Acousto-optic effect in nematic liquid crystals: Experimental evidence of an elastic regime

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We show that the experimental data for the action of ultrasonic waves on homeotropically aligned nematic-liquid-crystal cells reported by Kapustina, in Akust. Zh. **54**, 900 (2008) [Acoust. Phys. **54**, 778 (2008)] can be explained in the framework of the director-density coupling theory in the regime of low acoustic intensity. This result therefore provides support for the hypothesis that the interaction between sound and nematic liquid crystals is dominated by an elastic energy.

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I. INTRODUCTION

The acousto-optic effect in nematic liquid crystals [1–4] has been the focus of intense experimental and theoretical research. Shortly after its discovery [5], the effect was interpreted as a result of sound stresses [6,7]; nevertheless, the prediction of a threshold intensity for distorting the director orientation was soon recognized as a serious drawback of the theory. The model put forward by Dion and Jacob [8–10] relies on the anisotropy of ultrasonic absorption; the problematic nature of the theory, however, is manifested by the way in which the alignment torque is obtained. In parallel, a mechanism involving streaming due to gradients of acoustic radiation pressure was proposed and investigated [11–16].

Currently, two interesting scenarios are being invoked to explain the underlying physics of the acousto-optic effect. In the first one, the mechanism of molecular reorientation is a consequence of acoustical streaming arising from convective stresses [17–20], and a systematic study has been conducted to verify this possibility [4,21]. In the other possible scenario, the alignment action of ultrasound is of elastic nature and accordingly the action of the acoustic field upon the director is not mediated by flow. This hypothesis was first elaborated in Refs. [22,23], and was followed by extensive experimental and theoretical work [24–31]. More recently, a variant of this theory, in which liquid crystals are regarded as anisotropic Korteweg fluids, is being intensively investigated [32–37]. In particular, the theory has been applied to a flowless situation where the nematic is under the action of acoustic waves [35].

Within the framework of the director-density coupling theory [22], the optical transparency M of a homeotropically aligned nematic-liquid-crystal cell in the regime of low acoustic intensity J behaves as $M \propto J^2$ [30,31], which gives a good fit to experimental data (see also Fig. 1 below). The correctness of this behavior poses a difficulty to a theory based on the mechanism of streaming [17–20], since it predicts $M \propto J^4$. Despite this, in an effort to give support to the streaming theory, the quantity $J_{\max}(\varphi)$ [defined by the smallest root of $M(J) = 1$] for small angle of incidence φ was measured [21]. The agreement between theory and experiment was good but, and this is a point to be explored later, the significant deviations from the streaming-theory prediction, namely,

$$J_{\max} = \frac{H}{\varphi}, \tag{1}$$

where $H = 2.468\,\mathrm{rad\,mW/cm^2}$ is the best-fitting parameter obtained according to the least-squares method (see below), are entirely explained by the director-density coupling theory. As a consequence, we have come to the conclusion that the interaction between ultrasonic wave and nematic liquid crystals is in fact dominated by an elastic energy in the regime of low J. This is the main result of the paper and for this reason we invert the order of presentation; i.e., we derive the term of $O(J^3)$ for $M(J)$ in Sec. III but anticipate the use of $J_{\max}(\varphi)$ in Sec. II. Finally, we present our concluding remarks in Sec. IV.

II. ELASTIC REGIME

In this section, we should like to make an attempt to conciliate both scenarios presented in the Introduction. To accomplish this, we have extended our previous analysis [30,31] one step forward to obtain

$$M = AJ^2 + BJ^3 + O(J^4), \tag{2}$$

where

$$A = A_1 \sin^4(2\varphi), \tag{3}$$

$$B = B_1 \sin^4(2\varphi) + B_2 \sin^3(2\varphi)\sin(4\varphi) + B_3 \sin^2(2\varphi)\sin^2(4\varphi) + B_4 \sin^6(2\varphi). \tag{4}$$

Here the coefficients A_1 and B_i's do not depend on φ and therefore we need not specify their dependence on other variable factors. However, we note in passing that the coefficient A_1 can be inferred from Eq. (43) of Ref. [30]. Equation (2) formalizes the hypothesis of the purely aligning effect of ultrasound on liquid crystals in the limit of low acoustic intensity [22]. What makes this expansion special is that it differs considerably from the signature coming purely from the streaming theory [17–20]:

$$M = \sin^2(B\Lambda J^2) = CJ^4 - \tfrac{1}{3}C^2 J^8 + O(J^{12}), \tag{5}$$

where $C \equiv (B\Lambda)^2$.

