Optically Tunable and Thermally Erasable Terahertz Intensity Modulators Using Dye-Doped Liquid Crystal Cells with Metasurfaces

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Abstract: A terahertz metasurface that is imbedded into a dye-doped liquid crystal (DDLC) cell is fabricated in this work. After the metasurface-imbedded DDLC cell is irradiated with a linearly polarized pump beam, the irradiated cell is measured with a terahertz spectrometer. The irradiation of the pump beam causes the adsorption of the dye on one of the substrates of the cell, scattering incident terahertz waves and decreasing the transmittances of the terahertz metasurface at all the frequencies of its resonance spectrum. In addition, these transmittances decrease with an increase in the irradiation times of the pump beam. The adsorbed dye molecules are erased from the substrate after the cell is heated by a hot plate. The cell has similar spectra before the irradiation of the pump beam and after the heating of the hot plate. The aforementioned results reveal that the metasurface-imbedded DDLC cell is an optically tunable and thermally erasable terahertz intensity modulator. Therefore, this cell has the potential in developing intensity attenuators for terahertz imaging, frequency isolators for terahertz telecommunication, and spatial light modulators for terahertz information encryption and decryption.

Keywords: dye-doped liquid crystals; metasurface; spatial light modulators

1. Introduction

Azo dyes have attracted much attention due to their controllability on the orientations of liquid crystals (LCs) [1–4]. Methyl red dye can be used to reorient LCs in dye-doped LC (DDLC) cells [1–4]. The methyl red molecules in an LC cell sequentially undergo absorption, photoisomerization, diffusion, desorption and adsorption on the irradiated surface of the cell after the cell is irradiated with a linearly polarized green light [1–4]. The orientation of the LC director in the cell is determined by the competition between the desorption and the adsorption [1–4]. The LC director is perpendicular (parallel) to the polarized direction of the light as the adsorption (desorption) prevails over the desorption (adsorption) at a low (high) energy dose of the light [1–4]. Methyl red dye molecules exert relatively large torques on LC molecules in DDLC cells [2]. As a result, methyl red dye is an ideal candidate for the fabrication of advanced optical devices.

Metasurfaces have attracted great attention due to their unique response to the frequencies, intensities, polarizations and phases of incident electromagnetic waves. The manipulation of the resonance frequencies of metasurfaces can be achieved by photosensitive dyes [3–7]. Lee et al. reported that the light-induced adsorption of the dye in a DDLC cell reorients the director of the LC, forming a two-dimensional optical grating with a fishnet image [3]. A terahertz fishnet metasurface is fabricated after a photosensitive plastic substrate is exposed to the fishnet image. Therefore, the DDLC cell can be used to develop
passively tunable terahertz filters. However, the resonance frequency of the terahertz fishnet metasurface cannot be actively tuned by light. This drawback hinders the applications of the terahertz fishnet metamaterial. Atorf et al. reported that the light-induced adsorption of the dye in a DDLC cell reorients the LC director; rotating the polarized direction of a light that is incident to a near-infrared metasurface, and shifting its peak frequency from a resonance mode to another resonance mode [4]. However, the near-infrared metasurface only has two resonance frequencies before and after the director reorientation. In other words, the resonance frequency of the near-infrared metasurface cannot be continuously tuned. Therefore, the DDLC cell that is cascaded with the near-infrared metasurface is an optically switchable filter rather than an optically tunable filter. This drawback hinders the applications of this cell. Palto et al. reported that the irradiation of a pump light on a visible-light metasurface that is coated with a dye reorients the long axes of the dye molecules; inducing the birefringence of the dye, and shifting its resonance frequency [8]. However, the visible-light metasurface only has two resonance frequencies before and after the axis reorientation. In other words, the resonance frequency of the visible-light metasurface cannot be continuously tuned. Therefore, the visible-light metasurface that is coated with the dye is an optically switchable filter rather than an optically tunable filter. This drawback hinders the applications of this metasurface. Atorf et al. and Palto et al. manipulated the resonance frequencies of the metasurfaces by the adsorption-induced director reorientation and light-induced birefringence, and developed optically switchable filters that are operated in the short-wavelength region. It is interesting to manipulate the transmittances of metasurfaces at their resonance frequencies by other mechanisms of dyes and develop continuously tunable terahertz intensity modulators.

