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Sub-millisecond response phase modulator using a low crossover frequency dual-frequency liquid crystal

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Sub-millisecond response phase modulator using a low crossover frequency dual-frequency liquid crystal

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A high birefringence (Δn=0.292 at λ=633 nm, 25°C) and low crossover frequency (<1 kHz at T=25°C) dual-frequency liquid crystal (DFLC) mixture was developed. The high birefringence enabled us to use a thin liquid crystal cell, which is helpful for fast response time and low operating voltage. The initially low crossover frequency allowed us to operate the DFLC device at an elevated temperature, which significantly lowers the viscosity while keeping the crossover frequency in an acceptable range (<10 kHz). We demonstrated a 2π phase shifter at λ=633 nm using such a DFLC and obtained a sub-millisecond response time at T~45°C. This type of DFLC mixture together with elevated temperature operation opens a new way for achieving fast response time.

Keywords: dual-frequency liquid crystal; fast response; phase modulator; dielectric relaxation

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1. Introduction

The demand for fast response time on nematic liquid crystal (LC) electro-optic modulators is ever increasing. From a material perspective, high birefringence and a small visco-elastic coefficient are favourable for achieving a fast response time (*1*). Numerous compounds and mixtures have been developed toward these objectives (*2, 3*). Various LC operating modes, such as thin cell approach (*4–6*), π-cell (*7*) and dual-fringing field switching (*8*), have been developed. Overdrive and undershoot voltage methods (*9, 10*) have been commonly used in liquid crystal display (LCD) TVs for reducing motion picture image blurs. However, in the abovementioned approaches the LC relaxation process cannot be accelerated by the electric field; it is still governed by the restoring elastic force. As a result, the response time is still in the millisecond range. To obtain sub-millisecond response time, dual-frequency liquid crystal (DFLC) has proven useful (*11, 12*). Fast response DFLC devices have been developed for various applications, such as display (*13, 14*), variable optical attenuator (*15, 16*), spatial light modulator (*17, 18*), tuneable polarisation filter (*19*), and adaptive optics (*20, 21*).

Knowing how material parameters affect the DFLC response time provides useful guidelines for improving compound structures and for optimising device operating conditions. Under small angle approximation and neglecting flow effect, in a vertical alignment LC cell with strong anchoring energy, the DFLC director's response time is determined by (*22*)

$$\tau_{on} \sim \frac{\tau_o}{(V_h/V_{th,h})^2 - 1}, \tag{1}$$

$$\tau_{off} \sim \frac{\tau_o}{(V_l/V_{th,l})^2 - 1}. \tag{2}$$

where

$$\tau_0 = \gamma_1 d^2 / K_{33}\pi^2. \tag{3}$$

In Equations (1)–(3), V_h and V_l are the driving voltages at high and low frequencies, $V_{th,h}$ and $V_{th,l}$ are the corresponding threshold voltages, γ_1 is the rotational viscosity, K_{33} is the bend elastic constant, and d is the cell gap. Equations (1) and (2) tell us how the voltage switching ratio (V/V_{th}) affects the rise and decay times. Equation (3) shows that a smaller visco-elastic coefficient and a thinner cell gap are preferred for reducing the response time. A lower visco-elastic coefficient can be achieved by employing compounds with low viscosity or by increasing the operating temperature. When a certain phase retardation (δ) is needed, the minimum cell gap is determined by $\delta = 2\pi d\Delta n/\lambda$, where Δn is the birefringence of the DFLC at wavelength λ. Therefore, a higher birefringence is favoured, especially when the operating wavelength is long. The combined effect of birefringence and visco-elastic constant can be evaluated by the figure of merit (FoM) (*2*). In general, FoM depends on temperature. As the temperature increases, FoM increases first and then decreases sharply as the temperature approaches the clearing point. Therefore, operating a LC device at an elevated temperature helps to reduce response time. However, the crossover frequency of a DFLC increases almost exponentially as the temperature increases (*12*). A higher crossover frequency leads to a high operating frequency when the DFLC exhibits

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a negative $\Delta\varepsilon$. A too high operating frequency, say 100 kHz, leads to some undesirable effects: (1) higher cost in driving circuit design; (2) larger heat dissipation; and (3) stronger dielectric heating (*23, 24*). Therefore, the upper limit of operating temperature is determined by the temperature-dependent crossover frequency.