Armed with these expansions for $M(J)$, we have revisited the experimental data referred to above (see Table I) in order to explain the curve M versus J up to $J = J_{\max}$ (see, however, below). In Fig. 1 we plot Eqs. (2) and (5) with their parameters obtained by the method of least squares for $i = 1,2,3,4$, and 5. The coefficients are given by

$$A = 4.132 \times 10^{-3}\ (\mathrm{mW/cm^2})^{-2}, \tag{6}$$

$$B = 1.115 \times 10^{-4}\ (\mathrm{mW/cm^2})^{-3}, \tag{7}$$

$$C = 1.436 \times 10^{-4}\ (\mathrm{mW/cm^2})^{-4}. \tag{8}$$

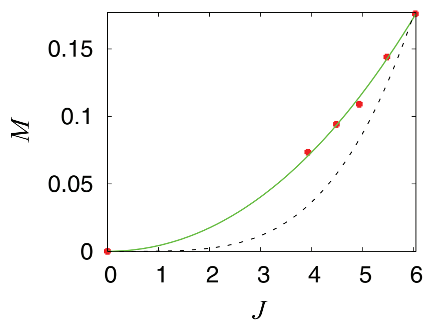

FIG. 1. (Color online) Optical transparency versus ultrasonic intensity in units of $\mathrm{mW/cm^2}$. The solid (dashed) curve is the plot of $M = AJ^2 + BJ^3$ ($M = CJ^4 - \frac{1}{3}C^2J^8$). The dots displayed were obtained from Table I. The best-fitting parameters are given in the text.

The agreement between theory, Eq. (2), and experiment displayed in this figure is impressive. Therefore, there can be no doubt regarding the elastic regime from the experimental point of view.

In the following, we show that the director-density coupling theory also provides the framework, via Eq. (2), for explaining the experimental data for $J_{\max}$ versus φ (see Table II). Keeping only terms of $O(\varphi^4)$, one gets

$$\varphi^4\left(A' J_{\max}^2 + B' J_{\max}^3\right) = 1, \tag{9}$$

where we have absorbed the constants arising from the use of Taylor expansion for $\sin(\cdots)$ into the new constants A' and B'. The solution of the above equation reads

$$J_{\max} = -\frac{A'}{3B'} + W - \frac{p}{3W}, \tag{10}$$

where

$$W = \sqrt[3]{-\frac{q}{2} - \sqrt{\frac{q^2}{4} + \frac{p^3}{27}}}, \tag{11}$$

TABLE I. Experimental data for optical transparency versus ultrasonic intensity in units of $\mathrm{mW/cm^2}$ [from Ref. [21], Fig. 2(a)]. The first five values of M_i in the fourth and fifth columns are calculated using the coefficients given by Eqs. (6)–(8).

i	J_i	M_i	M_i [Eq. (2)]	M_i [Eq. (5)]
1	3.93	0.0735	0.0706	0.0338
2	4.49	0.0941	0.0934	0.0572
3	4.94	0.109	0.114	0.0831
4	5.48	0.144	0.142	0.124
5	6.04	0.176	0.175	0.179
6	7.04	0.353		
7	8.07	0.541		
8	9.09	0.788		
9	9.66	0.882		
10	10.11	0.941		
11	10.57	1.00		

TABLE II. Experimental data for $J_{\max}$ in units of $\mathrm{mW/cm^2}$ versus angle of incidence φ in radians [from Ref. [21], Fig. 3(a)]. In the third and fourth columns we show the calculated values for $J_{\max}$.

