Interaction between graphene and metamaterials: split rings vs. wire pairs

Yanhong Zou,[1,2] **Philippe Tassin,**[1,*] **Thomas Koschny,**[1]
and Costas M. Soukoulis[1,3]

[1]*Ames Laboratory, U.S. DOE and Department of Physics and Astronomy,*
Iowa State University, Ames, Iowa 50011, USA
[2]*Key Laboratory for Micro/Nano Optoelectronic Devices, School of Information Science and*
Engineering, Hunan University, Changsha 410082, China
[3]*Institute of Electronic Structure and Lasers (IESL), FORTH, 71110 Heraklion, Crete, Greece*

[*]*tassin@ameslab.gov*

Abstract: We have recently shown that graphene is unsuitable to replace metals in the current-carrying elements of metamaterials. At the other hand, experiments have demonstrated that a layer of graphene can modify the optical response of a metal-based metamaterial. Here we study this electromagnetic interaction between metamaterials and graphene. We show that the weak optical response of graphene can be modified dramatically by coupling to the strong resonant fields in metallic structures. A crucial element determining the interaction strength is the orientation of the resonant fields. If the resonant electric field is predominantly parallel to the graphene sheet (e.g., in a complementary split-ring metamaterial), the metamaterial's resonance can be strongly damped. If the resonant field is predominantly perpendicular to the graphene sheet (e.g., in a wire-pair metamaterial), no significant interaction exists.

References and links

1. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, "Electric field effect in atomically thin carbon films," Science **306**, 666–669 (2004).
2. A. K. Geim and K. S. Novoselov, "The rise of graphene," Nature Mater. **6**, 183–191 (2007).
3. C. Lee, X. Wei, J. W. Kysar, and J. Hone, "Measurement of the elastic properties and intrinsic strength of monolayer graphene," Science **321**, 385–388 (2008).
4. C. Berger, Z. Song, X. Li, X. Wu, N. Brown, C. Naud, D. Mayou, T. Li, J. Hass, A. N. Marchenkov, E. H. Conrad, P. N. First, and W. A. de Heer, "Electronic confinement and coherence in patterned epitaxial graphene," Science **312**, 1191–1196 (2006).
5. S. De and J. N. Coleman, "Are there fundamental limitations on the sheet resistance and transmittance of thin graphene films," ACS Nano **4**, 2713–2720 (2010).
6. F. Bonaccorso, Z. Sun, T. Hasan, and A. C. Ferrari, "Graphene photonics and optoelectronics," Nature Photon. **4**, 611–622 (2010).
7. F. Xia, T. Mueller, X. Lin, A. Valdes-Garcia, and P. Avouris, "Ultrafast graphene photodetector," Nature Nanotech. **4**, 839–843 (2009).
8. M. Jablan, H. Buljan, and M. Soljacic, "Plasmonics in graphene at infrared frequencies," Phys. Rev. B **80**, 245435 (2009).
9. A. Vakil and N. Engheta, "Transformation optics using graphene," Science **332**, 1291–1294 (2011).
10. F. H. L. Koppens, D. E. Chang, and F. J. Garcia de Abajo, "Graphene plasmonics: A platform for strong light-matter interactions," Nano Lett. **11**, 3370–3377 (2011).

