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Magneto-optic transmittance modulation observed in a hybrid graphene–split ring resonator terahertz metasurface

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By placing a material in close vicinity of a resonant optical element, its intrinsic optical response can be tuned, possibly to a wide extent. Here, we show that a graphene monolayer, spaced a few tenths of nanometers from a split ring resonator metasurface, exhibits a magneto-optical response which is strongly influenced by the presence of the metasurface itself. This hybrid system holds promises in view of thin optical modulators, polarization rotators, and nonreciprocal devices, in the technologically relevant terahertz spectral range. Moreover, it could be chosen as the playground for investigating the cavity electrodynamics of Dirac fermions in the quantum regime. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4931704]

In the world of optical materials, graphene offers unique potentials like broadband operation, ultrafast response, large nonlinearities, and strong Faraday rotation, despite its monoatomic thickness. The key for some of these properties lies in its peculiar Dirac-like semimetal band structure, whose nature and whose consequences are at the center of systematic investigations taking place since the latest decade. Many efforts have been devoted to harness these potentials and to reflect the intriguing physical properties into functional devices.[1]

Graphene is acquiring an increasingly significant role also in terahertz (THz) photonics,[2] where certain key components like beam modulators, saturable absorbers, or polarization rotators suffer from poor efficiency or bulky dimensions. Large-area graphene flakes revealed potentials for transmittance or absorbance modulators, especially in conjunction with specifically patterned surfaces.[3–9]

It is thanks to surface patterns, which possibly feature subwavelength sizes, i.e., metasurfaces, that the intrinsic optical properties of a given material can be tuned to large extents. Graphene is an intrinsically magneto-optic material, whose response depends on the growth conditions and which inherits the quantum features of Dirac fermions in a magnetic field.[10–14] It has been recently shown how a proper engineering of the optical resonant components in vicinity of the graphene layer (or a patterning of the graphene layer itself) can result in very large Faraday or Kerr rotation angles,[15–21] in circular dichroism,[22] or in nonreciprocal behaviors.[23]

Among the photonic resonant structures which find applications in the terahertz spectral range, split-ring resonators (SRRs) are one of the most widespread due to their flexibility and ease of fabrication.[24–26] They have proven to be a versatile tool for tailoring the far- and near-field response of surfaces; in addition, proper combinations of the inductive and capacitive elements used in split-ring resonators allow for extremely low mode volumes. Nevertheless, hybrid graphene-split ring resonator metasurfaces have not yet been studied in great detail for what concerns the magneto-optic response. In this article, we present experimental and numerical results on such a structure, where the magnetic-field induced modification of the Drude-like response of Dirac fermions gives rise to resonant transmission modulation of up to 10%. We believe our concept holds promises, thanks to the tunability of the split ring resonator metasurface, in potential magneto-optic applications like non-reciprocal terahertz photonic devices, and, especially, for implementing frontier experiments aimed at investigating the cavity quantum electrodynamics of graphene cyclotron transitions.[27]

A schematic of the sample is sketched in Fig. 1(a). It consists of a semi-insulating gallium arsenide substrate, polished on both sides, which has been patterned through optical lithography and lift-off technique with an array of metallic SRRs. The deposited metals are Ti/Au, 10/100 nm in thickness; a scanning electron picture of the SRRs taken after the lift-off is reported in Fig. 1(b). Subsequently, a thin layer of silicon oxide ($\sim$30 nm) has been deposited through magnetron sputtering in Ar atmosphere. Finally, a wide area ($>4 \times 2\,\mathrm{mm}^2$) graphene film has been transferred onto the sample surface. Graphene growth has been performed on a separate host Cu foil by low-pressure chemical vapour deposition (CVD) using methane as a carbon precursor.[28] After chemically etching the Cu growth substrate, the graphene was transferred to the sample using a poly(methyl methacrylate) (PMMA) as a support film. Thanks to the presence of SiO_2, the graphene is electrically isolated from the SRRs,

