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Influence of the liquid-crystal splay-bend surface elastic constant on director fluctuations and light scattering

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The influence of the surface elastic constant K_{13} on director fluctuations in nematic liquid crystals is discussed. An explicit expression for the correlation function in the case of the homeotropically aligned cell is presented. It is shown that in this case K_{13} changes the anchoring strength from W_0 to $K_{33}W_0/(K_{33}-K_{13})$ where K_{33} is the Frank constant. The dependence of the angular distribution of the light-scattering intensity on K_{13} and W_0 is analyzed. A possibility of measuring K_{13} by means of an optical experiment is discussed.

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Physical properties of nematic liquid crystals are of interest from the fundamental as well as practical point of view. Studies of such systems require detailed accounting for surface phenomena. The starting point of the description is an expression for the free energy as a function of the director $\mathbf{n}$. There are two reasons why the behavior of a nematic liquid crystal in a confined volume differs from an infinite sample. First, there is an interaction between the confining surface and the director, which gives rise to a surface free energy. Second, there are surface elastic contributions to the free energy, which are characterized by the surface elastic constants K_{24} and K_{13}, that are negligible in the case of an infinite sample. Although the surface elastic constants were introduced long ago, measurements of the saddle-splay elastic constant K_{24} were made only recently [1]. According to the Landau–de Gennes expansion of the free energy in terms of the tensor order parameter, the difference between the splay and bend elastic constants $K_{11}-K_{33}$ and the unzero splay-bend elastic constant K_{13} appear in the same approximation [2]. In most nematic liquid crystals [3], $K_{33}>K_{11}$ is realized. The ratio of the elastic constants of nematic liquid crystals has been calculated using a simple inner-field model. The obtained result is $K_{22}:K_{24}:K_{13}=11:-9:-6$ [4]. Hence there is no reason to consider *a priori* K_{13} to be equal to zero. Unfortunately, until now there are no experimental determinations of K_{13}. An attempt to measure K_{13} has been made by the authors of Ref. [5]. Only the upper limit of $|K_{13}|$ for nematic 7CB has been determined. It has been shown that $|K_{13}|<0.7K_{33}$. All evaluations of K_{13} based on theories, which require a minimization of the free energy, contain a serious difficulty. The Euler-Lagrange differential equation, which is only an obligatory condition, does not provide the extremum, when K_{13} is incorporated into the expression for the free energy [6]. A distortion of the director field in a few molecular layers near the boundary surface is expected in the case if the surface director is obliquely oriented with respect to the surface normal [7].

The aim of this paper is to provide a theoretical analysis, which is not connected with the variational problem and can provide a method to measure K_{13} by means of an optical experiment. It is known that a thermal fluctuation spectrum in a thin sample of aligned liquid crystal depends on the surface contribution to the free energy. This dependence modifies the angular distribution of the scattered light intensity [8]. The mixed splay-bend contribution is commonly omitted due to a weakness of deformations [1]. This cannot be done when short-wavelength fluctuations are considered. In this Brief Report the correlation function of the fluctuations including K_{13} and the anchoring strength is presented for a homeotropic aligned nematic layer. The light-scattering intensity is analyzed.

Let the homeotropic aligned nematic layer be confined between the two planes situated at $z=\pm L/2$ in the Cartesian coordinate system. The starting point of the description is the expression for the energy, which contains the surfacelike terms

$$F=\frac{1}{2}\int d^3r[K_{11}(\mathrm{div}\mathbf{n})^2+K_{22}(\mathbf{n}\cdot\mathrm{curl}\mathbf{n})^2+K_{33}(\mathbf{n}\times\mathrm{curl}\mathbf{n})^2-K_{24}\mathrm{div}(\mathbf{n}\times\mathrm{curl}\mathbf{n}+\mathbf{n}\,\mathrm{div}\mathbf{n})+K_{13}\mathrm{div}(\mathbf{n}\,\mathrm{div}\mathbf{n})]$$
$$+\frac{W_0}{2}\int dr_\perp[\mathbf{n}_\perp^2(\mathbf{r}_\perp,L/2)+\mathbf{n}_\perp^2(\mathbf{r}_\perp,-L/2)]\ , \qquad (1)$$