φ	$J_{\max}$	$J_{\max}$ [Eq. (1)]	$J_{\max}$ [Eq. (10)]
0.07913	33.68	31.19	37.26
0.07183	39.79	34.36	39.47
0.06288	45.05	39.26	43.31
0.05366	49.47	46.00	49.29
0.04425	58.95	55.78	59.01
0.03523	75.37	70.06	74.81
0.02668	89.26	92.52	102.48
0.01757	134.1	140.49	169.89

$$p = -\frac{1}{3}\left(\frac{A'}{B'}\right)^2, \tag{12}$$

$$q = \frac{2}{27}\left(\frac{A'}{B'}\right)^3 - \frac{1}{B'\varphi^4}. \tag{13}$$

The plot of Eq. (10) is shown in Fig. 2 for $A'/B' = -30.2$ and $B' = 2.60$ (obtained manually). For the sake of comparison, we plot together Eq. (1) and highlight the experimental data. For $J_{\max} \leqslant 75\ \mathrm{mW/cm^2} \equiv J_S$, the agreement between theoretical prediction and experimental results is remarkable. On the other hand, for $J > J_S$, i.e, high acoustic intensity, the agreement is poor, indicating that the mechanism of streaming dominates the physics. Therefore, the validity of describing the acousto-optic effect using an elastic energy is restricted to the regime $J \leqslant \min(J_{\max}, J_S)$, where $\min(\cdots)$ is the notation for the minimum value between $J_{\max}$ and J_S.

III. OPTICAL TRANSPARENCY

In this section, we are interested in obtaining the term of $O(J^3)$ in Eq. (2) from the director-density coupling theory. In Fig. 3, it is shown a homeotropically aligned nematic-liquid-crystal cell of thickness a under the action of an incident monochromatic ultrasonic plane wave of wave vector $\mathbf{k}$ (making an angle φ with the z direction) and frequency ω. Under the same conditions described in Ref. [30], the dynamics of the liquid-crystal director $\hat{\mathbf{n}}(\mathbf{r}) = \hat{\mathbf{y}}\sin\theta + \hat{\mathbf{z}}\cos\theta$

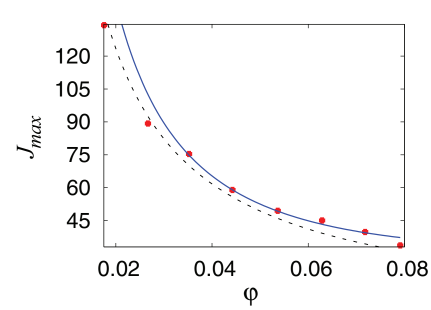

FIG. 2. (Color online) First maximum of $M(J)$ in units of $\mathrm{mW/cm^2}$ as a function of incidence angle in radians. The solid (dashed) curve is the plot of Eq. (10) [Eq. (1)]. The dots displayed were obtained from Table II.

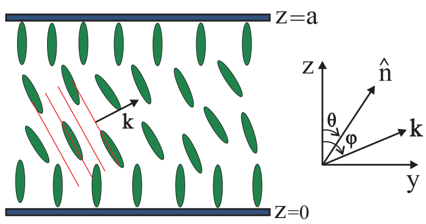

FIG. 3. (Color online) Cell of thickness a containing homeotropically aligned nematic liquid crystal in the presence of an incident ultrasonic wave. On the right, the coordinate system defines the angles θ and φ.

is determined by the equation

$$\gamma\frac{\partial\theta}{\partial t} = (K_1\sin^2\theta + K_3\cos^2\theta)\left(\frac{\partial^2\theta}{\partial z^2}\right) + \frac{1}{2}(K_1-K_3)\left(\frac{\partial\theta}{\partial z}\right)^2\sin(2\theta) + u_1(\Delta\rho)k^2\sin 2(\theta-\varphi)\sin(\omega t) + u_2(\Delta\rho)^2k^2\sin 2(\theta-\varphi)\cos^2(\omega t), \quad (14)$$

where γ is the rotational viscosity coefficient; K_1 and K_3 are, respectively, the Frank constants for splay and bend [38];