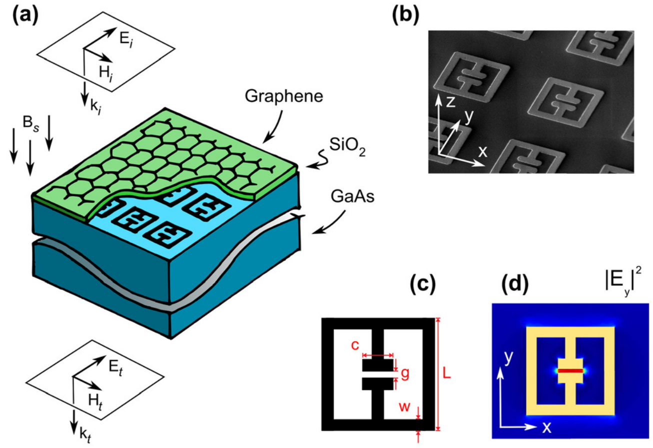

FIG. 1. (a) Schematic of the sample and of the experimental arrangement. An array of split-ring resonators (SRRs) is patterned on the surface of a gallium arsenide substrate. A graphene monolayer is transferred on the surface and isolated by a silicon oxide layer from the SRR. (b) Scanning electron micrography of the gold SRR. (c) Geometrical parameters of the SRR. (d) Resonant field excited in the gap of the SRR.

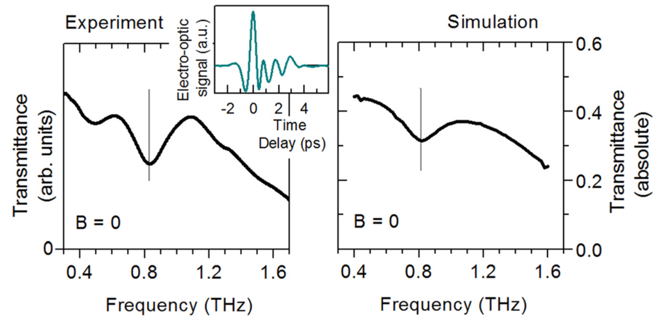

FIG. 2. Terahertz transmittance of the hybrid graphene-split ring resonator metasurface, both from experiment (left panel) and numerical simulation (right panel). The experimental curve reported in the main left panel is derived from the Fourier transform of the transient probe pulse, detected through electro-optic sampling, and reported in the inset.

whose capacitor section is hence not affected by a shunt resistance.

The SRR array implements a metasurface, featuring a resonance close to 1 THz which is coupled to the *y*-polarized electric field. The SRR design employed here is usually referred to as *electric SRR*, whose coupling to the far field is maximally achieved through the electric dipole moment.[24] In order to tune the resonance in the desired spectral range, consistently with the limitations imposed by optical contact lithography, the geometrical parameters are chosen to be $c = 7.5\,\mu m$, $g = 1.5\,\mu m$, $L = 26\,\mu m$, and $w = 3\,\mu m$ [see Fig. 1(c)]. At the resonance frequency, a strong near-field enhancement between the SRR "capacitor" plates occurs, as it can be observed in Fig. 1(d) which follows from a finite-element simulation. In detail, the graphene layer is modeled as a volume element whose conductivity is equal to the surface conductivity (Eq. (1)) divided by the volume element thickness. This thickness has been chosen to be 20 nm, a value sufficiently small to remain in the strongly subwavelength condition but large enough to allow a proper meshing without an unnecessary increase in the problem dimension. The whole simulation domain consists of a parallelepiped enclosing a single unit cell of the metastructure. On the vertical boundaries periodic conditions are set, while on the top and bottom surface boundaries the port condition has been employed. Ports excite and analyze plane waves at normal incidence.

