Consequences of director-density coupling theory for flexoelectricity in nematic liquid crystals

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We theoretically study how the measurements of the flexoelectric coefficients in nematic liquid crystals are affected by the inclusion of the director-density coupling energy. It is shown that this investigation is quite relevant for interpreting the data of experiments.

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I. INTRODUCTION

Most of the current interest in studying flexoelectricity derives from technological applications based on the flexoelectric-optic effect [1] and in the development of a zenithally bistable nematic device [2]. The flexoelectric effect in liquid crystals manifests itself as the appearance of spontaneous polarization induced by deformation [3], which in the nematic phase is given by

$$\mathbf{P}_{\mathrm{f}} = e_1\mathbf{n}(\nabla\cdot\mathbf{n}) + e_3[(\nabla\times\mathbf{n})\times\mathbf{n}]. \tag{1}$$

Here, $\mathbf{n}$ is the liquid crystal director, and e_1 and e_3 are, respectively, the flexoelectric coefficients for splay and bend distortions. Two mechanisms contribute to the flexoelectric couplings. The first is of a dipolar origin [3], arising from pear- and banana-shaped molecules with permanent dipoles; the second one relies on quadrupolar contributions [4], which are of a more general character. Much additional progress in our understanding of flexoelectric coefficients in terms of molecular parameters has been actively researched over the years [5–13].

In the early years after Meyer's discovery, experimental and theoretical works showed that the flexoelectric coefficients are of order of few pC/m [5,6,14,15], being subsequently confirmed by a great number of empirical evidences [16–29]. The announcement of giant flexoelectricity in bent-core nematic material [30,31], therefore, came as a surprise to most researchers. Some authors [32–35], however, have failed to reproduce the strength of the effect observed by Harden and collaborators. In addition, it appears to be impossible for conservation of energy reasons [36,37].

There are several complicating factors involved in the measurement of flexoelectric coefficients, such as ionic drift in experiments using dc voltage [26,38,39] and the influence of surface polarization [22,23,40]. An additional complication has been raised by many authors but has not yet been clarified. It is about the fact that for certain liquid crystal materials, different methods of measurement give conflicting results [16,18,41–43]. It is our purpose in this paper to discuss this problem with the aid of the elastic energy

$$f_{\mathrm{ddc}} = \sum_{i,j}\left[u_1\left(\frac{\partial^2\rho}{\partial x^i\partial x^j}\right) + u_2\left(\frac{\partial\rho}{\partial x^i}\right)\left(\frac{\partial\rho}{\partial x^j}\right)\right]n_i n_j + \frac{1}{2}B|\nabla\rho|^2, \tag{2}$$

introduced in Ref. [44] as a supplement to the Oseen-Frank energy density. According to this theory, inhomogeneities in the local mass density ρ couple directly with the liquid crystal director $\mathbf{n}$. Here, B, u_1, and u_2 are the coupling constants of the theory. The B term, originally missing, has been added in order to endow the theory with a minimum [45]. Also, it can be justified on general grounds [46]. When f_{ddc} is included in the total elastic energy of a nematic, it turns out that the static behavior of the resulting theory predicts a Fréedericksz transition accompanied by modulation in the mass density [45] (see also Ref. [47]). Besides the experimental support already given [44,48], there is convincing evidence [49–52] that the theory we are working with is able to interpret the experimental data on the acousto-optic effect reported in Ref. [53]. For this reason, it is worthwhile to investigate the implications of f_{ddc} on flexoelectricity with the aim towards shedding light on the problem of conflicting measurements alluded to above. We do this by calculating the effect of f_{ddc} in real experimental setups designed to measure e_3 and e_1 (in Secs. II and III, respectively) and the combinations $e_1 + e_3$ and $e_1 - e_3$ (in Secs. IV and V, respectively). We reserve Sec. VI for concluding remarks.

II. MEASURING e_3

Let us consider the following experiment designed to measure e_3. A nematic sample of thickness h is placed between two large parallel plates; the alignment is homeotropic and weakly anchored [see Fig. 1(a)]. The liquid crystal director reads

$$\mathbf{n}(z) = \hat{\mathbf{x}}\sin\theta(z) + \hat{\mathbf{z}}\cos\theta(z), \tag{3}$$

and $\rho = \rho(z)$ is assumed here (and throughout the paper). The total energy density is given by

