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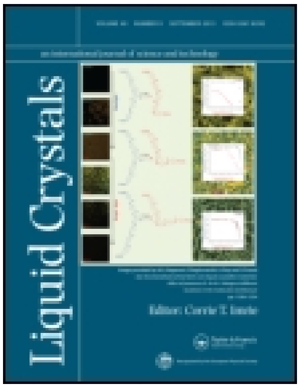

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# Ultrafast switchable wide angle negative refraction in novel dual-frequency liquid crystal mixture

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# Ultrafast switchable wide angle negative refraction in novel dual-frequency liquid crystal mixture

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Ultrafast switchable wide angle negative refraction in a novel dual-frequency liquid crystal mixture is demonstrated experimentally. Laterally fluorinated isothiocyanato phenyl-tolane liquid crystal compound is doped into Merck MLC-2048 to acquire the high birefringence dual-frequency liquid crystal. The maximum negative refraction angle achieved is about 12° and the switching time is only 25 ms for a 40-µm cell which is two orders faster than switchable negative refraction achieved before in nematic liquid crystals. Such properties provide a feasible approach to design negative refraction devices based on liquid crystals.

**Keywords:** negative refraction; fast switchable; high birefringence; dual-frequency liquid crystal

## 1. Introduction

Negative refraction (NR) is proposed conceptually by Veselago in 1968 [1]. Due to the unique applications such as super lens [2–4], optical cloaking [5,6] and reversed effects [1,7], negative refraction phenomena have been widely investigated in photonic crystals [8–11] and left-handed materials [12–20]. However, because of the inevitable energy losses and extremely complex fabrication process, realising NR in the visible wavelength is still a very tough mission. As a result, many researchers devote to achieving negative refraction in natural materials [21–26]. Uniaxial crystals are studied widely for negative refraction, but it cannot be turned because the optic axis is fixed. Nematic liquid crystal (NLC) also exhibits large optical anisotropy due to the molecules alignment and could be used for a variety of tunable optical devices. Pishnyak et al. [23] and Zhao et al. [24,25] have already observed negative refraction in NLC. According to the experiments reported in references [23–25], the maximum negative refraction angle (MNRA) is –7.7° and the switching speed is up to tens of seconds [23]. Recently extremely large MNRA (–14°) is realised by the high birefringence low viscosity nematic liquid crystal mixtures [27], but the switching speed is still slow. Larger negative refraction angle and faster switching speed are urgently required for practical use in negative refraction area.

In this work, negative refraction in novel dual-frequency liquid crystal (DFLC) mixture is investigated. Wide negative refraction angle and ultrafast switching speed are achieved in novel high birefringence ($\Delta n$) DFLC mixture which is synthesised successfully by our group. It is the first time that the tunable NR with so big MNRA and so fast switching speed is achieved in a dual-frequency liquid crystal experimentally.

## 2. Numerical method

The negative refraction can be realised in NLC due to the optical anisotropy effect. The theory has been illustrated in detail in references [23,24]; here, we directly show the formulas for refractive angle and critical angle. As Figure 1 shows, the LC cell lies in the X-Z plane and $Z = 0$ is the interface which separates the air and LC. The LC molecular (director **n**) is parallel to the cell substrates and the angle between alignment direction and Z axis is $\alpha$ which can be optimised through calculations. The incident beam is polarised in the figure plane and impinges the interface from air. The incident and refractive angles are $\theta_{\mathrm{i}}$ and $\theta_{\mathrm{r}}$, respectively. The refractive angle can be expressed as follows:

$$\theta_{\mathrm{r}} = \tan^{-1} \frac{2n_{\mathrm{i}} \sin\theta_{\mathrm{i}} + \sin 2\alpha (n_e^{-2} - n_o^{-2}) n_o n_e \sqrt{(n_o^{-2}\cos^2\alpha + n_e^{-2}\sin^2\alpha) n_o^2 n_e^2 - n_{\mathrm{i}}^2 \sin^2\theta_{\mathrm{i}}}}{2(n_o^{-2}\cos^2\alpha + n_e^{-2}\sin^2\alpha) n_0 n_e \sqrt{(n_o^{-2}\cos^2\alpha + n_e^{-2}\sin^2\alpha) n_o^2 n_e^2 - n_{\mathrm{i}}^2 \sin^2\theta_{\mathrm{i}}}}. \qquad (1)$$

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Chengliang Yang and Dan Jia contribute equally to this work.