$$J = \frac{v^3(\Delta\rho)^2}{2\rho_0} \quad (15)$$

is the acoustic intensity (here, ρ_0 is the average density and v is the sound velocity); and, finally, u_1 and u_2 are parameters of the theory. We are assuming strong-anchoring boundary conditions:

$$\theta(0,t) = \theta(a,t) = 0. \quad (16)$$

In the steady state and for $\varphi \neq 0$, $\theta(z,t)$ is expressed as a power series in $\Delta\rho$ [30]:

$$\theta(z,t) = (\Delta\rho)\theta^{(1)}(z,t) + (\Delta\rho)^2\theta^{(2)}(z,t) + (\Delta\rho)^3\theta^{(3)}(z,t) + O(\Delta\rho)^4. \quad (17)$$

We have already calculated the expressions for $\theta^{(1)}$ and $\theta^{(2)}$. For our purpose, it suffices to underline their dependence on φ and t:

$$\theta^{(1)}(z,t) = \sin(2\varphi)[f_1(z)\cos(\omega t) + f_2(z)\sin(\omega t)], \quad (18)$$

$$\theta^{(2)}(z,t) = \sin(2\varphi)f_3(z) + \sin(4\varphi)f_4(z) + [\sin(2\varphi)f_5(z) + \sin(4\varphi)f_6(z)]\cos(2\omega t) + [\sin(2\varphi)f_7(z) + \sin(4\varphi)f_8(z)]\sin(2\omega t). \quad (19)$$

Notwithstanding, the exact expressions for the functions f_i's can be easily obtained from Eqs. (21)–(23) and Eqs. (26)–(29) of Ref. [30] and further comparison with the expressions written above. We note that this expedient method will be used in order to simplify the calculation of $\theta^{(3)}$. In the stationary state, this function satisfies

$$\gamma\frac{\partial\theta^{(3)}}{\partial t} = K_3\frac{\partial^2\theta^{(3)}}{\partial z^2} + (K_1-K_3)(\theta^{(1)})^2\frac{\partial^2\theta^{(1)}}{\partial z^2} + (K_1-K_3)\theta^{(1)}\left(\frac{\partial\theta^{(1)}}{\partial z}\right)^2 + 2u_1k^2[\theta^{(2)}\cos(2\varphi) + 2(\theta^{(1)})^2\sin(2\varphi)]\sin(\omega t) + 2u_2k^2\theta^{(1)}\cos(2\varphi)\cos^2(\omega t). \quad (20)$$

We now insert Eqs. (18) and (19) into Eq. (20) and, after a little algebra, find that it can be cast in the form

$$\gamma\frac{\partial\theta^{(3)}}{\partial t} = K_3\frac{\partial^2\theta^{(3)}}{\partial z^2} + [h_1(z)\sin^3(2\varphi) + h_2(z)\sin(4\varphi) + h_3(z)\cos(2\varphi)\sin(4\varphi)]\cos(\omega t) + [h_4(z)\sin^3(2\varphi) + h_5(z)\sin(4\varphi) + h_6(z)\cos(2\varphi)\sin(4\varphi)]\sin(\omega t) + [h_7(z)\sin^3(2\varphi) + h_8(z)\sin(4\varphi) + h_9(z)\cos(2\varphi)\sin(4\varphi)]\cos(3\omega t) + [h_{10}(z)\sin^3(2\varphi) + h_{11}(z)\sin(4\varphi) + h_{12}(z)\cos(2\varphi)\sin(4\varphi)]\sin(3\omega t), \quad (21)$$

where the function h_i's are related to the functions f_i's. It will be sufficient, however, to observe that they can be expanded as Fourier series:

$$h_i(z) = \sum_{n=1}^{\infty} B_n^{(i)}\sin\left(\frac{n\pi z}{a}\right). \quad (22)$$