$$f = f_{\mathrm{of}} + f_{\mathrm{ddc}} + f_{\mathrm{flexo}}, \tag{4}$$

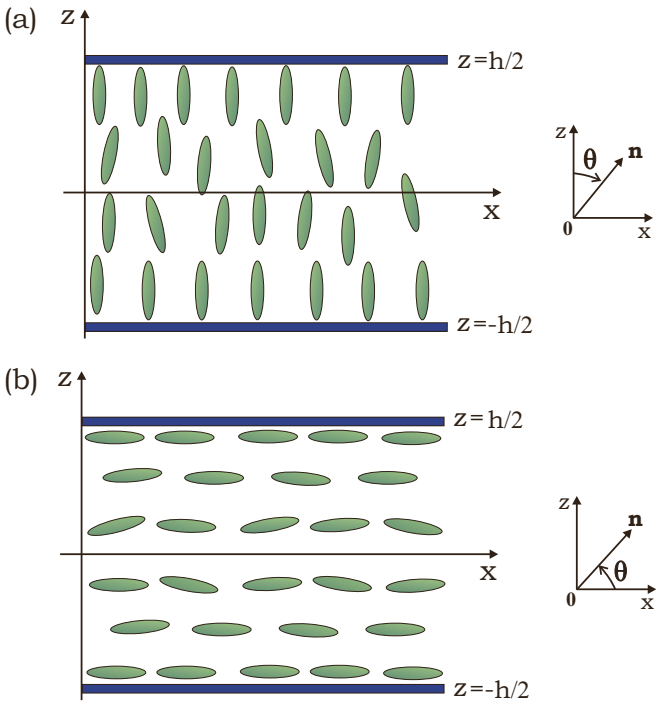

FIG. 1. Cell of thickness h containing a nematic liquid crystal: (a) Homeotropic alignment and (b) planar alignment. On the right, the coordinate system defines the angle θ for the director.

in which

$$f_{\rm of} = \tfrac{1}{2}K_1(\nabla\cdot\mathbf{n})^2 + \tfrac{1}{2}K_2[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2 + \tfrac{1}{2}K_3|\mathbf{n}\times(\nabla\times\mathbf{n})|^2, \quad (5)$$

$$f_{\rm flexo} = -\mathbf{P}_{\rm f}\cdot\mathbf{E}. \quad (6)$$

In the above, K_1, K_2, and K_3 are, respectively, the Frank constants for splay, twist, and bend, and $\mathbf{E}$ is the electric field applied to the nematic material. For the particular case in which $\mathbf{E} = -E\hat{\mathbf{x}}$, one gets

$$f_{\rm of} = \frac{1}{2}(K_1\sin^2\theta + K_3\cos^2\theta)\left(\frac{d\theta}{dz}\right)^2, \quad (7)$$

$$f_{\rm ddc} = \frac{1}{2}B\left(\frac{d\rho}{dz}\right)^2 + \left[u_1\left(\frac{d^2\rho}{dz^2}\right) + u_2\left(\frac{d\rho}{dz}\right)^2\right]\cos^2\theta, \quad (8)$$

$$f_{\rm flexo} = E(e_3\cos^2\theta - e_1\sin^2\theta)\left(\frac{d\theta}{dz}\right). \quad (9)$$

The dielectric term $f_{\rm diel}$ is of $O(E^2)$ and has been neglected (to be taken into account in Secs. IV and V). The equilibrium configuration is that one which minimizes the total energy $F = \int_{-h/2}^{h/2} f\,dz$ and satisfies the conservation of mass $M = \int_{-h/2}^{h/2}\rho(z)dz$. The solution of this isoperimetric calculus of variations problem [54] is found by solving the following set of equations,

$$\frac{d^2}{dz^2}\left(\frac{\partial\overline{f}}{\partial\rho_{zz}}\right) - \frac{d}{dz}\left(\frac{\partial\overline{f}}{\partial\rho_z}\right) + \frac{\partial\overline{f}}{\partial\rho} = 0, \quad (10)$$

$$\frac{d}{dz}\left(\frac{\partial\overline{f}}{\partial\theta_z}\right) - \frac{\partial\overline{f}}{\partial\theta} = 0, \quad (11)$$

where $\rho_{zz}\equiv d^2\rho/dz^2$, $\rho_z\equiv d\rho/dz$, $\theta_z\equiv d\theta/dz$, and $\overline{f} = f + \mu\rho$. Here, μ is a (constant) Lagrange multiplier. Since $\partial\overline{f}/\partial\rho = \mu$, it follows from Eq. (10) that

$$\frac{d}{dz}\left(\frac{\partial\overline{f}}{\partial\rho_{zz}}\right) - \frac{\partial\overline{f}}{\partial\rho_z} + \mu z = C. \quad (12)$$

Next, we substitute $\overline{f}$ in the above equation to obtain

$$-u_1\left(\frac{d\theta}{dz}\right)\sin(2\theta) - (B + 2u_2\cos^2\theta)\left(\frac{d\rho}{dz}\right) + \mu z = C. \quad (13)$$

