Submillisecond-response polymer network liquid crystal phase modulators at 1.06-μm wavelength

Jie Sun, Haiqing Xianyu, Yuan Chen, and Shin-Tson Wu[a]
College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816, USA

A fast-response and scattering-free polymer network liquid crystal (PNLC) light modulator is demonstrated at $\lambda = 1.06$ μm wavelength. A decay time of 117 μs for 2π phase modulation is obtained at 70 °C, which is $\sim 650 \times$ faster than that of the host nematic LCs. The major tradeoff is the increased operating voltage. Potential applications include spatial light modulators and adaptive optics. © 2011 American Institute of Physics. [doi:10.1063/1.3611031]

Fast-response liquid crystal (LC) phase modulators are useful for adaptive optics,[1] spatial light modulators,[2] and switchable displays.[3] The response time of a LC device is mainly determined by the cell gap, visco-elastic constant, and operating voltage.[4] For nematic LCs, the turn-on process can be accelerated by a high driving voltage while the turn-off process is governed by the restoring elastic torque which is relatively slow. Dual frequency LCs allow both rise and decay times to be accelerated by driving voltage; however, a complex driving process is required.[5–7] By introducing polymer into LC host,[8] polymer network liquid crystal (PNLC) exhibits a dramatically improved response time because of the increased elastic constant. At elevated temperatures, PNLCs exhibit even faster response time due to the reduced viscosity. However, very few studies on the temperature effect of PNLCs have been reported.

In this letter, we demonstrated a submillisecond-response and scattering-free PNLC phase modulator at $\lambda = 1.06$ μm. The measured response time is 117 μs for a 2π phase change at ~ 70 °C with an operating voltage of $V_{2\pi} = 90$ V. To understand the underlying physical mechanisms, we measured the temperature effects on birefringence (Δn), dielectric anisotropy ($\Delta\varepsilon$), splay elastic constant (K_{11}), and rotational viscosity (γ_1) of the PNLC. Our results indicate that the splay elastic constant of PNLC is increased by $\sim 950 \times$ which contributes to fast response time but also to the increased operating voltage.

To fabricate PNLC modulators, we first prepared a host nematic LC mixture by mixing 60% Merck E44 and 40% Merck E48 in order to obtain high birefringence and good miscibility with monomers. Light scattering is a serious problem for PNLCs. To suppress scattering, the domain size of PNLC should be smaller than the wavelength. Increasing polymer concentration would shrink the domain size which is favourable for suppressing scattering, but the operating voltage will increase dramatically. In our experiment, we have tried several monomers and different concentrations and found that following combination led to tolerable scattering ($< 1\%$) and minimum voltage: 7 wt. % bisphenol-A-dimethacrylate (M1) and 5 wt. % RM-257 (Merck). The reason for using M1 is because it helps to reduce the operating voltage and improve solubility.

We then filled the LC/monomer mixture into an empty cell with homogenous alignment and cured it with a UV lamp. The cell gap was controlled at 11.8 μm in order to obtain more than 2π phase change while keeping the operating voltage below 100 V. High UV exposure intensity helps to reduce domain sizes of polymer network. In our experiment, we controlled the UV intensity at ~ 300 mW/cm^2 and exposure time for 1 h.

To characterize the electro-optical properties of the PNLC cell, we measured its voltage-dependent transmittance (VT) with a laser beam at $\lambda = 1.06$ μm. The PNLC cell was sandwiched between two crossed polarizers, with the rubbing direction oriented at 45° to the polarizer's transmission axis. The driving frequency is 5 kHz. The clearing point of PNLC was measured to be $T_c = 92.0$ °C, which is slightly lower than that of the LC host ($T_c = 96.2$ °C).

Figure 1 depicts the measured VT curves of the PNLC cell (solid line) and the host LC cell (dashed lines) at $\lambda = 1.06$ μm. The total phase retardation of the PNLC cell is 3.0π and its corresponding voltage for $\delta = 2\pi$ occurs at $V_{2\pi} = 90$ V_{rms}. The threshold voltage of the PNLC cell is $V_{th} = 25.1$ V_{rms} while it is 0.72 V_{rms} for the host LC cell. The increased threshold voltage is attributed to the increased K_{11} because the LC domains are tightly anchored by the polymer networks. The peak transmittance of the PNLC cell is comparable to that of the host nematic LC cell, indicating that light scattering is negligible at this wavelength.