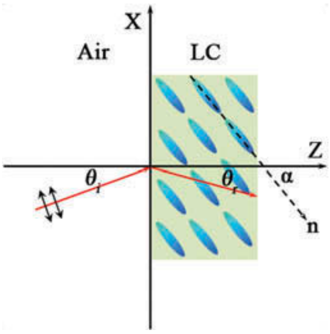

Figure 1. (colour online) The coordinate system and negative refraction in NLC cell.

From the formula, we can see that the refractive angle $\theta_r$ increases when the incident angle decreases for a fixed $\alpha$. When incident angle equals to zero and $\alpha = \tan^{-1}(n_e/n_o)$, the MNRA can be achieved as

$$\theta_{\mathrm{r\,max}} = -\tan^{-1}[(n_e^2 - n_o^2)/2n_o n_e]. \quad (2)$$

To the contrary, the corresponding incident angle is defined as the critical incident angle (CIA) when the refractive angle equals to zero:

$$\theta_{\mathrm{CIA}} = \arcsin \frac{\sin 2\alpha (n_e^2 - n_o^2)\sqrt{n_o^{-2}\cos^2\alpha + n_e^{-2}\sin^2\alpha}}{\sqrt{4 + \sin^2 2\alpha (n_e^{-2} - n_o^{-2})^2 n_o^2 n_e^2}}. \quad (3)$$

However, the NLC is not full angle negative refraction material, so MNRA and CIA are the most important parameters for NR in NLC. Increasing the optical anisotropic properties of NLC is a feasible way to realise larger MNRA and CIA.

## 3. Materials and experimental techniques

### *3.1 The LC materials*

In order to improve the MNRA and response time, a high birefringence low viscosity nematic liquid crystal mixture was used. In reference [27], –14° MNRA was achieved by the NLC mixture with a birefringence of 0.42. However, only the turn-on process can be accelerated by increasing the driving voltage; the turn-off process is still governed by the viscosity of material and surface anchoring energy. As a result, DFLC is used in this work. When a low frequency voltage is applied, the LC directors are reoriented to the field direction; to the contrary, a high frequency voltage will restore the LC back to the initial alignment. So both the turn-on and turn-off processes can be accelerated by applying the frequency-varied high voltage driving pulses [28–30].

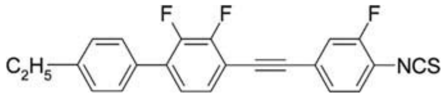

Figure 2. The molecular structure of FIPTLC.

In this work, MLC-2048 DFLC (Merck KGaA, Darmstadt, Germany) is employed, whose $\Delta n$ is about 0.21 ($\lambda = 633$ nm). With this DFLC, the response time (include both NR to positive refraction (PR) and PR to NR) is greatly improved at least two orders faster than NR in normal NLC reported before. But the MNRA and CIA are only –7.5° and 11.9°, respectively, according to formulas (2) and (3). As discussed in Section 2, when the rubbing direction is optimised ($\alpha = \tan^{-1}(n_e/n_o)$), the MNRA and CIA are governed by the optical birefringence $\Delta n$. In order to improve the MNRA and CIA properties, the high birefringence laterally fluorinated isothiocyanato phenyl-tolane liquid crystal (FIPTLC) compound which is studied first in [31,32] is synthesised by our group. The molecular structure is shown in Figure 2. There are two fluorine substitutions in the second and third positions of the middle phenyl ring and only one fluorine substitution at the last phenyl ring next to the isothiocyanato (NCS) group. Lateral fluorination is introduced on the phenyl-tolane in order to lower the melting point of the compound. The short alkyl chain is beneficial to the birefringence because of the higher molecular packing density which also affects the clearing temperature and stability of nematic phase. The birefringence of this FIPTLC compound is about 0.52 ($\lambda = 633$ nm) [31], which is very high among so many compounds have been studied; furthermore, there is no smectic phase for this compound [31]. The compound is dissolved into the MLC-2048 (about 35 wt% in this work). The measured birefringence for 532 nm is shown in Figure 3 as a function of temperature. The birefringence decreases gradually with increasing temperature. The mixture exhibits

