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Determination of the flexoelectric coefficient (e_1–e_3) in nematic liquid crystal by using fully leaky optical-guided mode

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Fully leaky optical-guided mode was employed to determine the difference in the splay and bend flexoelectric coefficient (e_1–e_3) in negative nematic liquid crystal MS-N01300-000. The experimental curves of reflectivity versus internal angle (angle of incident light to the liquid crystal) were obtained when a laser beam passed through the hybrid-aligned nematic in-plane switching liquid crystal cell; the cell was embedded in pyramid-coupled waveguide with different alternating current (AC) and direct current (DC) voltages. The curves of the applied DC with voltage similar to that of AC shift to the left or the right. Experimental results were then compared with theoretical results derived from elastic continuum theory and multi-layer optical theory of liquid crystals. The approximate value of the flexoelectric coefficient (e_1–e_3) of MS-N01300-000 is 9.0×10^{-11} C/m. [http://dx.doi.org/10.1063/1.4942050]

I. INTRODUCTION

A strain-free nematic is invariant with respect to the inversion of its director $\boldsymbol{n}$ to $-\boldsymbol{n}$. The curvature deformations of the splay and bend types can break the symmetry and lead to a non-vanishing local polarization in liquid crystal (LC) systems.[1] This phenomenon involves linear physical coupling between mechanical deformation and electric polarization and is analogous to distortion-induced polarization of piezoelectricity in certain classes of crystal; this phenomenon is known as the flexoelectric effect or flexoelectricity.[2,3] Flexoelectricity has attracted much research interest since it was discovered by Meyer in 1969.[4] Following Meyer's sign convention, the flexoelectric polarization $\boldsymbol{P}$ induced by a nematic director field is given as follows:

$$\boldsymbol{P} = e_1 (\nabla \cdot \boldsymbol{n})\,\boldsymbol{n} + e_3 (\nabla \times \boldsymbol{n}) \times \boldsymbol{n}, \tag{1}$$

where e_1 and e_3 are the flexoelectric coefficients corresponding to splay and bend deformations, respectively. The magnitude of the flexoelectric coefficient has been experimentally found to be within the order of 10^{-11} C/m in many rod-like LC molecules.[5] However, Harden *et al.*[6] proposed that the flexoelectric coefficient in bent-core nematics is 10^3 times higher than that in rod-like systems; the discrepancy in the results provides another reason for examining the flexoelectric coefficient.[7–13] Various experimental techniques have been developed to measure flexoelectric coefficients;[14–22] most of these techniques use the hybrid-aligned nematic (HAN) cell because the LC

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material filled in HAN cell contains splay and bend distortions in the initial state and thus present evident flexoelectric properties.[5,16–22]

The flexoelectric effect induces changes in the LC director under an external applied voltage and is the primary cause of flexoelectricity in the LC layer. Director deformation changes the optical guided-wave mode when a laser beam passes through the LC waveguide. The accurate measurement of LC-guided mode can determine the flexoeletric coefficient. Fully leaky guided mode is an electro-optic technique that can easily provide sufficient information compared with other similar techniques.[23,24] This method was used to measure director profiles in various types of LC cells. Fully leaky guided mode uses a thin LC layer, called the LC waveguide, between two glass plates with refractive indices lower than the principal indices of the LC. Controlling linearly polarized light (p or s light) with different incident angles leads to different optical field distributions and optical waveguide modes. The sets of the signals of the polarization conservation (pp or ss, which refers to the input polarization and the output, respectively) and polarization conversion (ps or sp) components of both reflectivity (R) and transmittance (T) as functions of internal angle can be detected. These signals are sensitive to the director of LC. Several material parameters of LC can be determined, such as the flexoelectric coefficient, by comparing experimental and theoretical results derived from elastic continuum theory and multi-layer optical theory of LC.

In this paper, we measured the difference in the splay and bend flexoelectric coefficient (e_1–e_3) in negative nematic LC MS-N01300-000 by using fully leaky optical-guided mode. We introduced the theory for the theoretical calculation of the guided mode of LC waveguide in section II. Experiment installation set-up and process, as well as the parameters of negative nematic MS-N01300-000 and hybrid-aligned nematic in-plane switching (HAN-IPS) cell, were provided in section III. The sets of experimental and theoretical curves of R as a function of the internal angle were compared in section IV. The approximate value of the flexoelectric coefficient (e_1–e_3) of MS-N01300-000 is 9.0×10^{-11} C/m. Conclusion were given in section V.