Note that the conditions $\theta(0) = 0$ and $(\frac{d\rho}{dz})_{z=0} = 0$ [by virtue of $\rho(z) = \rho(-z)$] automatically imply $C = 0$. The substitution of $\overline{f}$ in Eq. (11), on the other hand, leads to

$$(K_1\sin^2\theta + K_3\cos^2\theta)\left(\frac{d^2\theta}{dz^2}\right) + \frac{1}{2}(K_1 - K_3)\left(\frac{d\theta}{dz}\right)^2\sin(2\theta) + \left[u_1\left(\frac{d^2\rho}{dz^2}\right) + u_2\left(\frac{d\rho}{dz}\right)^2\right]\sin(2\theta) = 0. \quad (14)$$

It is reasonable to seek a solution of the form

$$\theta(z) = E\theta_1(z) + \cdots, \quad (15)$$

$$\rho(z) = \rho_0 + E^2\rho_2(z) + \cdots, \quad (16)$$

where ρ_0 is the average mass density and $\rho(z)$ is an even function of E. Hence $\mu = \mu_2E^2 + \cdots$ and μ_2 is field independent. Consequently, the linearized form of Eq. (14) reads $d^2\theta_1/dz^2 = 0$, which immediately furnishes $\theta_1 = \lambda z$ since $\theta(0) = 0$. In order to calculate the constant of integration λ, we first return to Eq. (13), using our solution for θ_1 to get

$$\frac{d\rho_2}{dz} = \frac{(\mu_2 - 2u_1\lambda^2)z}{B + 2u_2}. \quad (17)$$

We now evaluate the corresponding energy cost by means of Eq. (4),

$$f = \left(\frac{1}{2}K_3^*\lambda^2 + e_3\lambda + \frac{u_1\mu_2}{B + 2u_2}\right)E^2 + \cdots, \quad (18)$$

where

$$K_3^* \equiv K_3 - \frac{4u_1^2}{B + 2u_2}. \quad (19)$$

In the Appendix, it is argued that $B \sim 2u_2$ and $u_1^2/u_2 \sim K_3$. We should expect, therefore, that $f_{\rm ddc}$ brings a significant correction to K_3^*. If one assumes that K_3^* is positive, it is clear that f (and then F) becomes minimum when $\lambda = -e_3/K_3^*$. In this case, a distortion of the nonthreshold character is observed, i.e.,

$$\theta(z) = -\left(\frac{e_3E}{K_3^*}\right)z + \cdots. \quad (20)$$

Note that the only effect of $f_{\rm ddc}$, Eq. (2), is to renormalize the Frank constant K_3. The experiment we are discussing, therefore, gives the ratio e_3/K_3^* (presumably correct) instead

of the classical result e_3/K_3 [55]. In light of this, the agreement between experimental data with a theoretical prediction (ignoring the effect of $f_{\rm ddc}$) is achieved through the enhanced flexoelectric coefficient

$$e_3^{\rm eff} = \left(\frac{K_3}{K_3^*}\right) e_3. \tag{21}$$

At this point, it is interesting to observe that the presence of surface polarization m_p has the effect of replacing e_3 with $e_3 + m_p$ [40].

III. MEASURING e_1

We now consider an experiment intended to measure the flexoelectric coefficient e_1. The nematic material is sandwiched between two glass plates and is planarly aligned [see Fig. 1(b)]. As in the prior section, the anchoring is also supposed to be very weak. The liquid crystal director is written as $\mathbf{n}(z) = \hat{\mathbf{x}}\cos\theta(z) + \hat{\mathbf{z}}\sin\theta(z)$. In this case, when the external field $\mathbf{E} = E\hat{\mathbf{x}}$ is applied, the total energy density is obtained by replacing $\theta \to \pi/2 - \theta$ and $E \to -E$ in Eqs. (7)–(9). If we now substitute $\overline{f}$ into Eq. (12), we find that

$$u_1\left(\frac{d\theta}{dz}\right)\sin(2\theta) - (B + 2u_2\sin^2\theta)\left(\frac{d\rho}{dz}\right) + \mu z = C. \tag{22}$$

This is just the equation analogous to Eq. (13). Again, $C = 0$ (for the same reason) and we assume the validity of the expansions given by Eqs. (15) and (16). Inspection of Eq. (8) (after making the substitution $\cos\theta \to \sin\theta$) and Eq. (22) reveals that the contribution of $f_{\rm ddc}$ to f is of $O(E^4)$. Hence, in contrast to the previous case, no correction is predicted to the established result that $\theta(z) = (e_1E/K_1)z + \cdots$ [55], so that one has $e_1^{\rm eff} = e_1$.

