Fluctuations and light scattering in free-standing smectic-C films

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The Goldstone mode of the director fluctuations in free-standing smectic-C liquid crystal films is studied theoretically. The system considered is three dimensional but of finite thickness. The response function of the system and the correlation function of the fluctuations are presented in a compact form. The theory is used to analyze light scattering experiments. It is shown that a two-dimensional model for very thin films can be obtained from one for a bulk system by minimizing over q_z rather than setting $q_z = 0$. This leads to some correction to the effective bend elastic constant of a film.

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I. INTRODUCTION

In studying the elastic and dynamical properties of liquid crystals, light scattering can be an enormously useful and powerful tool [1]. This is because of the optical anisotropy and very strong thermal orientational fluctuations associated with liquid crystals. Such techniques have long been used to investigate bulk systems, but more recently considerable progress has been made in applying these methods to bounded liquid crystals such as cells and films.

A free-standing smectic-C film can be imagined as a stack of layers, each of which constitutes a two-dimensional liquid consisting of long molecules. The director $\mathbf{n}$ (the preferred orientation of the molecules) is tilted with respect to the layer normal by a polar angle θ. Random thermal deviations of θ from its average are small. An azimuthal angle φ defines an orientation of the tilt plane and its fluctuations are of the Goldstone type. These fluctuations destroy the long-range order in a two-dimensional system.

This system is attractive from a fundamental point of view. First, the number of layers can be varied from 2 to several hundred allowing one to study experimentally the dependence of the physical properties on the thickness. Second, there are well-developed fluctuations of the director that cause strong light scattering [2].

Free-standing films have been under extensive experimental as well as theoretical study since the 1970s. The main attention has been focused on chiral smectic-C films. Apart from molecular ordering they exhibit spontaneous polarization. The polarization vector is perpendicular to the tilt plane. In addition to elastic energy there is also long-range interaction between charges created by nonuniform fluctuations. The first measurement of the polarization, elastic constants, and viscosities was carried out by means of light scattering by Rosenblatt *et al.* [3]. It was shown that the elastic properties of the film can be characterized by two constants K^S and K^B, responsible for splay and bend deformations, respectively, and that the long-range interaction effectively gives rise to a term in the elastic energy linear in q. The magnitude of the two elastic constants and their dependence on the thickness of the film L, apart from very thin films, can be estimated as $K^{S,B} \approx LK$, with K a typical bulk elastic constant.

The behavior of free-standing smectic-C films in the vicinity of the phase transition to smectic-A liquid crystal has been studied theoretically using the Kosterlitz-Thouless model by Heinekamp and Pelcovits [4]. These predictions have been checked by Amador and Pershan [5] via light-scattering and ellipsometry studies. Light scattering related to the azimuthal angle fluctuations have been studied by Lu *et al.* [6] using a chiral smectic film thick enough for finite-size effects to be irrelevant. Angular dependence in this case is characterized by four elastic constants B_1, B_2, B_3, and B_{13}. These authors showed that the long-range interaction between induced charges leads to an effective change in the elastic coefficient B_1.

A detailed experimental and theoretical study of nonchiral smectic films has been presented by Sprunt *et al.* [7]. The case of small film thickness was discussed in terms of natural modes of out-of-plane distortions. It is shown that if the thickness is less than the wavelength of the light, only the main natural distortion need be taken into account. In fact, this contribution depends on the boundary conditions at the interfaces. To the author's best knowledge, such a dependence has not yet been studied.

There have been many recent studies of surface phenomena in free-standing smectic films. An unusual surface stripe state has been observed by Demikhov [8]. This state is believed to be due to the chiral symmetry breaking at the interfaces [9]. The surface tension has been measured via x-ray scattering from thermal layer undulations by Shindler *et al.* [10].

In view of these studies, interesting theoretical avenues

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include the following. First, the issue of how the three-dimensional system, described by the four elastic constants, evolves into a two-dimensional one described only by two elastic constants. Second, it is instructive to assess the way in which the interfaces affect the correlation function describing fluctuations of the c director. Third, one expects that light scattering data will exhibit interesting surface-driven effects.

We note in addition that the correlation function is connected by the fluctuation-dissipation theorem [11] to the response function (generalized susceptibility) describing reaction of the system to external action. Thus the correlation function is of importance in undertaking external-field related applications.

The plan of the paper is as follows. In Sec. II the correlation of azimuthal angle fluctuations is described in terms of the natural modes as well as the response function. In Sec. III, which is devoted to the light scattering process, compact expressions for angular and frequency distributions of scattered light intensity are derived. The results obtained in this paper are summarized in Sec. IV.

