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Thermodielectric Bistability in Dual Frequency Nematic Liquid Crystal

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We report on a thermodielectric bistability in dual frequency nematic liquid crystals (LCs) caused by the anisotropic nature of dielectric heating and director reorientation in an electric field. The bistability is a result of the positive feedback loop: director reorientation → anisotropic dielectric heating → dielectric anisotropy → director reorientation. We demonstrate both experimentally and theoretically that two states with different temperature and director orientation, namely, a cold planar state and a hot homeotropic state coexist in a LC cell for a certain frequency and amplitude range of the applied voltage.

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The phenomenon that two stable states of a system coexist under the same external conditions is known as bistability. Bistability is found in atomic systems [1], gases [2], semiconductors [3], liquid crystals [4–8], biological cells [9], etc. Bistabilities in liquid crystals (LCs) are of special interest due to their applications in LC displays [5,7], spatial light modulators [4], optical information storage [6], etc. The bistabilities in LCs are caused by a balance of two different agents, such as an electric field \( E \) that orients the LC director \( \hat{n} \) in the bulk, and surface anchoring that keeps the director aligned at the bounding substrates. In this work, we demonstrate a novel effect, in which the bistability originates from the electric field alone, through the balance of two different mechanisms of anisotropic coupling between \( E \) and LC, namely, director reorientation and anisotropic dielectric heating.

When an electric field \( E \) is applied to a LC, it exerts a torque on the director \( \hat{n} \) that depends on dielectric anisotropy \( \Delta \varepsilon = \text{Re}(\varepsilon_{||} - \varepsilon_{\perp}) \), where \( \varepsilon_{||} \) and \( \varepsilon_{\perp} \) are the dielectric permittivity components corresponding to \( \hat{n} \parallel E \) and \( \hat{n} \perp E \), respectively. For LCs with \( \Delta \varepsilon > 0 \), \( \hat{n} \) reorients parallel to \( E \); if \( \Delta \varepsilon < 0 \), then \( \hat{n} \perp E \). In the so-called dual frequency nematic (DFN) LC, \( \Delta \varepsilon \) changes sign from \( \Delta \varepsilon > 0 \) at \( f < f_c \), to \( \Delta \varepsilon < 0 \) at \( f > f_c \), as the function of the applied field frequency \( f \). The crossover frequency \( f_c \) corresponds to the dispersion region of the parallel component of dielectric permittivity \( \varepsilon_{||} \); \( f_c \) is a strong monotonically increasing function of temperature. In addition to the dielectric reorienting torque [10], the electric field causes heating of the LC, especially in the dispersion region. As the result, the heating is essentially an anisotropic effect: the temperature increase is maximum when \( \hat{n} \parallel E \) and minimum when \( \hat{n} \perp E \). The two mechanisms of coupling between \( E \) and LC thus set up a possibility of a bistability of two states that can coexist at a given \( E \): a cold planar (CP) state and a hot homeotropic (HH) state. The idea can be explained by the following gedanken experiment. One starts an experiment with a DFN sandwiched between two flat electrodes. A modest electric voltage \( U \) is applied at \( f > f_c \) (so that \( \Delta \varepsilon < 0 \)) to stabilize the CP state, \( \hat{n} \perp E \). An increasing \( U \) would increase the LC temperature \( T \), up to the point \( T_c \) where the inversion frequency \( f_c(T) \) exceeds \( f \) and \( \Delta \varepsilon \) becomes positive. With \( \Delta \varepsilon > 0 \), the director reorients into the HH state, \( \hat{n} \parallel E \). The voltage \( U_{CP-HH} \) of the CP → HH transition is relatively high because in the CP state the dielectric heating is minimum. In the HH state, however, the dielectric heating is more efficient than in the CP state. The threshold \( U_{HH-CP} \) of the reverse HH → CP transition, caused by decreasing \( U \) to reach \( T_c \) from above in the HH state, should be lower than \( U_{CP-HH} \). Therefore, because of the anisotropic nature of both dielectric heating and dielectric reorientation of the director, one would expect a bistability, i.e., a coexistence of the CP and HH states in the certain range of voltages, \( U_{HH-CP} < U < U_{CP-HH} \). Below, we first demonstrate the phenomenon experimentally and then describe it with a quantitative model that, in addition to the consideration of the bulk dielectric reorientation and heating, takes into account the surface anchoring effects at the bounding plates (which hinders the occurrence of bistability rather than causes it, as in other known cases [4–8]).

We used a DFN mixture MLC2048 (EM Industries). The real \( (\varepsilon_{||}, \varepsilon_{\perp}) \) and imaginary \( (\varepsilon_{||}^*, \varepsilon_{\perp}^*) \) components of the dielectric permittivity tensor of MLC2048 were measured using the Impedance/Gain-Phase analyzer SI-1260 (Schlumberger Inc.) in the range \( f = (1–500) \) kHz at \( T_0 = 24.0^\circ \text{C} \), Fig. 1. The sign reversal of \( \Delta \varepsilon \) is caused by the dispersion of \( \varepsilon_{||} \) and the maximum value of \( \varepsilon_{||}^* \) is reached at \( f_c \).

