Temperature Control of Terahertz Metamaterials With Liquid Crystals

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(Invited Paper)

***Abstract*—We design and experimentally characterize terahertz (THz) electric split-ring-resonator metamaterials infiltrated with nematic liquid crystals (LCs). We demonstrate that the resonant response of such metamaterials can be tuned by temperature due to the change of the LC arrangement from nematic to isotropic phase. In particular, we show that the frequency tuning is strongly dependent on the initial LC orientation, with resonant frequency shifting towards lower frequencies for LC director perpendicular to the gap and towards higher frequencies for LC director parallel to the gap of the electric split ring resonators. Our results open new opportunities for the design of tunable THz filters based on LCs.**

***Index Terms*—Liquid crystals (LCs), metamaterials, terahertz, thermal tuning.**

I. Introduction

METAMATERIALS have attracted significant attention in the scientific community because they provide a pathway for obtaining exotic electromagnetic properties not found in natural materials [1]. Metamaterials stimulated the development of novel concepts such as negative refraction [2], perfect lenses [3], perfect absorbers [4], as well as transformation optics [5]. At the same time, the development of real-life applications of such materials is hindered by a range of factors, including losses and narrowband response. Indeed, metamaterials are based on resonant elements, and therefore they exhibit their desired properties only in a narrow frequency range. This spectral range is generally determined by the geometry of the structure and the properties of the constituent materials. While the geometry of the composite structure is often defined at the manufacturing stage, any tunability has to be achieved through the external change of the properties of the constituent materials.

Tunability of metamaterials is particularly important for applications in the terahertz (THz) frequency range, where the technology for THz modulators, absorbers, and tunable filters is not yet mature. The use of metamaterials in THz technologies promises to enable novel functional THz devices with enhanced tunability over a large spectral range. As such, various approaches for active tuning and modulation of THz metamaterial properties have been explored. These include THz metamaterial tunability through photo- and electro-excitation of free carriers [6]–[8], mechanical movement of elements [9], phase-change [10] and superconducting materials [11], as well as liquid crystal (LC) infiltration [12], [13]. The LC-tuning approach, however, has proven to be the most versatile due to its easy post-fabrication implementation and several physical tuning mechanisms. Such mechanisms are based on tuning of the LC refractive index through external electric and magnetic fields or temperature [14].

Many ideas for LC tuning of metamaterials were first explored at optical and microwave frequencies. Examples include LC infiltrated metamaterials tunable by electric [15] or magnetic [16] fields at microwaves. However, at microwave frequencies, the near-field of metamaterial modes extends over several millimeters, which requires the use of large amounts of LC, making such devices impractical in terms of cost. In optics, thermal [17], all-optical [18] and electrooptic tunability [19] of metamaterials infiltrated with LCs have also been demonstrated. However, due to the strong anchoring of the LC to the surfaces of the metamaterials, at optical frequencies the tunability is largely suppressed. Because of this strong anchoring, the LC layer adjacent to the surface (~100 nm thick, which largely coincides with the near-field zone of the optical metamaterials) appears 'frozen' and no significant reorientation of the LC molecules occurs within this layer [19].

In contrast, the near-field of THz metamaterials extends over 20–100 μm, well beyond the anchoring layer of the LC. In addition, only a small volume of LC is required to fill the entire near-field region of the metamaterial, which is similar to the typical size of an LC cell in commercial TV displays. As such, THz metamaterials seem ideal for integration with LC and the demonstration of LC tunable metamaterial devices at THz is of the utmost importance.

Here, we study and demonstrate experimentally the thermal tunability of a LC infiltrated electric split-ring resonator (eSRR) metamaterial operating at THz frequencies. The metamaterial is

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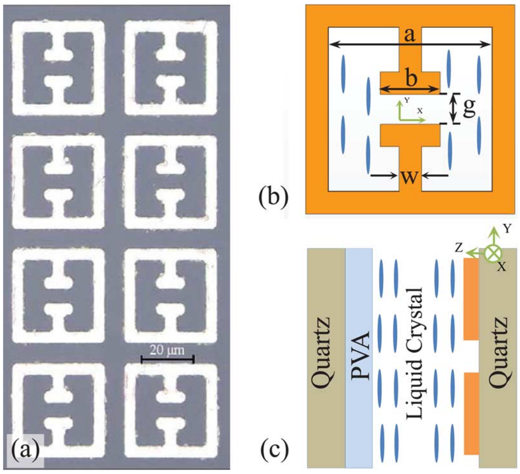

