Reconfigurable liquid crystal droplets using a dielectric force

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Dielectric force-induced shape change of liquid crystal (LC) droplets which are randomly dispersed in a liquid polymer is demonstrated. By applying a sufficiently high voltage, the isolated spherical LC droplets are coalesced to form a planar layer, while the cylindrical LC droplets which make contact on both substrate surfaces cannot be deformed easily. On the contrary, each cylindrical LC droplet functions as a swollen well to unite the adjacent LC droplets. The dynamic behavior for flattening the spherical LC droplets is also studied. Potential applications of these reconfigurable LC droplets for various photonic devices are discussed. © 2009 American Institute of Physics.
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Polymer-dispersed liquid crystals (PDLCs) have been widely used as light shutters,[1,2] phase modulators,[3–6] and other photonic devices.[7–12] A PDLC is usually formed by polymerizing a LC/monomer mixture. After polymerization, LC is phase separated from polymer to form micron-sized droplets in the polymer matrix. In the voltage-off state, the randomly oriented LC droplets scatter light due to the refractive index mismatch between the LC droplets and the polymer matrix. As a result, the PDLC appears translucent. As the applied voltage increases, the LC molecules within the droplets are reoriented along the electric field direction so that their refractive index mismatch gradually vanishes and the cell becomes transparent. However, due to the solid polymer matrix the surface of the LC droplets cannot be changed by an external voltage.

Different from conventional (UV and thermal) phase separation methods, West *et al.*[13] proposed to use dielectric force to induce phase separation for preparing polymer walls. This method is attractive because the phase separation between LC and polymer is induced by a dielectric force; a UV light is used only to fix the formed structures. Based on similar idea, we demonstrated a variable phase separation structure using lateral electric fields.[14] In comparison to conventional methods, dielectric force is a useful approach to control or reconstruct the phase separation structure. However, very little work has been reported about the impact of dielectric force on the shape change of LC droplets after phase separation.

In this letter, we demonstrate that the LC droplet shape can be altered by a dielectric force even after thermal-induced phase separation. In experiment, we show that a spherical LC droplet can be flattened to form a thin layer by an electric field. An example using such a reconfigurable LC thin layer for phase modulation is illustrated.

When a spherical LC droplet is formed in a liquid monomer, it either suspends in the liquid monomer or makes contact to one substrate surface. Usually, the droplet contacting substrate surface is more stable due to the pinning effects of the surface anchoring. To keep the discussion general, Fig.

1(a) shows a LC droplet in a liquid monomer. In the voltage-off state, the LC droplet exhibits a spherical shape. At a voltage V, the LC droplet experiences an electric field as follows:

$$E_h \approx \frac{V/\varepsilon_{LC}}{\left(\dfrac{2h}{\varepsilon_{LC}} + \dfrac{d-2h}{\varepsilon_P} \right)}, \tag{1}$$

where ε_{LC} denotes the average dielectric constant of the LC, ε_P represents the dielectric constant of the liquid polymer, d is the cell gap, and h is the semiheight of the LC droplet along y-axis.

Equation (1) is valid only in the case with a LC droplet covered by liquid monomer. On the contact border, there is a sudden dielectric constant change. From this equation, the electric field can be reduced to $E_b \sim V\varepsilon_P/d\varepsilon_{LC}$ at droplet border $h \sim 0$ (in the LC side), since ε_{LC} is much larger than ε_P, near the border the electric field is the weakest. Because the surface of the LC droplet changes continuously, the electric field has a gradient distribution. At the droplet border $h \rightarrow 0$ (in the polymer side), ε_{LC} should be substituted by ε_P, and the electric field is reduced to $E = V/d$. From Eq. (1), only the LC at the contact border ($h=0$) experiences the highest electric field gradient.

