Resonance enhancement of terahertz metamaterials by liquid crystals/indium tin oxide interfaces

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Abstract: This work fabricates a terahertz (THz) metamaterial device. whose structure consists of split ring resonator array/ plastic substrate/ Indium Tin Oxide (ITO) film/ liquid crystals/ ITO film/ plastic substrate. Experiment results show that the resonance of the THz metamaterial device can be enhanced as voltage is applied to the liquid crystals. The enhancement will be more significant as higher voltage applied. The resonance enhancement is attributed to the fact that the liquid crystals/ITO interfaces exhibit the large difference in terms of refractive index between the two materials in THz regime. The interfaces reflect the incident electromagnetic wave and cause the reflected wave to enhance the resonance of the metamaterials. As those frequency-tunable metamaterial devices show different resonant transmittance at different frequencies, which is undesired, the liquid crystals/ITO interfaces can improve those frequency-tunable metamaterial devices with a constant transmittance at different frequencies.

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References and links

- H. T. Chen, J. F. O'Hara, A. K. Azad, A. J. Taylor, R. D. Averitt, D. B. Shrekenhamer, and W. J. Padilla, 1 "Experimental demonstration of frequency-agile terahertz metamaterials," Nat. Photonics 2(5), 295–298 (2008).
- W. J. Padilla, M. T. Aronsson, C. Highstrete, M. Lee, A. J. Taylor, and R. D. Averitt, "Electrically resonant 2. terahertz metamaterials: theoretical and experimental investigations," Phys. Rev. B 75(4), 041102 (2007).
- H. Tao, C. M. Bingham, D. Pilon, K. Fan, A. C. Strikwerda, D. Shrekenhamer, W. J. Padilla, X. Zhang, and R. D. Averitt, "A dual band terahertz metamaterial absorber," J. Phys. D Appl. Phys. 43(22), 225102 (2010).
- N. R. Han, Z. C. Chen, C. S. Lim, B. Ng, and M. H. Hong, "Broadband multi-layer terahertz metamaterials 4. fabrication and characterization on flexible substrates," Opt. Express 19(8), 6990-6998 (2011).
- D. Shrekenhamer, W. C. Chen, and W. J. Padilla, "Liquid crystal tunable metamaterial perfect absorber," 5. arXib:1206.4214v1 (2012).
- F. Zhang, W. Zhang, Q. Zhao, J. Sun, K. Qiu, J. Zhou, and D. Lippens, "Electrically controllable fishnet metamaterial based on nematic liquid crystal," Opt. Express 19(2), 1563-1568 (2011).
- Q. Zhao, L. Kang, B. Du, B. Li, J. Zhou, H. Tang, X. Liang, and B. Zhang, "Electrically tunable negative 7. permeability metamaterials based on nematic liquid crystals," Appl. Phys. Lett. 90(1), 011112 (2007).
- F. Zhang, Q. Zhao, W. Zhang, J. Sun, J. Zhou, and D. Lippens, "Voltage tunable short wire-pair type of metamaterial infiltrated by nematic liquid crystal," Appl. Phys. Lett. **97**(13), 134103 (2010).
- 9 S. A. Jewell, E. Hendry, T. H. Isaac, and J. R. Sambles, "Tuneable Fabry-Perot etalon for terahertz radiation," New J. Phys. 10(3), 033012 (2008).
- 10. Y. S. Jin, G. J. Kim, and S. G. Jeon, "Terahertz dielectric properties of polymers," J. Korean Phys. Soc. 49(2), 513-517 (2006).
- 11. C. L. Pan and R. P. Pan, "Characterization and applications of liquid crystals in the THz frequency range," Proc. SPIE 8279, 82790I (2012).
- 12. E. Hecht, Optics (Addison Wesley, 2002), Chap. 4.