This work fabricates a terahertz metasurface that is imbedded into a DDLC cell. The irradiation of a pump beam on the cell induces the adsorption of the dye, scattering incident terahertz waves and decreasing the transmittances of the terahertz metasurface at all the frequencies of its resonance spectrum. In addition, these transmittances decrease with an increase in the irradiation times of the pump beam. Therefore, the metasurface-imbedded DDLC cell is a continuously tunable terahertz intensity modulator. This cell exhibits promising applications on terahertz telecommunication, terahertz imaging and terahertz biosensing.

2. Materials and Methods

Figure 1 shows the photoisomerization of a methyl red dye molecule. The dye molecule has trans and cis forms. When the dye molecule is excited by a pump light with a short wavelength of \( \lambda_s \), the molecular conformation is changed from trans form to cis form. The cis form reconverts to the trans form by irradiating a pump light with a long wavelength of \( \lambda_l \) or increasing its temperature \( (T) \). The trans–cis conversion by light is a reversible process, and called photoisomerization.

![Photoisomerization of methyl red dye molecule.](image)

Figure 1. Photoisomerization of methyl red dye molecule.

Figure 2a presents the design of the split-ring resonators (SRRs) of two silver metasurfaces. Each of the SRRs has a linewidth \( (\omega) \), split gap \( (g) \), inner side length \( (l_i) \), outer side length \( (l_o) \), period in the y direction and period in the x direction of 12 \( \mu \)m, 18 \( \mu \)m, 25 \( \mu \)m,
40 µm, 86 µm and 98 µm, respectively. Each of the silver metasurfaces is deposited on a polyethylene terephthalate (PET) substrate with a thickness of 188 µm using photolithography, metal evaporation and lift-off process, and that metasurface has a thickness of 200 nm. A 700 nm-thick polyimide layer (AL-1426CA, Daily Polymer corporation, Kaohsiung City, Taiwan (R.O.C)) is coated on each metasurface as a homogenous alignment layer, and is rubbed parallel to the gaps of the SRRs using a rubbing machine. Figure 2b presents the optical microscope image of one of the silver metasurfaces. The SRRs in the metasurface have the same geometrical dimensions. Therefore, the metasurfaces are reliable in this work.

Figure 2a presents the design of the split-ring resonators (SRRs) of two silver metasurfaces. Figure 2b shows the optical microscope image of one of the silver metasurfaces.

Figure 3a presents the schematic drawing of the two DDLC cells that are imbedded with the silver metasurfaces before the irradiation of a linearly polarized pump beam. Each of the DDLC cells consists of a bare PET substrate and PET substrate with a polyimide-coated metasurface. The two PET substrates are separated by two plastic spacers with a thickness of 100 µm. Two DDLC mixtures are prepared by mixing an azo dye (methyl red, Merck) with a nematic LC (HTW114300-100, Fusol Material, Tainan City, Taiwan (R.O.C)). The HTW114300-100 LC is a nematic mixture, and has a clearing temperature at 106 °C. The HTW114300-100 LC has an extraordinary (ordinary) refractive index of 1.65 (1.52) in the terahertz region [9]. One of the mixtures has a mixing ratio of azo dye: LC = 1:99 by weight, and the other exhibits a mixing ratio of azo dye: LC = 2:98 by weight. The two DDLC mixtures are used in the two cells.

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A linearly polarized pump beam from a semiconductor laser (wavelength = 532 nm) is normally incident to each of the metasurface-imbedded DDLC cells from its bare PET substrate. After the pump beam with an intensity of 1.3 mW/mm² irradiates each cell, that cell is placed in the chamber of a terahertz spectrometer (TPS 3000, TeraView, Cambridge,
UK) for measuring the resonance spectrum of the metasurface. Terahertz waves are polarized in a direction parallel to the x axis of Figure 3a and are normally incident to the DDLC cells from their bare PET substrates.

3. Results and Discussion

Figure 4a displays the experimental terahertz spectra of the two PET substrates with the metasurfaces. The metasurfaces have an identical resonance frequency of 0.703 THz and exhibit an identical transmittance of −9 dB at the resonance frequency. The metasurfaces have the same resonance frequency and transmittance, so they are used to study the effect of the methyl red dye of the DDLC cells on the electromagnetic resonance of the metasurfaces.

![Figure 4](image)

Figure 4. (a) Experimental terahertz spectra of two PET substrates with metasurfaces. (b) Simulated terahertz spectrum of two PET substrates with metasurfaces. The insert presents the near-field distribution of the simulated SRR at 0.703 THz.