II. THEORY

Nematic LC confined in the HAN-IPS cell is considered. LC in the cell is homogeneously aligned on one surface and homeotropical on the other surface. The anchoring at the boundaries is assumed to be strong. The rubbing direction of the two substrates is set along 45° from the x-axis. In the Cartesian coordinate, the director $\boldsymbol{n}$ can be written as $\boldsymbol{n} = (\cos\theta\cos\varphi, \cos\theta\sin\varphi, \sin\theta)$, where θ is the tilt angle of the director measured from the substrate, and φ is the azimuthal angle of the director from the x-axis. Angles θ and φ are the functions of the position z (perpendicular to the substrate), that is, $\theta = \theta(\mathrm{z})$ and $\varphi = \varphi(\mathrm{z})$. U is the external applied voltage, L is the electrode-separated gap, and d is the cell thickness. Given that $L >> d$, only the horizontal component of electric field is considered. Hence, the structure of the HAN-IPS cell, where an electric field is applied along the y-axis, is shown in Fig. 1. The vector of the electric field $\boldsymbol{E}$ can be written as $E = (0, U/L, 0)$.

The elastic continuum theory of LC states that the total free energy density of the system is the sum of elastic, dielectric, and flexoelectric contributions, as follows:

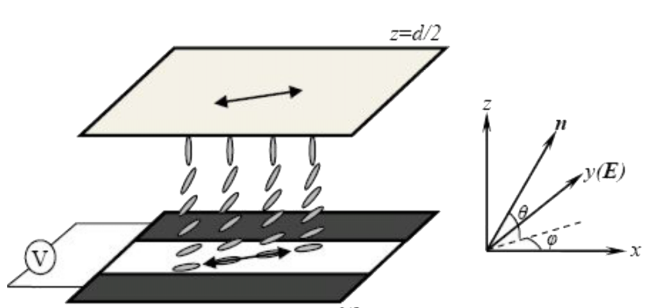

FIG. 1. Structure of HAN-IPS cell and the coordinate system.

$$f = f_{\rm el} + f_{\rm d} + f_{\rm p} = \frac{1}{2}\left(K_{11}\cos^2\theta + K_{22}\sin^2\theta\right)\left(\frac{d\theta}{dz}\right)^2 + \frac{1}{2}\left(K_{22}\cos^2\theta + K_{33}\sin^2\theta\right)\cos^2\theta\left(\frac{d\varphi}{dz}\right)^2$$
$$-\frac{1}{2}\varepsilon_0(\varepsilon_\perp + \Delta\varepsilon\cos^2\theta\sin^2\varphi)\left(\frac{U}{L}\right)^2 - \left(e_1\cos^2\theta - e_3\sin^2\theta\right)\sin\varphi\frac{U}{L}\frac{d\theta}{dz} - e_3\sin\theta\cos\theta\cos\varphi\frac{U}{L}\frac{d\varphi}{dz} \quad (2)$$

where K_{11}, K_{22}, and K_{33} are the splay, twist, and bend elastic constants, respectively; ε_0 is the vacuum dielectric constant; $\Delta\varepsilon = \varepsilon_{//}–\varepsilon_\perp$ is the dielectric anisotropy; and $\varepsilon_{//}$ and $\varepsilon_\perp$ are the extraordinary and ordinary dielectric constants of LC, respectively.

For a given applied voltage, the equilibrium configuration of LC is such that the total free energy has a minimal value subject to the following boundary conditions: $\theta(-d/2) = 0$, $\theta(d/2) = \pi/2$, $\varphi(-d/2) = \varpi/4$, and $\varphi(d/2) = \pi/4$. The equilibrium equations for θ and φ are written as follows:

$$\left(K_{11}\cos^2\theta + K_{33}\sin^2\theta\right)\frac{d^2\theta}{dz^2} - (K_{11} - K_{33})\sin\theta\cos\theta\left(\frac{d\theta}{dz}\right)^2 + \left(2K_{22}\cos^2\theta + K_{33}\sin^2\theta - K_{33}\cos^2\theta\right)$$
$$\sin\theta\cos\theta\left(\frac{d\varphi}{dz}\right)^2 - \varepsilon_0\Delta\varepsilon\sin\theta\cos\theta\sin^2\varphi\left(\frac{U}{L}\right)^2 - (e_1 - e_3)\cos^2\theta\cos\varphi\frac{U}{L}\frac{d\varphi}{dz} = 0, \quad (3)$$