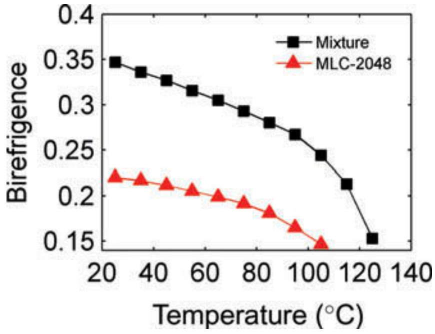

Figure 3. (colour online) Temperature-dependent birefringence of mixture (square) and pure MLC-2048 (triangle) for 532 nm. The dots are experimental data and the lines are connecting lines.

significantly higher birefringence than pure MLC-2048. The melting point temperature is less than –50°C and the clearing point is about 126°C for the mixture. The crossover frequency is 15 kHz which is measured in 20°C room temperature. With this novel DFLC mixture, the ultrafast switchable wide angle negative refraction can be achieved.

### 3.2 *Experimental set-ups*

In experiment, a linearly polarised 532 nm solid laser (CNI-MGL-532 nm 100 mw) is used as the light source. The set-up is shown in Figure 4(a). The half waveplate (HWP) and polariser (P) are used to control the light intensity and polarisation. After the beam passes the polariser, the polarisation direction is in the X-Z plane which means P polarisation for LC cell. A long focal distance lens ($f = 200$ mm) is placed behind the polariser in order to focus the beam on the interface ($Z = 0$) of LC cell. The focus point on the interface is about 20 μm which is smaller than the thickness of the cell (40 μm). The purpose of such configurations is to make sure that the beam can be coupled into the cell efficiently and can propagate a long distance before diverging into the substrates. The cell is made up of two parallel glass substrates whose inner surfaces are coated with the transparent conductive indium-tin-oxide (ITO) electrodes. On the ITO film, the polyimide (PI) alignment film is coated and the optimised rubbing directions are $\alpha = 49°$ and 51° for MLC-2048 and DFLC mixture, respectively, which can be calculated in Section 2.The cell is mounted on the goniometer sample stage to control the incident angle accurately. A signal generator (AFG-3022, Tektronix Incorporation, Beaverton, OR, USA) and voltage amplifier (HA-400, Pintek Electronic Limited, Taipei, Taiwan) are connected to the ITO electrodes to apply the driving fields. The beam propagation route in the cell can be observed by human eye directly and recorded by the CCD camera. The set-up for response time measurement is shown in Figure 4(b). The cell is placed perpendicular to the beam. The polariser (P1) is placed before the cell and an analyser (P2) is crossed behind the cell. The intensity of transmitted beam is measured by the New Focus detector (Model-2031) and displayed on the oscilloscope (Tektronix MSO 3032).

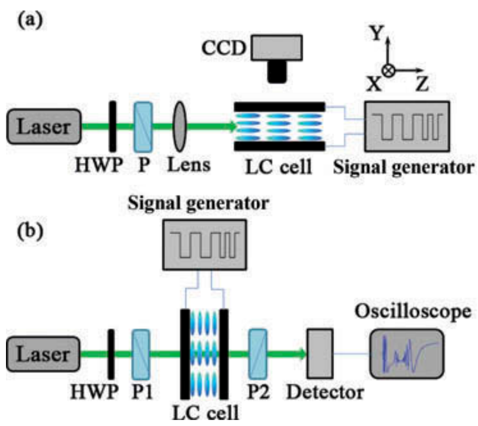

Figure 4. (colour online) The experimental set-ups. (a) Switchable negative refraction in NLC cell. (b) Response time measurement.

