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Polarization-independent phase modulation of a homeotropic liquid crystal gel

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A homeotropic liquid crystal (LC) gel with submicron domain size is fabricated and its phase-only modulation property evaluated. The LC gel is highly transparent in the voltage-off state and exhibits pure phase modulation before light scattering occurs. Compared to a nanosized polymer-dispersed liquid crystal (nano-PDLC), our gel possesses a larger phase change but at a lower operating voltage because of a higher LC concentration. Similar to a nano-PDLC, our gel also exhibit submillisecond response time, hysteresis-free, and polarization-independent phase change. © 2005 American Institute of Physics. [DOI: 10.1063/1.2126107]

Liquid crystal (LC)-based phase modulators are useful for photonic devices, optical information processing, optical communications, and three-dimensional displays. To realize phase-only modulation, several approaches such as homogeneous LC,¹ twisted-nematic LC,^{2,3} polymer network LC,⁴ sheared or stressed liquid crystal,^{5,6} polymer-dispersed liquid crystal (PDLC),^{7–9} and nanosized PDLC^{10–14} (nano-PDLC) have been investigated. Among these approaches, the nano-PDLC is particularly attractive because of its polarization independence and submillisecond response time. However, in a nano-PDLC, the LC concentration is ~30% and the LC molecules are strongly anchored by the polymer walls. As a result, nano-PDLC exhibits a small phase shift and requires a high operating voltage (~10 V/ μ m). To enlarge phase shift by using a thick nano-PDLC layer would further increase the operating voltage.

Recently, we demonstrated a polarization-independent phase modulator using a conventional PDLC layer.¹⁵ To bypass the light scattering regime, a bias voltage is applied to the PDLC layer. Because the average LC droplet size is larger than the visible wavelength, the operating voltage is relatively low (3 V/ μ m). Meanwhile, the response time is fast (~1 ms) benefiting from the bias voltage effect. However, due to the bias voltage effect the remaining tunable phase change is relatively small (~0.04 π). Although the observed small phase change is still usable for microphotonic devices, it is highly desirable to increase the phase shift and decrease the operating voltage while keeping a fast response time.

In this letter, we demonstrate a homeotropic LC gel whose phase shift is larger but operating voltage is lower than a nano-PDLC. The larger phase change and lower operating voltage originate from the higher LC concentration in our gel. Different from a conventional LC gel,¹⁶ our LC domain size is in submicron region. In the voltage-off state, the light scattering is completely suppressed because of the good refractive index match between the LC and the polymer network.

Figures 1(a), 1(b), and 1(c) show the schematic configurations of the LC directors and polymer networks in the voltage-off (V=0), threshold (V_{th}) for directors reorientation, and light scattering states ($V_s > V_{th}$) of the homeotropic LC gel. In Fig. 1(a) where V=0, the LC directors are aligned nearly perpendicular to the substrates and stabilized by polymer gels. The polymer networks are formed along the same orientation as the LC directors. Because of the small domain sizes and good index match, the LC gel is highly transparent. As the applied voltage exceeds a threshold, the LC directors begin to tilt away from the electric field direction, as shown in Fig. 1(b), because the LC has a negative dielectric anisotropy ($\Delta \varepsilon < 0$). Further increasing voltage to V_s , light scattering occurs because the refractive index mismatch between the LC and the polymer gel, as shown in Fig. 1(c). From Fig. 1(b) to 1(c), phase-only modulation is expected. Due to the random reorientation of the LC directors in the polymer networks, the phase shift is polarization independent for the normally incident light.

To prepare a homeotropic LC gel with small LC domain sizes, we mixed 20 wt % of a Merck photocurable LC diacrylate monomer RM257 in a negative nematic LC MLC-



FIG. 1. Schematic diagrams of polymer and LC directors orientations of a homeotropic LC gel: (a) V=0, (b) $V=V_{\rm th}$ where the LC directors reorientation starts, and (c) $V=V_{\rm s}$ where the light scattering takes place.

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FIG. 2. Voltage-dependent transmittance of the LC gel. $d=23 \ \mu\text{m}$. An unpolarized He–Ne laser was used for this measurement.