and

$$\left(K_{22}\cos^4\theta + K_{33}\sin^2\theta\cos^2\theta\right)\frac{d^2\varphi}{dz^2} - 2\left(2K_{22}\cos^2\theta + K_{33}\sin^2\theta - K_{33}\cos^2\theta\right)\sin\theta\cos\theta\frac{d\theta}{dz}\frac{d\varphi}{dz}$$
$$-\varepsilon_0\Delta\varepsilon\cos^2\theta\ \sin\varphi\cos\varphi\left(\frac{U}{L}\right)^2 - (e_1 - e_3)\cos^2\theta\cos\varphi\frac{U}{L}\frac{d\varphi}{dz} = 0. \quad (4)$$

The director distributions in different conditions are determined by solving equilibrium equations through difference iterative numerical method.[11,14] The curves of the polarization conservation (pp, ss) and polarization conversion (ps, sp) components of R and T as functions of internal angles can be simulated using multi-layer optical theory based on director distributions.[25–28]

III. EXPERIMENT

The HAN-IPS cell was filled with negative nematic LC material MS-N01300-000. The fully leaky optical-guided mode technique utilizes the LC layer as the guiding layer. A discrete set of guided modes was excited in the LC layer, and the guided modes depend on the director distribution of the LC.[23,24] The geometric structure of the pyramid-coupled waveguide is shown in Fig. 2. Two low-refractive index pyramids ($n = 1.52$) with matching fluid ($n = 1.52$) were used to couple light into and out of the LC cell. The cell consists of two standard glass substrates with transparent indium-tin-oxide (ITO) coatings and alignment layers polyimide (PI). The matching fluid (Cargille Labs, USA) allows the cell to be rotated with respect to the pyramid. The internal angle $\theta_{\rm p}$ was varied in the experiment, and the coupling to the waveguide was monitored. The observed features

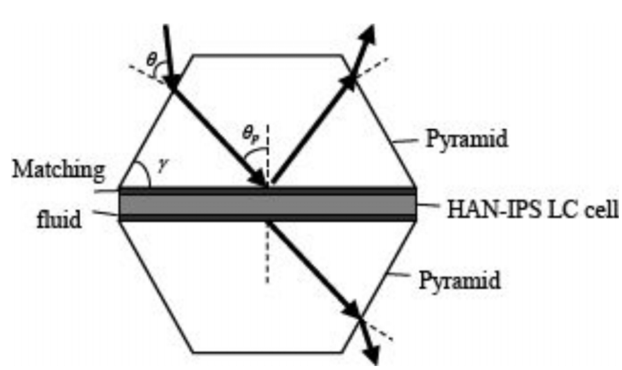

FIG. 2. Geometric structure of pyramid-coupled waveguide.

TABLE I. Parameters of negative nematic LC MS-N01300-000 and HAN-IPS cell.

MS-N01300-000	HAN-IPS cell	
$K_{11} = 24.1$ pN	Thickness of cell	$d = 3.685 \pm 0.005$ µm
$K_{22} = 9.81$ pN	Electrode-separated gap	$L = 15.0$ µm
$K_{33} = 17.0$ pN	ITO	$n_o = n_e = 1.897$
$\varepsilon_{//} = 3.27$		$d = 28.8 \pm 0.2$ nm
$\varepsilon_{\perp} = 5.66$	PI of the lower substrate	$n_o = n_e = 1.432$
$\Delta\varepsilon = -2.39$		$d = 65 \pm 0.2$ nm
$n_o = 1.494 \pm 0.005$	PI of the upper substrate	$n_o = 1.637$, $n_e = 1.486$
$n_e = 1.590 \pm 0.005$		$d = 65 \pm 0.2$ nm

that correspond to the resonant mode coupling reflect the mode structure. According to Snell's law, the relation of the internal angle θ_p and the external angle θ can be written as follows:[25]

$$\theta = \sin^{-1}\left[\frac{n_p}{n_0}\sin\left(\frac{\pi}{2} - \theta_p - \frac{\gamma}{2}\right)\right], \tag{5}$$

where γ is the apex angle of the symmetric pyramid; and n_p and n_0 are the refractive indices of the pyramid and air, respectively. The following values were considered in the experiment: $\gamma = 60°$, $n_p = 1.52$, and $n_0 = 1$.