Due to the gradient electric field, the LC droplet is subject to a dielectric force[13,15]

$$\vec{F} = \tfrac{1}{2}\varepsilon_0(\varepsilon_{LC} - \varepsilon_P) \nabla (\vec{E} \bullet \vec{E}), \tag{2}$$

where ε_0 represents the permittivity of free space and E denotes the electric field on the curved droplet surface. Since the dielectric force impacting on the LC droplet border is the
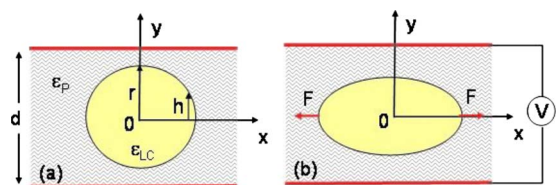

FIG. 1. (Color online) A LC droplet dispersed in a liquid polymer: (a) spherical shape at $V=0$ and (b) elongated shape induced by dielectric force along axial direction.

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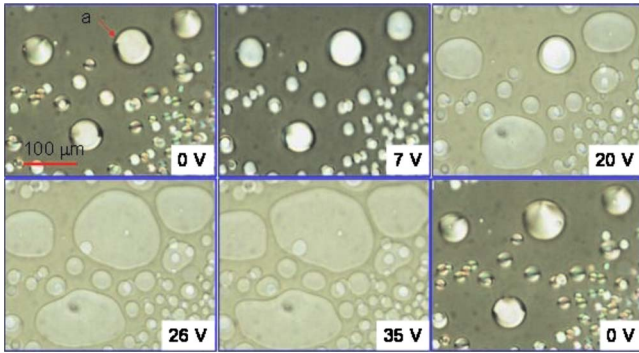b)Electronic mail: swu@mail.ucf.edu.

FIG. 2. (Color online) Phase separation morphologies of LC/monomer mixture at $V=0$ and the impact of different voltages on the shape change of LC droplets.

FIG. 3. (Color online) A large LC droplet surrounded by small droplets at $V=0$ (left), 50 V_{rms} (middle), and 70 V_{rms} (right).

strongest and LC has a larger dielectric constant, the LC droplet has tendency to spread out to the strong electric field region in axial direction, as shown in Fig. 1(b).

Usually there are many droplets dispersed in liquid polymer. When the applied voltage is sufficiently high, the adjacent LC droplets may touch each other due to the shape deformation and then grow into a larger droplet. If such a large droplet makes contact on one substrate surface, then the LC droplet position will be fixed due to the pinning effect from substrate surface. By applying a high voltage, this LC droplet will be flattened to form a thin layer.

In experiment, to disperse LC droplets in a liquid polymer we first filled an empty cell with a mixture consisting of 35 wt % nematic LC BL-038 ($\Delta\varepsilon=16.4$, $\varepsilon_{\parallel}=21.7$) and 65 wt % NOA65 (Norland Optical Adhesive, $\varepsilon_p \sim 5$ at 1 kHz). The cell gap was measured to be ~ 15 μm. Both ITO electrode surfaces were over-coated with a thin (~ 80 nm) polyimide layer and rubbed in one direction in order to increase surface anchoring. The mixture was capillary-filled into the cell in an isotropic state and then gradually cooled to room temperature (~ 21 °C). Because the LC and liquid polymer were almost immiscible, phase separation was observed at room temperature.

To study the impact of dielectric force on the LC droplet, we used an optical microscope to observe the phase separation morphologies. The observed results were recorded by a digital camera mounted on top of the microscope. The cell was placed on a microscope stage and sandwiched between two crossed polarizers. Figure 2 shows the phase separation morphologies of the LC cell at different voltages. At $V=0$, the droplets are observed clearly due to the LC birefringence. The LC droplets were randomly dispersed in the liquid monomer and their sizes were not uniform. The biggest droplet was formed after coalescing with several adjacent droplets.