IV. MEASURING THE COMBINATION $e_1 + e_3$

Returning to Fig. 1(a), we are now assuming that there is a surface free charge density $\sigma = \hat{\mathbf{z}}\cdot\mathbf{D}$ on the bottom surface of the nematic cell (there is no other external source of electric field). However, there are no free charge in the medium, so that

$$\nabla\cdot\mathbf{D} = 0. \tag{23}$$

In the above, $\mathbf{D} = \varepsilon_0[\varepsilon_\perp\mathbf{E} + (\Delta\varepsilon)(\mathbf{E}\cdot\mathbf{n})\mathbf{n}] + \mathbf{P}_{\rm f}$ is the electric displacement, $\mathbf{E} = E(z)\hat{\mathbf{z}}$, ε_0 is the permittivity of vacuum, $\varepsilon_\perp$ and $\varepsilon_\parallel$ are, respectively, dielectric susceptibilities perpendicular and parallel to the director, and $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$ denotes the dielectric anisotropy [55]. We confine ourselves to a negative dielectric material, $\Delta\varepsilon = -|\Delta\varepsilon|$. The aim is to describe an experiment where a Fréedericksz transition takes place [56]. Thus, a strong-anchoring boundary condition is required,

$$\theta(-h/2) = \theta(h/2) = 0, \tag{24}$$

along with [45]

$$\left(\frac{d\rho}{dz}\right)_{z=-h/2} = \left(\frac{d\rho}{dz}\right)_{z=h/2} = 0. \tag{25}$$

From Eq. (23) it follows that the z component of $\mathbf{D}$ is constant and

$$\mathbf{E}(z) = \frac{(\sigma - P_{\rm fz})\hat{\mathbf{z}}}{\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)}, \tag{26}$$

where

$$P_{\rm fz} = -\frac{(e_1+e_3)}{2}\left(\frac{d\theta}{dz}\right)\sin(2\theta) \tag{27}$$

is the z component of $\mathbf{P}_{\rm f}$. Here, $f_{\rm of}$ and $f_{\rm ddc}$ are given by the same expressions as Eqs. (7) and (8). For the remaining terms we have

$$f_{\rm flexo} = -\frac{P_{\rm fz}(\sigma - P_{\rm fz})}{\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)} \tag{28}$$

and the dielectric term

$$f_{\rm diel} = \frac{1}{2}\mathbf{D}\cdot\mathbf{E} = \frac{\sigma(\sigma - P_{\rm fz})}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)}. \tag{29}$$

The equilibrium configuration is found by solving Eqs. (13) [in this case, Eqs. (24) and (25) together imply $\mu = C = 0$] and

$$\begin{aligned}&(K_1\sin^2\theta + K_3\cos^2\theta)\left(\frac{d^2\theta}{dz^2}\right) + \frac{1}{2}(K_1 - K_3)\left(\frac{d\theta}{dz}\right)^2\\ &\times\sin(2\theta) + \frac{\sigma^2|\Delta\varepsilon|\sin(2\theta)}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)^2} + \frac{(e_1+e_3)^2}{4\varepsilon_0}\\ &\times\left[\frac{2\sin(2\theta)}{\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta}\left(\frac{d^2\theta}{dz^2}\right)\right.\\ &\left.+\frac{4(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)\cos(2\theta) - |\Delta\varepsilon|\sin^2(2\theta)}{(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)^2}\left(\frac{d\theta}{dz}\right)^2\right]\\ &\times\sin(2\theta) + \left[u_1\left(\frac{d^2\rho}{dz^2}\right) + u_2\left(\frac{d\rho}{dz}\right)^2\right]\sin(2\theta) = 0,\end{aligned} \tag{30}$$

which follows from Eq. (11). We now substitute Eq. (13) into Eq. (30) and the resulting equation is then multiplied by $d\theta/dz$ (the trick for exactly integrating [55]). After the calculation is finished, one obtains

$$\frac{d}{dz}\left[\frac{1}{2}\left(\frac{d\theta}{dz}\right)^2 f(\theta) - \frac{\sigma^2}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)}\right] = 0, \tag{31}$$

where

$$\begin{aligned}f(\theta) = (K_1\sin^2\theta + K_3\cos^2\theta) - \frac{u_1^2\sin^2(2\theta)}{B + 2u_2\cos^2\theta}\\ + \frac{(e_1+e_3)^2\sin^2(2\theta)}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)}.\end{aligned} \tag{32}$$

Hence the integration can be easily performed, leaving us with

$$\frac{1}{2}\left(\frac{d\theta}{dz}\right)^2 f(\theta) - \frac{\sigma^2}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta)} = C. \tag{33}$$

The constant of integration

$$C = -\frac{\sigma^2}{2\varepsilon_0(\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta_m)} \tag{34}$$

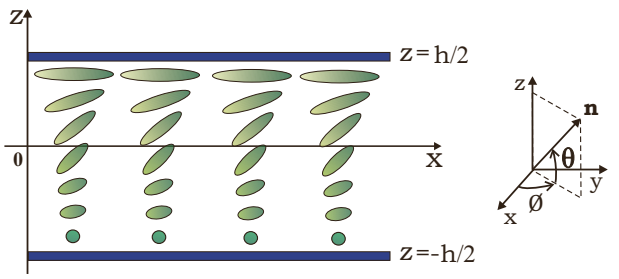

FIG. 2. Twisted nematic cell of thickness h. On the right, the coordinate system defines the angles θ and ϕ for the director.