In this paper, we report a DFLC mixture, UCF-A, with a high birefringence (Δn=0.292 at λ=633 nm, 25°C) and very low crossover frequency (<1 kHz at 25°C). The high birefringence makes it possible to employ a thin cell gap, which effectively reduces the response time. As a result of the low crossover frequency, this mixture can be operated at an elevated temperature, which means a lower visco-elastic constant and thereby a shorter response time. A DFLC optical phase modulator is demonstrated using this material. By operating the device at 45°C and applying 17 V overdrive voltage pulses, the measured rise and decay time is less than 1 ms for a phase change of 2π at λ=633 nm.

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2. Experiment and results

2.1. Mixture formulation

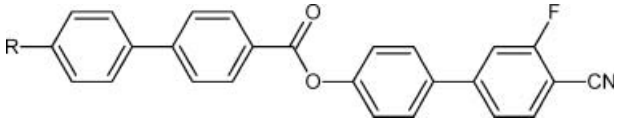
A DFLC mixture consists of two types of compounds: (1) positive compounds whose dielectric anisotropy ($\Delta\varepsilon$) is positive at low frequency but decreases as the driving frequency increases because of dielectric relaxation; and (2) negative compounds whose $\Delta\varepsilon$ is always negative and stays almost constant when the driving frequency is below the MHz range. In order to achieve a low crossover frequency and high birefringence, fluorinated single ester cyanate compounds with low dielectric relaxation frequency and high birefringence were developed and employed as the positive part in UCF-A. The structure of positive compounds is listed in Table 1. The negative part of the mixture consists of laterally fluorinated tolanes with alkoxy end groups and laterally fluorinated phenyl tolanes, as presented in Table 1. The fluorinated tolanes were chosen because of their large negative $\Delta\varepsilon$, relatively high birefringence (Δn) and low melting temperature. The fluorinated phenyl tolanes were adopted owing to their high birefringence and relatively low visco-elastic coefficient.

2.2. Mixture properties

The performance of a LC in electro-optical devices is greatly affected by the operating temperature (*12, 25*). Therefore, the properties of UCF-A were characterised at various temperatures. Results are presented in Table 2. These data can be utilised to optimise the operating condition of devices employing UCF-A.

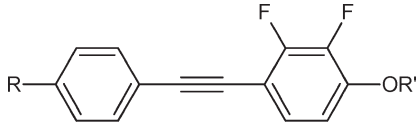
Table 1. Molecular structure of the compounds employed in UCF-A.

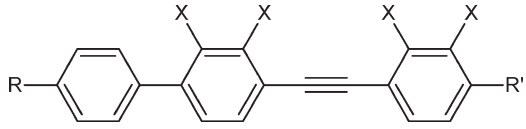
Positive compounds:



R: alkyl end group

Negative compounds:





R, R′: alkyl or alkoxy end groups;
X: at least two neighbouring X on a phenyl ring are fluorine, others are hydrogen.

The new DFLC mixture shows a high birefringence, Δn=0.292 at λ=633 nm and 25°C. The birefringence decreases with the increase of temperature, as depicted in Figure 1(a), but still remains 0.28 at 55°C. The visco-elastic coefficient, on the other hand, decreases faster with the increase of temperature as shown in Figure 1(b). The value at 45°C is only 40% of the value at 25°C. Therefore, the performance of the mixture, evaluated by the FoM$\equiv K_{33}(\Delta n)^2/\gamma_1$ (*2*), improves substantially at an elevated temperature, as presented in Figure 1(c).

Table 2. Electro-optical properties and dielectric relaxation properties of UCF-A at various temperatures.

	Temperature			
	25°C	35°C	45°C	55°C
Δn (633 nm)	0.292	0.288	0.286	0.282
γ_1/K_{11} (ms μm^{-2})	31.88	18.1	12.89	8.88
FoM (μm^2 s^{-1})	2.67	4.60	6.34	8.93
$\Delta\varepsilon$ (0)	6.48	6.00	5.39	5.00
$\Delta\varepsilon$ (∞)	−4.13	−3.89	−3.68	−3.45
Crossover frequency (kHz)	0.78	2.06	5.01	11.95