A simulation is performed using commercial software based on finite-difference time-domain method to verify the experimental spectra of the two PET substrates with the metasurfaces. A simulated SRR has the same geometrical dimensions as the SRR of Figure 2a and is deposited on a 188 µm-thick PET substrate with an area of 70 µm × 80 µm. The PET substrate has a permittivity of 3.0 in this work, and was obtained from its time-domain spectrum. A periodical boundary condition is set in the simulation, and the conductivity of silver in the simulated SRR is 6.30 \times 10^7 S/m. Figure 4b displays the simulated spectrum of the two PET substrates with the metasurfaces. The metasurfaces have a simulated peak at 0.703 THz. Therefore, the peak frequency of the simulated spectrum of Figure 4b verifies that of the experimental spectra of Figure 4a. The inset in Figure 4b presents the near-field distribution of the simulated SRR at 0.703 THz. The near field of this SRR exhibits the maximum strength at its gap. This result reveals that the electromagnetic resonance of the metasurfaces is an inductive-capacitive mode at 0.703 THz. An SRR can be considered as an inductor-capacitor circuit, and its resonance frequency is given by [10].

\[
f = \frac{1}{2\pi \sqrt{LC}} \approx \frac{1}{n},
\]

where \( L \) is an inductance of the inductor and \( C \) is a capacitance of the capacitor. \( L \) is determined by the enclosed area of the SRR and \( C \) is proportional to the permittivity of a medium that is deposited on the SRR. Equation (1) depicts that the resonance frequency of the inductive-capacitive mode of a metasurface is inversely proportional to the refractive index \( n \) of a dielectric layer that is deposited on the metasurface. Therefore, the resonance frequency of the metasurface will be sensitive to a change in the refractive index of the dielectric layer.

The geometrical dimensions of part of the SRRs differ from those of the designed SRR. Therefore, the discrepancy between the experimental and simulated bandwidths of the metasurface arises from the fabrication tolerance. Efforts are being made at the authors’ laboratory to improve the fabrication of a metasurface, the results of which will be published in the near future.
Figure 5a,b display the experimental spectra of the metasurface-imbedded cells with the 1.0 wt% and 2.0 wt% DDLC mixtures, respectively, at various irradiation times of the pump beam. The resonance transmittance of the metasurface in the 1.0 wt% cell decreases with an increase in the irradiation time, and its resonance frequency remains at the same value after the irradiation of the pump beam. The transmittance modulation in Figure 5a arises from the scattering of the incident terahertz waves. As the methyl red dye molecules in the 1 wt% cell absorb the pump beam, they undergo photoisomerization. The dye molecules sequentially diffuse and adsorb on the inner surface of the bare PET substrate following the photoisomerization. The adsorbed dye molecules on the inner surface scatter the incident terahertz waves, decreasing the transmittances of the metasurface at all the frequencies of its resonance spectrum. In addition, these transmittances decrease with an increase in the irradiation times of the pump beam. Therefore, the transmittances of the metasurface at all the frequencies of its resonance spectrum are tuned by the irradiation times of the pump beam. In other words, the metasurface-imbedded LC cell with the 1 wt% DDLC mixture is an optically tunable terahertz intensity modulator. This cell exhibits promising applications on terahertz telecommunication, terahertz imaging and biosensing.

![Experimental spectra of metasurface-imbedded cells with (a) 1.0 wt% and (b) 2.0 wt% DDLC mixtures at various irradiation times of pump beam. (c) Time-dependent resonance transmittance curves of metasurfaces of 1.0 wt% and 2.0 wt% DDLC cells.](image)

The role that the metasurface plays a band-stop filter. The band-stop filter decreases the transmittance of the metasurface at its resonance frequency. Therefore, it is necessary to use the metasurface to form a band-stop spectrum. The transmittance of the metasurface at its resonance frequency can be tuned by its geometrical structure and conductivity [11], but they are constant in this work. This work uses the adsorption-induced wave scattering to tune not only the transmittance of the metasurface at its resonance frequency but also the transmittances of the metasurface at the other frequencies \( f \). The metasurface has no electromagnetic resonance at \( f \) while the adsorption of the dye molecules tunes the transmittances at \( f \). This result reveals that the adsorption-induced wave scattering will reduce the transmittance of non-resonating metallic structures.