The experimental HAN-IPS cell was obtained from Hebei Jiya Electronics Co. Ltd., and the negative nematic LC MS-N01300-000 was provided by Hebei Milestone Electronic Material Co. Ltd. This kind of LC material is a kind of mixture composed of 12 kinds of LC monomer and has high contrast and better transmittance. Under normal circumstances, it is in the uniform nematic phase and mainly applied in vertical alignment (VA) mode to fabricate the car LCD monitor. The parameters of LC material MS-N01300-000 and HAN-IPS cell are shown in Table I, where n_o and n_e are the ordinary and extraordinary indices, respectively.

The reflected signal was used to measure the flexoelectric coefficient (e_1–e_3) in the experiment, and the experiment installation set-up is shown in Fig. 3. The beam source is He-Ne laser ($\lambda = 632.8$), and the mechanical chopper modulates the laser beam at 18.6 kHz to allow phase-sensitive detection. The adjustable attenuator modulates the intensity of incident light. Polarizer 1 and the 1/4 wave plate were used to obtain circularly polarized light. Polarizers 2 and 3 were adjusted to choose p- or s-polarized state of the incident and reflected lights, respectively. The glass plate reflects ~2% of the incident light into detector 1, which was used to compensate for drift in source intensity. The fully leaky LC waveguide was placed on the θ–2θ rotation table controlled by a computer. The signal of the reflective light was detected by detector 2.

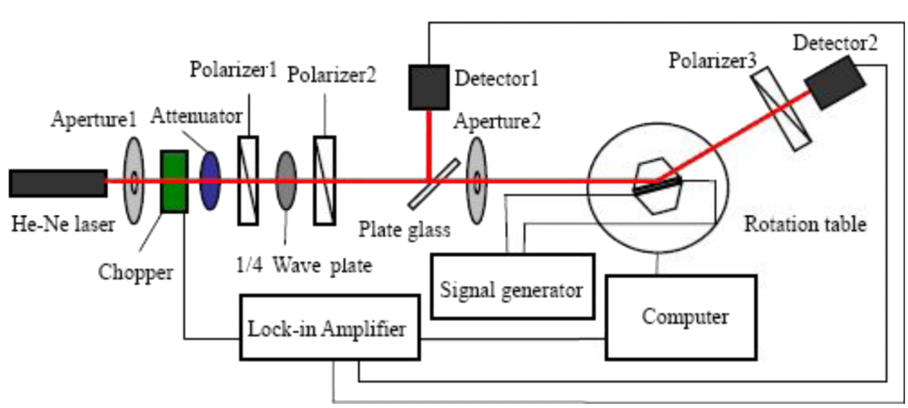

FIG. 3. Schematic of the experiment installation.

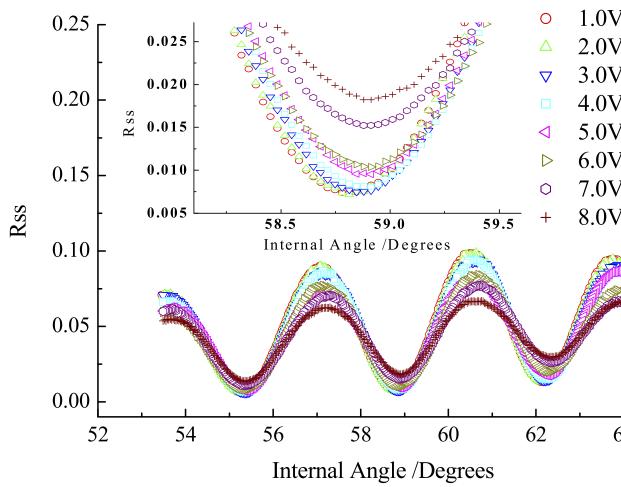

FIG. 4. Experimental curves of Rss versus internal angle with different 1kHz AC voltages from 1.0 V to 8.0 V.

