspectives in view of ultrafast-controllable magneto-optic terahertz devices, such as optically-controllable non-reciprocal polarization rotators and optical isolators.

ACKNOWLEDGEMENTS

The work was supported in part by the European Union Graphene Flagship under grant agreement n° 604391.

REFERENCES

[1] A. Ferrari, F. Bonaccorso, V. Fal'ko, K. S. Novoselov, S. Roche, P. Boggild, S. Borini, F. H. L. Koppens, V. Palermo, N. Pugno, et al., "Science and technology roadmap for graphene, related two-dimensional crystals, and hybrid systems", Nanoscale **7**, 4598 (2015).

[2] A. Tredicucci and M. S. Vitiello, "Device concepts for graphene-based terahertz photonics", IEEE Journal of Selected Topics in Quantum Electronics, **20**, 8500109 (2014).

[3] R. Degl'Innocenti, D. S. Jessop, Y. D. Shah, J. Sibik, J. A. Zeitler, P. R. Kidambi, S. Hofmann, H. E. Beere, and D. A. Ritchie , "Low-bias terahertz amplitude modulator based on split-ring resonators and graphene", ACS Nano **8**, 2548 (2014).

[4] B. Sensale-Rodriguez, T. Fang, R. Yan, M. M. Kelly, D. Jena, L. Liu, and H. Xing, "Unique prospects for graphene-based terahertz modulators", Appl. Phys. Lett. **99**, 113104 (2011).

[5] B. Sensale-Rodriguez, R. Yan, M. Zhu, D. Jena, L. Liu, and H. Grace Xing, “Efficient terahertz electro-absorption modulator employing graphene plasmonic structures”, Appl. Phys. Lett. **101**, 261115 (2012).
[6] S. H. Lee, M. Choi, T.-T. Kim, S. Lee, M. Liu, X. Yin, H. K. Choi, S. S. Lee, C.-G. Choi, S.-Y. Choi, et al., “Switching terahertz waves with gate-controlled active graphene metamaterials”, Nature Materials **11**, 936 (2012).
[7] F. Valmorra, G. Scalari, C. Maissen, W. Fu, C. Schoenenberg, J. W. Choi, H. G. Park, M. Beck, and J. Faist, “Low-bias active control of terahertz waves by coupling large-area CVD graphene to a terahertz metamaterial”, Nano Lett. **13**, 3193 (2013).
[8] B. Vasic and R. Gajic, “Graphene induced spectral tuning of metamaterial absorbers at mid-infrared frequencies”, Appl. Phys. Lett. **103**, 261111 (2013).
[9] I. Crassee, J. Levallois, A. L. Walter, M. Ostler, A. Bostwick, E. Rotenberg, T. Seyller, C. Van Der Marel, and A. B. Kuzmenko, “Giant Faraday rotation in single- and multilayer graphene”, Nature Physics **7**, 48 (2011).
[10] R. Shimano, G.Yumoto, J. Y. Yoo, R. Matsunaga, S. Tanabe, H. Hibino, T. Morimoto, and H. Aoki, “Quantum Faraday and Kerr rotations in graphene”, Nature Communications **4**, 1841 (2013)
[11] H. Da, Q. Bao, R. Sanaei, J. Teng, K. P. Loh, F. J. Garcia-Vidal, and C.-W. Qiu, “Monolayer graphene photonic metastructures: giant Faraday rotation and nearly perfect transmission”, Phys. Rev. B **88**, 205405 (2013).
[12] H. Da and G. Liang, “Enhanced Faraday rotation in magnetophotonic crystal infiltrated with graphene”, Appl Phys. Lett. **98**, 261915 (2011).
[13] N. Ubrig, I. Crassee, J. Levallois, I. O. Nedoliuk, F. Fromm, M. Kaiser, T. Seyller, and A. B. Kuzmenko, “Fabry-Pérot enhanced Faraday rotation in graphene”, Optics Express **21**, 24736 (2013).
[14] A. Fallahi and J. Perruisseau-Carrier, “Manipulation of giant Faraday rotation in graphene metasurfaces”, Appl. Phys. Lett. **101**, 231605 (2012).
[15] Y. Hadad, A. R. Davoyan, N. Engheta, and B. Z. Steinberg, “Extreme and quantized magneto-optics with graphene meta-atoms and metasurfaces”, ACS Photonics **1**, 1068 (2014).
[16] M. Tymchenko, A. Y. Nikitin, and L. Martin-Moreno, “Faraday rotation due to excitation of magnetoplasmons in graphene microribbons”, ACS Nano **7**, 9780 (2013).
[17] M. Wang, Y. Wang, M. Pu, C. Hu, X. Wu, Z. Zhao, and X. Luo, “Circular dichroism of graphene-based absorber in static magnetic field”, Journal of Appl. Physics **115**, 154312 (2014).
[18] M. Tamagnone, A. Fallahi, J. R. Mosig, and J. Perruisseau-Carrier, “Fundamental limits and near-optimal design of graphene modulators and non-reciprocal devices”, Nature Photonics **8**, 556 (2014).
[19] M. Tamagnone, C. Moldovan, J.-M. Poumirol, A. B. Kuzmenko, A. M. Ionescu, J. R. Mosig, and J. Perruisseau-Carrier, “Near Optimal Graphene Terahertz Non-Reciprocal Isolator.” Nature Communications **7**, 11216 (2016).
[20] D. Shurig, J. J. Mock, and D. R. Smith, “Electric-field coupled resonators for negative permittivity metamaterials”, Appl. Phys. Lett. **88**, 041109 (2006).
[21] A. K. Azad, A. J. Taylor, E. Smirnova, and J. F. O’Hara, “Characterization and analysis of terahertz metamaterials based on rectangular split-ring resonators”, Appl. Phys. Lett. **92**, 011119 (2008).
[22] V. Miseikis, D. Convertino, N. Mishra, M. Gemmi, T. Mashoff, S. Heun, N. Haghighian, F. Bisio, M. Canepa, V. Piazza, et al., “Rapid CVD growth of millimeter-sized single-crystal graphene using a cold-wall reactor”, 2D Mater. **2**, 014006 (2015).
[23] S. Zanotto, C. Lange, T. Maag, A. Pitanti, V. Miseikis, C. Coletti,R. Degl'Innocenti, L. Baldacci, R. Huber, and A. Tredicucci, “Magneto-optic transmittance modulation observed in a hybrid graphene–split ring resonator terahertz metasurface”, Appl. Phys. Lett. **107**. 121104 (2015).
[24] A. M. Witowski, M. Orlita, R. Stepniewski, A. Wysmolek, J. M. Baranowski, W. Strupinski, C. Faugeras, G. Martinez, and M. Potemski, “Quasiclassical cyclotron resonance of Dirac fermions in highly doped graphene”, Phys. Rev. B **82**, 165305 (2010).
[25] I. Crassee, M. Orlita, M. Potemski, A. L. Walter, M. Ostler, T. Seyller, I. Gaponenko, J. Chen, and A. B. Kuzmenko, “Intrinsic terahertz plasmons and magnetoplasmons in large scale monolayer graphene”, Nano Letters **12**, 2470 (2012).