We filled MLC2048 into a cell of a thickness \( d = 11 \mu \text{m} \). The indium tin oxide electrodes at the inner surfaces of the cell’s bounding glass plates were coated with a polyimide layer SE-7511 (Nissan Chemicals) to set \( \hat{n} \) perpendicular to the bounding plates; this homeotropic alignment allows us to identify unambiguously the HH state as a “black” state in observations with crossed polarizers. The cell was placed inside a LTS-350 hotstage (Linkam) permitting temperature stabilization at \( T_0 = 24.00 \pm 0.01^\circ \text{C} \). To improve the uniformity of the thermal
environment around the cell, we created an air gap between the cell and the hot stage, by separating them with two wood rods of 0.5 mm diameter. We measured the temperature $T_g$, at the outer surface of the glass plate bounding the LC, by an attached thermocouple. According to our previous studies [11], (a) the heat flux $Q_{\text{out}}$ at the interface between the cell and the surrounding medium obeys the Newton’s cooling law; i.e., $Q_{\text{out}} = k(T_g - T_0)$, where $k$ is the heat transfer coefficient of the glass-air interface, and (b) the temperature gradient across the LC layer is small, less than $0.01^\circ$C. Thus the average temperature $T$ of the LC layer at the thermal equilibrium is related to $T_g$ as [12],

$$T - T_0 = (1 + \text{Bi})(T_g - T_0),$$

where $\text{Bi} = kL/G$ is the Biot number, $L = 1.1$ mm and $G = 1$ W m$^{-1}$ K$^{-1}$ are the thickness and thermal conductivity [13] of the glass plate, respectively. With the typical $k = 10\text{–}50$ W m$^{-1}$ K$^{-1}$ [14], $\text{Bi}$ is very small, 0.01–0.05, and thus, according to Eq. (1), $T_g$ yields a good quantitative measure of $T$.

We applied a harmonic voltage $\ddot{U} = \sqrt{2}U \cos2\pi ft$ with varying rms voltage $U$. We have chosen $f = 20$ kHz, because $T_c$ for this frequency is $26^\circ$C, i.e., slightly larger than $T_0$, Fig. 2. The rms voltage was changed in steps of 0.2 V with 400 s equilibration time; the latter is enough to achieve an equilibrium temperature within 0.01$^\circ$C [11], the accuracy of the temperature control. The state of the cell placed between two crossed polarizers of a microscope was monitored through the textural (Fig. 3) and light transmittance changes (Fig. 4); the polarizers were aligned to maximize the transmittance in the CP state.

The initial homeotropic state transforms into a state with $\hat{n}$ tilted away from $E$ when the applied voltage exceeds the Fredericks threshold $U^0_F = \pi(K_{33}/\varepsilon_0\Delta\varepsilon(T_0))^{1/2} = 6$ V; this state is an analog of the CP state in the gedanken experiment, the only difference is that $\hat{n}$ is not strictly parallel to the bounding plates (because of the surface anchoring at the bounding plates). We will continue to label this “cold tilted” state as the CP state. The CP state appears as a bright texture under the crossed polarizers (insets 1 and 2 in Fig. 3); light transmission is high, Fig. 4. Small increments of voltage lead to small increments of the
measured temperature \( T_e \). However, at some voltage \( U_{CP-HH} \), the CP structure transforms abruptly into a dark HH texture (inset 3 in Figs. 3 and 4) with a substantial increase of \( T_e \). Further voltage increases produce relatively small temperature increases, Fig. 3. In the reverse process of lowering the voltage, the HH state persists until the smallest positive root of the equation \( q_1 \tan q_1 = Bi \) [11]. Because \( \tau_f \) is much larger than the director reorientation time \( \tau \approx 10^{-2} \) s, the director configuration can be found as an equilibrium state of a cell with elastic and dielectric properties that correspond to the current temperature.

Let us start with the director orientation at the bounding plates is not fixed by surface anchoring, to show the essence of the effect and the importance of dielectric heating anisotropy. In the anchoring-free case, the equilibrium state is either strictly homeotropic, \( \hat{n} \parallel E \), when \( \Delta \varepsilon(T) > 0 \), or strictly planar, \( \hat{n} \perp E \), when \( \Delta \varepsilon(T) < 0 \). Thus, both transitions should start at \( T = T_c \) and the bistability range \( U_{HH-CP} < U < U_{CP-HH} \) should be determined by anisotropy of the dielectric heating, as

\[
U_{HH-CP} = U_0/(e^e_1)^{1/2} \quad \text{and} \quad U_{CP-HH} = U_0/(e^e_1)^{1/2},
\]

where \( U_0 = [(T_c - T_0)kd/(\pi(1 + Bi)f\varepsilon_0)]^{1/2} \). Note that the anisotropy of dielectric heating must be positive, i.e., \( e^e_1 > e^e_1 \), for the effect to take place.