IV. RESULTS AND ANALYSIS

The sets of experimental signals, namely, Rpp, Rss, Rsp, and Rps, with different AC voltages were tested to obtain the parameters of the LC material and cell. The frequency of AC voltages used in this experiment was 1kHz. Experimental results show that the signals of Rps and Rsp can be easily disturbed because they are very small. The curve of Rss is smoother than that of Rpp. Thus, the signal of Rss was used to determine the difference in the splay and bend flexoelectric coefficient (e_1–e_3). The experimental curves of Rss as functions of internal angle with different AC voltages from 1.0 V to 8.0 V are shown in Fig. 4. The curves of Rss shift rightward with increasing applied AC voltage.

Typical voltage of 3.0 V was used to determine flexoelectric coefficients (e_1–e_3) because the Rss signals for 0.0, 1.0, and 2.0 V AC voltages almost overlapped. The corresponding profiles of the tilt angle θ and the azimuthal angle φ with different flexoelectric coefficients (e_1–e_3) are shown in Figs. 5 and 6. The director profiles differ with distinct values of the flexoelectric coefficient (e_1–e_3). Therefore, the flexoelectric coefficients (e_1–e_3) has an important influence on LC director profiles, especially the profiles of the azimuthal angle φ, in the strong anchoring HAN-IPS cell under applied external voltages.

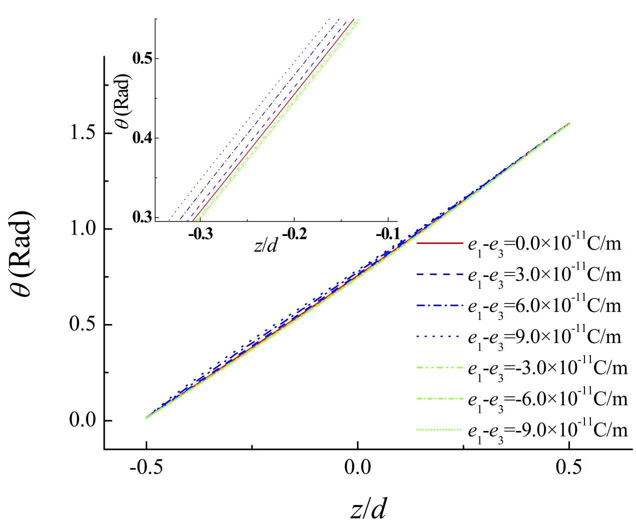

FIG. 5. Profiles of the tilt angle θ with different flexoelectric coefficients (e_1–e_3).

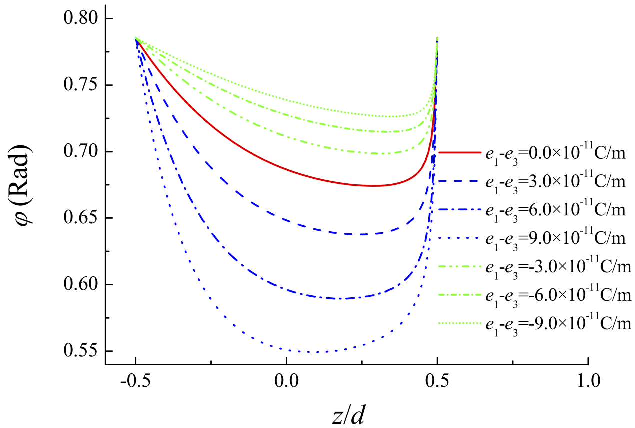

FIG. 6. Profiles of the azimuthal angle φ with different flexoelectric coefficients (e_1–e_3).

The experiment results of the reflectivity Rss as a function of the internal angle in the strong anchoring HAN-IPS cell under external applied 1kHz AC and DC voltages with similar values of 3.0 V are shown in Fig. 7. When DC voltage was applied to the HAN-IPS cell, the anode and the cathode could be reversed, denoted by DC and –DC, respectively. A shift to the left or the right was observed in the curves in which DC relative to the applied AC in the experimental results. These results are the same as the theoretical results simulated based on elastic continuum theory and multi-layer optical theory of LC. The approximate value of difference in the splay and bend flexoelectric coefficient (e_1–e_3) of MS-N01300-000 is 9.0×10^{-11} C/m, which was obtained by comparing the theoretical results with experimental data (Fig. 8).