## 4. Results and discussion

Liquid crystals are widely used for tunable optical devices because of its large optical anisotropy due to the alignment of its rod-like molecules under electric and magnetic fields, which makes it a perfect choice to realise switchable NR. When the 50 KHz square-driving field is applied on the ITO electrodes, the director is parallel to the cell substrates. The incident beam is *e* light for the LC and the NR occurs in the interface of the air and LC as Figure 5(a) shows (solid white line is the interface, dashed white line is the normal and red lines indicate the incident and refractive directions). In the figure, we can also see the reflected beam which can be used to confirm the position of interface. When the driving frequency is changed to 1 KHz, the LC directors are reoriented to the field direction, which means the incident beam is *o* light for LC now. The negative refraction is switched to PR as Figure 5(b) shows. The switch between NR and PR is achieved by controlling the frequency of driving field. The MNRA for MLC-2048 is about –7.5° which is quite small for practical use. As discussed in material section, FIPTLC compound is synthesised by our group in order to improve the MNRA property and expand the NR range. The birefringence of the DFLC mixture is about 0.35 for 532 nm that

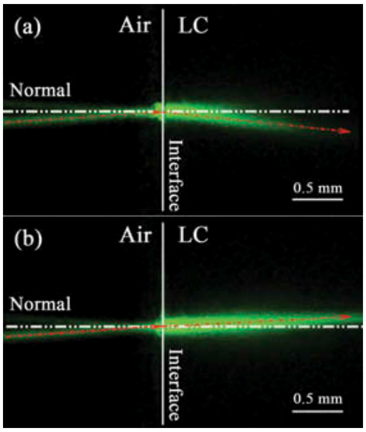

Figure 5. (colour online) The photograph of switchable negative refraction in DFLC. (a) The negative refraction for 50 KHz driving field. (b) The positive refraction for 1 KHz driving field.

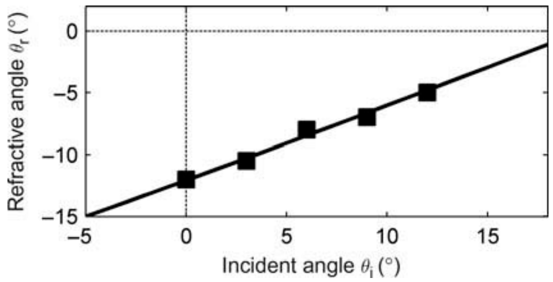

Figure 6. The relation between refractive angles and incident angles in DFLC mixture.

is measured at 20°C temperature. In the experiment, a group of the refractive angles for different incident angles are recorded. The relation between incident and refractive angles are shown in Figure 6. The discrete dots are experimental data and the corresponding theory prediction calculated by Equation (1) is indicated by the line. In the calculation, the measured ordinary and extraordinary refractive indices are used ($n_o = 1.54$ and $n_e = 1.89$ for 532 nm; 20°C) and the rubbing direction $\alpha = 51°$. From Figure 6, we can see that the experimental data agree very well with the theoretical prediction. There is a range of incident angles $0 < \theta_\mathrm{i} < \theta_\mathrm{CIA}$ for which refraction is negative. In this range, the refraction angle is monotonously decreased with the increase in incident angle. When $\theta_\mathrm{i} = 0$, the MNRA of DFLC mixture is achieved as –12.1°, which is smaller than the value in [27], but is still much larger than NR achieved before in DFLC to our knowledge. The range for NR ($\theta_\mathrm{CIA}$) is also improved greatly to 19.7°, which expands the possible application region of NR in optical devices.