In a cell with \( \hat{n} \) fixed by surface anchoring at the plates, the equilibrium field-induced distortion of \( \hat{n} = \hat{n}(z) \) has a well-known analytical solution [16]. For small \( e^e_1 - e^e_1 < 0 \), and for one constant approximation of the LC elasticity, this solution relates \( e^e_1z, U \) and the maximum angle \( \theta_m \) in the middle of the LC layer:

\[
U = 2\left(\frac{K}{\varepsilon_0 \Delta \varepsilon(T)}\right)^{1/2} \mathbf{K}(\sin^2 \theta_m),
\]

where \( K \) is elastic constant of the LC, \( \mathbf{K}(\sin^2 \theta_m) \) and \( \mathbf{E}(\sin^2 \theta_m) \) are the complete elliptic integrals of the first and second kind, respectively [17].

To describe the bistability, in addition to Eqs. (3) and (4), one needs to specify the dependence \( \Delta \varepsilon(T) \); Fig. 2 shows that it follows a linear behavior,

\[
\Delta \varepsilon(T) = a(T_c - T),
\]

where \( a = 0.316^\circ \text{C}^{-1}, T_c = 26.0^\circ \text{C} \).

Equations (3)–(5) allow us to find an analytical solution describing \( T(U) \) and \( \theta_m(U) \), Fig. 5. The bistability is controlled by the anisotropy of dielectric heating, \( \Delta e^e = e^e_1 - e^e_1 \). In Fig. 5, we fix \( e^e_1 = 3.5 \) (which is characteristic of MLC2048, Fig. 2) and analyze the scenarios for three different \( e^e_1 \)’s, corresponding to \( \Delta e^e = 0 \) (curve 1), \( \Delta e^e = 0.93 \) (curve 2), and \( \Delta e^e = 3.1 \) (curve 3). Curve 1 describes the “usual” monostable sequence characteristic of zero heating anisotropy. The LC layer, originally in the homeotropic state, experiences a regular Frederiks transition into a tilted CP state \( \theta_m > 0 \) above some modest threshold \( U_f \), Fig. 5, curve 1. A further voltage increase leads to an increase of \( \theta_m \) but it also increases \( T \). Because of the latter, \( \Delta \varepsilon \) decreases to zero and at some \( U_{f2} \), the cell experiences a reverse Frederiks (RF) transition, back to \( \theta_m = 0 \).
To conclude, we observed and interpreted the thermoelectric bistability that is caused by a competition of dielectric heating and director reorientation of the LC in an electric field. For the effect to occur, the dielectric heating should be anisotropic (i.e., dependent on the director orientation with respect to the applied electric field), a feature that has not been appreciated much in the literature so far. The thermoelectric bistability might be of practical importance, because (a) the optical contrast between the HH and CP states is extremely sharp, as the director reorients substantially, from $\theta_m = 0^\circ$ to $\theta_m = 70^\circ$ at the HH-CP transition, Fig. 5, and (b) the states can be switched between with a very low power when the frequency of the applied voltage is only slightly above the crossover frequency.

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FIG. 5 (color online). The simulation of thermoelectric bistability exhibited in $\theta_m(U)$ and $T(U)$ for CP (dashed curves), HH (solid curves), and unstable (dotted curves) states in a homeotropic cell with different $\varepsilon_\perp$: (1) 0.45, (2) 2.57, (3) 3.2. The other parameters correspond to MLC2048 and the experiment presented above. The heat transfer coefficient of the air-gap interface $k = 20$ $\text{W m}^{-2}\text{K}^{-1}$ is chosen to make curve 3 close to the experimental data.

Decreasing $U$ would result in the reverse sequence of transitions without any hysteresis. The scenario changes completely if the heating anisotropy is positive and large enough, curve 3. The original state $\theta_m = 0$ is converted into the CP state $\theta_m > 0$ at the same $U_{RF}$, but because the heat production in this state is relatively low ($\varepsilon_\perp \ll \varepsilon_\parallel$), the dielectric anisotropy $\Delta \varepsilon$ remains positive and the CP state is retained even for voltages above $U_{RF}$: it transforms abruptly into the HH state only at some $U_{\text{CP-HH}} > U_{RF}$. Upon reduction of the voltage, the HH state returns into the CP state at $U_{\text{HH-CP}} = U_{RF}$, hence a bistability. Finally, curve 2 is a critical regime separating the monostable and bistable behaviors.

The model (curve 3) describes the experimental data very closely, as evident from the predicted voltage and temperature range of the bistability; compare Figs. 3 and 5. Clearly, some model simplifications, such as one-constant elasticity, the exact value of $k$, and cell imperfections, such as director distortions around the spacers or finite electric conductivity of the cell, might cause quantitative discrepancies, but the overall agreement is good.

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