- Z. Schlesinger and A. J. Sievers, "IR surface-plasmon attenuation coefficients for Ge-coated Ag and Au metals," Phys. Rev. B 26(12), 6444–6454 (1982).
- 14. J. Han, X. Lu, and W. Zhang, "Terahertz transmission in subwavelength holes of asymmetric metal-dielectric interfaces: the effect of a dielectric layer," J. Appl. Phys. **103**(3), 033108 (2008).
- 15. K. Y. Lo, C. Y. Huang, T. H. Chu, C. J. Hsu, C. H. Lin, and A. Y. G. Fuh, "Variation of nematic liquid crystal on a silver surface," J. Opt. A, Pure Appl. Opt. 8(6), 501–506 (2006).
- A. Boltasseva and V. M. Shalaev, "Fabrication of optical negative-index metamaterials: recent advances and outlook," Metamaterials (Amst.) 2(1), 1–17 (2008).
- 17. J. F. O'Hara, R. Singh, I. Brener, E. Smirnova, J. Han, A. J. Taylor, and W. Zhang, "Thin-film sensing with planar terahertz metamaterials: sensitivity and limitations," Opt. Express **16**(3), 1786–1795 (2008).
- V. G. Chigrinov, V. M. Kozenkov, and H. S. Kwok, *Photoalignment of Liquid Crystalline Materials: Physics and Applications* (Wiley, 2008).
- F. Zhang, L. Kang, Q. Zhao, J. Zhou, X. Zhao, and D. Lippens, "Magnetically tunable left handed metamaterials by liquid crystal orientation," Opt. Express 17(6), 4360–4366 (2009).
- X. Wang, D. H. Kwon, D. H. Werner, I. C. Khoo, A. V. Kildishev, and V. M. Shalaev, "Tunable optical negative-index metamaterials employing anisotropic liquid crystals," Appl. Phys. Lett. 91(14), 143122 (2007).
- B. Zhu, Y. Feng, J. Zhao, C. Huang, and T. Jiang, "Switchable metamaterial reflector/absorber for different polarized electromagnetic waves," Appl. Phys. Lett. 97(5), 051906 (2010).

1. Introduction

Because of their negative index property in the terahertz (THz) frequency regime, THz metamaterials has attracted great attention recently [1–5], while frequency tunability and board-band capability of THz metamaterials remain as technical challenges. Liquid crystals have received considerable interest in the development of tunable THz metamaterial devices due to the large birefringence of liquid crystals [5–8]. In these studies, liquid crystals are used to act as the surrounding media of the metamaterials, as electrical field, magnetic field or heat is applied to the metamaterial devices embedded with liquid crystals, the resonance frequency of the metamaterials is shifted because the probe electromagnetic wave is experienced the variation in the permittivity or the permeability of the liquid crystals. As a drawback, when resonance frequency shifting, the magnitude of resonance transmittance is changed as well which is undesirable for commercial tunable electro-optic devices. It is of great interests that researchers want to develop an approach to a constant resonance intensity of the THz metamaterials even the resonance frequency of the metamaterials even the resonance frequency of the metamaterial devices are tuned by the aforementioned methods.

This work fabricates a THz metamaterial device, where split ring resonator (SRR) array is deposited on a liquid crystal cell which is formed by two inward-facing indium tin oxide (ITO)-coated plastic substrates, to study the effect of the liquid crystals/ITO interfaces on the resonance of the SRR array. The reason for choosing ITO is due to its good conductivity so that we can bias liquid crystals by applying voltage and its large refractive index in THz regime. The liquid crystals are used because their refractive index can be controlled by varying the applied voltage to it. Experiment results show that the resonance of the SRR array is enhanced as a voltage is applied to the liquid crystal cell, and that an increase in the voltage makes the resonant transmittance lower. The resonance enhancement results from the coupling of the surface-plasmon-polarition-like mode and the electromagnetic wave reflected from the liquid crystals/ITO interfaces. The liquid crystals/ITO interfaces can be used to keep the resonant transmittance of the proposed metamaterial devices constant by external voltages [5–8].