6608 (Δn =0.083, $\Delta \varepsilon$ =-4.2). The LC/monomer mixture was injected into an empty cell in the isotropic state. The inner surfaces of the indium-tin-oxide (ITO) glass substrates were coated with thin homeotropic polyimide alignment layers and rubbed in orthogonal directions in order to reduce the polarization dependency originating from the boundary layers. The pretilt angle is ~88° on each surface. The filled cell was then slowly cooled down to room temperature and exposed to UV light (λ ~365 nm, I~10 mW/cm²). The UV curing time for the cell was ~30 min. The cell gap was controlled at ~23 μ m using two stripes of mylar film.

The electro-optic properties of the LC gel was characterized using an unpolarized He–Ne laser beam (λ =633 nm). The transmitted light was measured by a photodiode detector which was placed at ~30 cm behind the sample. A computer controlled LabVIEW data acquisition system was used for driving the sample and recording the light transmittance. The response time of the LC gel was recorded by a digital oscilloscope.

The LC gel sample appears slightly bluish as visually observed in the reflection state, which implies that the formed LC domain size is comparable to a blue wavelength ($\lambda \sim 400$ nm). The LC gel is highly transparent at the He–Ne laser wavelength ($\lambda \sim 633$ nm).

Figure 2 plots the voltage-dependent transmittance of the LC gel. The LC gel is highly transparent at V=0. Below $180V_{\rm rms}$, the transmittance remains at ~88%. The ~12% light loss mainly originates from the interface reflections between the ITO-glass substrates and the air. The refractive index of ITO is ~1.90. As the voltage exceeds $180V_{\rm rms}$, the transmittance begins to decline due to light scattering. Therefore, we define $180V_{\rm rms}$ as the V_s of the LC gel. The relatively high V_s is due to the small $\Delta \varepsilon$ and small Δn of the LC mixture employed, and the submicron domain sizes due to high (20%) polymer concentration. The hysteresis of the voltage-dependent transmittance of the LC gel was also measured. Results indicate that the forward and backward curves overlap very well, which means the hysteresis is completely suppressed.

To examine the phase modulation capability of the LC gel, we measured the transmittance $(T_{\parallel} \text{ and } T_{\perp})$ at $\lambda = 633$ nm between parallel and crossed polarizers with the beam normally incident on the sample surface. We set the polarization axis of the analyzer parallel to one rubbing direction of the cell. Results are shown in Fig. 3. Due to the homeotropic alignment of the LC gel, $T_{\perp} \sim 0$ at V=0. As the applied voltage exceeds the threshold voltage $V_{\text{th}}=130V_{\text{rms}}$, T_{\parallel} decreases but T_{\perp} increases with voltage gradually. This means phase-only modulation exists. At $V > 180V_{\text{rms}}$, light scattering takes place which is not desir-



FIG. 3. Voltage-dependent transmittance of the LC gel between parallel (T_{\parallel}) and crossed (T_{\perp}) polarizers. Cell gap $d=23 \ \mu$ m, $f=1 \ \text{kHz}$, $\lambda=633 \ \text{nm}$, and $T\sim21 \ ^{\circ}\text{C}$.

able for the phase-only modulation. To validate whether the gel is polarization dependent, we rotated the sample in the azimuthal direction. Results remain unchanged. This indicates that the phase-only modulation is independent of incident light polarization.

The phase change in the range from $0 \sim 180V_{\rm rms}$ can be calculated by taking the ratio of T_{\parallel}/T_{\perp} .¹⁷ Figure 4 plots the voltage dependent phase change of the LC gel at $\lambda = 633$ nm and $T \sim 21$ °C. As the voltage increases, the phase increases gradually because the LC directors inside the domains are reoriented randomly away from the electric field, leading to an increased effective birefringence. From 130 to $180V_{\rm rms}$, the phase shift is $\Delta \delta \sim 0.08\pi$ (for the 23 μ m LC gel) which is $2 \times$ larger than our previous results using a conventional PDLC.¹⁵

In our LC gel, the effective refractive index at V=0 can be expressed as

$$n_{o\text{-eff}} = \frac{\nu_{\text{LC}} n_o + \nu_p n_p}{\nu_{\text{LC}} + \nu_p},\tag{1}$$

where n_o is the ordinary refractive index of the LC, ν_{LC} , and ν_p are the LC and polymer volume fractions, and n_p is the refractive index of the polymer. By applying a voltage across the LC gel, the LC directors tend to reorient themselves perpendicular to the electric field because of the negative $\Delta \varepsilon$. Therefore, light impinging on the sample at normal incidence will see an average refractive index increased from n_o to $\overline{n}(V)$. In this case, the effective refractive index of the LC gel can be rewritten as



FIG. 4. Measured phase shift of the LC gel at different voltages. $d=23 \ \mu m$ and $\lambda=633 \ nm$.