We now write $\theta^{(3)}$ in the steady state as

$$\theta^{(3)}(z,t) = \sum_{n=1}^{\infty}[J_n\cos(\omega t) + L_n\sin(\omega t) + M_n\cos(3\omega t) + N_n\sin(3\omega t)]\sin\left(\frac{n\pi z}{a}\right) \quad (23)$$

and substitute this expression, together with Eq. (22), into Eq. (21). After some calculation, we find that

$$X_n = X_n^{(1)}\sin^3(2\varphi) + X_n^{(2)}\sin(4\varphi) + X_n^{(3)}\cos(2\varphi)\sin(4\varphi). \quad (24)$$

Here, X stands for J,L,M, and N. Inserting these formulas back into Eq. (23), it is easy to verify that

$$\theta^{(3)}(z,t) = [f_9(z)\sin^3(2\varphi) + f_{10}(z)\sin(4\varphi) + f_{11}(z)\cos(2\varphi)\sin(4\varphi)]\cos(\omega t) + [f_{12}(z)\sin^3(2\varphi) + f_{13}(z)\sin(4\varphi) + f_{14}(z)\cos(2\varphi)\sin(4\varphi)]\sin(\omega t) + [f_{15}(z)\sin^3(2\varphi) + f_{16}(z)\sin(4\varphi) + f_{17}(z)\cos(2\varphi)\sin(4\varphi)]\cos(3\omega t) + [f_{18}(z)\sin^3(2\varphi) + f_{19}(z)\sin(4\varphi) + f_{20}(z)\cos(2\varphi)\sin(4\varphi)]\sin(3\omega t). \quad (25)$$

We are now in a position to calculate the optical transparency, which is defined by [38]

$$M(t) = \sin^2(\Gamma/2), \quad (26)$$

where

$$\Gamma(t) = \frac{2\pi}{\lambda}\int_0^a(\Delta n_{\text{eff}})\,dz \quad (27)$$

is the retardation; λ is the wavelength of the light;

$$\Delta n_{\text{eff}} = \frac{n_o n_e}{\sqrt{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta}} - n_o = \frac{n_o\left(n_e^2 - n_o^2\right)}{2n_e^2} \times \left[\sin^2\theta + \frac{3\left(n_e^2 - n_o^2\right)}{4n_e^2}\sin^4\theta + O(\sin^6\theta)\right] \quad (28)$$

is the effective birefringence; and n_o and n_e are, respectively, the ordinary and extraordinary refractive index. In this expression, one has

$$\sin^2\theta = (\Delta\rho)^2(\theta^{(1)})^2 + 2(\Delta\rho)^3\theta^{(1)}\theta^{(2)} + (\Delta\rho)^4\left[2\theta^{(1)}\theta^{(3)} + (\theta^{(2)})^2 - \tfrac{1}{3}(\theta^{(1)})^4\right] + O(\Delta\rho)^5, \quad (29)$$

$$\sin^4\theta = (\Delta\rho)^4(\theta^{(1)})^4 + O(\Delta\rho)^5. \quad (30)$$

In the following we insert Eq. (28) into Eq. (27) and evaluate the integration with respect to z. Thus by keeping the aim of neglecting irrelevant information, the final result is

$$\Gamma(t) = (\Delta\rho)^2\Gamma_1(\varphi,t) + (\Delta\rho)^3\Gamma_2(\varphi,t) + (\Delta\rho)^4\Gamma_3(\varphi,t) + O(\Delta\rho)^5, \quad (31)$$

where

$$\Gamma_1(\varphi,t) = \sin^2(2\varphi)[\gamma_1 + \gamma_2\cos(2\omega t) + \gamma_3\sin(2\omega t)], \quad (32)$$

$$\begin{aligned}\Gamma_2(\varphi,t) = {} & [\gamma_4\sin^2(2\varphi) + \gamma_5\sin(2\varphi)\sin(4\varphi)]\cos(\omega t) \\ & + [\gamma_6\sin^2(2\varphi) + \gamma_7\sin(2\varphi)\sin(4\varphi)]\sin(\omega t) \\ & + [\gamma_8\sin^2(2\varphi) + \gamma_9\sin(2\varphi)\sin(4\varphi)]\cos(3\omega t) \\ & + [\gamma_{10}\sin^2(2\varphi) + \gamma_{11}\sin(2\varphi)\sin(4\varphi)]\sin(3\omega t),\end{aligned} \quad (33)$$