where $\mathbf{n}_\perp$ and $\mathbf{r}_\perp$ are the in-plane components of $\mathbf{n}$ and $\mathbf{r}$. The last term, proposed by Rapini and Papoular [9], accounts for the surface anchoring, which is assumed to be the same at both sides. Furthermore, we assume the equilibrium director $\mathbf{n}^0$ to be normal to the surfaces. This assumption is equal to one that a homeotropically aligned cell exists and does not contradict the statement made in Ref. [7]. Let us consider only small fluctuations of the director $\delta\mathbf{n}(\mathbf{r})=\mathbf{n}(\mathbf{r})-\mathbf{n}^0$. Since $\mathbf{n}$ is a unit vector, only two components, namely, δn_x and δn_y, are independent. The aim is to calculate the correlation function $G_{\alpha\beta}(\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')=\langle\delta n_\alpha(\mathbf{r})\delta n_\beta(\mathbf{r}')\rangle$. Here the statistical average $\langle\ \rangle$ is taken with respect to $\delta\mathbf{n}(\mathbf{r})$, i.e.,

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$$\langle \cdots \rangle = \int D\,\delta\mathbf{n} \cdots \times \exp\left[-\frac{\Delta F}{k_B T}\right] \Big/ \int D\,\delta\mathbf{n} \exp\left[-\frac{\Delta F}{k_B T}\right], \quad (2)$$

where ΔF is the deviation of the free energy associated with $\delta\mathbf{n}(\mathbf{r})$. Using the Gaussian approximation we must keep the quadratic contributions to ΔF and drop the others. Equation (1) allows one to obtain the expression

$$\Delta F = \frac{1}{2}\int d^2 r_\perp \left\{ \int_{-L/2}^{L/2} dz [K_{11}(\mathrm{div}\,\delta\mathbf{n}_\perp)^2 + K_{22}(\partial_x \delta n_y - \partial_y \delta n_x)^2 - K_{33}\delta\mathbf{n}_\perp \cdot \partial_z^2 \delta\mathbf{n}_\perp] + W_0[\delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, L/2) + \delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, -L/2)] + (K_{33}-K_{13})[\delta\mathbf{n}_\perp(\mathbf{r}_\perp, L/2)\cdot\partial_z \delta\mathbf{n}_\perp(\mathbf{r}_\perp, L/2) - \delta\mathbf{n}_\perp(\mathbf{r}_\perp, -L/2)\cdot\partial_z\delta\mathbf{n}_\perp(\mathbf{r}_\perp, -L/2)] \right\}. \quad (3)$$

Such terms as $\partial_x \delta\mathbf{n}(\mathbf{r}_\perp, \pm L/2)$ and $\partial_y \delta\mathbf{n}(\mathbf{r}_\perp, \pm L/2)$ have been omitted in Eq. (3), since they are small and therefore negligible, when the thickness L is much smaller than the extension of the sample in the xy plane. The term proportional to K_{24} in Eq. (1) does not effect ΔF for small $\delta\mathbf{n}$ due to this contribution being negligible by the same argument. It should be noted that if we used the common assumption $(\delta\mathbf{n}\cdot\mathbf{n}^0)=0$, we would get a wrong contribution to ΔF from the surfacelike terms and the right ones from the others. To get the right contribution one should set $\delta n_z = (1-\mathbf{n}_\perp^2)^{1/2}$.