11. P. Tassin, T. Koschny, M. Kafesaki, and C. M. Soukoulis, "A comparison of graphene, superconductors and metals as conductors for metamaterials and plasmonics," Nature Photon. **6** 259–264 (2012).
12. D. R. Smith, J. B. Pendry, and M. C. K. Wiltshire, "Metamaterials and negative refractive index," Science **305**, 788–792 (2004).
13. N. M. Litchinitser and V. M. Shalaev, "Photonic metamaterials," Laser. Phys. Lett. **5**, 411–420 (2008).
14. C. M. Soukoulis and M. Wegener, "Optical metamaterials—more bulky and less lossy," Science **330**, 1633–1634 (2010).
15. C. M. Soukoulis and M. Wegener, "Past achievements and future challenges in the development of three-dimensional photonic metamaterials," Nature Photon. **5**, 523–530 (2011).
16. R. A. Shelby, D. R. Smith, and S. Schultz, "Experimental verification of a negative index of refraction," Science **292**, 77–79 (2001).
17. T. Koschny, P. Markos, E. N. Economou, D. R. Smith, D. C. Vier, and C. M. Soukoulis, "Impact of inherent periodic structure on effective medium description of left-handed and related metamaterials," Phys. Rev. B **71**, 245105 (2005).
18. D. R. Smith, W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, "Composite medium with simultaneously negative permeability and permittivity," Phys. Rev. Lett. **84**, 4184–4187 (2000).
19. E. Plum, J. Zhou, J. Dong, V. A. Fedotov, T. Koschny, C. M. Soukoulis, and N. I. Zheludev, "Metamaterial with negative index due to chirality," Phys. Rev. B **79**, 035407 (2009).
20. J. Zhou, J. Dong, B. Wang, T. Koschny, M. Kafesaki, and C. M. Soukoulis, "Negative refractive index due to chirality," Phys. Rev. B **79**, 121104 (2009).
21. N. Papasimakis, V. A. Fedotov, N. I. Zheludev, and S. L. Prosvirnin, "A metamaterial analog of electromagnetically induced transparency," Phys. Rev. Lett. **101**, 253903 (2008).
22. P. Tassin, L. Zhang, T. Koschny, E. N. Economou, and C. M. Soukoulis, "Low loss metamaterials based on classical electromagnetically induced transparency," Phys. Rev. Lett. **102**, 053901 (2009).
23. J. B. Pendry, "Negative refraction makes a perfect lens," Phys. Rev. Lett. **85**, 3966–3969 (2000).
24. N. Engheta, "An idea for thin subwavelength cavity resonators using metamaterials with negative permittivity and permeability," IEEE Ant. Wireless Prop. Lett. **1**, 10 (2002).
25. P. Tassin, X. Sahyoun, and I. Veretennicoff, "Miniaturization of photonic waveguides by the use of left-handed materials," Appl. Phys. Lett. **92**, 203111 (2008).
26. A. Alu, N. Engheta, A. Erentok, and R. W. Ziolkowski, "Single-negative, double-negative and low index metamaterials and their electromagnetic applications," IEEE Antennas Prop. Mag. **49**, 23–36 (2007).
27. U. Leonhardt, "Optical conformal mapping," Science **312**, 1777–1780 (2006).
28. J. B. Pendry, D. Schurig, and D. R. Smith, "Controlling electromagnetic fields," Science **312**, 1780–1782 (2006).
29. M. Rahm, D. Schurig, D. A. Roberts, S. A. Cummer, D. R. Smith, and J. B. Pendry, "Design of electromagnetic cloaks and concentrators using form-invariant coordinate transformations of Maxwell's equations," Photon. Nanostruct.: Fundam. Appl. **6**, 87–95 (2008).
30. M. Dalarsson and P. Tassin, "Analytical solution for wave propagation through a graded index interface between a right-handed and a left-handed material," Opt. Express **17**, 6747–6752 (2009).
31. N. Papasimakis, Z. Luo, Z. X. Shen, F. De Angelis, E. Di Fabrizio, A. E. Nikolaenko, and N. I. Zheludev, "Graphene in a photonic metamaterial," Opt. Express **18**, 8353–8359 (2010).
32. F. Schedin, E. Lidorikis, A. Lombardo, V. G. Kravets, A. K. Geim, A. N. Grigorenko, K. S. Novoselov, and A. C. Ferrari, "Surface-enhanced Raman spectroscopy of graphene," ACS Nano **4**, 5617–5626 (2010).
33. J. Zhou, T. Koschny, M. Kafesaki, E. N. Economou, J. B. Pendry, and C. M. Soukoulis, "Saturation of the magnetic response of split-ring resonators at optical frequencies," Phys. Rev. Lett. **95**, 223902 (2005).