Fig. 1. (a) Optical microscope image of the fabricated metamaterial before infiltration with liquid crystal. (b) Schematic top view of the unit cell of the metamaterial, containing an eSSR and LC. Ellipses show the orientation direction of the LC molecules with respect to the resonator. (c) Schematic cross section of the LC cell containing the metamaterial on a quartz substrate, cover slip with PVA alignment layer, and LC in between.

infiltrated with a nematic LC, having two possible orientations (predefined by the top alignment layer), either perpendicular or parallel to the gap of the eSRRs. Because the state of polarization of the LC molecules can be switched by temperature between the anisotropic and isotropic states, thus changing the effective index of the LC, we are able to demonstrate increase and decrease of the resonant frequency of the metamaterial. We note that, while an LC electrically tunable absorber [12] and bandstop filter [13] have been previously demonstrated, here we study, for the first time, to the best of our knowledge, the *thermal tuning of THz metamaterial transmission* coupled to the LCs.

II. Sample Design and Fabrication

Our metamaterial consists of periodically arranged eSRRs, as shown in Fig. 1. The eSRRs are made from 100 nm thick gold deposited on a 1 mm-thick quartz substrate. The eSRRs form a square lattice with a lattice constant of 44 μm. The overall structure size is 5 mm by 5 mm. An additional 20 nm layer of titanium provides adhesion between the gold layer and the quartz substrate. The resonators are fabricated using ultra-violet lithography and lift-off. Fig. 1(a) shows an optical microscope image of the fabricated structure before infiltration with LC. The parameters of the SRR, as indicated in Fig. 1(b) are $a = 36$ μm, $b = 11$ μm, $g = 6$ μm and $w = 4$ μm.

We use E7 LC from Merck, which is infiltrated between the metamaterial and an additional 1-mm-thick quartz cover slip [see the schematic cross section of the structure in Fig. 1(c)]. We study two different alignment directions of the LC's extraordinary axis relative to the metamaterial. The first is perpendicular to the eSRR gap shown in Fig. 1(b) (y-orientation), and the second is parallel to the gap (x-orientation). To pre-align the molecules of the LC, we spin-coat a polyvinyl alcohol (PVA) layer onto the top cover slip [Fig. 1(c)] and rub it with microfiber cloth in the direction perpendicular or parallel to the gap of the eSRRs. This ensures that, at low temperature, a significant proportion of molecules are oriented along the selected directions. The thickness of the LC layer is determined by two dielectric spacers and is approximately 100 μm. A 10-Ω resistor mounted on a metal plate touching the top quartz cover and a dc power supply are used for heating the LC-metamaterial cell up to 71 °C. A thermocouple is used for monitoring the temperature of the cell. In its anisotropic phase, our LC is characterized by an ordinary index $n_o = 1.59$ and an extraordinary index $n_e = 1.74$ at 1 THz [20], [21]. By increasing the temperature, we increase the internal energy of the LC molecules, and the LC undergoes a transition to the isotropic phase with random orientation of the molecules. The refractive index of the LC in this phase does not depend on the polarization or temperature and is described by $n = 2n_o/3 + n_e/3 = 1.64$ [20].

III. Metamaterial Tuning: Measurements and Simulations

The transmission through the LC-metamaterial cell is measured using a THz time-domain spectroscopy system from EKSPLA. The polarization of the incident field is aligned across the gaps of the eSRRs (y-direction). For the orthogonal polarization, our structure does not have any resonant features in the frequency range of interest, and therefore we do not expect any tunability. The system is excited by a short THz pulse, and the transmission response is sampled in time. By performing a Fourier transform, one may obtain both the amplitude and the phase of the transmission coefficient. The multilayered structure of our sample produces multiple ripples in the spectrum of the transmitted signal due to Fabry-Perot resonances. To eliminate this effect we use time gating, zeroing the part of the temporal signal which corresponds to multiple scattering inside the sample. This leaves us with the spectrum of the signal directly transmitted through the sample, which is shown with a solid line in Fig. 2(c). The results are further normalized to the transmission spectrum through two quartz plates and LC without the metamaterial [see Fig. 2(c)]. The normalized results are presented in Fig. 2(b).