The usual routine for a calculation of the correlation function is to take the Fourier transform with respect to $\mathbf{r}$ and then to apply the equipartition theorem. Unfortunately, the surface terms in Eq. (3) do not permit us to use this theorem. The same situation is considered in Ref. [10]. The idea of how to overcome this obstacle can be formulated as follows. One should find a class of functions $\delta\mathbf{n}(\mathbf{r})$, such that $\Delta F = \frac{1}{2}(\delta\mathbf{n}, \hat{A}\,\delta\mathbf{n})$ with $\hat{A}$ being a self-adjoint operator acting on these functions. It does not imply any restriction on the consideration of the fluctuations, because any function can be approximated by a function from the class with arbitrary accuracy (the eigenfunctions of $\hat{A}$ comprise a basis). When ΔF is a quadratic form, the integrals in Eq. (2) can be calculated in standard manner, which leads to $\langle \delta\mathbf{n}\otimes\delta\mathbf{n}\rangle = K_B T \hat{A}^{-1}$. If $\mathbf{n}^0$ is an equilibrium field, $\hat{A}^{-1}$ exists.

If the fluctuation $\delta\mathbf{n}(\mathbf{r})$ satisfies the boundary conditions

$$w\,\delta\mathbf{n}_\perp(\mathbf{r}_\perp, \pm L/2) \pm \partial_z \delta\mathbf{n}_\perp(\mathbf{r}_\perp, \pm L/2) = 0, \quad (4)$$

with $w = W_0/(K_{33}-K_{13})$, then the contribution of the surface terms vanishes and Eq. (3) transforms into a quadratic form. Now it is convenient to take the Fourier transform of $\delta\mathbf{n}(\mathbf{r}_\perp, z)$ with respect to $\mathbf{r}_\perp$:

$$\delta\mathbf{n}(\mathbf{v}, z) = \int d^2 r_\perp \exp(-i\mathbf{r}_\perp\cdot\mathbf{v})\delta\mathbf{n}(\mathbf{r}_\perp, z). \quad (5)$$

ΔF can be presented as the integral over $\mathbf{v}$,

$$\Delta F = \frac{1}{(2\pi)^2}\int d^2 v\, \Delta F_{\mathbf{v}}, \quad (6)$$

where $\Delta F_{\mathbf{v}}$ is the contribution to the free energy associated with the fluctuation $\delta\mathbf{n}(\mathbf{v}, z)$

$$\Delta F_{\mathbf{v}} = \frac{1}{2}\int_{-L/2}^{L/2} dz\,(n_x^*, n_y^*)\hat{A}\begin{bmatrix} n_x \\ n_y \end{bmatrix}, \quad (7)$$

with

$$\hat{A} = \begin{bmatrix} v_x^2 K_{11} + v_y^2 K_{22} - K_{33}\partial_z^2 & (K_{11}-K_{22})v_x v_y \\ (K_{11}-K_{22})v_x v_y & v_y^2 K_{11} + v_x^2 K_{22} - K_{33}\partial_z^2 \end{bmatrix}. \quad (8)$$

One can prove that the operator $\hat{A}$ acting on the functions which satisfy the boundary conditions (4) is a self-adjoint one [11]. The Fourier component $\hat{G}(\mathbf{v}, z, z')$ of $\hat{G}(\mathbf{r}_\perp - \mathbf{r}'_\perp, z, z')$ must satisfy the equation

$$\hat{A}\hat{G} = k_B T \hat{I}\delta(z-z'), \quad (9)$$

where $\hat{I}$ is the identity matrix. Without any restriction we can assume $\mathbf{v}$ to be normal to the y axis and denote the corresponding correlation function by $\hat{G}'(\mathbf{v}, z, z')$. $\hat{G}(\mathbf{v}, z, z')$ for arbitrary direction of $\mathbf{v}$ is a result of rotation round the z axis. Combining Eqs. (8) and (9) and taking into account the boundary conditions (4) we get that the nondiagonal elements of the new matrix $\hat{G}'$ are equal to zero while the diagonal elements $G'_{11}(i=1,2)$ must satisfy the equations

$$(\alpha_i^2 - \partial_z^2)G'_{ii}(\mathbf{v}, z, z') = k_B T \delta(z-z'), \quad (10)$$

with $\alpha_i = (K_{ii}/K_{33})^{1/2} v$. The associated boundary conditions are

$$w G'_{ii}(\mathbf{v}, \pm L/2, z') \pm \partial_z G'_{ii}(\mathbf{v}, \pm L/2, z') = 0. \quad (11)$$