Decay time of the PNLC cell at 70 °C which was initially biased at $V_{2\pi} = 90$ V was measured. The biased
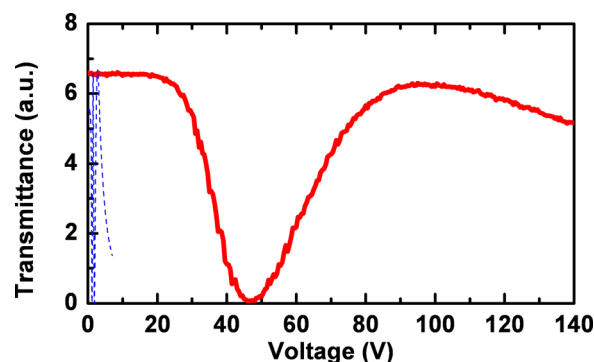

FIG. 1. (Color online) Measured VT curve of host nematic LC (dash lines) and PNLC (solid line) cells. The cell gap for both cells is 11.8 μm. Wavelength $\lambda = 1.06$ μm and T = 70 °C.

[a]Electronic mail: swu@mail.ucf.edu.

voltage was removed abruptly at t = 0 and then the transient relaxation process recorded by a photodiode. The experimental data fit well (not shown) with following exponential phase decay equation: $\delta(t) = 2\pi \exp(-t/\tau)$, where τ is the decay time constant.[9] From fitting, we obtain $\tau = 50$ μs. If the decay time is defined from $100 \rightarrow 10\%$ phase change, then we find $t_{relax} = 117$ μs. The rise time t_{rise} $(0 \rightarrow 90\%)$ was measured to be 45 μs. For the host nematic LC cell, its time constant τ is ~ 35 ms, and the corresponding decay time and rise time is ~ 77 ms and ~ 104 ms, respectively. Our PNLC improves the decay time by $\sim 658 \times$ at the expense of increasing $V_{2\pi}$ from 1.42 V to 90 V. Figure 2 shows the temperature-dependent decay time of our PNLC cell while keeping phase modulation at 2π. As the operating temperature increases, the response time gets faster because of the reduced visco-elastic constant.

Figure 3 shows the temperature dependent Δn of the PNLC cell. As the temperature increases, the birefringence decreases gradually. The filled and open circles represent the experimental data for the PNLC and host LC, respectively, while lines represent the fittings with Haller's equations:[10,11] $\Delta n = \Delta n_0 \cdot S$ and $S = (1 - T/T_c)^\beta$; where Δn_0 is the birefringence at T = 0, β is a material constant and S is the order parameter.

From Fig. 3, the PNLC shows lower Δn_0 and β ($\Delta n_0 = 0.235$ and $\beta = 0.20$) than the host LC ($\Delta n_0 = 0.325$ and $\beta = 0.25$). The Δn_0 of PNLC is only 72% (instead of 88%) of that of the host LC. This discrepancy is believed to result from the disturbed LC alignment surrounded by the polymer networks. Although each substrate surface has an alignment layer, the polymer networks may not follow perfectly the surface rubbing direction. Moreover, the employed M1 polymer is not rod-like so that its formed networks will have bend structure, which further reduces the birefringence of the PNLC composite. Since some LC molecules are anchored tightly by the surrounding polymer networks, the birefringence of PNLC is less sensitive to the temperature change than the host LC. This explains why the β value decreases from 0.25 for the host LC to 0.20 for the PNLC.

We also measured the temperature dependent V_{th} through the VT curves and $\Delta\varepsilon$ of PNLC by the conventional two-cell method.[9] The $\Delta\varepsilon$ results (not shown) fit with $\Delta\varepsilon = C_0 \cdot S$ well, where C_0 is a material constant and S is the

FIG. 3. (Color online) Temperature-dependent birefringence of PNLCs and LC host. Filled circles and solid lines are the experimental and fitting results for PNLC, respectively. Open circles and dashed lines are the experimental and fitting results for LC host, respectively.

order parameter. For PNLC, $C_0 = 13.5$ and for host nematic LC, $C_0 = 22.0$. By using the obtained V_{th} and $\Delta\varepsilon$, we then calculate K_{11} according to the following equation:[12]

$$V_{th} = \pi\sqrt{K_{11}/\varepsilon_0 \Delta\varepsilon}. \tag{1}$$

At $70\,^\circ$C, the splay elastic constant of our PNLC is $K_{11} = 1.63 \times 10^4$ pN, which is $\sim 920 \times$ larger than that of the LC host, $K_{11} = 17.7$ pN. This is because the strong polymer network anchoring helps to increase the elastic constant of the PNLC. On the positive side, a large elastic constant is favorable for achieving faster response. But on the down side, it increases the operating voltage, as Eq. (1) indicates.