The metasurface in the DDLC cell with the 1.0 wt% DDLC mixture has a resonance transmittance of \(-27.26\) dB before the irradiation of the pump beam. The pump laser is tuned off after the pump beam irradiates this cell for 150 min. The cell is put in a dry cabinet for preservation. After 60 days, the cell is measured with the terahertz spectrometer. The metasurface has a resonance transmittance of \(-27.26\) dB after the 60 days. This result reveals that the \(\text{cis} \rightarrow \text{trans}\) photoisomerization is tiny at room temperature and so does not interfere with the irradiation-induced effect.

Figure 5c presents the time-dependent resonance transmittance curves of the metasurfaces of the 1.0 wt% and 2.0 wt% DDLC cells. Figure 5c depicts that the sensitivity of the resonance transmittance of the metasurface to the irradiation time of the pump beam is increased as the concentration of the DDLC mixture is increased from 1.0 wt% to 2.0 wt%. This result arises from the fact that the amount of the adsorbed dye molecules at a given irradiation time is larger than in the 2 wt% DDLC cell than in the 1.0 wt% DDLC cell. The key performance parameter for a controllable intensity modulator is to have a
large modulation depth \( D \). \( D \) is given by an equation of 
\[
D = 100\% \times \left( T_{\text{max}} - T_{\text{min}} \right)/T_{\text{max}},
\]
where \( T_{\text{max}} \) and \( T_{\text{min}} \) are the maximum and minimum transmittances of a metasurface at a specific frequency, respectively [12]. The metasurface in the 1.0 wt% DDLC cell has a maximum linear (decibel) transmittance of 0.0060 (−22.24 dB) at its resonance frequency, and exhibits a minimum linear (decibel) transmittance of 0.0019 (−27.26 dB) at the frequency. Therefore, the metasurface in the 1.0 wt% DDLC cell has a modulation depth of 68%. The metasurface in the 2.0 wt% DDLC cell has a maximum linear (decibel) transmittance of 0.0056 (−22.51 dB) at its resonance frequency, and exhibits a minimum linear (decibel) transmittance of 0.0017 (−27.66 dB) at the frequency. As a result, the metasurface in the 2.0 wt% DDLC cell has a modulation depth of 70%. The modulation depths of the metasurfaces increase with an increase in the concentrations of the DDLC mixtures. As the concentration of the DDLC mixture is increased from 2% to 3%, part of the dye powder cannot dissolve in the LC (data not shown herein). A situation in which other LCs are used in this work may increase the solubility of the dye powder in the LCs. Efforts are being made at the authors’ laboratory to develop metasurface-imbedded DDLC cells with large modulation depths, the results of which will be published in the near future.

After the irradiation of the pump beam, a metasurface-imbedded cell with a 1.0 wt% DDLC mixture is placed on a hot plate at 80 °C for various periods. The HTW114300-100 LC has a clearing temperature at 106 °C. Therefore, the DDLC mixture has a nematic phase at 80 °C. Figure 6a presents the spectra of this cell before the irradiation of the pump beam, after irradiation of the pump beam and after the heating of the hot plate for 1 h, 3 h and 5 h. This result reveals that the cell is reusable. In other words, the metasurface-imbedded cell with the 1.0 wt% DDLC mixture is an optically tunable and thermally erasable terahertz intensity modulator.

![Figure 6](image-url)

**Figure 6.** (a) Spectra of metasurface-imbedded cell with 1.0 wt% DDLC mixture before irradiation of pump beam, after irradiation of pump beam, and after heating of hot plate for 1 h, 3 h and 5 h. Photos of metasurface-imbedded cell with 1.0 wt% DDLC mixture (b) before irradiation of pump beam, (c) after irradiation of pump beam, and (d) after heating of hot plate for 5 h.

Figure 6b–d present the photos of the metasurface-imbedded cell with the 1.0 wt% DDLC mixture before the irradiation of the pump beam, after the irradiation of the pump beam, and after the heating of the hot plate for 5 h. Figure 6b,c verify that the dye molecules are adsorbed on the substrate without a polyimide layer. Figure 6d depicts that the adsorbed dye molecules are erased from this substrate after the heating of the hot plate for 5 h, and return to the DDLC mixture. This result verifies that the metasurface-imbedded
cell with the 1.0 wt% DDLC mixture is an optically tunable and thermally erasable terahertz intensity modulator.