The value of coefficient (e_1–e_3) of MS-N01300-000 is rather high as compare to other materials reported by S. Kaur *et al.*[29] Since LC MS-N01300-000 is uniform nematic phase, the flexoelectric coefficients should be satisfied with the fundamental limit to the conventional order of magnitude from the conservation of energy considerations.[30,31] Using inequalities (5) in Ref. 30 and the LC material parameters in Table I, the absolute value of splay and bend flexoelectric coefficient are $|e_1| = 2.641 \times 10^{-11}$ C/m and $|e_3| = 2.918 \times 10^{-11}$ C/m, respectively. Considering the sign of two coefficients, the value range of (e_1–e_3) is from -5.559×10^{-11} C/m to 5.559×10^{-11} C/m. The experiment value of coefficient (e_1–e_3) is clearly not within that limit proposed by Castles F *et al.* The possible reason is that LC MS-N01300-000 is a mixture, rather than it becomes unstable to the formation of a modulated phase composed of either a twist-bend or splay-bend phase.[32–35]

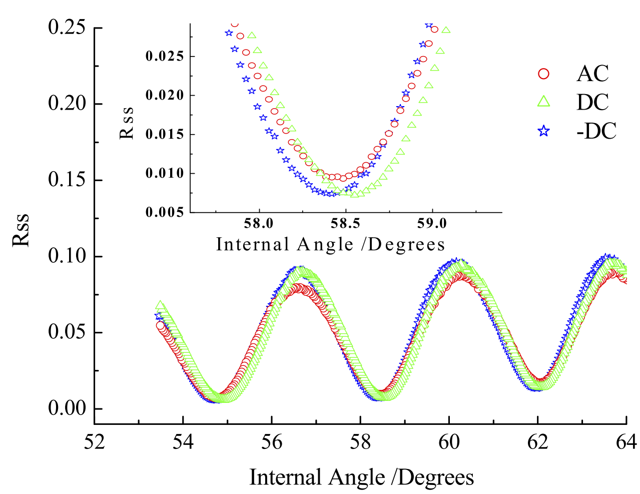

FIG. 7. Experimental curves of Rss versus internal angle for 1kHz AC and DC voltages of 3.0 V.

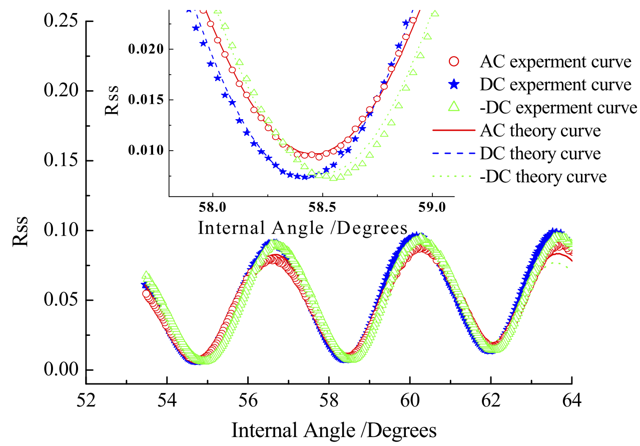

FIG. 8. Experimental curves of Rss versus internal angle for 1kHz AC and DC voltages of 3.0 V and the fitted theoretical ones for $(e_1 - e_3) = 9.0 \times 10^{-11}$ C/m.

V. CONCLUSION

We explored the flexoelectric coefficient (e_1–e_3) in negative nematic LC MS-N01300-000 by using fully leaky optical-guided mode. The curves of R as a function of the internal angle in the strong anchoring HAN-IPS LC cell under similar values of external applied AC and DC voltages were tested. The theoretical curves were calculated using elastic theory and multi-layer optical theory of LC. The approximate value of the flexoelectric coefficient (e_1–e_3) of MS-N01300-000 is 9.0×10^{-11} C/m, which was obtained by comparing the experimental data with numerically simulated results. Although LC MS-N01300-000 is uniform nematic phase, not other twist-bend or splay-bend phase, the value of flexoelectric coefficient (e_1–e_3) is not within the theoretical limit and rather high as compare to that reported for many other LC materials. We think that the blending of 12 kinds of LC monomer is the main reason to increase the flexoelectric coefficient. The fully leaky optical-guided mode technique can also be used to the measure flexoelectric coefficient (e_1+e_3) of this material, as well as the (e_1–e_3) and (e_1+e_3) coefficients of other materials.

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