Figure 4 depicts K_{11} of our PNLC against S^2. Symbols stand for the experimental data and dashed lines are the fittings according to Maier-Saupe theory:[13] $K_{11} = A_0 \cdot S^2$, where A_0 is a material constant. Through fittings, we get $A_0 = 68.5$ pN for the LC host and $A_0 = 4.7 \times 10^4$ pN for the PNLC. However, it is found that K_{11} of PNLC no longer fits well with the mean field theory because of the involvement of polymer network. Therefore, we propose a modified equation for the K_{11} of PNLCs

$$K_{11} = A_0 \cdot S^\alpha, \tag{2}$$
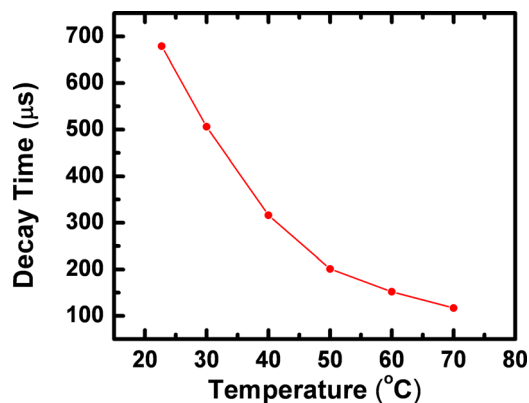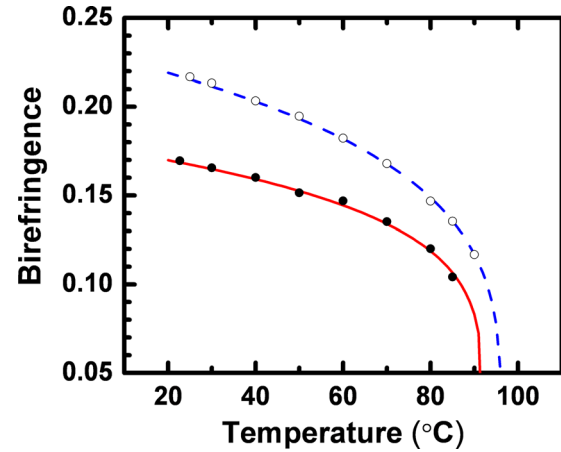

FIG. 2. (Color online) Temperature-dependent decay time of the PNLC cell. d = 11.8 μm. Filled circles are the measured decay time for 2π phase change.
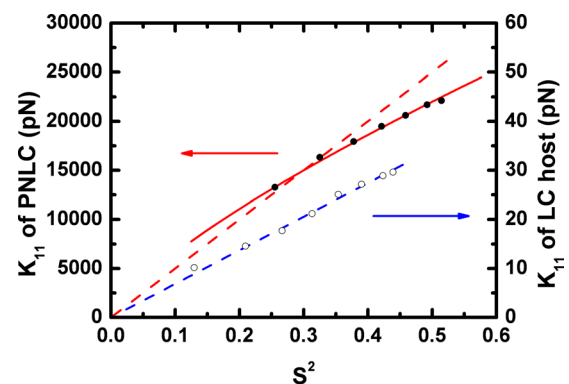