As the metasurface has the same resonance transmittance before the irradiation and after the heating, the time (temperature) for removing the adsorbed methyl red dye molecules from the irradiated surface is defined as the erasing time (temperature) of the DDLC cell. Figure 6 presents the spectra of the metasurface-imbedded cell with the 1.0 wt% DDLC mixture before the irradiation of the pump beam, after the irradiation of the pump beam, and after the heating of the hot plate for 1 h, 3 h and 5 h. The cell has an initial resonance transmittance before the irradiation of the pump beam. As the erasing time is smaller than 5 h, the resonance transmittance of the metasurface does not return to its initial value. This result reveals that an incomplete recovery of the initial state of the DDLC cell is obtained at a short erasing time.

The erasing time and temperature of the DDLC cell are 5 h and 80 °C, respectively. Efforts are being made at the authors’ laboratory to develop metasurface-imbedded DDLC cells with short erasing times and low erasing temperatures, the results of which will be published in the near future.

The manipulation of the intensities of incident electromagnetic waves can be achieved by applying voltage [14], light [15] and heat [16] to terahertz metasurfaces. The electrical, optical and thermal energies are continuously supplied to the terahertz metasurfaces because the intensities of the electromagnetic waves will return to their initial values after the removal of the voltage, light and heat. This drawback hinders the applications of the terahertz metasurfaces. This work uses the metasurface-imbedded DDLC cells to manipulate the intensities of the terahertz waves. The transmittances of the metasurfaces remain at the same values after the removal of the pump beam because the adsorption of the dye is permanent in the cells [1–4]. This result reveals that the terahertz metasurfaces exhibit low power consumption during long-term use.

Following the heating of the hot plate, the metasurface-imbedded cell with the 1.0 wt% DDLC mixture is irradiated with the pump beam for 150 min. The director reorientation is verified by inspecting this cell via a polarizing optical microscope. The transmission axis of the polarizer was set parallel to the rubbing direction (the x axis of Figure 2a) of the poly-imide layer, and a white light was incident from the rubbed surface. Figure 7a,b present the intensity images of the metasurface-imbedded cell with the 1.0 wt% DDLC mixture under parallel polarizers and crossed polarizers, respectively. Figure 7a reveals that the irradiated (non-irradiated) region appears dark (bright) as the transmission axis of the analyzer is parallel to that of the polarizer. Figure 7b reveals that the irradiated (non-irradiated) region appears bright (dark) as the transmission axis of the analyzer is perpendicular to that of the polarizer. In addition, the transmitted intensity of the white light is increased from a minimum value to a maximum value as the transmission axis of the analyzer is gradually rotated from parallel polarizers to crossed polarizers. The results in Figure 7a,b display that the adsorption of the methyl red dye molecules in the 1.0 wt% DDLC cell reorients the LC director from a homogeneous orientation to a twist nematic orientation after the irradiation of the pump beam, and the adsorbed dye molecules twist the LC director of the 1.0 wt% DDLC cell by an angle of 90°.
is the twist angle of the LC director if the Mauguin condition is satisfied. The LC crystals 

Figure 7. Intensity images of metasurface-imbedded cell with 1.0 wt% DDLC mixture under (a) parallel polarizers and (b) crossed polarizers.

Figure 5a presents that the metasurface has a constant resonance frequency of 0.557 THz before and after the irradiation of the pump beam. Figure 7a,b reveal that the adsorption of the dye causes the director reorientation in the 1.0 wt% DDLC cell. Therefore, the director reorientation in this cell does not change the resonance frequency of the metasurface. The constant resonance frequency of the metasurface is due to the Mauguin condition of the cell. Consider that a linearly polarized light is normally incident to a twist nematic LC cell, and the polarized direction of the light is parallel to or perpendicular to the surface director of one of the substrates of the cell. The Mauguin condition of the twist nematic LC cell is given by [17]