1. Introduction

Graphene, a single-atom thick layer of covalently bonded carbon atoms [1, 2], has recently emerged as an alternative for conducting materials in optical systems. Graphene derives its unusual current transport properties from the Dirac cones in its band structure at the six corners of the first Brillouin zone, which can be directly related to the arrangement of the carbon atoms in a two-dimensional honeycomb structure [2]. Having a linear dispersion relation at energies close to the cone's apex, the charge carriers are relativistic quasi-particles (also called Dirac fermions), resulting in superior low-frequency electronic [1] and mechanical properties [3] for a sheet only a single atom thick.

Advances in the fabrication of graphene using chemical vapour deposition or by epitaxial growth on silicon carbide or metals have paved the way towards optical applications [4]. One promising example is the use of graphene for transparent electrodes, owing to its relatively large DC conductivity—compared to transparent conducting oxides like indium tin oxide—and high

transparency at optical frequencies [5], but graphene has also been advertised as a versatile material for opto-electronics and terahertz technology, [6], e.g., in solar cells, light-emitting devices, display technology, and ultrafast photodetectors [7]. Graphene was also suggested as a platform for infrared surface plasmon polaritons (SPP) [8, 9, 10]. Nevertheless, although graphene indeed supports tightly bound SPPs with micrometer-scale wavelength in the infrared, our recent research efforts have demonstrated that the propagation length of such plasmons does not exceed a few SPP wavelengths at room temperature [11].

Recently, we got interested in the use of graphene in metamaterials. Metamaterials are artificially structured materials in which small, subwavelength electric circuits replace atoms as the basic unit of interaction with electromagnetic radiation [12, 13, 14, 15]. The design of appropriate constituents, such as split-ring resonators (SRR) [16], cut wires, and fishnets, allows for effectively homogeneous media [17] with exotic material response, e.g., magnetism at terahertz and optical frequencies, simultaneous negative permittivity and negative permeability (the so-called left-handed materials) [18], giant chirality [19, 20], and slow-light media [21, 22]. They may enable lenses with subwavelength resolution [23], optical systems going beyond the diffraction limit [24, 25, 26], and reflectionless photonic devices [27, 28, 29, 30].

Due to its high AC surface resistivity, graphene is unsuitable as a direct replacement for metals in the current-carrying constituents of metamaterials [11]. Instead of replacing metals by graphene, researchers at the University of Southampton have deposited a graphene layer onto a complementary split-ring metamaterial using chemical vapour deposition [31]. They find strong modification of the metamaterial's resonances reflected in a reduction of the transmission and significant broadening of the spectral features, a surprising finding given that a standalone graphene layer transmits more than 97% of infrared radiation. In this paper, we want to clarify how graphene can have such a dramatic effect on the optical response of a metamaterial with a study of the interaction of graphene with the metallic constituents of the metamaterial.