$$\begin{aligned}\Gamma_3(\varphi,t) = {} & \gamma_{12}\sin^4(2\varphi) + \gamma_{13}\sin^2(4\varphi) \\ & + \gamma_{14}\sin^2(2\varphi) + \gamma_{15}\sin(2\varphi)\sin(4\varphi) \\ & + [\gamma_{16}\sin^4(2\varphi) + \gamma_{17}\sin^2(4\varphi) + \gamma_{18}\sin^2(2\varphi) \\ & + \gamma_{19}\sin(2\varphi)\sin(4\varphi)]\cos(2\omega t) \\ & + [\gamma_{20}\sin^4(2\varphi) + \gamma_{21}\sin^2(4\varphi) + \gamma_{22}\sin^2(2\varphi) \\ & + \gamma_{23}\sin(2\varphi)\sin(4\varphi)]\sin(2\omega t) \\ & + [\gamma_{24}\sin^4(2\varphi) + \gamma_{25}\sin^2(4\varphi) + \gamma_{26}\sin^2(2\varphi) \\ & + \gamma_{27}\sin(2\varphi)\sin(4\varphi)]\cos(4\omega t) \\ & + [\gamma_{28}\sin^4(2\varphi) + \gamma_{29}\sin^2(4\varphi) + \gamma_{30}\sin^2(2\varphi) \\ & + \gamma_{31}\sin(2\varphi)\sin(4\varphi)]\sin(4\omega t).\end{aligned} \quad (34)$$

The knowledge of $\Gamma(t)$ allows us to calculate $M(t)$ via Eq. (26):

$$M(t) = \tfrac{1}{4}\left[(\Delta\rho)^4\Gamma_1^2 + 2(\Delta\rho)^5\Gamma_1\Gamma_2 + (\Delta\rho)^6\left(2\Gamma_1\Gamma_3 + \Gamma_2^2\right) + O(\Delta\rho)^7\right]. \quad (35)$$

All we need do now is to take the time average of this quantity, which is defined by

$$\langle M\rangle \equiv \lim_{T\to\infty}\frac{1}{T}\int_0^T M(t)\,dt. \quad (36)$$

Using this definition, we find

$$\left\langle\Gamma_1^2\right\rangle = \overline{\gamma}_1\sin^4(2\varphi), \quad (37)$$

$$\langle\Gamma_1\Gamma_2\rangle = 0, \quad (38)$$

$$\left\langle 2\Gamma_1\Gamma_3 + \Gamma_2^2\right\rangle = \overline{\gamma}_2\sin^4(2\varphi) + \overline{\gamma}_3\sin^3(2\varphi)\sin(4\varphi) + \overline{\gamma}_4\sin^2(2\varphi)\sin^2(4\varphi) + \overline{\gamma}_5\sin^6(2\varphi). \quad (39)$$

We thus conclude the proof by eliminating $\Delta\rho$ in favor of J, Eq. (15), and by expressing the result in terms of the constants defined by Eqs. (3) and (4).

IV. CONCLUDING REMARKS

In conclusion, the aim of this paper is to confirm the assumption that the acousto-optic effect in nematic liquid crystals can be modeled by an elastic energy in the regime of low acoustic intensity. To fulfill this task, we have derived a Taylor expansion for $M(J)$ up to third order in J within the framework of the director-density coupling theory and stressed that it differs greatly from the corresponding version obtained under streaming condition. Next we have made use of experimental data for validating the theoretical prediction that $M \sim J^2$ and consequently for verifying that the mechanism of streaming plays minor role in the limit $J \to 0$. Moreover, we have used experimental data for J_{max} versus φ in order to investigate the range of validity of a purely elastic description of the acousto-optic effect. Thus we have found that above a certain critical value of the acoustic intensity, the underlying physics is dominated by the mechanism of streaming. Finally, we also have tentatively estimated the critical value of J at $\min(J_{\max}, J_S)$. However, we note that J_S cannot be obtained in the context of the director-density coupling theory.

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