$$2\pi \Delta nd / \lambda \gg \phi,$$

where $\Delta n$ is the birefringence of the LC; $d$ is the thickness of the LC layer in the cell; $\lambda$ is the wavelength of the linearly polarized light, and $\phi$ is the twist angle of the LC director in the cell. Equation (2) reveals that the polarized direction of the light that passes through the twist nematic LC cell is rotated by $\phi$ if the Mauguin condition is satisfied. The LC has a birefringence of 0.13 in the metasurface-imbedded cell with the 1.0 wt% DDLC mixture [9], and the LC layer has a thickness of 100 $\mu$m in the cell. The metasurface has a wavelength of 539 $\mu$m at its resonance frequency, and the twist angle of the LC director is $\pi/2$. Substituting $\Delta n = 0.13$, $d = 100 \mu$m, $\lambda = 539 \mu$m and $\phi = \pi/2$ into Equation (2) reveals that the Mauguin condition is unsatisfied in this work. This result reveals that the terahertz waves that pass through the LC layer of the metasurface-imbedded cell have the same polarized direction before and after the director reorientation. In addition, the LC molecules near the metasurface have the same orientation before and after the irradiation of the pump beam because the polyimide layer has a strong surface anchoring energy. Therefore, the near field of the metasurface “experiences” the constant refractive index of the LC before and after the director reorientation. Equation (1) reveals that a metasurface has the same resonance frequency at a constant $n$. As a result, the metasurface in the 1.0 wt% DDLC cell has the same resonance frequency before and after the director reorientation.

A DDLC cell with the same geometrical structure as that in Figure 3a but without a metasurface is fabricated to verify that the adsorption of the dye in the metasurface-imbedded cell with the 1.0 wt% DDLC mixture scatter the incident terahertz wave. The cell without a metasurface is filled with a 1.0 wt% DDLC mixture. The linearly polarized pump beam from the semiconductor laser is normally incident to the DDLC cell from its substrate without a polyimide layer. After the pump beam with an intensity of 1.3 mW/mm² irradiates the cell, the irradiated cell is placed in the chamber of the terahertz spectrometer for measuring its spectrum. Figure 8 displays the terahertz spectra of the DDLC cell without a metasurface at various irradiation times of the pump beam. The results in Figure 8 reveal that the adsorbed dye molecules on the substrate without a polyimide layer scatter the incident terahertz waves, decreasing the transmittances of the cell at all the frequencies of its spectrum. Therefore, the adsorbed dye molecules in the metasurface-imbedded cell with the 1.0 wt% DDLC mixture scatter the incident terahertz waves.
Silver nanoslits with a period of 470 nm are imbedded in a DDLC cell. Each of the silver nanoslits has a width and height of 200 nm and 70 nm, respectively. Figure 9 presents the terahertz spectrum of the nanoslit-imbedded DDLC cell. The silver nanoslits have no resonance in the terahertz region. The adsorption of the dye decreases the transmittances of the nanoslit-imbedded DDLC cell at all the frequencies of its spectrum after the cell is exposed to the pump beam for 120 min. Therefore, the adsorption-induced wave scattering reduces the transmittance of the non-resonating silver nanoslits.

4. Conclusions

This work made the metasurface-imbedded cell with the 1.0 wt% DDLC mixture. The transmittances of the metasurface at all the frequencies of its resonance spectrum are reduced by the dye molecules that are adsorbed on the substrate without a polyimide layer, and these transmittances decrease with the increase in the irradiation times of the pump beam. Therefore, the metasurface-imbedded cell is a continuously tunable terahertz intensity modulator. The transmittances of the metasurface at all the frequencies of its resonance spectrum remain at the same values after the removal of the pump beam because the adsorption of the dye is permanent on the substrate. Therefore, the terahertz metasurface that is imbedded into the DDLC cell exhibits low power consumption during long-term use. The adsorbed dye molecules are erased from the substrate after the cell is...
heated by the hot plate. The cell has similar spectra before the irradiation of the pump beam and after the heating of the hot plate, so is an optically tunable and thermally erasable terahertz intensity modulator. The metasurface-imbedded DDLC cell can be used to develop filters, absorbers, sensors and spectral imagers.

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**References**

1. Ouskova, E.; Fedorenko, D.; Reznikov, Y.; Shiyano, S.V.; Su, L.; West, J.L.; Kuksenok, O.V.; Francescangeli, O.; Simoni, F. Hidden photoalignment of liquid crystals in the isotropic phase. *Phys. Rev. E-Stat. Phys. Plasmas Fluids Relat. Interdiscip. Top.* 2001, 63, 021701. [CrossRef] [PubMed]

2. Simoni, F.; Francescangeli, O. Effects of light on molecular orientation of liquid crystals. *J. Phys. Condens. Matter* 1999, 11, R439–R487. [CrossRef]