2. Graphene on a complementary split-ring metamaterial

We start this study with the complementary split-ring metamaterial of Ref. [31], which consist of an SRR-like incision in a 65 nm-thick gold film [see Fig. 1(a)] on a 102 nm-thick Si_3N_4 substrate. We have calculated the scattering parameters of this metamaterial with a time-domain electromagnetics solver (CST Microwave Studio), using periodic boundary conditions in the
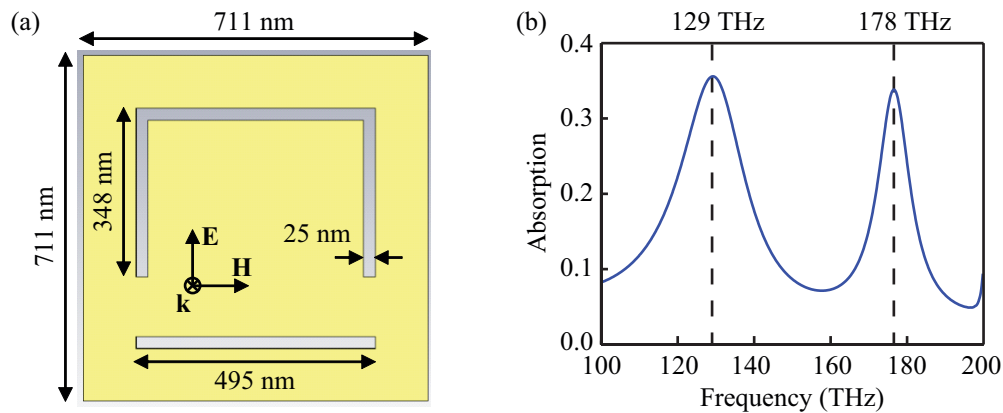

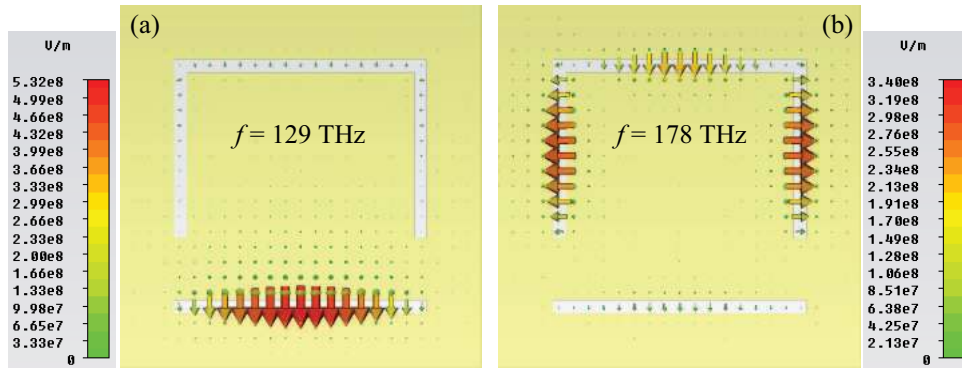Fig. 1. (a) Unit cell of the complementary SRR metamaterial. (b) Absorption spectrum of the same metamaterial.

Fig. 2. (a) Electric field patterns of the resonances of the complementary SRR metamaterial shown in Fig. 1 (without graphene). (a) Fields concentrated in the slit at $f = 129$ THz. (b) Fields concentrated in the U-shaped ring at $f = 178$ THz.
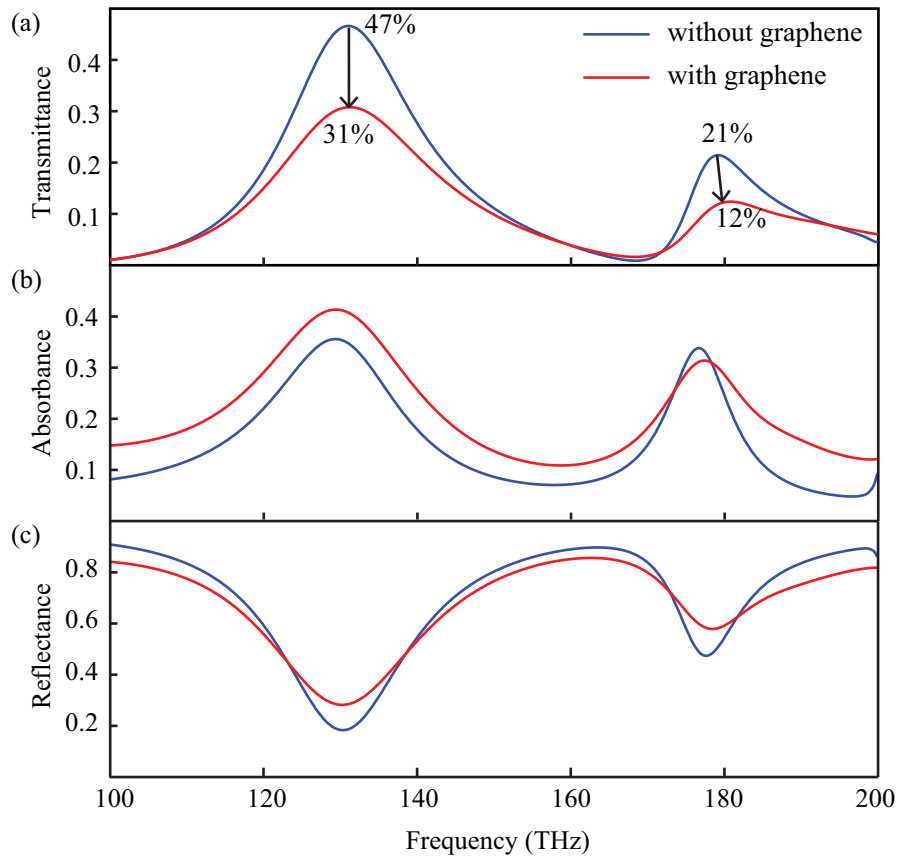