2. Preparation of samples

Figure 1(a) depicts the configuration of the THz metamaterial device. An empty cell is fabricated using two ITO-coated polyester (PET) substrates, which are separated by two 100- μ m-thick spacers. The thicknesses (permittivities) of the ITO films and PET substrates, d_{ITO} and d_{PET} (ε_{ITO} and ε_{PET}), are 100 nm and 175 μ m (6400 [9] and 3.0 [10]), respectively. On the top PET substrate of the cell is deposited a 200-nm-thick aluminum SRR array using lift-off process. The empty cell is filled with E7 liquid crystals (Merck, $\varepsilon_{\parallel} = n_e^2 = 2.90$ and

 $\varepsilon_{\perp} = n_o^2 = 2.48$ at THz [11]), then sealed with polymer gel. In order to achieve free polarization for the liquid crystals, no aligning layer has been adopted in the case. The reason of preferring liquid crystals with free polarization is to make the characterization simpler since no polarized light is needed as well. Figure 1(b) shows the dimensions of the SRR, where the line-width (*T*), gap (*G*), width (*W*) and length (*L*) are 6, 20, 50 and 60 µm, respectively. A reflective optical microscopy image of the SRR array is shown in Fig. 1(c). Both of the periods along *x* and *y* directions are 100 µm. As shown in Fig. 1(d), the flexibility and transparency of the metamaterial device are demonstrated by pasting it on a cylindrical plastic bottle.

This study will use external voltages to modify the permittivity of the liquid crystals, ε_{LC} . Owing to the orientation of the liquid crystals in the THz metamaterial device is isotropic, ε_{LC} at zero applied voltage is given by $\varepsilon_{iso} = (\varepsilon_{\parallel} + 2\varepsilon_{\perp})/3 = 2.62$ for a non-polarized THz electromagnetic wave which is normally incident to the metamaterial device. When the applied voltage is high enough, ε_{LC} will equal $\varepsilon_{\perp} = 2.48$ since the electrical field of the electromagnetic wave makes an angle of 90° with respect to the director. Therefore, the permittivity of the liquid crystals decreases with the increasing applied voltage.



Fig. 1. (a) Configuration of metamaterial device. (b) Dimensions of SRR unit: $T = 6 \mu m$, $G = 20 \mu m$, $W = 50 \mu m$ and $L = 60 \mu m$. (c) Optical microscope image for the SRR array. (d) Photograph of the metamaterial device, which is pasted on a transparent cylindrical plastic bottle.

3. Results and discussion

Figure 2 shows the transmission spectrums of the metamaterial device at applied voltages of 0 V, 4 V and 10 V. These spectrums are obtained by THz time-domain spectroscopy (TeraView, TPS 3000) in transmission mode, where the non-polarized THz electromagnetic wave as the probe light is normally incident to the metamaterial device from the side of the SRR array. In Fig. 2, the resonant transmittance decreases from -16 dB to -19 dB as a voltage of 4V is applied to the metamaterial device. In addition, an increase in the voltage makes the resonant transmittance lower. The two results reveal that the resonance of the SRR array is enhanced with the voltage. The resonance enhancement results from the liquid crystals/ITO interfaces. The liquid crystals/ITO interfaces exhibit high difference in refractive index between these two materials. For the normally incident THz electromagnetic wave, the reflectance (*R*) by the liquid crystals/ITO interfaces can be written as [12]

$$R = \left(\frac{n_{\rm ITO} - n_{\rm LC}}{n_{\rm ITO} + n_{\rm LC}}\right)^2,\tag{1}$$

where n_{ITO} and n_{LC} are the refractive indexes of the ITO and liquid crystals, respectively. If there is no applied voltage, R = 0.922 is yielded by substituting $n_{\text{LC}} = n_{\text{iso}} = (n_e + 2n_o)/3 = 1.62$ and $n_{\text{ITO}} = 80$ into Eq. (1). For a voltage is applied to the liquid crystal layer, R equals 0.924 by substituting $n_{\text{LC}} = n_o = 1.58$ and $n_{\text{ITO}} = 80$ into Eq. (1). Therefore, the increase in the voltage makes R higher, which subsequently gives rise to the increase in the intensity of the reflected wave. The reflected wave couples with the surfaceplasmon-polarition-like mode and enhances the resonance of the SRR array. In next paragraph, respective simulations are performed to confirm that the resonance enhancement of the SRR array arises from the liquid crystals/ITO interfaces. The experimental result in Fig. 2 reveals that the metamaterial device is an intensity-switchable band-stop filter in THz regime.