Numerical simulations are performed using the commercial electromagnetic solver CST Microwave Studio. In our simulations, we have used normally incident waves polarized across the gap of the metamaterial resonator. Fig. 2(a) shows the calculated transmission coefficients for the metamaterial before and after infiltrating with the LC. In the absence of the LC, a resonant dip in transmission appears at 1.351 THz. This resonant dip at the room temperature of 24 °C shifts to a lower frequency of 1.162 THz when a 100-μm-thick LC layer is aligned in x-direction and 1.147 THz when the LC is aligned in y-direction of the metamaterial. Fig. 2(b) shows the corresponding experimental transmission coefficients of the metamaterials without and with the LC layer in both orientations. A resonant dip appears at 1.291 THz without the LC. After infiltrating the metamaterial with LC, the resonant dip shifts to 1.161 THz for the x-orientation and 1.154 THz for the y-orientation.

Next, we study the change of the LC-metamaterial resonance with increasing LC temperature. The sample is heated in steps of 5 °C to 10 °C, and thermal equilibrium is reached before the transmission is measured. The experimentally measured transmission through the sample of LC aligned in the x- and y-directions is shown in Fig. 3(a) and 4(a), respectively, for temperatures of 24 °C, 34 °C, 52 °C, and 71 °C. As the temperature increases, the resonant dip in the x-orientation moves to lower frequencies, whereas the resonant dip moves to higher frequencies in the y-orientation. We also extract the phase of both samples at each temperature as shown in Figs. 3(b) and

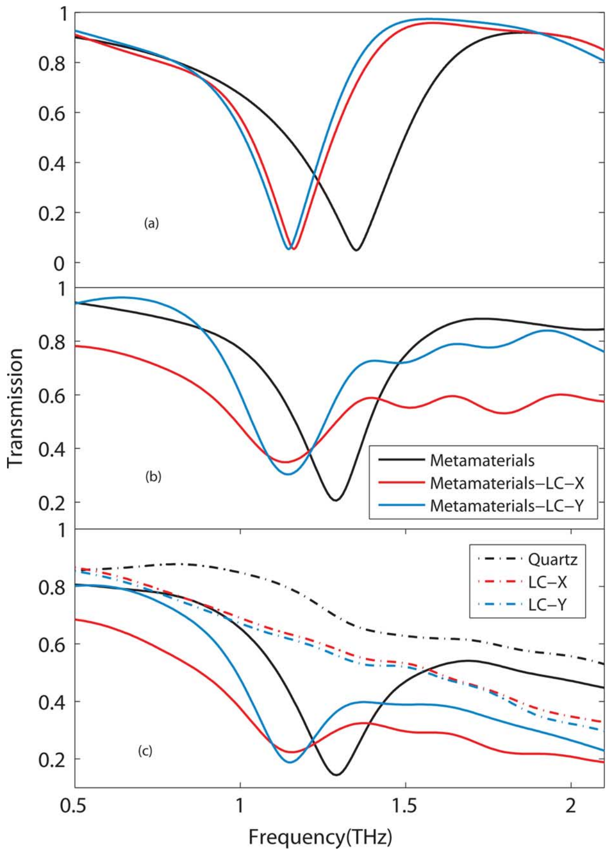

Fig. 2. Transmission coefficients as a function of frequency before and after infiltration for both orientations of the LC. (a) Numerically calculated and (b) normalized experimentally measured coefficients. (c) Transmission coefficients for the metamaterial before and after infiltration (solid lines), as well as transmission through the pair of quartz substrates, and LC cells without metamaterial for two different pre-alignment orientations, which are used for normalization.