Fig. 3. Comparison between the scattering properties of the complementary SRR metamaterial with and without graphene. (a) Transmittance. (b) Absorbance. (c) Reflectance.

lateral dimensions to model the periodic array and absorbing boundary conditions in the propagation directions. Within the frequency range of interest, we observe from the absorption plotted in Fig. 1(b) that this metamaterial has two resonances—the complement of the $\lambda/2$-resonance of the "wire" with resonant fields concentrated in the bottom slit at $f = 129$ THz and the complement of the $3\lambda/2$-resonance of the U-shaped ring with resonant fields mainly in the U-shaped slit at $f = 178$ THz. The resonant field patterns at those frequencies are plotted in Fig. 2.

We now consider the same structure with a charge-neutral graphene layer deposited on top—this may require the use of a gate voltage to cancel the effect of the substrate or metal on the Fermi level. The graphene layer is modelled by a layer of thickness t and conductivity $\sigma = 6.08 \times 10^{-5}$ S$/t$, and we have subsequently decreased t until we found that the limit $t \to 0$ is sufficiently converged in our simulations at a thickness of $t = 1$ nm (results shown in Fig. 3). The transmittance of the metamaterial with and without graphene is shown in Fig. 3(a). We observe that the transmittance drops from 47% to 31% at the lower resonance frequency and from 21% to 12% at the higher resonance frequency, in agreement with experimental findings [31]. These transmission contrasts are much larger than the few percents observed for a standalone graphene sheet.