In order to study the magneto-optic response of the hybrid metasurface, the sample has been mounted in a helium flow cryostat equipped with optical access for terahertz radiation and a superconducting magnet for biasing up to 5 T in the direction perpendicular to the surface sample. The sample transmission is measured by time-domain spectroscopy, where a probe transient pulse is generated by optical rectification of a transform-limited 35 fs pulse centered at 800 nm through a ZnTe crystal. The transmitted pulse is then detected by electro-optic sampling, resulting in the time-domain trace reported in the inset of Fig. 2. From the Fourier transform of this transient, the frequency-domain transmittance spectrum reported in the main left panel of Fig. 2 is retrieved. This spectrum features a clear dip at about 0.8 THz, whose contrast and linewidth are consistent with the predictions of a numerical simulation, which is reported in Fig. 2, right panel. While the numerical data are reported in absolute scale (with reference to Fig. 1, the transmittance is $T = |E_t|^2/|E_i|^2$), due to experimental limitations it was not possible to present the measured data in an analogous way. This was due to the limited space available in the cryostat, which was partially cutting the beam while recording the reference spectrum (measured through a sample region which consisted of the bare GaAs wafer covered by the SiO_2). However, this only provides a scale factor and does not affect the features, i.e., resonance position, linewidth, and contrast, of the main dip. The presence in the experimental data of a smaller feature at 0.45 THz could instead be attributed to a slight drift of the setup occurred between the measurements on the metamaterial and on the reference region. This fact is consistent with the observation that in the B-field induced transmittance variation (to be analyzed in the following), which were collected in a more restricted time interval, there is no feature at 0.45 THz.

The spectra reported in Fig. 2 are obtained at zero magnetic field bias; when the latter is turned on, a modulation of the spectrum in the vicinity of the resonance is observed. In order to better read out this modulation, the relative transmittance variations $T(B)/T(B = 0)$ are plotted in Fig. 3. When the magnetic biasing exceeds $\sim$1 T, a dip feature close to 0.8 THz emerges from the noise floor which in the present experiment is about $\sim$1% on the relative transmittance curves. As the magnetic field reaches 5 T, a clear dip around 0.8 THz is observed, together with a shoulder on the high frequency side. The simultaneous presence of a negative and a positive transmittance variation in a narrow spectral range around the SRR resonance frequency proves that the observed phenomenon is due to a non-trivial interplay between the resonant metasurface and the graphene film. Indeed, the effect of a 5 T magnetic field on the transmittance of an unpatterned graphene/SiO_2/GaAs layer stack is around 1%, without strongly dispersive features in the spectral range under consideration.

Support to the experimental data is provided by numerical simulations, which reproduce with good accuracy the

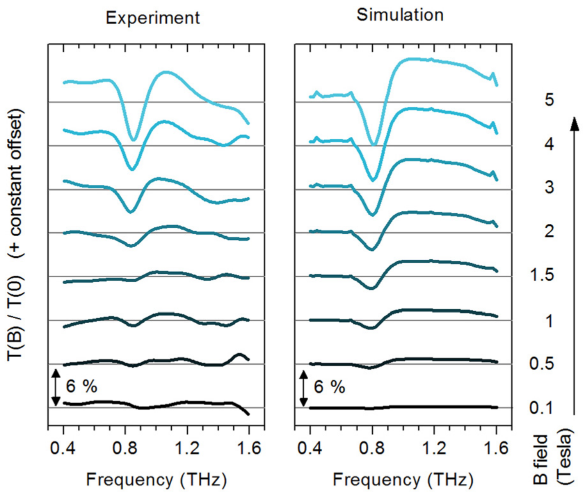

FIG. 3. Magneto-optic transmission modulation effect observed at terahertz frequencies from the hybrid graphene/split ring resonator metasurface. Left panel, measured data (from time-domain spectroscopy); right panel, simulated data (from finite-element calculations). Modulation contrasts up to about 10% are observed at a magnetic field of 5 T.

measured data (Fig. 3, right panel). In the numerical simulation, the only sample component which is assumed to respond to the magnetic field is graphene, hence supporting the picture that the magneto-optic response of the metasurface is actually due to the interplay of magnetically biased Dirac fermions and the split-ring resonance. However, in order to get a good agreement between experiment and theory, it turned out that it is crucial to choose a proper model for the magneto-optic response of the graphene. Indeed, since no spectral features whose frequency scales as $\sqrt{B}$ is observed, the graphene response appears to be incompatible with the quantum magneto-optic regime, characterized by discrete transitions between Landau levels.[10] Instead, the correct description appears to be that of a semiclassical Drude gas, whose conductivity is modulated by the magnetic field. This picture is known to correctly describe the far infrared optical properties of a highly doped graphene layer, where classical cyclotron resonances dominate the optical spectra.[11,12] In formulas, the two-dimensional conductivity which describes the active cyclotron transition is given by