is determined by imposing symmetric distortion around $z = 0$, namely,

$$\left(\frac{d\theta}{dz}\right)_{z=0} = 0, \quad \theta(0) = \theta_m, \tag{35}$$

where θ_m is the maximum angle. We can express the solution of Eq. (33) with $d\theta/dz \geqslant 0$ as follows,

$$\int_0^{\theta} \sqrt{\frac{f(\alpha)}{g(\alpha)}}d\alpha = z + \frac{h}{2}, \tag{36}$$

$$\int_0^{\theta_m} \sqrt{\frac{f(\alpha)}{g(\alpha)}}d\alpha = \frac{h}{2}, \tag{37}$$

where

$$g(\theta) = \frac{\sigma^2}{\varepsilon_0}\left[\frac{1}{\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta} - \frac{1}{\varepsilon_\parallel + |\Delta\varepsilon|\sin^2\theta_m}\right]. \tag{38}$$

For $0 < z \leqslant h/2$, one has $\theta(-z) = -\theta(z)$. It is now easy to see that due to the presence of the second term in Eq. (32), the fit between experiment and theory becomes problematic. To better illustrate this point, let us admit that contributions of $O(\theta^4)$ can be neglected. In this case, it is the combination

$$\frac{(e_1 + e_3)^2}{2\varepsilon_0\varepsilon_\parallel} - \frac{u_1^2}{B + 2u_2} \tag{39}$$

that emerges from the experimental work. Let us assume, for continuing discussion, that the combination is positive. Then the counterpart of Eq. (21) reads

$$(e_1 + e_3)^2_{\text{eff}} = (e_1 + e_3)^2 - \frac{2\varepsilon_0\varepsilon_\parallel u_1^2}{B + 2u_2}. \tag{40}$$

It should be noticed that for positive dielectric anisotropy, Eq. (31) describes a Fréedericksz transition undergone by a nematic material planarly aligned after the replacements $K_1 \to K_3, \varepsilon_\parallel \to \varepsilon_\perp$, $|\Delta\varepsilon| \to \Delta\varepsilon$, and $u_2\cos^2\theta \to u_2\sin^2\theta$. In view of the new form of $f(\theta)$, the analogous to Eq. (40) lacks the term involving u_2:

$$(e_1 + e_3)^2_{\text{eff}} = (e_1 + e_3)^2 - \frac{2\varepsilon_0\varepsilon_\perp u_1^2}{B}. \tag{41}$$

V. MEASURING THE COMBINATION $e_1 - e_3$

Let us now consider a twisted nematic sample (pitch infinite) and subjected to the electric field $\mathbf{E} = E\hat{\mathbf{x}}$ (see Fig. 2). The director is written as $\mathbf{n} = \hat{\mathbf{x}}\cos\theta\cos\phi + \hat{\mathbf{y}}\cos\theta\sin\phi + \hat{\mathbf{z}}\sin\theta$, where θ and ϕ are assumed to be functions of z only and also to satisfy $\phi(-h/2) = \phi_1$, $\phi(h/2) = \phi_2$, and $\theta(-h/2) = \theta(h/2) = 0$ [55]. Using $f_{\text{diel}} = -\frac{1}{2}\varepsilon_0(\Delta\varepsilon)(\mathbf{E}\cdot\mathbf{n})^2$ for the dielectric energy, the total energy density then takes the form

$$\begin{aligned} f = {} & \frac{1}{2}(K_1\cos^2\theta + K_3\sin^2\theta)\left(\frac{d\theta}{dz}\right)^2 + \frac{1}{2}K_2\left(\frac{d\phi}{dz}\right)^2\cos^4\theta + \frac{1}{2}K_3\left(\frac{d\phi}{dz}\right)^2\sin^2\theta\cos^2\theta \\ & + e_3E\left[\left(\frac{d\theta}{dz}\right)\sin^2\theta\cos\phi + \frac{1}{2}\left(\frac{d\phi}{dz}\right)\sin(2\theta)\sin\phi\right] - e_1E\left(\frac{d\theta}{dz}\right)\cos^2\theta\cos\phi + \frac{1}{2}B\left(\frac{d\rho}{dz}\right)^2 \\ & + \left[u_1\left(\frac{d^2\rho}{dz^2}\right) + u_2\left(\frac{d\rho}{dz}\right)^2\right]\sin^2\theta - \frac{1}{2}\varepsilon_0(\Delta\varepsilon)E^2\cos^2\theta\cos^2\phi. \end{aligned} \tag{42}$$