II. CORRELATION FUNCTION

In addition to the c-director fluctuations mentioned above, there are two other fluctuation modes of the director, that play an important role in the enormously strong light scattering effects typical of liquid crystals. There are the director deviations from equilibrium caused by undulations of the layers and the deviations of the polar angle θ. The former has been studied in detail by Shalaginov and Romanov [12], while the interaction between the two types has been studied Spector *et al.* [13]. Here we leave aside both of these types and consider only the c-director fluctuations, which, being of the Goldstone type, dominate.

We choose a Cartesian coordinate system such that the smectic-C film of thickness L is confined between the two outer layers at $z = \pm L/2$. The x axis is defined along the projection of $\mathbf{n}$ in the plane of the layers. We recall that the director is defined by the azimuthal φ and polar θ angles. Assuming θ to be fixed and deviations of φ from zero to be small, we arrive at a free energy expression [14]

$$F = \frac{1}{2}\int d^3r\left[B_1\left(\frac{\partial\varphi}{\partial x}\right)^2 + B_2\left(\frac{\partial\varphi}{\partial y}\right)^2 + B_3\left(\frac{\partial\varphi}{\partial z}\right)^2 + 2B_{13}\frac{\partial\varphi}{\partial x}\frac{\partial\varphi}{\partial z}\right], \tag{1}$$

where the integration is carried out over the volume of the film and the B coefficients are elastic constants. The system is stable if $B_i > 0$ $(i = 1,2,3)$ and $B_{13}^2 < B_1 B_3$. To make a connection to the well-known elastic constants K_{ii}, we can use $\mathbf{n}(\varphi) = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta)$ in the Frank energy expression

$$K_{11}(\nabla\cdot\mathbf{n})^2 + K_{22}[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2 + K_{33}(\mathbf{n}\times\nabla\times\mathbf{n})^2 \tag{2}$$

and rewrite it in terms of φ, giving the relations [15]

$$\begin{aligned} B_1 &= \left(K_{22}\cos^2\theta + K_{33}\sin^2\theta\right)\sin^2\theta, \\ B_2 &= K_{11}\sin^2\theta, \\ B_3 &= \left(K_{33}\cos^2\theta + K_{22}\sin^2\theta\right)\sin^2\theta, \\ B_{13} &= (K_{33} - K_{22})\sin^3\theta\cos\theta. \end{aligned} \tag{3}$$

Equations (3) automatically guarantee stability of the system and show how the elastic constants depend on θ. These expressions are consistent with a qualitative statement made by Carlsson *et al.* [16], according to which B_i $(i = 1,2,3)$ are even in θ, while B_{13} is odd in θ. All these coefficients must vanish with vanishing θ. It is clear that B_{13} is negligible compared to the others if θ is small. This happens, for example, in the vicinity of the transition to the smectic-A phase. By contrast, deep in the smectic-C phase all the elastic constants have the same order of magnitude.

To obtain the correlation function in the reciprocal space representation for an infinite sample, one can take the Fourier transform over all space and then apply the equipartition theorem. Clearly, a more complicated analysis is required in the case of a bounded system, since boundary conditions must then be taken into account. We shall follow two approaches to the problem. Of these, the first, based on a description in terms of the natural modes, although a plausible formulation, leads to a somewhat intractable formula. The second gives the correlation function in a form that is easier to analyze and is computationally more efficient.

A. Description in terms of natural modes

The idea of this approach is to find a full series of orthogonal functions (natural modes), providing a basis in which the free energy takes a diagonal form, and then to apply the equipartition theorem. In order to find such a series, we take a Fourier transform over the xy coordinates and rewrite the free energy using the new variable $\tilde{\varphi}(\mathbf{q}_\perp, z) \equiv \exp(ib_{13}q_x z)\varphi(\mathbf{q}_\perp, z)$. After integrating by parts we arrive at

$$F = \frac{B_3}{2(2\pi)^2}\int d^2q_\perp\left\{\int_{-L/2}^{L/2} dz\tilde{\varphi}^*(\mathbf{q}_\perp, z) \times\left[-\frac{\partial^2}{\partial z^2} + (b_1 - b_{13}^2)q_x^2 + b_2 q_y^2\right]\tilde{\varphi}(\mathbf{q}_\perp, z) + \tilde{\varphi}^*(\mathbf{q}_\perp, z)\frac{\partial}{\partial z}\tilde{\varphi}(\mathbf{q}_\perp, z)\Big|_{-L/2}^{L/2}\right\}, \tag{4}$$

where $b_1 = B_1/B_3$, $b_2 = B_2/B_3$, and $b_{13} = B_{13}/B_3$. A fundamental condition that must be satisfied in order that arbitrary φ can be expanded into a full series of eigenfunctions is that the operator $-\partial^2/\partial z^2$ should be self-adjoint. This property is determined from the choice of boundary conditions. In line with [17], in which fluctuations in a homeotropically aligned nematic cell are considered, we choose

$$\frac{\partial}{\partial z}\tilde{\varphi}(\mathbf{q}_\perp, z = \pm L/2) = 0. \tag{5}$$