As the applied voltage increases gradually, the LC directors in the droplets are reoriented along the electric field direction. At $V=7$ V_{rms} all the droplets respond to the electric field but there is no evidence about the surface profile change of each droplet. As the applied voltage continues to increase, the surface profiles of some small droplets coalesce first. At 20 V_{rms}, some large droplets are formed. Some small LC droplets also coalesce but remain relatively small. At 26 V_{rms}, some LC droplets grow significantly like a pie. At $V=35$ V_{rms}, the adjacent large LC droplets are almost connected together to form a layer. Our results indicate that
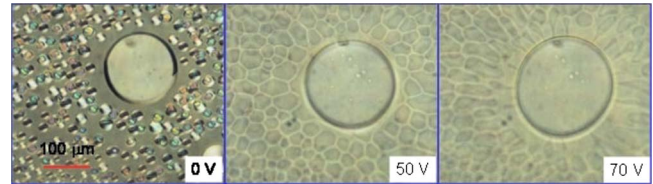
the large droplets make contact only to one substrate surface. By removing the voltage, these deformed LC droplets relax to their original spherical shapes quickly, as Fig. 2 shows.

In comparison to the spherical droplets at $V=0$, the surface area of a large LC droplet, say droplet-**a** as marked in Fig. 2, is expanded by $\sim$6x at $V=35$ V_{rms}. Further expansion is possible just by increasing the voltage. To confirm the laminated droplets at 35 V_{rms} are in planar state, a simple method is to observe its focusing effect. Our result (not given) shows that at $V=0$ the droplet-**a** indeed exhibits focusing effect due to its curved surface. However, at 35 V_{rms} the reconfigured LC droplet completely loses its focusing property. This implies that the surface of the LC droplet at 35 V_{rms} is approximately planar.

As a LC droplet keeps on growing, it will eventually touch both substrate surfaces and form a cylindrical domain. Two ways may form a cylindrical domain as follows: one is the natural coalescence of neighboring LC droplets, and the other is induced by the external voltage. If many small LC droplets are flattened by an applied voltage, the droplets can connect together and form a large flattened droplet. By removing the applied voltage, the large LC droplet may restore to its relaxing state due to the surface tension. If the droplet touches both substrates, then it becomes a cylindrical shape. According to Eq. (1), there is almost no gradient electric field generated on the droplet in vertical direction, and the shape of the droplet will not be deformed by the external voltage. A big LC droplet was found as shown in Fig. 3. At $V=0$, the diameter of the large LC droplet was about 140 μm. It was surrounded by many small LC droplets. To analyze the droplet shape, we simply applied a voltage to the LC cell. By applying a voltage, some small droplets were coalesced and spread out in axial directions, while the large droplet did not respond to the voltage. As the coalesced droplets touched the large droplet, they were swallowed by the large droplet. This is due to the action of surface tension. Therefore, the volume of the large droplet is increased gradually, as shown at 50 V_{rms}. As the voltage was increased to 70 V_{rms}, more adjacent LC droplets were merged and the volume of the LC droplet was expanded significantly. Such a result implies that the large LC droplet exhibits a cylindrical shape. Because the cylindrical LC domain cannot be reconfigured by a voltage, it is less useful for practical applications. To avoid forming cylindrical LC droplets, one can increase the cell gap and control the LC concentration in the liquid polymer, as well as the phase separation condition.

When the applied voltage is high enough, every LC droplet can be merged to a very thin layer except the cylindrical type. To briefly explain the reason, we choose a big LC droplet (diameter ~ 55 μm) for analysis, as shown in the left side of Fig. 4(a), and its geometrical structure is depicted in the right side. The droplet contacts the bottom substrate surface and LC molecules present random orientation inside the
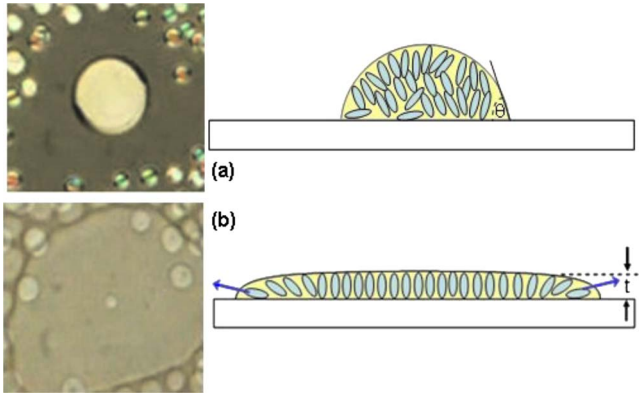
FIG. 4. (Color online) A LC droplet contacting on one substrate surface and its side-view structure: (a) relaxing state, and (b) flattening with a voltage.