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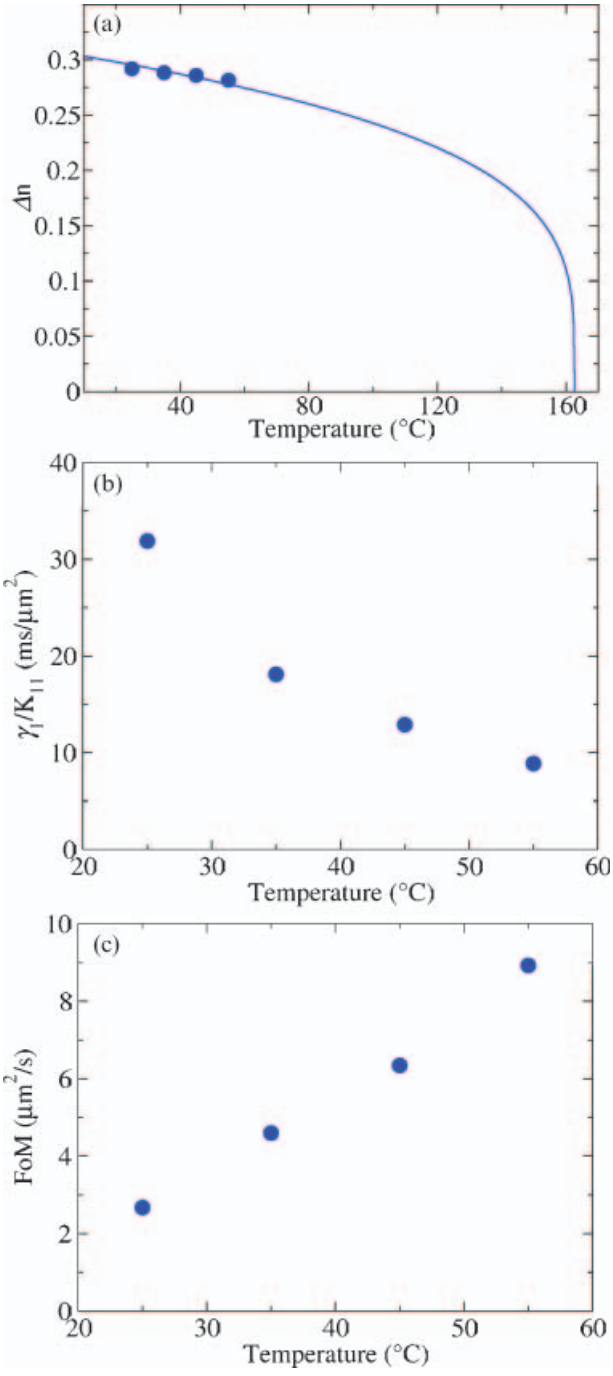

Figure 1. Temperature-dependent (a) birefringence, (b) visco-elastic coefficient and (c) figure of merit of UCF-A.

We used the simple Debye model to fit the dielectric relaxation of UCF-A at various temperatures and the results are also listed in Table 2. Δε as a function of frequency measured at 25°C, 45°C and 55°C is displayed in Figure 2(a) and the temperature dependence of crossover frequency is shown in Figure 2(b). The crossover frequency of the DFLC mixture increases almost exponentially with the

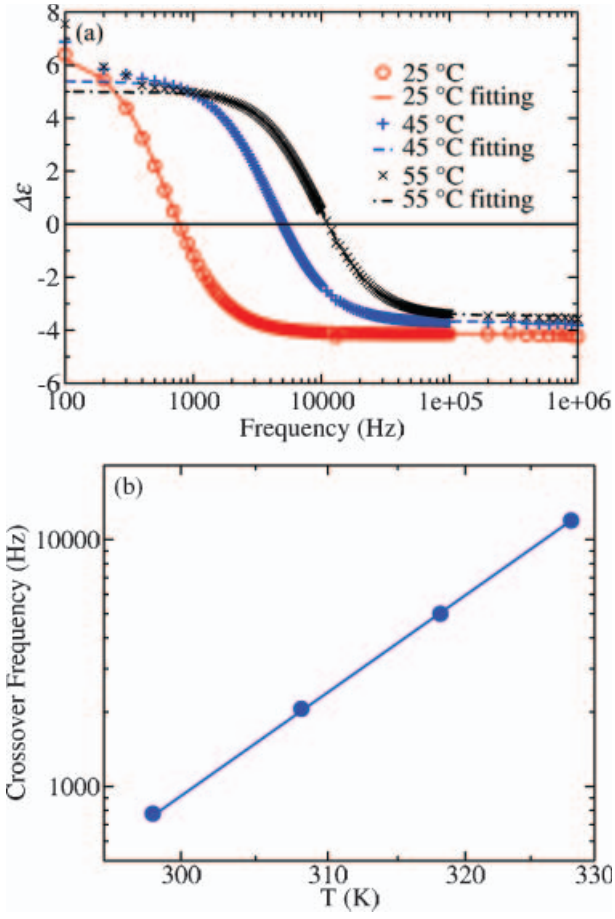

Figure 2. (a) Dielectric relaxation of UCF-A at 25°C, 45°C and 55°C; (b) the crossover frequency as a function of temperature (thermal dynamic temperature).