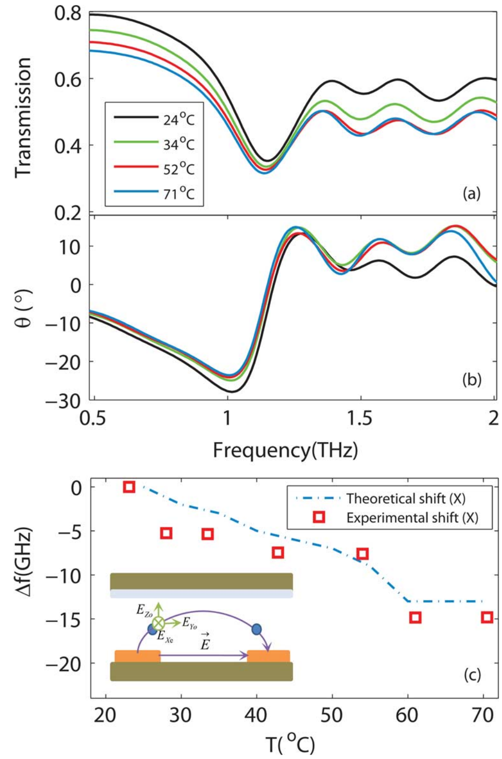

Fig. 3. Experimentally measured results with the LC aligned in the x-direction. (a) Extracted transmission curves, (b) extracted phase curves, and (c) theoretical shift (dashed line) of the resonant frequency and experimental shift (points) estimated from transmission and phase as a function of temperature. The inset shows the dominant field component in the eSRR gap relative to the LC orientation.

4(b). It shows the same trend with increasing temperature as the transmission. To find the resonant frequency at each temperature, we use a Lorentz oscillator model to simultaneously fit the amplitude and phase of the experimental transmission data over a 0.3-THz bandwidth. A nonlinear optimization is performed to minimize χ^2, which parametrizes the difference between the model and experimental data. The uncertainty in frequency was calculated based on the fitting error and the sensitivity of the fitting measure χ^2 to errors in the measured data. In all cases, the resulting uncertainty in resonant frequency (using the criterion of two standard deviations) is less than 1 GHz.

The extracted dependence of the frequency shift $\Delta f = f(T) - f(24\,^{\circ}\mathrm{C})$ is shown in Fig. 3(c) for x-orientation and in Fig. 4(c) for y-orientation of the LC molecules. In Figs. 3(c) and 4(c), the points show the experimentally measured frequency shift as a function of temperature, whilst the dashed curve shows the results of numerical simulation. For numerical simulations, the temperature dependence of the ordinary and extraordinary indices of refraction for the anisotropic phase, and the average value of the isotropic phase are taken from [20]. At room temperature, the molecules of the LC are predominantly oriented along the alignment marks in the top PVA layer, however their orientation becomes random due to the increase of temperature and the subsequent thermal agitation of molecules above the critical temperature (~60 °C for E7 LC).

IV. Discussion

The numerical simulations show an interesting result: the maximum frequency shift induced by the variation of the LC index is -12.3 GHz for the x-orientation while it is only 2.7 GHz for the y-orientation. We observe that the resonant frequency shift is approximately four times larger when the LC is aligned in the x-direction than in the y-direction. To understand this strong asymmetry in the frequency tuning of the metamaterial, we resort to a simple toy model. The resonant frequency of the metamaterial can be roughly described by [22] $f_0 = 1/(2\pi\sqrt{LC})$, where L and C are the effective inductance and capacitance. The tunability of the eSRR metamaterial comes from the tunable effective capacitance, which is proportional to the effective permittivity $\varepsilon_{\mathrm{eff}}$ [23]. The orientation of the LC affects the effective permittivity $\varepsilon_{\mathrm{eff}}$, and therefore modifies the resonant frequency.

We consider the orientation of the LC director relative to the strongest local field components in the gaps of the eSRRs. When the LC is aligned in the x-direction, the electric field component E_{zo} in the z-direction and E_{yo} in the y-direction will only experience the ordinary LC refractive index, as shown in the schematic in Fig. 3(c). Therefore, it is reasonable to assume that the average permittivity is equal to the ordinary component of the permittivity tensor, so that $\varepsilon_{\mathrm{eff}} = n_o^2$. With increasing temperature, $\varepsilon_{\mathrm{eff}}$ increases due to the increase of the refractive index of the medium along z- and y-directions. Therefore, the effective capacitance increases, leading to the continuous decrease of resonant frequency shown in Fig. 3(c).