FIG. 4. (Color online) Elastic constant (K_{11}) vs. S^2. Filled circles and empty circles are the experimental results for PNLC and LC host, respectively. The dashed lines are fitting results using $K_{11} = A_0 \cdot S^2$, while the solid line is the fitting using Eq. (2) with $\alpha = 1.5$.
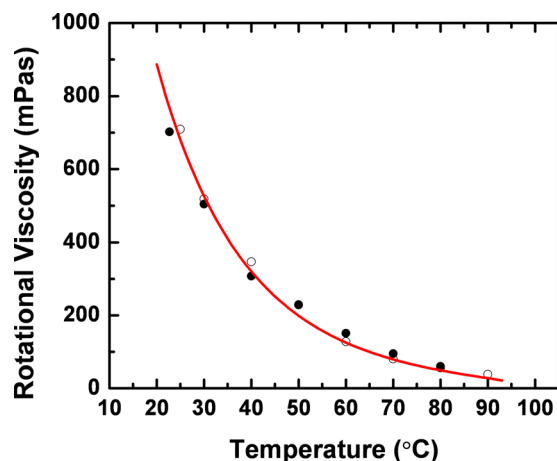
FIG. 5. (Color online) Rotational viscosity (γ_1) vs. temperature. Filled circles and empty circles are the experimental results for PNLC and LC host, respectively. The solid line represents the fitting results with Eq. (3).

where α is a material constant related to polymer network. For nematics, $\alpha = 2$. But for our PNLC, we found $A_0 = 3.7 \times 10^4$ pN and $\alpha = 1.5$ (solid line).

In order to get the rotational viscosity of PNLCs, we first obtained visco-elastic constant (γ_1/K_{11}) by measuring the free relaxation time of the sample cell and then calculated γ_1 based on the K_{11} values we already obtained. Figure 5 shows the temperature dependent rotational viscosity of PNLC (filled circles) and nematic host LCs (open circles). The solid line is the fitting results according to the modified Arrhenius model[14,15]

$$\gamma_1 = bS \exp(E_a/k_B T), \tag{3}$$

where b is the proportionality constant, E_a is the activation energy, and K_B is the Boltzmann constant. From Fig. 5, the measured viscosity of PNLC is very close to that of LC host; the small variation results from the minor difference in their clearing temperatures. The fitting parameters are $b = 5.0 \times 10^{-4}$ mPa $\cdot$ s and $E_a = 372.0$ meV.

In conclusion, we have demonstrated a submillisecond-response and scattering-free PNLC phase modulator for $\lambda = 1.06$ μm. A decay time of 117 μs is obtained at 70 °C for 2π phase modulation. Our results show that the temperature dependent birefringence, dielectric anisotropy, and rotational viscosity of PNLCs still follow the models for nematic LCs. The rotational viscosity of PNLC remains basically the same as that of the LC host. However, since the splay elastic constant (K_{11}) is largely dependent on the polymer network, some deviation from nematic LCs is observed.

[1]M. Langlois, C. D. Saunter, C. N. Dunlop, R. Myers, and G. D. Love, Opt. Express **12**, 1689 (2004).
[2]U. Efron, *Spatial Light Modulator Technology: Materials, Devices, and Applications* (Marcel Dekker, New York, 1994).
[3]M. G. H. Hiddink, S. T. de Zwart, O. H. Willemsen, and T. Dekker, SID Symp. Digest **37**, 1142 (2006).
[4]S. T. Wu, Appl. Opt. **28**, 48 (1989).
[5]H. K. Bücher, R. T. Klingbiel, and J. P. VanMeter, Appl. Phys. Lett. **25**, 186 (1974).
[6]M. Schadt, Mol. Cryst. Liq. Cryst. **66**, 319 (1981).
[7]J. Sun, H. Xianyu, S. Gauza, and S. T. Wu, Liq. Cryst. **36**, 1401 (2009).
[8]Y. H. Fan, Y. H. Lin, H. Ren, S. Gauza, and S. T. Wu, Appl. Phys. Lett. **84**, 1233 (2004).
[9]S. T. Wu and C. S. Wu, Phys. Rev. A **42**, 2219 (1990).
[10]I. Haller, Prog. Solid State Chem. **10**, 103 (1975).
[11]S. T. Wu, Phys. Rev. A **33**, 1270 (1986).
[12]I. C. Khoo and S. T. Wu, *Optics and Nonlinear Optics of Liquid Crystals* (World Scientific, Singapore, 1993).
[13]W. Maier and A. Saupe, Z. Naturforsch. Teil A **15**, 287 (1960).
[14]W. H. De Jeu, Mol. Cryst. Liq. Cryst. **63**, 83 (1981).
[15]S. T. Wu and C. S. Wu, Liq. Cryst. **8**, 171 (1990).