increase of temperature (*12*). However, the crossover frequency of UCF-A is less than 1 kHz at 25°C but only increases to 5 kHz at 45°C. Therefore, it is possible to benefit from the much reduced visco-elastic coefficient by operating UCF-A at an elevated temperature. The frequency of the high frequency signal for turning off the cell remains low enough provided that the operating temperature is not too high (<50°C). The dielectric anisotropy at both low frequency limit (Δε(0)) and the dielectric anisotropy at high frequency limit (Δε(∞)) decrease as the temperature increases. A small Δε leads to a high threshold voltage and high operating voltage. This is another trade-off for a high operating temperature.

Operating UCF-A at an elevated temperature may raise some concerns about the thermal stability of our mixture. In the past, tolane liquid crystals have been utilised to fabricate heat-resistant light modulators (*26*). For practical reasons, we will not operate our mixture higher than 60°C. Therefore, tolane compounds should be sufficiently stable.

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Previous study of the thermal stability of isothiocyanato-tolanes indicates that phenyl tolane compounds are reasonably stable when operating at elevated temperatures (*27*). However, thermal stability of the four ring single ester compounds has not been tested yet. Further investigation of operating UCF-A at elevated temperatures and extended periods of time is still necessary.

2.3. *Fast response phase modulator*

To evaluate the response time of UCF-A, we prepared a phase modulator. The DFLC was filled into a homogeneous cell with cell gap $d=5.08\,\mu m$. The LC director was set at 45° between two crossed polarisers. A helium-neon laser beam ($\lambda=633\,nm$) was used to probe the voltage-dependent transmittance of the cell. Through the measured voltage-dependent transmittance, we could obtain the phase retardation at any voltage.

We considered several factors when choosing the operating temperature of this device. We chose 30 kHz as the high frequency signal to drive UCF-A as a negative $\Delta\varepsilon$ LC. A too high driving frequency has several shortcomings as mentioned earlier. We plotted the FoM vs. crossover frequency in Figure 3. This figure elucidates how the high driving frequency put a limit on the achievable FoM, provided that the operating temperature is not an issue. The crossover frequency of UCF-A is around 12 kHz and the dielectric relaxation is almost finished at 30 kHz when operated at 55°C. The dielectric anisotropy at 30 kHz is −2.76. Therefore, operating the DFLC cell at 55°C is still possible. However, when the operating temperature exceeds 55°C, the crossover frequency is so high that the dielectric relaxation cannot finish at 30 kHz. Moreover, the $\Delta\varepsilon$ at 30 kHz is too small, which implies a fairly high turn-off voltage. On the other hand, the dielectric anisotropy decreases as the operating temperature increases. A too high operating temperature also raises concerns on the reliability of the driving electronics. In order to balance all these factors, we determined to operate the phase modulator at 45°C.

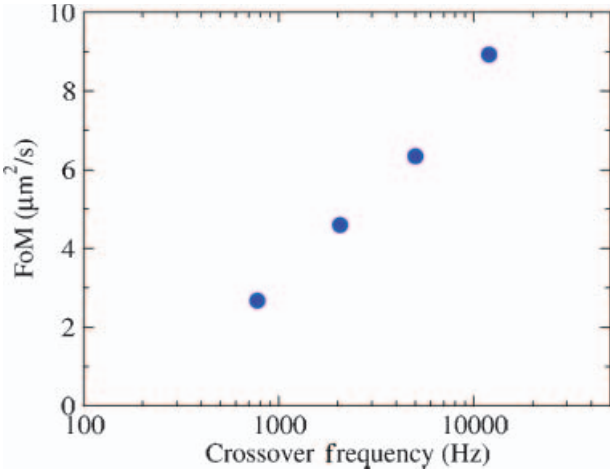

Figure 3. Figure of merit *vs.* crossover frequency of UCF-A.