If w is negative, Eq. (10) with the right-hand side equal to zero has a solution from the defined class for some α_i. It means that the homeotropic alignment is not stable, when W_0 and $K_{33}-K_{13}$ have opposite signs. When $z \neq z'$ we get zero in the right-hand side of Eq. (10). There are only two independent solutions $\exp(\alpha_i z)$ and $\exp(-\alpha_i z)$. The solution to this problem can be expressed through solutions to Eq. (10) with zero right-hand side $u_+^{(i)}$ and $u_-^{(i)}$, which satisfy Eqs. (11) when $z = L/2$ and $z = -L/2$, respectively [12]

$$\hat{G}'_{ii}(\mathbf{v},z,z')=\frac{k_B T}{K_{33}(u_+^{(i)}\partial_z u_-^{(i)}-u_-^{(i)}\partial_z u_+^{(i)})}\times\begin{cases}u_+^{(i)}(z)u_-^{(i)}(z') & \text{if } z>z'\\ u_-^{(i)}(z)u_+^{(i)}(z') & \text{if } z<z' .\end{cases} \tag{12}$$

Now it is not difficult to find functions $u_\pm^{(i)}(z)$. For example, the following expressions are suitable:

$$u_\pm^{(i)}(z)=(\alpha_i\mp w)\exp[\alpha_i(z\mp L/2)]+(\alpha_i\pm w)\exp[-\alpha_i(z\mp L/2)] . \tag{13}$$

Using Eq. (13) in (12), we have

$$G'_{ii}(\mathbf{v},z,z')=\frac{k_B T}{2\alpha_i K_{33}\Delta_i}\{(\alpha_i^2-w^2)\cosh[\alpha_i(z+z')] + [(\alpha_i^2+w^2)\cosh(\alpha_i L)+2\alpha_i w\sinh(\alpha_i L)]\cosh[\alpha_i(z-z')]-\Delta_i\sinh(\alpha_i|z-z'|)\} , \tag{14}$$

where

$$\Delta_i=(\alpha_i^2+w^2)\sinh(\alpha_i L)+2w\alpha_i\cosh(\alpha_i L) .$$

It is easy to notice that expression (14) evolves into an expression for an infinite nematic, when the thickness L is large.

To analyze the influence of the mixed splay-bend free energy on the light-scattering process, let us consider the director-associated fluctuations $\delta\varepsilon_{\alpha\beta}(\mathbf{r})$ of the dielectric tensor

$$\delta\varepsilon_{\alpha\beta}(\mathbf{r})=\varepsilon_a(n_\alpha^0\delta n_\beta(\mathbf{r})+n_\beta^0\delta n_\alpha(\mathbf{r})) , \tag{15}$$

where $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$, $\varepsilon_\parallel$ and $\varepsilon_\perp$ being the permittivities along and transverse to $\mathbf{n}$. The intensity of the scattered light can be expressed in terms of the function $\langle E'_\alpha(\mathbf{r})E'^*_\beta(\mathbf{r})\rangle$, which in the Born approximation is given by

$$\langle E'_\alpha(\mathbf{r})E'^*_\beta(\mathbf{r})\rangle=\frac{\omega^4}{c^4}\int d^3r'd^3r''T_{\alpha\gamma}(\mathbf{r},\mathbf{r}')T^*_{\beta\lambda}(\mathbf{r},\mathbf{r}'')\langle\delta\varepsilon_{\gamma\mu}(\mathbf{r}')\delta\varepsilon_{\lambda\nu}(\mathbf{r}'')\rangle \times E_\mu^0E_\nu^0\exp[\mathbf{k}_i(\mathbf{r}'-\mathbf{r}'')] . \tag{16}$$