3. Lee, C.R.; Lin, S.H.; Wang, S.M.; Lin, J.-D.; Chen, Y.S.; Hsu, M.C.; Liu, J.K.; Mo, T.S.; Huang, C.Y. Optically controllable photonic crystals and passively tunable terahertz metamaterials using dye-doped liquid crystal cells. *J. Mater. Chem. C* 2018, 6, 4959–4966. [CrossRef]

4. Atorf, B.; Mühlenbernd, H.; Zentgraf, T.; Kitzerow, H. All-optical switching of a dye-doped liquid crystal plasmonic metasurface. *Opt. Express* 2020, 28, 8898–8908. [CrossRef] [PubMed]

5. Silalahi, H.M.; Chen, Y.-P.; Shih, Y.-H; Chen, Y.-S.; Lin, X.-Y.; Liu, J.-H.; Huang, C.-Y. Floating terahertz metamaterials with extremely large refractive index sensitivities. *Photons Res.* 2021, 9, 1970–1978. [CrossRef]

6. Bi, K.; Yang, D.; Chen, J.; Wang, Q.; Wu, H.; Lan, C.; Yang, Y. Experimental demonstration of ultra-large-scale terahertz all-dielectric metamaterials. *Photons Res.* 2019, 7, 457–463. [CrossRef]

7. Ashalley, E.; Acheampong, K.; Besteiro, L.V.; Yu, P.; Neogi, A.; Govorov, A.O.; Wang, Z.M. Multitask deep-learning-based design of chiral plasmonic metamaterials. *Photons Res.* 2020, 8, 1213–1225. [CrossRef]

8. Palto, S.P.; Draginda, Y.A.; Artemov, V.V.; Gorkunov, M.V. Optical control of plasmonic grating transmission by photoinduced anisotropy. *J. Opt.* 2017, 19, 074001. [CrossRef]

9. Shih, Y.H.; Lin, X.Y.; Silalahi, H.M.; Lee, C.R.; Huang, C.Y. Optically tunable terahertz metasurfaces using liquid crystal cells coated with photoalignment layers. *Crystals* 2021, 11, 1100. [CrossRef]

10. O’Hara, J.P.; Singh, R.; Brener, I.; Smirnova, E.; Han, J.; Taylor, A.J.; Zhang, W. Thin-film sensing with planar terahertz metamaterials: Sensitivity and limitations. *Opt. Express* 2008, 16, 1786–1795. [CrossRef] [PubMed]

11. Chiang, W.F.; Silalahi, H.M.; Chiang, Y.C.; Hsu, M.C.; Zhang, Y.S.; Liu, J.-H.; Yu, Y.; Lee, C.R.; Huang, C.Y. Continuously tunable intensity modulators with large switching contrasts using liquid crystal elastomer films that are deposited with terahertz metamaterials. *Optik* 2020, 202, 27676–27687. [CrossRef] [PubMed]

12. Zhou, Z.; Wang, S.; Yu, Y.; Chen, Y.; Feng, L. High performance metamaterials-high electron mobility transistors integrated terahertz modulator. *Opt. Express* 2017, 25, 17832–17840. [CrossRef] [PubMed]

13. Lee, C.R.; Mo, T.S.; Cheng, K.T.; Fu, T.L.; Fuh, A.Y.G. Electrically switchable and thermally erasable biphotonic holographic gratings in dye-doped liquid crystal films. *Appl. Phys. Lett.* 2003, 83, 4285–4287. [CrossRef]

14. Hu, F.; Rong, Q.; Zhou, Y.; Li, T.; Zhang, W.; Yin, S.; Chen, Y.; Han, J.; Jiang, G.; Zhu, P.; et al. Terahertz intensity modulator based on low current controlled vanadium dioxide composite metamaterial. *Opt. Commun.* 2019, 440, 184–189. [CrossRef]

15. Heyes, J.E.; Withayachumnankul, W.; Grady, N.K.; Chowdhury, D.R.; Azad, A.K.; Chen, H.T. Hybrid metasurface for ultra-broadband terahertz modulation. *Appl. Phys. Lett.* 2014, 105, 181108. [CrossRef]

16. Sanphuang, V.; Ghaličhechian, N.; Nahar, N.K.; Volakis, J.L. Reconfigurable THz Filters Using Phase-Change Material and Integrated Heater. *IEEE Trans. Terahertz Sci. Technol.* 2016, 6, 583–591. [CrossRef]

17. Yeh, P.; Gu, C. *Optics of Liquid Crystal Displays*; Wiley: Hoboken, NJ, USA, 2009.