Fig. 2. Transmission spectrums of the metamaterial device at applied voltages of 0 V, 4 V and 10 V.

To study the effect of the liquid crystals/ITO interfaces on the resonance enhancement of the SRR array, a simulation, in which the configuration of a metamaterial device is the same as that in Fig. 1(a) while the device has no ITO films, is carried out by commercial finiteintegration time-domain software. The material parameters that are used in the simulation refer to Section 2. Under $\varepsilon_{LC} = \varepsilon_{iso} = 2.62$ (i.e., at zero voltage), the resonant transmittance is lower in the metamaterial device with ITO films than the case without ITO films, as shown in Fig. 3(a). This result reveals that the ITO films of the metamaterial device play a key role in the resonance enhancement of the SRR array. Figure 3(b) shows the simulated transmittance spectrums of the metamaterial device with the ITO films under various ε_{LC} . These spectrums are used to study the responses of the metamaterial device at various voltages. In Fig. 3(b), the resonant transmittance decreases with the decreasing $\varepsilon_{\rm LC}$ (i.e., the increasing voltage). This simulated result is consistent with the experimental result of Fig. 2, and demonstrates that high difference between $\varepsilon_{\rm LC}$ and $\varepsilon_{\rm TO}$ results in the enhancement of the resonance of the SRR array. Moreover, as a material whose refractive index is higher than ITO in THz regime is substituted for the ITO films of Fig. 1(a), the resonant transmittance of the metamaterial device with the high-permittivity material ($\varepsilon_{\rm M} = 10^4$) decreases in comparison with that of the metamaterial device with the ITO films, as shown in Fig. 3(c). The simulated results in Figs. 3(a), 3(b) and 3(c) confirm that the resonance enhancement of the metamaterial device with the ITO films results from the liquid crystals/ITO interfaces. Therefore, the difference of refractive index at the interfaces has the capability to modify the resonant transmittance of a THz metamaterial device.



Fig. 3. (a) Simulated transmission spectrums of metamaterial devices with and without ITO films under $\varepsilon_{LC} = \varepsilon_{iso} = 2.62$ (i.e., at zero voltage). (b) Simulated transmission spectrums of metamaterial device with ITO films with various ε_{LC} . (c) Simulated transmission spectrums of metamaterial device with a high-permittivity material ($\varepsilon_M = 10^4$).

In Fig. 2, the resonant frequency of the metamaterial device does not shift under the applied voltage. This is due to the fact that the thickness of the top PET substrate, $d_{\text{PET}} = 175$ µm, is greater than its critical thickness, d_c [13,14]. d_c can be written as

$$d_{\rm c} = \frac{c}{4f\sqrt{\varepsilon_{\rm PET} - 1}},\tag{2}$$

where *c* is the speed of the surface-plasmon-polarition-like mode in vacuum, and *f* refers to the resonant frequency. Substituting f = 0.62 THz and $\varepsilon_{\text{PET}} = 3.0$ into Eq. (2) yields $d_c = 81.6$ µm. Since the condition $d_{\text{PET}} > d_c$ is satisfied, the electric field of the surface-plasmon-polarition-like mode almost confines in the top PET substrate. Therefore, the resonant frequency of the metamaterial device depends mainly on the permittivity of the top PET substrate and remains constant under the applied voltages.

4. Application

Before the application of the liquid crystals/ITO interfaces is demonstrated, a metamaterial device whose configuration is slightly different from the metamaterial device of Fig. 1(a) in terms of the SRR-deposited surface is designed, as shown in Fig. 4(a). In Fig. 4(a), the SRR array of the metamaterial device is deposited on the ITO film of the top PET substrate. Consequently, the liquid crystals are regarded as the surrounding media of the SRR array. This study uses thermal evaporation and lift-off process to fabricate the metal SRRs, as shown in Fig. 1(c). The thermal evaporation should induce the surface roughness of the metal thin film to be larger than the dimensions of the LC molecules [15]. Besides, the lift-off process makes the sidewall of the metal SRRs rough [16]. Therefore, the orientation of the LC molecules in Fig. 4(a) is regarded as isotropy at no applied voltage. From Fig. 4(b), both the simulated resonant frequency and transmittance vary with the decreasing the permittivity of the liquid crystals, ε_{LC} . The shift of the resonant frequency is attributed to the fact that a SRR unit can be considered as an inductive-capacitive resonator circuit, and its resonant frequency (f) is given by $f = 1/2\pi\sqrt{LC}$, where the inductance L is determined by the effective enclosed area of the SRR unit, and the capacitance C is related to the permittivity of the surrounding medium and the gap size [17]. Figure 4(b) indicates that the resonant frequency of the metamaterial device is tunable as the liquid crystals are applied external voltages.