To find the minimum, we have to solve the following set of equations,

$$\begin{aligned} & (K_1\cos^2\theta + K_3\sin^2\theta)\left(\frac{d^2\theta}{dz^2}\right) + \frac{1}{2}(K_3 - K_1)\left(\frac{d\theta}{dz}\right)^2\sin(2\theta) + \left(\frac{d\phi}{dz}\right)^2\left[2K_2\cos^3\theta\sin\theta - \frac{K_3}{4}\sin(4\theta)\right] \\ & - \left[u_1\left(\frac{d^2\rho}{dz^2}\right) + u_2\left(\frac{d\rho}{dz}\right)^2\right]\sin(2\theta) + (e_1 - e_3)E\left(\frac{d\phi}{dz}\right)\cos^2\theta\sin\phi - \frac{1}{2}\varepsilon_0(\Delta\varepsilon)E^2\sin(2\theta)\cos^2\phi = 0, \end{aligned} \tag{43}$$

$$\begin{aligned} & K_2\left[\left(\frac{d^2\phi}{dz^2}\right)\cos^4\theta - 4\cos^3\theta\sin\theta\left(\frac{d\phi}{dz}\right)\left(\frac{d\theta}{dz}\right)\right] + K_3\left[\frac{1}{4}\left(\frac{d^2\phi}{dz^2}\right)\sin^2(2\theta) + \frac{1}{2}\left(\frac{d\phi}{dz}\right)\left(\frac{d\theta}{dz}\right)\sin(4\theta)\right] \\ & - \frac{1}{2}\varepsilon_0(\Delta\varepsilon)E^2\cos^2\theta\sin(2\phi) + (e_3 - e_1)E\left(\frac{d\theta}{dz}\right)\cos^2\theta\sin\phi = 0, \end{aligned} \tag{44}$$

and Eq. (22) with $\mu = C = 0$ [after use of Eqs. (24) and (25)]. It is clear that Eq. (44) comes from Eq. (11) [this applies equally to Eq. (43)] when θ is changed to ϕ. Instead of proceeding with the calculation, we are interested only in

pointing out the relevance of $f_{\rm ddc}$ in this kind of experiment. Indeed, if we assume that the equilibrium configuration can be written as

$$\theta(z;E) = E\theta^{(1)}(z) + E^2\theta^{(2)}(z) + E^3\theta^{(3)}(z) + \cdots, \quad (45)$$

$$\phi(z;E) = \phi^{(0)}(z) + E\phi^{(1)}(z) + E^2\phi^{(2)}(z) + E^3\phi^{(3)}(z) + \cdots, \quad (46)$$

then it can be seen from Eq. (43) that both terms, the dielectric and the one coming from $f_{\rm ddc}$, are of $O(E^3)$. This means, in essence, that $f_{\rm ddc}$ should not be ignored while maintaining the dielectric term.

VI. CONCLUDING REMARKS

It has been recorded in the literature that for certain liquid crystal materials, different methods of measuring the flexoelectric coefficients give rise to conflicting results. In an effort to shed light on this issue, we have studied the effect of the director-density coupling theory in real experimental situations designed to measure the flexoelectric coefficients e_3 and e_1, and the combinations $e_1 + e_3$ and $e_1 - e_3$. It is worthwhile to recall that this theory has passed a crucial test [52] and, therefore, we believe our findings are of great interest to the ongoing debate on flexoelectricity in nematic liquid crystals.

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APPENDIX: CONJECTURE ON THE MAGNITUDE OF u_1

Unfortunately, only the magnitude of u_2 is known [44,48]; what follows provides a conjectural approach to get the order of magnitude of u_1. The argument is based on the results of Ref. [52]. If our explanation in this reference is correct, then the optical transparency M behaves as $M = AJ^2 + BJ^3$. Here, J is the acoustic intensity (assumed to be low) and A and B are constants. To derive this, it was assumed that the action of a monochromatic ultrasonic plane wave of wave vector $\mathbf{k}$ and frequency ω causes a rapid fluctuation in the local mass density of the form

$$\rho(\mathbf{r},t) = \rho_0 + (\Delta\rho)\sin(\mathbf{k}\cdot\mathbf{r} - \omega t), \quad (A1)$$

where

$$J = \frac{v^3(\Delta\rho)^2}{2\rho_0}. \quad (A2)$$

Here, v is the sound velocity. If we insert Eq. (A1) into Eq. (2), we obtain

$$f_{\rm ddc} = \frac{1}{2}Bk^2(\Delta\rho)^2\cos^2(\mathbf{k}\cdot\mathbf{r} - \omega t) - u_1(\Delta\rho)(\mathbf{n}\cdot\mathbf{k})^2\sin(\mathbf{k}\cdot\mathbf{r} - \omega t) + u_2(\Delta\rho)^2(\mathbf{n}\cdot\mathbf{k})^2\cos^2(\mathbf{k}\cdot\mathbf{r} - \omega t). \quad (A3)$$