100

Voltage, Vms

150

200

50

0.00

0



FIG. 5. Measured response time of the LC gel: (a) $\tau_{\rm rise} \sim 0.59$ ms and (b) $\tau_{\rm decay} \sim 0.15$ ms. The square waves in each figure represent the voltage bursts at f=1 kHz.

As a result, the field-induced phase shift can be expressed as

$$\Delta \delta = \frac{2\pi}{\lambda} d(n_{\rm eff} - n_{o-\rm eff}), \qquad (3)$$

where *d* is the cell gap and λ is the incident wavelength. By combining Eqs. (1) and (3), we rewrite the phase shift as follows:

$$\Delta \delta = \left(\frac{\nu_{\rm LC}}{\nu_{\rm LC} + \nu_p}\right) \frac{2 \pi d(\bar{n}(V) - n_o)}{\lambda}.$$
(4)

From Eq. (4), the LC volume fraction $(\nu_{\rm LC})$ is an important parameter contributing to the phase change because $\nu_{\rm LC} + \nu_p$ does not change. In our gel system, the LC concentration is ~80% which is much higher than that in a nano-PDLC (~30%). As a result, our LC gel should exhibit a larger phase shift than the nano-PDLC, if the same Δn is employed.

From Fig. 4, the 0.08π phase change was obtained at $V=180V_{\rm rms}$ which corresponds to 7.8 V/ μ m. Two factors affecting the on-state voltage are $\Delta \varepsilon$ of the LC mixture and LC concentration. Negative LC mixtures tend to have a smaller $\Delta \varepsilon$ than their positive counterparts. A nano-PDLC device usually uses a positive, large $\Delta \varepsilon$ LC mixture in order to suppress its operating voltage. Although our LC gel uses a negative, small $\Delta \varepsilon$ LC mixture, its required electric field strength is still lower than that of a nano-PDLC because of the higher LC concentration involved. A higher LC concentration not only leads to a slightly larger LC domain size but also decreases the contact interface between the polymer binder and the LC molecules. As a result, the operating voltage is reduced. The strong anchoring force that the polymer binders exert to the LC directors is responsible for the observed fast response time.

If we operate the gel in reflective mode, the phase change will be doubled. Although the achievable phase change is small ($\delta \sim 0.08 \pi$), it is still quite useful for the polarization-independent microlens and microprism applications. From Eq. (4), to increase phase shift we could either enlarge the LC cell gap or use a higher birefringence LC material.¹⁸ The latter is preferred because increasing cell gap would lead to a higher operating voltage. However, for a given domain size, increasing LC birefringence would also enhance light scattering capability. Therefore, an optimal LC birefringence should exist before light scattering takes place. On the other hand, to lower the on-state voltage we could use a higher $\Delta \varepsilon$ LC mixture.

Response time is another important parameter for LC based phase modulator. We measured the response time of the LC gel using square voltage bursts at f=1 kHz between 0 and $180V_{\rm rms}$. Results are shown in Fig. 5. The measured rise time is $\tau_{\rm rise} \sim 590 \ \mu$ s and decay time $\tau_{\rm decay} \sim 150 \ \mu$ s at room temperature. Such a fast response time results from the small LC domain size as well as the strong polymer stabilization effects. Based on the measured decay time, we estimate the domain size is around 300 nm. This is consistent to the very weak bluish appearance of the LC gel.

In conclusion, we have demonstrated a homeotropic LC gel whose phase modulation is polarization insensitive. Such a phase modulation is free from light scattering and hysteresis. Its response time is submillisecond at room temperature and its operation stability is excellent. The obtainable phase change is $2 \times$ larger than that of a nano-PDLC system, but at a lower operating voltage.

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