The free energy under such boundary conditions in this basis takes a diagonal quadratic form. The eigenvalues of the operator are $\lambda_0 = 0$, $\lambda_n = (\pi n/L)^2$ $(n = 1, 2, ...)$ with eigenfunctions

$$\tilde{\varphi}_0(z) = \frac{1}{\sqrt{L}}, \tag{6}$$

$$\tilde{\varphi}_n(z) = \frac{1}{\sqrt{2L}} \left[\exp(i\pi nz/L) + (-1)^n \exp(-i\pi nz/L)\right].$$

Now, arbitrary fluctuations can be expanded in a full series of eigenfunctions

$$\varphi(\mathbf{q}_\perp, z, t) = \exp(-ib_{13}q_x z) \sum_{n=0}^{\infty} c^{(n)}(\mathbf{q}_\perp, t)\tilde{\varphi}_n(z). \tag{7}$$

Let us consider dynamical properties of the system. We adopt the simplest feasible dynamical model, neglecting coupling between the director and hydrodynamic flow. In this approximation the coefficients in Eq. (7) must satisfy the equations

$$\gamma \frac{\partial}{\partial t} c^{(n)}(\mathbf{q}_\perp, t) = -\Gamma^{(n)} c^{(n)}(\mathbf{q}_\perp, t) \tag{8}$$

with γ being a viscosity coefficient and

$$\Gamma^{(n)} = \frac{B_3}{\gamma} \left[(b_1 - b_{13}^2)q_x^2 + b_2 q_y^2 + \lambda_n\right]. \tag{9}$$

The correlation function

$$G(\mathbf{q}_\perp, z, z', t) \equiv \langle \varphi(\mathbf{q}_\perp, z, t)\varphi^*(\mathbf{q}_\perp, z', 0)\rangle \tag{10}$$

can be written as

$$G(\mathbf{q}_\perp, z, z', t) = \sum_{n=0}^{\infty} \exp\left[ib_{13}(z' - z)q_x - \Gamma^{(n)}|t|\right] \times \langle c^{(n)}(\mathbf{q}_\perp, 0)c^{(n)*}(\mathbf{q}_\perp, 0)\rangle \tilde{\varphi}_n(z)\tilde{\varphi}_n^*(z'), \tag{11}$$

where the single time correlation coefficients are given by

$$\langle c^{(n)}(\mathbf{q}_\perp, 0)c^{(n)*}(\mathbf{q}_\perp, 0)\rangle = \frac{k_B T}{\gamma \Gamma^{(n)}}. \tag{12}$$

In the $(\mathbf{q}_\perp, z, z', \omega)$ representation, which is convenient from the light scattering perspective, we have

$$G(\mathbf{q}_\perp, z, z', \omega) = \frac{2k_B T}{\gamma} \sum_{n=0}^{\infty} \frac{1}{\omega^2 + \Gamma^{(n)2}} \tilde{\varphi}_n(z)\tilde{\varphi}_n^*(z') \times \exp\left[ib_{13}(z' - z)\right]. \tag{13}$$

In the limit of $L = 0$ only the main mode with $n = 0$ contributes to Eq. (13). As one can see from Eq. (9), that contribution corresponds to the Goldstone mode of a pure two-dimensional system described by the two effective elastic coefficients $B_1 - B_{13}^2/B_3$ and B_2.

It is worth mentioning that a similar description is used by Sprunt *et al.* [7], but there is some difference. First, these authors implicitly assume that the free energy takes a diagonal quadratic form in the basis of functions $\exp(inz/L)$ $(n = 0, \pm 1, \pm 2, ...)$. Here we do not make this assumption, developing our argument instead from a full set of orthogonal functions that follows directly from the free energy expression. Second, they truncate the summation over the natural modes by the number of the layers in the film. Such a cutoff is reasonable, because length scales less than the order of the layer spacing are beyond the focus of such a model.