On the other hand, it is experimentally verified that for $J \geqslant 75$ mW/cm^2 (see Fig. 2), the mechanism of streaming dominates the physics. We thus argue that for this acoustic intensity, the amplitude of the oscillating terms in Eq. (A3) are of same order, so that $B \sim 2u_2$ and

$$u_1 \sim u_2(\Delta\rho). \quad (A4)$$

Combining Eqs. (2) and (A4), we get

$$\frac{u_1^2}{u_2} \sim \frac{2\rho_0 J u_2}{v^3} = 0.58 \times 10^{-11}\ \text{N} \sim K_3, \quad (A5)$$

with the use of $\rho_0 = 1.0$ g/cm^3, $J = 75$ mW/cm^2, $v = 1500$ m/s, and $u_2 = 1.30 \times 10^3$ cm^7 g^{-1} s^{-2} [44,48].

[1] J. S. Patel and R. B. Meyer, Phys. Rev. Lett. **58**, 1538 (1987).
[2] G. P. Bryan-Brown, C. V. Brown, J. C. Jones, E. L. Wood, I. C. Sage, P. Brett, and J. Rudin, SID Digest, Vol. 18, pp. 37–40 (1997); G. P. Bryan-Brown, C. V. Brown, J. C. Jones, E. L. Wood, I. C. Sage, P. Brett, and J. Rudin, Proceedings of SID International Display Research Conference, SID 4th International Display Research Conference, Nagoya, Japan, 1997 [see also Ref. [6] of Phys. Rev. E **73**, 011713 (2006)].
[3] R. B. Meyer, Phys. Rev. Lett. **22**, 918 (1969).
[4] J. Prost and J. P. Marcerou, J. Phys. (Paris) **38**, 315 (1977).
[5] W. Helfrich, Z. Naturforsch. A **26**, 833 (1971).
[6] W. Helfrich, Phys. Lett. A **35**, 393 (1971).
[7] A. Derzhanski and A. G. Petrov, Phys. Lett. A **36**, 483 (1971).
[8] M. A. Osipov, Zh. Eksp. Teor. Fiz. **85**, 2011 (1983) [Sov. Phys. JETP **58**, 1167 (1983)].
[9] J. Stelzer, R. Berardi, and C. Zannoni, Chem. Phys. Lett. **299**, 9 (1999).
[10] J. Stelzer, R. Berardi, and C. Zannoni, Mol. Cryst. Liq. Cryst. **352**, 187 (2000).
[11] A. V. Zakharov and R. Y. Dong, Eur. Phys. J. E **6**, 3 (2001).
[12] S. Dhakal and J. V. Selinger, Phys. Rev. E **81**, 031704 (2010).
[13] S. M. Shamid, S. Dhakal, and J. V. Selinger, Phys. Rev. E **87**, 052503 (2013).
[14] D. Schmidt, M. Schadt, and W. Helfrich, Z. Naturforsch. A **27**, 277 (1972).
[15] W. Helfrich, Mol. Cryst. Liq. Cryst. **26**, 1 (1974).
[16] J. Prost and P. S. Pershan, J. Appl. Phys. **47**, 2298 (1976).
[17] Y. P. Bobylev, V. G. Chigrinov, and S. A. Pikin, J. Phys. (Paris) **40**, C3-331 (1979).
[18] I. Dozov, Ph. Martinot-Lagarde, and G. Durand, J. Phys. (Paris) Lett. **43**, 365 (1982).
[19] I. Dozov, Ph. Martinot-Lagarde, and G. Durand, J. Phys. (Paris) Lett. **44**, 817 (1983).
[20] L. A. Beresnev, L. M. Blinov, S. A. Davidyan, S. G. Kononov, and S. B. Yablonskii, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 592 (1987) [JETP Lett. **45**, 755 (1987)].
[21] D. S. Hermann, P. Rudquist, K. Ichimura, K. Kudo, L. Komitov, and S. T. Lagerwall, Phys. Rev. E **55**, 2857 (1997).
[22] L. M. Blinov, M. I. Barnik, H. Ohoka, M. Ozaki, and K. Yoshino, Phys. Rev. E **64**, 031707 (2001).