We can understand what happens by looking at the current density in the graphene layer, shown in Fig. 4. The current density simply follows the resonant electric field, $\mathbf{J}_g = \sigma_g \mathbf{E}_{res}$. This results in a dissipative power density of $\mathbf{J}_g \cdot \mathbf{E}_{res} = \sigma_g \|\mathbf{E}_{res}\|^2$. Even though the surface conductivity of graphene is small, the absorption can become significant when the resonant electric field of the metamaterial is large enough, e.g., by field enhancement in the vicinity of metals. (Note that a similar field enhancement effect has recently been observed in surface-enhanced Raman spectroscopy of graphene with plasmonic nanoparticles [32].) It is worth to note that the resonances of the metallic constituents are damped by the dissipation in the graphene layer (note the increased linewidth of the resonances when graphene is present), but there is no qualitative change in the resonant field patterns otherwise. There is, however, a difference in the response between the lower and the higher resonance frequencies. At $f = 129$ THz, the absorption [Fig. 3(b)] increases by 5% due to the additional dissipation driven by the resonant fields in the graphene layer. At $f = 178$ THz, on the other hand, the absorption decreases from 34% to 31%. The damping of this resonance is large enough to suppress the amplitude of the resonant field and, hence, there is less dissipation. Nevertheless, the dominating effect behind the transmission reduction here is not the absorption but rather the change in impedance of
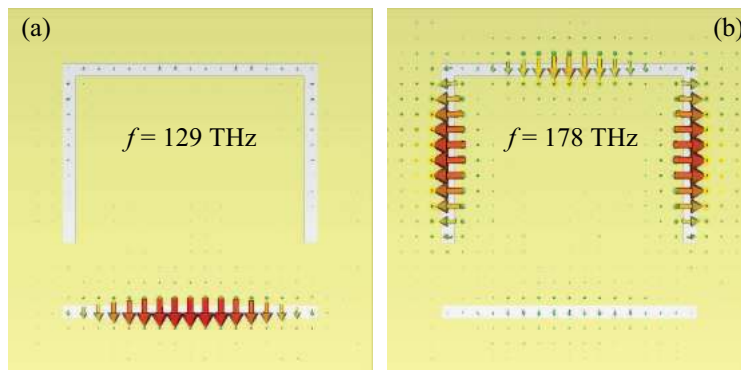

Fig. 4. Current density in the graphene sheet. The current density simply follows the resonant electric field of the resonances of the complementary SRR structure.
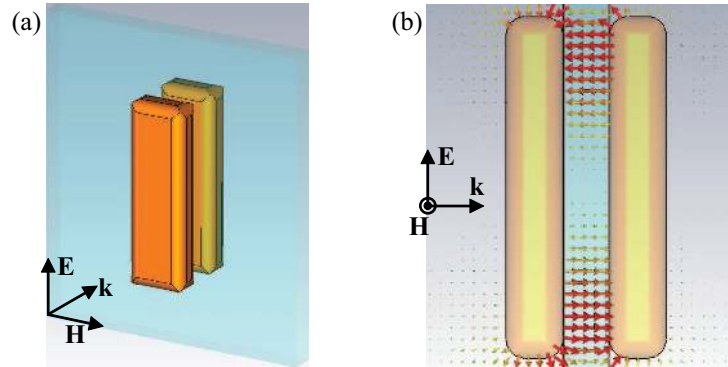
Fig. 5. (a) Unit cell of the wire-pair metamaterial. (b) Electric field at the resonance frequency of the magnetic dipole mode. Note that the large resonant electric field is mostly perpendicular to the substrate.

the metamaterial. We indeed see that the reflectance is approximately 10% larger when the resonances are damped by graphene [see Fig. 3(c)].

3. Graphene on a wire-pair metamaterial

Another important prototype of metamaterials is the wire-pair metamaterial. Wire pairs ease the fabrication and characterization of (negative-index) metamaterials at optical frequencies and also increase the saturation frequency [33]. One might, therefore, want to deposit graphene onto a wire-pair structure. The unit cell of the wire-pair metamaterial we have simulated contains two 525 nm-long, 164 nm-wide, 87.5 nm-thick gold wires deposited on a 70 nm-thick substrate with dielectric constant $\varepsilon = 2.25$, as shown in Fig. 5(a). This structure has a magnetic dipole mode with resonance frequency $f = 149.5$ THz. The resonant electric field of this mode is shown in Fig 5(b).

We now add a layer of graphene to the bottom of the gap between the wire pairs (the metamaterial is illuminated from the top). We find there is hardly any change between the transmittance spectra of the wire-pair metamaterial with and without the graphene layer present [see Fig. 6(a)]. The largest reduction in transmittance is slightly less than 4%. From the absorbance spectra in Fig. 6(b), we see that the resonance is only minimally broadened. We can understand the insensitivity of the wire pair to graphene from the topology of the resonant electric field of the wire pair [see Fig. 5(b)]. The large resonant electric field between the charges at the end of the wires is mainly perpendicular to the graphene sheet and, hence, cannot accelerate the charge carriers in graphene. The graphene sheet simply attenuates the incident field, but does not interact with the metamaterial. This is also confirmed by the reflectance spectra in Fig. 6(c), which show that the effective wave impedance due to the resonance is unaltered. The wire-pair metamaterials thus confirms the theory of interaction between metamaterials and graphene described above.