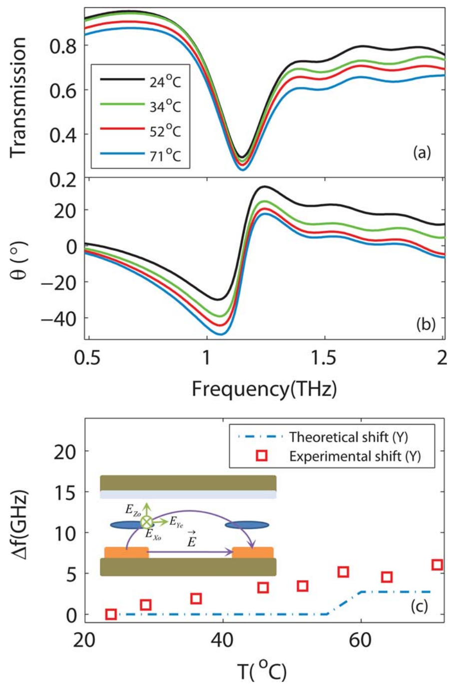

Fig. 4. Experimentally measured results with the LC aligned in the y-direction. (a) Extracted transmission curves, (b) extracted phase curves, and (c) theoretical shift (dashed line) of the resonant frequency and experimental shift (points) estimated from transmission and phase as a function of temperature. The inset shows the dominant field component in the eSRR gap relative to the LC orientation.

With the LC aligned in the y-direction, the electric field component E_{zo} still experiences the ordinary LC refractive index. However, the electric field component E_{ye} experiences the extraordinary LC refractive index shown in schematic of Fig. 4(c). Thus, the average permittivity is $\varepsilon_{\rm eff} = n_o^2 n_e^2/(n_o^2 \cos^2\theta + n_e^2 \sin^2\theta)$, where θ is the angle between the electric field vector and the LC director. In principle the effective permittivity of the eSRR mode can be obtained by integrating this quantity if the field and director distribution are known precisely. However, a simple examination of the schematic of the fields in the inset of Fig. 4(c) suggests that the contributions of n_o and n_e are of similar order. Because n_o increases while n_e decreases with increasing temperature, these changes oppose each other, such that there is almost no change in the effective capacitance and the resonant frequency. Below the phase transition temperature of the LC, the average permittivity can be roughly estimated as $\varepsilon_{\rm eff} = 2.78$. Above the transition temperature it is reduced to $\varepsilon_{\rm eff} = 2.64$. In the numerical results in Fig. 4(c) we see essentially no frequency shift below the phase transition temperature, then a small frequency increase in the isotropic phase.

Experimentally, we measure a maximum frequency shift of -14.8 GHz for the x-orientation, and 6 GHz for the y-orientation. We attribute the small difference between our simulations and experiments to the imperfect alignment of the LC molecules at room temperature. Because of the imperfect alignment of LC, the increase of n_o does not cancel the effects of decreasing n_e exactly. Indeed a visual inspection of the sample sandwiched between two cross polarizers shows that the LC layer is not fully aligned above the metamaterial. This can be explained because the metal structure of the eSRRs imposes a nonuniform alignment of the LC on the bottom quartz substrate [19], which competes with the alignment imposed by the top PVA layer.

In the experiment, we also observe an increase in background absorption which increases with frequency, as well as some variation in the resonance profile. We attribute both of these phenomena to the different absorption of the LC in the ordinary and extraordinary orientations [21]. In the anisotropic phase the extraordinary index dominates the response, and it has a lower imaginary part. In the isotropic phase, we introduce a contribution from the ordinary index, with the corresponding higher absorption. As both absorption coefficients increase with frequency at different rates [21], we see a corresponding frequency dependence in the background absorption. Overall, the experimental results and the numerical simulations show a good agreement.

V. Conclusion

We have shown that an LC THz eSSR-based metamaterial can be tuned by temperature, with tunability depending on the LC orientation. A frequency decrease of 14 GHz was achieved with the liquid crystal aligned in the x-direction, whereas a frequency increase of 6 GHz is observed with the LC aligned in the y-direction. We envisage that a larger tuning range could be achieved by optimizing the structural geometry and/or by engineering of Fano-type resonances based on metamaterials. The achieved thermal tunability in our structure is quite small, and most likely it will not find any applications on its own. At the same time, temperature stability is an important issue for any device operation, even when LC-infiltrated metamaterial is tunable by a different mechanism such as electric field, and our study shows the scale of the variation of material parameters induced by temperature change.

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