The maximum phase retardation of our DFLC cell at $\lambda=633\,nm$ is more than 4π at 45°C. For a phase modulator, we need 2π phase change. We chose the 4π retardation state as the off state, and the 2π retardation state as the on state. Owing to the low crossover frequency, we were able to take advantage of the low visco-elastic coefficient without using a too high frequency signal to turn off the cell. To turn on the device, we applied a 1-ms low frequency (1 kHz)

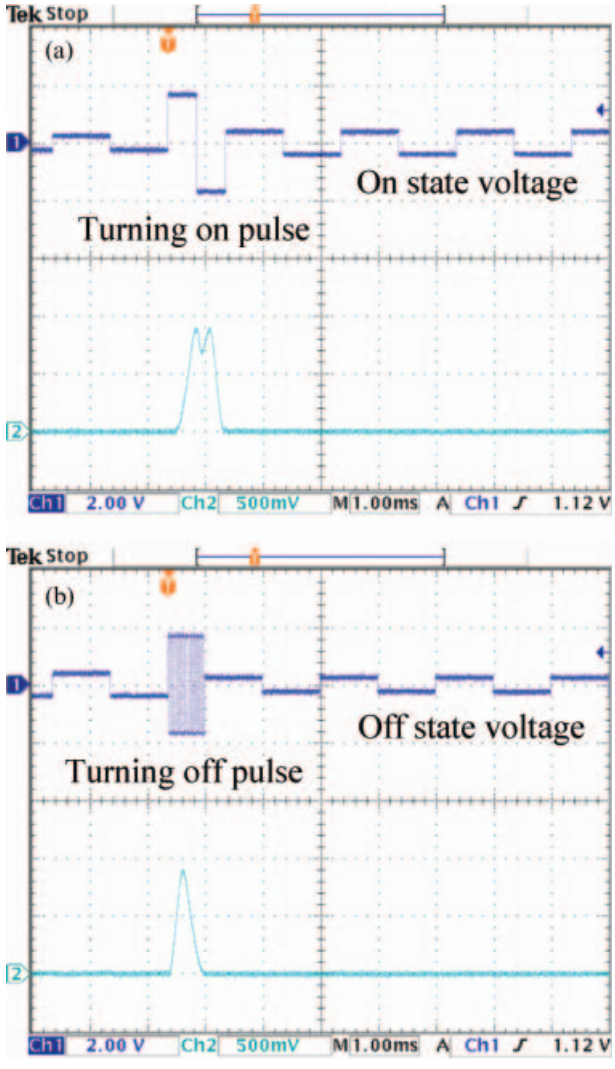

Figure 4. (a) Turn-on and (b) turn-off of the dual-frequency liquid crystal phase modulator with high-voltage pulses. The phase retardation changes between 4π (off) and 2π (on). Therefore, the transmittance changes from 0 to 0.

voltage (17 V_{rms}) pulse to quickly overdrive the cell to the on state, as illustrated in Figure 4(a). While turning off the device, a 634 μs high frequency (30 kHz) pulse (17 V_{rms}) was utilised to force the LC directors falling back to the off state, as depicted in Figure 4(b). A 500-Hz signal was employed to maintain the cell at the on and off states after the overdrive and turn-off pulses. The turn-on time, defined by the 10% to 10% transmittance change (because the involved phase change is 2π in this case) is 708 μs. Similarly, the 10% to 10% off time is 490 μs. The 'ripple' near the transmittance peak in Figure 4(a) is due to the high frequency components in the driving signal at the point when the overdrive pulse changes sign (*28*).

3. Conclusion

We have developed a DFLC mixture, UCF-A, with high birefringence and a low crossover frequency. High birefringence enables a thinner cell gap to be used, which not only reduces the response time, but also lowers the driving voltage. This is especially appreciated in electro-optical devices working in the infrared region. On the other hand, the initially low crossover frequency permits us to operate the device at an elevated temperature for reducing visco-elastic coefficient. This is particularly important because most DFLC mixtures have a relatively high viscosity due to the highly polar compounds. In a phase modulator employing UCF-A and operating at 45°C, we demonstrated sub-millisecond turn-on and turn-off time with 17 V overdrive pulse for 2π phase change at $\lambda = 633$ nm. This type of DFLC and design concept opens a new door for high-speed DFLC phase modulators.

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