[23] A. Mazzulla, F. Ciuchi, and J. R. Sambles, Phys. Rev. E **64**, 021708 (2001).
[24] S. A. Jewell and J. R. Sambles, J. Appl. Phys. **92**, 19 (2002).
[25] C. V. Brown and N. J. Mottram, Phys. Rev. E **68**, 031702 (2003).
[26] R. A. Ewings, C. Kischka, L. A. Parry-Jones, and S. J. Elston, Phys. Rev. E **73**, 011713 (2006).
[27] T. Tóth-Katona, N. Éber, A. Buka, and A. Krekhov, Phys. Rev. E **78**, 036306 (2008).
[28] S. J. Elston, Phys. Rev. E **78**, 011701 (2008).
[29] C. L. Trabi, C. V. Brown, A. A. T. Smith, and N. J. Mottram, Appl. Phys. Lett. **92**, 223509 (2008).
[30] J. Harden, B. Mbanga, N. Éber, K. Fodor-Csorba, S. Sprunt, J. T. Gleeson, and A. Jákli, Phys. Rev. Lett. **97**, 157802 (2006).
[31] J. Harden, R. Teeling, J. T. Gleeson, S. Sprunt, and A. Jákli, Phys. Rev. E **78**, 031702 (2008).
[32] P. Kumar, Y. G. Marinov, H. P. Hinov, U. S. Hiremath, C. V. Yelamaggad, K. S. krishnamurthy, and A. G. petrov, J. Phys. Chem. B **113**, 9168 (2009).
[33] K. V. Le, F. Araoka, K. Fodor-Csorba, K. Ishikawa, and H. Takezoe, Liq. Cryst. **36**, 1119 (2009).
[34] P. S. Salter, C. Tschierske, S. J. Elston, and E. P. Raynes, Phys. Rev. E **84**, 031708 (2011).
[35] R. Balachandran, V. P. Panov, J. K. Vij, A. Lehmann, and C. Tschierske, Phys. Rev. E **88**, 032503 (2013).
[36] F. Castles, S. M. Morris, and H. J. Coles, AIP Adv. **1**, 032120 (2011).
[37] F. Castles, S. M. Morris, and H. J. Coles, AIP Adv. **3**, 019102 (2013).
[38] S. Ponti, P. Ziherl, C. Ferrero, and S. Zumer, Liq. Cryst. **26**, 1171 (1999).
[39] G. Barbero and L. R. Evangelista, Phys. Rev. E **68**, 023701 (2003).
[40] A. Derzhanski, A. G. Petrov, and M. D. Mitov, J. Phys. (Paris) **39**, 273 (1978); Y. G. Marinov, G. B. Hadjichristov, A. G. Petrov, S. Sridevi, U. S. Hiremath, C. V. Yelamaggad, and S. K. Prasad, J. Phys.: Conf. Ser. **253**, 012060 (2010); S. Sridevi, U. S. Hiremath, C. V. Yelamaggad, A. G. Petrov, and S. K. Prasad, Bulg. J. Phys. **39**, 3 (2012).
[41] N. V. Madhusudana and G. Durand, J. Phys. (Paris) Lett. **46**, 195 (1985).
[42] B. Valenti, C. Bertoni, G. Barbero, P. Taverna-Valabrega, and R. Bartolino, Mol. Cryst. Liq. Cryst. **146**, 307 (1987).
[43] L. M. Blinov, G. Durand, and S. V. Yablonsky, J. Phys. (Paris) II **2**, 1287 (1992).
[44] J. V. Selinger, M. S. Spector, V. A. Greanya, B. T. Weslowski, D. K. Shenoy, and R. Shashidhar, Phys. Rev. E **66**, 051708 (2002).
[45] C. Vitoriano, Phys. Rev. E **90**, 032502 (2014).
[46] E. G. Virga, Phys. Rev. E **80**, 031705 (2009).
[47] G. De Matteis and G. Napoli, J. Appl. Math. **73**, 882 (2013).
[48] V. A. Greanya, M. S. Spector, J. V. Selinger, B. T. Weslowski, and R. Shashidhar, J. Appl. Phys. **94**, 7571 (2003); A. P. Malanoski, V. A. Greanya, B. T. Weslowski, M. S. Spector, J. V. Selinger, and R. Shashidhar, Phys. Rev. E **69**, 021705 (2004); V. A. Greanya, A. P. Malanoski, B. T. Weslowski, M. S. Spector, and J. V. Selinger, Liq. Cryst. **32**, 933 (2005).
[49] C. Sátiro and C. Vitoriano, Phys. Rev. E **84**, 041702 (2011).
[50] C. Sátiro and C. Vitoriano, Phys. Rev. E **86**, 011701 (2012).
[51] C. Vitoriano and C. Sátiro, Phys. Rev. E **86**, 061702 (2012).
[52] C. Vitoriano, Phys. Rev. E **88**, 032501 (2013).
[53] O. A. Kapustina, Akust. Zh. **54**, 900 (2008) [Acoust. Phys. **54**, 778 (2008)].
[54] See, e.g., I. M. Gelfand and S. V. Fomin, *Calculus of Variations* (Prentice-Hall, Englewood Cliffs, NJ, 1963).
[55] D.-K. Yang and S.-T. Wu, *Fundamentals of Liquid Crystal Devices* (Wiley, Hoboken, NJ, 2006).
[56] H. J. Deuling, Solid State Commun. **14**, 1073 (1974).