4. Discussion

From the complementary SRR metamaterial, we conclude that—despite its small surface conductivity—graphene can significantly damp a resonance of a quasistatic or plasmonic element when it overlaps with the strong resonant electric fields generated by the currents in the metallic elements. From the wire-pair results, we find that this effect does not occur if the
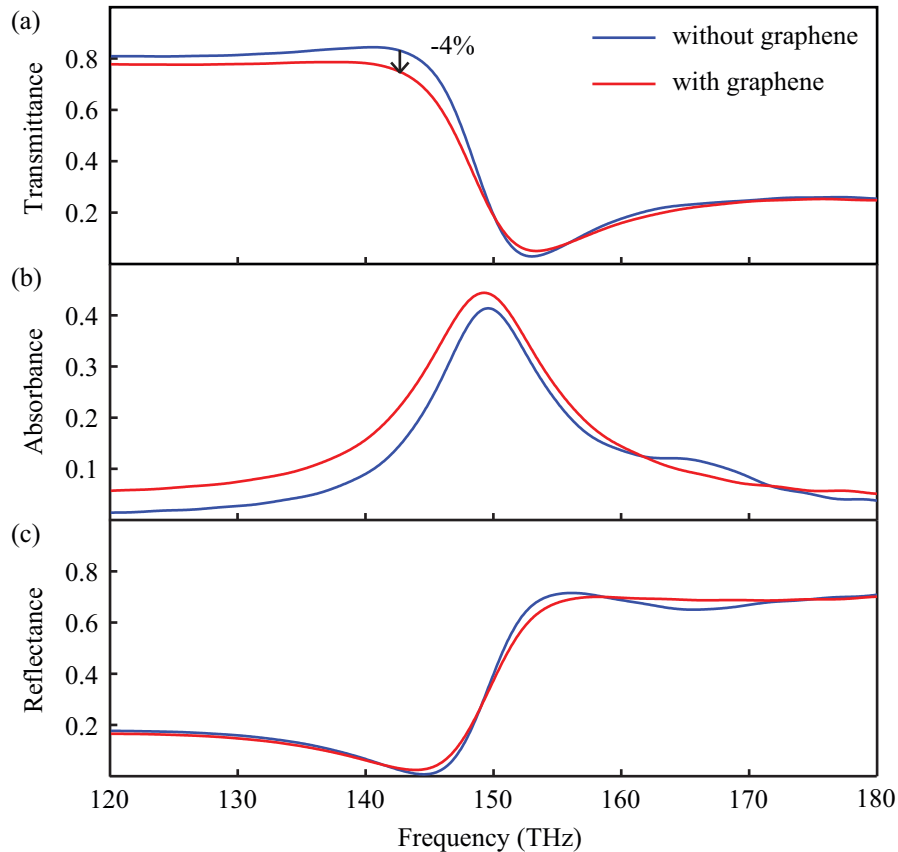
Fig. 6. Comparison between the scattering properties of the wire-pair metamaterial with and without graphene. (a) Transmittance. (b) Absorbance. (c) Reflectance.

resonant electric field is perpendicular to the plane of the graphene sheet. These conclusions may be useful in the design of tunable metamaterials. Graphene can indeed be biased by an electric potential, allowing for electro-optic control over the metamaterial's response with potentially very high modulation rates [7]. This approach will only work for metamaterials of the SRR-type with resonant electric fields predominantly parallel to the graphene flake, as opposed to wire-pair/fishnet metamaterials that cannot interact with graphene because their resonant electric field is predominantly perpendicular to the planar structure.

Acknowledgments