Fig. 4. (a) Configuration of metamaterial device, where liquid crystals are regarded as the surrounding media of SRR array. (b) Simulated transmittance spectrums of the device under various ε_{LC} .

The resonant transmittance, for the metamaterial device which adopts the liquid crystals/ITO interfaces to enhance the resonance, decreases with decreasing $\varepsilon_{\rm LC}$, as shown in Fig. 3(b). Meanwhile, the resonant transmittance, for the metamaterial device which utilizes the liquid crystals to tunable the resonant frequency, increases with decreasing $\varepsilon_{\rm LC}$, as shown in Fig. 4(b). The two results may be helpful to develop a frequency-tunable metamaterial device with a constant transmittance at different frequencies. Consider a metamaterial device, as shown Fig. 5(a), which is formed by stacking the two metamaterial devices in Figs. 1(a)and 4(a) and then substituting a double-side ITO-coated PET substrate for the two single-side ITO-coated PET substrates. Figure 5(b) shows the simulated transmittance spectrums of the m for the top and bottom liquid crystal layers with different permittivities. In Fig. 5(b), the four resonant spectrums exhibit the same resonant transmittance. It is attributed the fact that the liquid crystals/ITO interfaces adjacent to the bottom liquid crystal layer makes the resonant transmittance of the SRR array constant. The liquid crystals/ITO interfaces has the ability to develop a frequency-tunable metamaterial device with a constant transmittance at different frequencies. The liquid crystals/ITO interfaces can be stacked on absorbers, cloaking and filters due to the flexible PET substrates, hence exhibiting its wide range of applications. Moreover, the liquid crystals/ITO interfaces have features such as easy fabrication and low cost. For future study, photoalignment which has the capability of modifying the permittivity of liquid crystals could be used in such metamaterial structures so as to achieve opticallycontrollable metamaterial devices [18].

Liquid crystals have been used to modify the resonant frequency of the SRRs in the previous works [19,20], but they have not been utilized to tunable the resonant transmittance of SRRs. Moreover, previous work used an electromagnetic resonant structure embedded with PIN diodes to fabricate a transmittance-tunable metamaterial device [21]. It is difficult for this device to work in high frequencies such as THz and visible regimes because the diodes are very large for THz and visible metamaterials (micro- and nano-scale structures). In contrast to the PIN diodes, the PET/ITO/liquid crystals/ITO/PET structure can be stacked on other metamaterials due to the planar design. Therefore, the planar structure which resembles a sticker exhibits high convenience on application.



Fig. 5. (a) Schematic drawing of stacked metamaterial device. (b) Simulated transmission spectrums of the device with various permittivities of the liquid crystals. $\varepsilon_{LC_{top}}$ and $\varepsilon_{LC_{bot}}$ are the permittivities of the top and bottom liquid crystal layers, respectively.

5. Conclusions

In conclusion, we report the THz metamaterial device whose resonant transmittance decreases with increasing voltages while its resonant frequency remains at the same value. The decrease in the resonant transmittance results from the liquid crystals/ITO interfaces with large difference in refractive index between these two materials in THz regime. The fixed resonant frequency is attributed to the fact that the electric field of the surface-plasmon-polarition-like mode almost confines in the top PET substrate and therefore the frequency depends mainly on the permittivity of the top PET substrate. By leveraging the switchable resonant transmittance and the fixed resonant frequency, we propose the improved method for those frequency-tunable metamaterial devices so that they can exhibit constant transmittance at different frequencies. Owing to that no aligning layer is needed in the THz metamaterial device, this device has a feature of free polarization for using in band-stop filters.

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