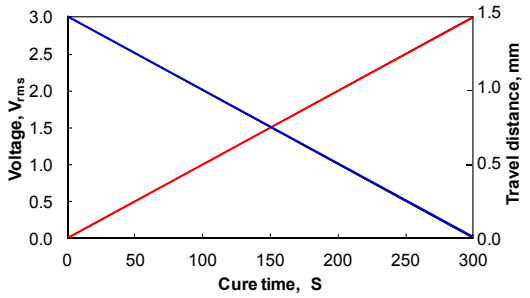

Fig. 5. The voltage applied to the cell (blue), the movement of the cell (red), and the curing time.

At V = 0, the observed color stripes of the PNLC under POM are shown in Fig. 6(a). The top-left region (red color) was cured at 3 V_{rms} and the bottom-right region (red color) was cured at V = 0. Along the diagonal direction, this PNLC shows nearly two cycles of the color change (from red to red) and the color changes continuously in each cycle, implying that the PNLC presents a gradient phase (or refractive index) difference. The transmittance (normalized to two parallel polarizers) of the PNLC cell between two crossed polarizers is expressed by [21]:

$$T = \sin^2 \frac{\pi d (n_\theta - n_o)}{\lambda}, \tag{4}$$

From Eq. (4), the red stripes will appear when the effective refractive index (with tilt angle θ) satisfies the following relation:

$$n_\theta = n_o + (p - \frac{1}{2}) \frac{\lambda_{red}}{d}, \tag{5}$$

where p is an integer and λ_{red} is the wavelength of the red light. If λ_{red} = 0.65 μm, d = 6.7 μm, when p = 1, 2, and 3, n_θ is calculated to be 1.58, 1.67, and 1.77, respectively. These three refractive indices are obtainable for LCs oriented with a small or zero tilt angle (BL038 n_o = 1.527, n_e = 1.799). Also from Eq. (5), the phase difference between two adjacent red stripes is 2π, so the phase difference within three red stripes at V = 0 is ~4π, which is very close to that shown in Fig. 2 (~4.2π phase retardation when the voltage increases from 0 to 3 V_{rms}). Compared to the previous fabrication procedures of gradient PNLC, in which the gradient refractive index distribution is formed by exposing UV light through a patterned photomask [8] or hole-patterned electrodes [22], our approach offers a possibility to get PNLC with the largest refractive index change (i.e., from n_e to n_o) under a given precursor.

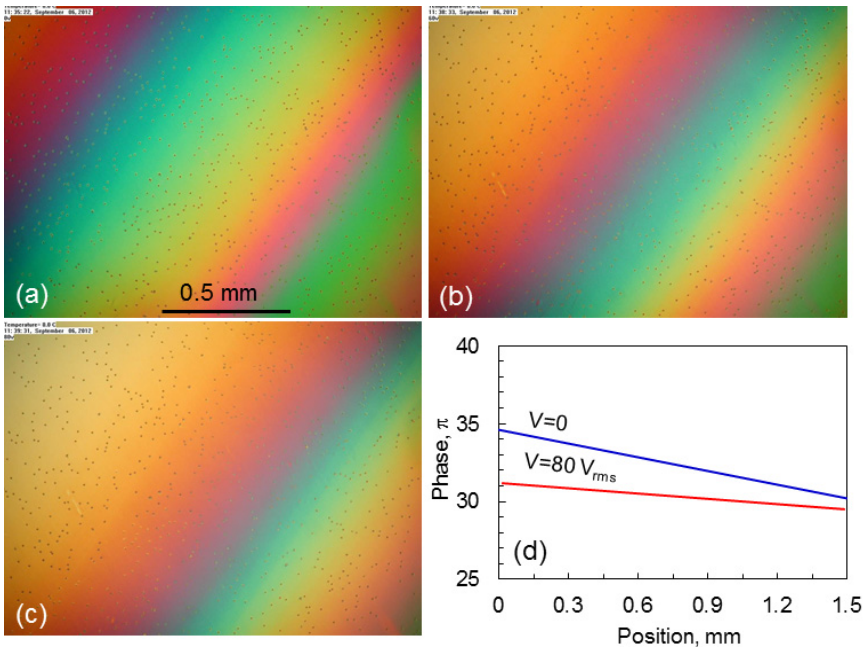

Fig. 6. The observed PNLC color at (a) V = 0, (b) V = 50 V_{rms} (c) V = 80 V_{rms} (Media 3), and (d) the measured phase difference across the cured region with a 1.5-mm width (travel distance).