The same truncation scheme is used by Poniewierski to study layer undulations [18]. Unfortunately, it leads to nonphysical oscillations in the dependence of the correlation function on z and z', which turns out to be quite large for films that are only a few layers thick.

We prefer to keep the tail of the sum because it ensures smooth dependence across the film. In the opposite limit of an infinite sample the distance between neighboring λ_n tends to zero (i.e., the spectrum of the operator $-\partial^2/\partial z^2$ becomes continuous) and all modes must be taken into account.

B. Resolvent function and response function

Rather than carry out the summation in Eq. (13), we adopt in this section an alternative approach that affords a compact analytical expression for the correlation function. Another goal of this section is to derive a response function that describes the reaction of our system to an external action. In accordance with the fluctuation-dissipation theorem, the correlation function of an infinite system must be equal to the imaginary part of the response function multiplied by $2k_B T/\omega$. We will show that this statement is in fact also valid for our bounded system, where surface conditions play an essential role.

If an external field $h(\mathbf{r}, t)$ acts on the film, then $\varphi(\mathbf{r}, t)$, according to the our choice of dynamical model, obeys

$$\gamma \frac{\partial \varphi(\mathbf{r}, t)}{\partial t} = -\frac{\delta F}{\delta \varphi(\mathbf{r}, t)} + h(\mathbf{r}, t), \tag{14}$$

with the boundary conditions of Eq. (5), which, in terms of φ, are

$$\left(\frac{\partial}{\partial z} + ib_{13}q_x\right) \varphi(\mathbf{r}_\perp, \pm L/2, t) = 0. \tag{15}$$

A solution to Eq. (14) in the $(\mathbf{q}_\perp, z, \omega)$ representation is

$$\varphi(\mathbf{q}_\perp, z, \omega) = \int_{-L/2}^{L/2} dz' R_\lambda(z, z')h(\mathbf{q}_\perp, z', \omega), \tag{16}$$

where the response function R is a solution to the equation

$$B_3 \left[-\left(\frac{\partial}{\partial z} + ib_{13}q_x\right)^2 - \lambda\right] R_\lambda(z, z') = \delta(z - z') \tag{17}$$

with λ dependent on $\mathbf{q}_\perp$ and ω:

$$\lambda = i\omega \frac{\gamma}{B_3} - (b_1 - b_{13}^2)q_x^2 - b_2 q_y^2. \tag{18}$$

Equation (17) does not uniquely define the response function. The correct form of R_λ in Eq. (16) must give φ satisfying boundary conditions Eq. (15). Using $\tilde{\varphi}$ instead of φ again reduces the problem to a solved one (see, for example, Ref. [12]). Equation (17) can also be solved as follows. For $z \neq z'$, the differential equation (17) is homogeneous and can be readily solved. By joining the solutions for the regions $z > z'$ and $z < z'$, taking into the account the boundary conditions Eq. (15), we obtain

$$R_\lambda(z,z') = \frac{\exp[-ib_{13}q_x(z-z')]}{2B_3\sqrt{-\lambda}\sinh(\sqrt{-\lambda}L)}\left\{\cosh\left[\sqrt{-\lambda}(z+z')\right] + \cosh\left[\sqrt{-\lambda}(|z-z'|-L)\right]\right\}. \quad (19)$$

Equation (19), together with Eq. (16), allows one to find the response function.

Another consequence of Eq. (19) is that the single time correlation function can be written as

$$G(\mathbf{q}_\perp, z, z', 0) = k_B T R_{\lambda(\mathbf{q}_\perp,0)}(z,z'). \quad (20)$$

According to the dynamical model chosen here, the two-time correlation function $G(\mathbf{q}_\perp, z, z', t)$ must satisfy the equation

$$\gamma\frac{\partial}{\partial t}G(\mathbf{q}_\perp, z, z', t) = -B_3\left[(b_1 - b_{13}^2)q_x^2 + b_2 q_y^2 - \left(\frac{\partial}{\partial z} + ib_{13}q_x\right)^2\right] \times G(\mathbf{q}_\perp, z, z', t). \quad (21)$$

Taking the half Fourier transform

$$G^{(+)}(\mathbf{q}_\perp, z, z', \omega) \equiv \int_0^\infty dt e^{i\omega t} G(\mathbf{q}_\perp, z, z', t),$$

we obtain

$$B_3\left[-\left(\frac{\partial}{\partial z} + ib_{13}q_x\right)^2 - \lambda(\mathbf{q}_\perp, \omega)\right] G^{(+)}(\mathbf{q}_\perp, z, z', \omega) = \gamma G(\mathbf{q}_\perp, z, z', 0). \quad (22)$$