The response time is also very important which can be measured by the set-ups as Figure 4(b) shows. Due to the property of DFLC, the switch between NR and PR can be controlled by changing the frequency of driving field. For a DFLC cell, the turn-on (rise) and turn-off (decay) speed are governed by the balance of elastic torque, viscous torque and electric torque. Under some assumptions, the director's response time can be expressed as [28,29].

$$\tau_\mathrm{rise} = \frac{\tau_0}{(V_l/V_{\mathrm{th},l})^2 - 1}, \tag{4}$$

$$\tau_\mathrm{decay} = \frac{\tau_0}{(V_h/V_{\mathrm{th},h})^2 - 1}. \tag{5}$$

In Equations (4) and (5), $\tau_0 = \gamma_1 d^2/\mathrm{K}_{11}\pi^2$ is the directors' free relaxation time, $\gamma_1$ is the rotational viscosity, $\mathrm{K}_{11}$ is the elastic constant, $V_l$ and $V_h$ are the driving voltages for rise and decay process, and $\mathrm{V}_{\mathrm{th,l}}$ and $\mathrm{V}_{\mathrm{th,h}}$ are the threshold voltage for low and high frequencies. From these equations, we can see that both rise and decay processes can be accelerated by the driving voltage. The higher the voltage, the shorter is the response time. In this work, the response time of the rise, decay and free relaxation all are measured, which are shown in Figure 7. From Figure 7(a), the rise time of this novel DFLC mixture cell is about 4 ms; that is to say, it will only take 4 ms to complete the switch from NR to PR for this DFLC cell. Generally the rise process is much faster than decay process for normal LCs, whether the fast switch can be achieved or not mainly depends on the decay process. As Figure 7(b) shows, the decay time for 50 KHz driving field is about 25 ms which is longer than the rise time but is still two orders faster than the free relaxation process (shown in Figure 7(c)). The driving voltages for rise and decay process are the same, so the slow response for decay is mainly due to the higher threshold voltage for high frequency. To our knowledge, it is the first time that the NR is achieved in dual-frequency liquid crystal mixtures with so big MNRA and so fast switching speed experimentally. From Equations (4) and (5), the response time can be further reduced through the use of a thinner cell gap. A lower threshold voltage and smaller viscosity coefficient which can be accomplished by additional molecular structural modifications are also helpful to achieve even faster switching speed. Furthermore, the overdrive method [33–35] also can be used to shorten the response time during both rise and decay process. Under these optimisations, the response time can be improved to submillisecond.

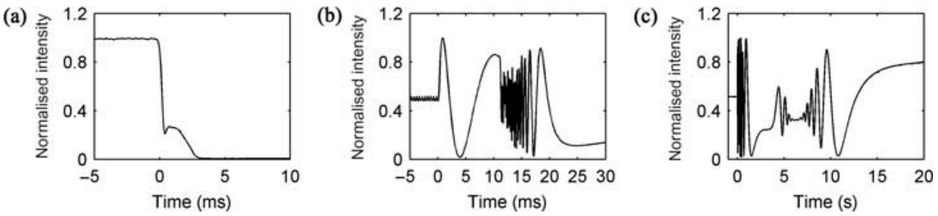

Figure 7. The output signal of detector illustrating response times for (a) 50 KHz to 1 KHz driving field, (b) 1 KHz to 50 KHz driving field and (c) 1 KHz to no driving field.

## 5. Conclusions

In conclusion, we have demonstrated ultrafast switchable wide angle negative refraction in novel high birefringence DFLC mixture. Through doping the high birefringence laterally fluorinated isothiocyanato phenyl-tolane liquid crystal compound into the MLC-2048, the MNRA is improved to 12.1° and the critical angle reaches 19.7° which are both the biggest for NR in DFLC. Both the rise and decay processes are accelerated by the driving fields and 25-ms switching speed between NR and PR has been realised in the 40-μm cell. The large NR angle and fast switch properties may be used in the design of NR optical devices.

## Acknowledgement

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