$$\sigma = \sigma_0 \frac{i\gamma}{\omega - \omega_c + i\gamma}, \tag{1}$$

where σ_0 is the DC conductivity at zero magnetic field, $\omega_c = eB/2\pi m^*$ is the classical cyclotron frequency, and γ is the scattering rate.[29] The DC conductivity can be expressed as

$$\sigma_0 = \frac{2e^2}{h}\frac{E_F}{\gamma}, \tag{2}$$

where $2e^2/h$ is the conductance quantum, and E_F is the Fermi energy. In the above expressions, the effective mass m^* is found; this, for Dirac fermions, is not a well-defined quantity. Indeed, in the semiclassical approximation, this has to be expressed in terms of the Einstein relation, $E_F = m^* v_F^2$, where $v_F \approx 10^6$ m/s is the Fermi velocity characterizing the graphene electronic bands. The only free parameters are hence the Fermi energy and the scattering rate, which strongly depend on the growth and manipulation techniques that the graphene undergoes. For the present case, the optical spectra are correctly reproduced assuming $E_F = 50$ meV and $\gamma = 100$ meV, which lie within the bounds for epitaxially grown, transferred graphene.

To gain further insights into the behaviour of hybrid graphene-split ring metasurfaces, we study the role of the graphene parameters E_F and γ on the optical response of the device. Consider the spectra reported in Fig. 4: the different panels correspond to different values of the Fermi energy, while the different colors of the traces identify the scattering rate. It clearly results that the scattering rate is the crucial parameter governing the optical contrast, which can reach very large values of $\sim$35% when $\gamma = 10$ meV and $E_F = 50$ meV. The Fermi energy is instead connected to the absolute values of the relative transmittance curve, which is translated upwards as E_F increases. In terms of metasurface response, the simultaneous presence of contrast modulation, baseline shift, and baseline tilt can be attributed to the interplay between the modulations induced by the magnetic field on both real and imaginary parts of graphene conductivity, eventually connected to the position and to the strength of the SRR resonance.

What is notable in hybrid metasurfaces is that their properties can be controlled independently with electric and magnetic degrees of freedom. Indeed, the electric actuation can be achieved in an integrated fashion: the electrostatic biasing needed to tune the Fermi energy of graphene can be applied in metamaterial surfaces exploiting the patterned metal itself, provided that it is arranged in a connected geometry (thanks to the complementary metasurface approach, this is not an issue even for SRR structures[30,31]). This effect, in conjunction with the non-diagonal conductivity response of magnetostatically biased graphene, could lead to functional devices working towards the quantum regime where certain figures of merit like the Faraday rotation angle may reach anomalously large values.[20]

In conclusion, we designed, fabricated, and studied with experiments and numerical simulations a hybrid metasurface consisting of coupled graphene–split ring resonator thin layers. This surface exhibits a strong electric dipole

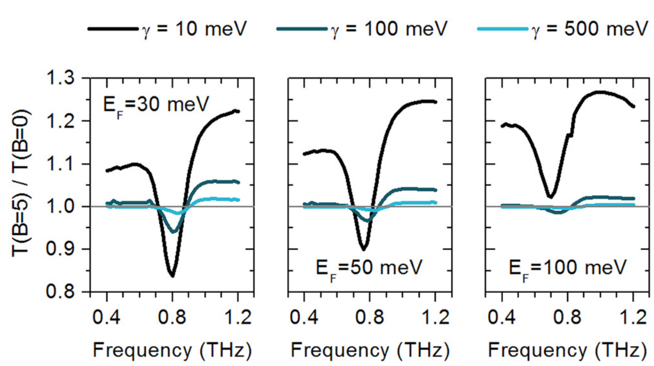

FIG. 4. Effect of Fermi energy and of electron scattering rate on the transmittance modulation of the graphene/split ring resonator hybrid metasurface. The dominant contribution to the optical contrast is that of the scattering rate.

resonance in the terahertz spectral range, which can be controlled through magnetic and electric biasing. We believe that, thanks to its flexibility in design and operation, the hybrid metasurface concept may be exploited in view of numerous applications, from the field of optoelectronic devices like modulators, polarization controllers, and nonreciprocal elements, to the field of cavity quantum electrodynamics of Dirac fermions.

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