Here $\mathbf{E}^0$ and $\mathbf{k}_{(i)}$ are the amplitude and wave vector of the incident light, $\hat{T}(\mathbf{r},\mathbf{r}')$ is the Green's function for an optically anisotropic medium, taking the boundaries into account. The detailed analysis of the multireflection and the optical anisotropy effects in the case of a homeotropically aligned liquid-crystal cell has already been presented [14]. We assume the Green's function to be one for a far zone in an infinite medium with permittivity $\varepsilon=(\varepsilon_\perp+\varepsilon_\parallel)/2$ in order to simplify our calculation

$$T_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|}\exp(ik|\mathbf{r}-\mathbf{r}'|)(\delta_{\alpha\beta}-s_\alpha s_\beta) , \tag{17}$$

where $k=\sqrt{\varepsilon}\omega/c$ and $\mathbf{s}=(\mathbf{r}-\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|$.

Keeping in mind that ordinary waves are not scattered into ordinary ones by director fluctuations and scattering of extraordinary waves to extraordinary ones is strongest in nematic liquid crystals [13], we consider only case where the incident wave and the scattered waves are polarized in the scattering plane. We assume $\mathbf{s}=(\sin\theta_s,0,\cos\theta_s)$ and $\mathbf{k}_i=k(\sin\theta_i,0,\cos\theta_i)$. The angles θ_i and θ_s show the propagation directions in the xz plane of the incident and the scattered waves, respectively. The extraordinary scattered waves are polarized along $\mathbf{m}=(\cos\theta_s,0,-\sin\theta_s)$. The intensity I of these waves can be written as follows:

$$I=\frac{c\sqrt{\varepsilon}}{8\pi}m_\alpha m_\beta\langle E'_\alpha E'^*_\beta\rangle . \tag{18}$$

Using Eqs. (15) and (18) in Eq. (16) and carrying out the integration over $\mathbf{r}'_\perp$, $\mathbf{r}''_\perp$, taking into account that $|\mathbf{r}|\gg|\mathbf{r}'|,|\mathbf{r}''|$ gives rise to

$$I=I_0\frac{\omega^4V\varepsilon_a^2}{c^4(4\pi r)^2}\sin^2(\theta_i+\theta_s)\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''\exp[-iq_z(z'-z'')]G'_{11}(\mathbf{q}_\perp,z,z') , \tag{19}$$

where I_0 is the incident wave intensity, the scattering vector $\mathbf{q}=k\mathbf{s}-\mathbf{k}_{(i)}$ and has components $q_x=k(\sin\theta_s-\sin\theta_i)$, $q_y=0$, and $q_z=k(\cos\theta_s-\cos\theta_i)$; V is the illuminated volume. The correlation function in Eq. (19) is defined by Eq. (14). The double integral in the right-hand side of Eq. (19) can be calculated analytically for arbitrary K_{ii}, K_{13}, and W_0. The final result is

$$I(\theta_s)=I_0C\sin^2(\theta_i+\theta_s)\{(w+g)[2q_z^2-2wg+L(w+g)(g^2+q_z^2)]+4ge^{-gL}[(w^2-q_z^2)\cos(Lq_z)-2wq_z\sin(Lq_z)] + (g-w)e^{-2gL}[2wg+2q_z^2+L(w-g)(g^2+q_z^2)]\} \times\{L(g^2+q_z^2)^2[(w+g)^2-(w-g)^2e^{-2gL}]\}^{-1} , \tag{20}$$

where

$$g=(K_{11}/K_{33})^{1/2}q_\perp ,$$
$$C=\frac{k_B T V\omega^4\varepsilon_a^2}{K_{33}c^4(4\pi r)^2} .$$

Consider separately the case of normal incidence ($\theta_i=0$). In this case the angle θ_s is equal to the scattering angle. When L is sufficiently large, expression (20) becomes the well-known expression for an infinite sample [13],

$$I(\theta_s)=I_0\frac{K_{33}C\sin^2(\theta_s)}{K_{33}q_z^2+K_{11}q_\perp^2} . \tag{21}$$