By applying various voltages to the PNLC cell, the phase gradient of the PNLC can be changed as well. At V = 10 V_{rms}, the color starts to shift along the direction from top-left to bottom-right. At V = 50 V_{rms}, a distinct color shift is observed (Fig. 6(b)), indicating that the phase gradient of the PNLC is decreased. At V = 80 V_{rms} the gradient of the PNLC is largely decreased as a much loose color distribution is observed (Fig. 6(c)). LCs in the top-left region is highly reoriented along the electric field, so this region presents a mutual color. Media 3 shows the dynamic color change of the PNLC when it is switched between 0 and 80 V_{rms}. Similar to the first PNLC cell, the second cell can be driven cycle by cycle with good color stability, fast response time and negligible light scattering. The measured phase gradient of the PNLC is shown in Fig. 6(d). At V = 0, the PNLC exhibits the largest phase gradient. At V = 80 V_{rms}, the phase gradient is largely decreased. If the voltage is continuously increased, the phase gradient can be further decreased.

In Fig. 6(a), the color distribution in the PNLC across the 1.5-mm-width of the cured region is quite loose. To contract the color distribution, one approach is to decrease the cell's traveling distance and increase the voltage gradient during UV exposure. Another approach is to increase the cell gap, as depicted in Eq. (1). As a result, the phase difference ($\Delta\phi$) across the cured region can be increased, but the voltage to tune the gradient of the PNLC will increase accordingly. Depending on the photomask patterns, our gradient PNLC can be used to prepare various adaptive photonic devices, such as prism gratings and Fresnel-zone/circular/lenticular lenses. Due to high diacrylate monomer concentration, the formed PNLC presents good stability, fast response and negligible light scattering.

Exposure time, UV intensity and LC concentration do affect the electro-optical properties (response time, operating voltage) and the gradient refractive index distribution (stripe colors) of the formed PNLCs. In our experiments, since the UV intensity is ~20 mW/cm^2 the exposure time is already long enough to fully polymerize the monomer and fix the oriented LC molecules. A longer curing time will not cause too much difference in the polymerization. But if the UV intensity is too high, the operating voltage will increase due to a thinner and denser polymer network. If it is too low, the monomers cannot be fully polymerized and the

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polymer network structure will not be stable. Moreover, LC (or monomer) concentration is also a critical parameter for the polymer network structure. Generally speaking, a high LC concentration will induce large polymer domains along with light scattering and performance degradation. While a low LC concentration will lead to fast relaxation, high operating voltage and small effective phase shift. In our experiments, ~88 wt% BL038 concentration is close to the optimal condition. Detailed investigation about the impact of UV curing time, UV intensity and LC concentration is certainly important, but it is beyond the scope of this paper.

For a given precursor, the number of steps, distance of each step and applied voltage during UV exposure play important roles in establishing the gradient of the refractive index change across the curing range. Here we just give two examples of the proposed approach: 1) 6 steps of 0.4 mm each (Fig. 3) and 2) continuous traveling (Fig. 5). The voltage applied to the cell during UV exposure can be determined according to the VT curve shown in Fig. 2 and it affects the maximal phase change of the PNLC. As shown in Figs. 5 and 6, when the applied voltage deceases from 3 V_{rms} to zero, the maximal phase change of the PNLC decreases to ~4π. Therefore, our approach offers more freedom to design the moving trail along with the voltage applied to the cell during UV exposure, based on the specific requirements. The cell's travelling speed itself does not affect the formed PNLC morphology. However, given a fixed total traveling time a faster travelling speed leads to a smoother gradient of the refractive index change. While given a fixed travelling distance, a faster travelling speed leads to a shorter curing time. It is not a concern if the UV intensity is high enough to fully polymerize the monomers, otherwise, the formed polymer network structure will not be stable. Because the phase separation process is rather complicated, it is challenging to optimize all the parameters (e.g. LC/monomer concentration, curing time/intensity, moving trail/speed, applied voltage) at once.

5. Conclusion

We have demonstrated a simple approach to prepare a gradient PNLC with large refractive index change, in which the LCs are first reoriented by voltages and then stabilized by the UV-induced polymer network. The spatial refractive index change of such a PNLC depends on the applied voltage along with the cell's movement during UV exposure. Based on this approach, various PNLCs can be prepared by using different photomasks. Due to uniform distribution and high density of polymer network, our PNLC shows good stability and fast response. To enlarge the refractive index change of PNLC, two simple methods can be considered: decreasing the cell's travel distance during UV exposure and increase the cell gap. Our PNLC with a large refractive index change has attractive applications in prism gratings, adaptive lenses, Fresnel zone lenses, and other adaptive photonic devices.

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