droplet. This droplet has a contact angle on the substrate surface. When a high voltage is applied to the LC droplet, the droplet is coalesced noticeably, as shown in the left side of Fig. 4(b). Correspondingly, its side view structure is shown in the right side of Fig. 4(b). At a high voltage, most LC molecules are reoriented along the electric field direction. Usually the contact angle of the LC droplet cannot be suppressed totally due to the surface tension, even though the central area of the droplet could be flat. Because the electric field direction on the border surface deviates largely from the vertical direction, thus LC molecules at the border area will not be reoriented in vertical direction. Therefore, the average dielectric constant of the LC at the border is much larger than that of the liquid polymer. From Eq. (2), LC molecules near the border always experience a net dielectric force. This force causes the LC droplet to spread out until it is balanced by the surface tension, as shown in Fig. 4(b). The thickness of the laminated LC droplet in its central area is t.

From Fig. 4(b), when we vary the applied voltage while keeping the central area flat, the thickness of the central area will change accordingly. This film can be used as a phase modulator with a phase shift $\delta = 2\pi \Delta t (n_{LC} - n_P)/\lambda$, where n_{LC} and n_P represent the refractive indices of LC and polymer, respectively, Δt is the thickness change, and λ is the wavelength of the incident light. At a high voltage, n_{LC} can be approximated by the ordinary refractive index of the LC. To increase the phase shift, one can increase either Δt or $n_{LC} - n_P$ or both.

Response time is an important parameter for reconfiguring LC droplets. To estimate the response time, we recorded the transient LC droplet shape change through a digital camera attached to an optical microscope. When a gated square wave of 35 V_{rms} pulse amplitude was applied to the lens cell, for all LC droplets the coalescing time was ~200 ms, while the relaxing time was between ~100 and 500 ms, depending on the droplet size. A larger droplet needs a longer time to restore its spherical state.

From Fig. 4, the diameter of the large droplet can be scaled up to millimeter level while still maintaining a spherical-like shape in relaxing state. When it is reconfigured, a significant shape change would occur. For a larger LC droplet, the cell gap is thicker so that the required operating voltage is higher. Due to the pinning effect of the substrate surface, the reconfigured LC droplets can relax to their original states. Different from the devices in which liquid droplets are actuated from relaxing to contracting state,[16,17] our LC droplets are driven from relaxing to merging state. If the employed liquid polymer is photopolymerizable, then a thin LC/polymer composite film can be fabricated by UV curing when the LC droplets are flattened to a thin layer. This technique is simpler than the anisotropic polymerization method[18] for making LC/polymer composite films. The LC droplets with reconfigurable shape have potential applications in photonic devices, such as phase modulator, tunable filter, electronic lens, and optical attenuators.

In conclusion, we have studied the impact of dielectric force on the shape change of LC droplets. In our LC cell, the LC droplets themselves have the ability to convert a homogeneous electric field to an inhomogeneous one. Owing to the inhomogeneous electric field, the generated dielectric force can laminate the LC droplets into a thin layer. By removing the dielectric force, the deformed LC droplets can be restored to their original shape. This is due to the pinning effect of the substrate surface and the interfacial surface tension between droplets and liquid monomer. Potential applications of these devices are foreseeable in optical communications, tunable filter, electronic lens, and phase modulators.

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