According to Eq. (21) the $I(0)$ (forward scattering) is finite due to the Goldstone fluctuations. A different situation takes place in the case of the finite cell. When the anchoring strength is not equal to zero, $\langle n_x(\mathbf{q}_\perp,z')n_x^*(\mathbf{q}_\perp,z'')\rangle$ is finite at the point $\mathbf{q}_\perp=0$; therefore the coefficient $\sin^2(\theta_s)$ in Eq. (20) leads to zero forward scattering. It happens only if $\theta_i=0$. Such a situation is described in detail in Refs. [8,14]. The intensity angular distribution $I(\theta_s)$ is presented in Fig. 1 for $\theta_i=\pi/4$, $kL=100$, $K_{11}/K_{33}=0.7$, $W_0=5.0\times10^{-3}$ dyne/cm, and $K_{13}/K_{33}=-1.0,0.0,1.0$. It is seen that the variation of the intensity with the scattering angle depends strongly on the ratio K_{13}/K_{33}. The scattering profile in Fig. 1 for $K_{13}/K_{33}=1.0$ according to Eq. (4) is that of the strong regime. This case hardly takes place in practice because such a magnitude of K_{13} corresponds to the boundary of the stability.

To estimate the possibility of measuring K_{13} by means of a light-scattering experiment, it is appropriate to analyze qualitatively the variation of the intensity with W_0 and K_{13}. One can see from the boundary conditions (4) the introduction of the splay-bend term into the free energy leads to the change of the anchoring strength from W_0 to $K_{33}W_0/(K_{33}-K_{13})$. Hence K_{13} is important only if W_0 is finite. Meanwhile, the less the anchoring strength is, the larger the range of the variation of w. Hence the angular distribution is more sensitive to K_{13} for small W_0 than for large. Unfortunately, the anchoring strength must be determined by an independent experiment. If the light-scattering experiment is ideal, i.e., it gives the precise magnitude of w, the errors of K_{13} and W_0 are related by $\Delta K_{13}=\Delta W_0/w$.

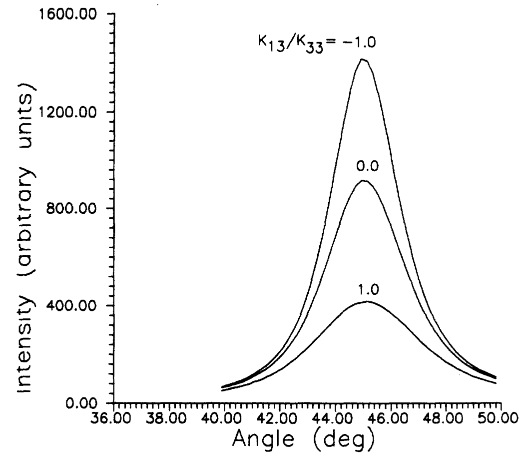

FIG. 1. Graph of the scattered-light intensity vs angle θ_s for a homeotropically aligned sample with finite size in the z direction and finite anchoring strength. Calculation was carried out on the basis of Eq. (20) for $\theta_i=\pi/4$, $W_0=5.0\times10^{-3}$ dyne/cm, $K_{11}/K_{33}=0.7$, $K_{33}=10^{-6}$ dyne, $k=10^5$ cm^{-1}, $L=10^{-3}$ cm.

The presented theory is for cells homogeneous in the xy plane. Apart from the thermal fluctuations there are stationary inhomogeneities of the anchoring strength and the thickness in a real cell. They also give a contribution to the light-scattering intensity, but this contribution is time independent and can be separated from the one of interest by a statistical routine. The question is when can we consider this contribution as an additive one. An exact answer to this question requires a detailed analysis, a few interesting aspects of which can be found in Ref. [15]. In a successful experiment the random deviations of the anchoring strength and the thickness must be much less than their average magnitudes.

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