The solution to Eq. (22) can also be expressed through the resolution function

$$G^{(+)}(\mathbf{q}_\perp, z, z', \omega) = k_B T\gamma\left(\widehat{R}_{\lambda(\mathbf{q}_\perp,\omega)} \cdot \widehat{R}_{\lambda(\mathbf{q}_\perp,0)}\right)(z,z'). \quad (23)$$

For the full Fourier transform we have

$$G(\mathbf{q}_\perp, z, z', \omega) = G^{(+)}(\mathbf{q}_\perp, z, z', \omega) + \overline{G^{(+)}(\mathbf{q}_\perp, z', z, \omega)}. \quad (24)$$

Using the following property of resolvent functions of self-adjoint operators [19]:

$$\widehat{R}_\mu \cdot \widehat{R}_\nu = \frac{1}{B_3(\mu-\nu)}(\widehat{R}_\mu - \widehat{R}_\nu),$$

we get the usual relation [11] between the correlation and response functions

$$G(\mathbf{q}_\perp, z, z', \omega) = \frac{2k_B T}{\omega}\mathrm{Im}R_{\lambda(\mathbf{q}_\perp,\omega)}(z,z'). \quad (25)$$

The pair of expressions (19) and (25) is more computationally efficient than Eq. (13). In the limit $L \to \infty$ it gives, as expected, the known result (see, for example, [6]) for the infinite sample and in the limit of $L = 0$ it reduces to the contribution of the Goldstone mode mentioned in the previous Sec. II A.

III. LIGHT SCATTERING

It has been shown by Galerne *et al.* [2] that the fluctuations of the dielectric tensor that are associated with the c director give the main contribution to the light scattering intensity. Leaving aside scattering from the layers undulations, considered in detail in [12], and a contribution from fluctuations of the tilt angle, we can assume

$$\delta\epsilon_{\alpha\beta} = \epsilon_a\left[n_\alpha(0)\frac{\partial n_\beta(0)}{\partial\varphi} + n_\beta(0)\frac{\partial n_\alpha(0)}{\partial\varphi}\right]\varphi, \quad (26)$$

where ϵ_a is the optical anisotropy of the smectic liquid crystal. The intensity of the scattered light in the Born approximation is given by

$$I(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}, \mathbf{q}, \omega) \sim V\epsilon_a^2 f(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}) g(\mathbf{q}, \omega). \quad (27)$$

Here V is illuminated volume. The geometric factor f is defined as

$$f(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}) = \{e_\alpha^{(s)}[n_\alpha(0)\partial_\varphi n_\beta(0) + n_\beta(0)\partial_\varphi n_\alpha(0)]e_\beta^{(i)}\}^2, \quad (28)$$

where $\mathbf{e}^{(i)}$ and $\mathbf{e}^{(s)}$ are unit polarization vectors of incident and scattered light. The scattering vector $\mathbf{q}$ has components $(\mathbf{q}_\perp, q_z)$, ω is a frequency shift, and we define

$$g(\mathbf{q}, \omega) = \frac{1}{L}\int_{-L/2}^{L/2} dz \int_{-L/2}^{L/2} dz' \exp\left[-iq_z(z-z')\right] \times G(\mathbf{q}_\perp, z, z', \omega). \quad (29)$$

A prefactor dependent on the intensity of the incident light and its wavelength has been dropped in Eq. (27) for brevity. We have also neglected the difference in wavelength of ordinary and extraordinary waves, reflection and refraction at the interfaces. These optical corrections are beyond the scope of this paper and are considered in detail elsewhere [12].

As an example of application of the formula, let us consider the geometry in which the incidence plane coincides with the tilt plane, incident light is polarized perpendicular to it, but the polarization vector of the scattered light lies in it. The components of the incident and scattered wave vectors can be written as $\mathbf{k}^{(i)} = k_0(\sin\theta_i, 0, \cos\theta_i)$, and $\mathbf{k}^{(s)} = k_0(\sin\theta_s, 0, \cos\theta_s)$, where k_0 is the wave number of the incident light. Here θ_i,θ_s are the angles between the z axis and, respec-

tively, the wave vectors of the incident and scattered waves. The components of the scattering vector are now $\mathbf{q} = k_0(\sin\theta_s - \sin\theta_i, 0, \cos\theta_s - \cos\theta_i)$. This geometry yields $f(\mathbf{e}^{(s)}, \mathbf{e}^{(i)}) = \sin^2\theta \sin^2(\theta_s - \theta)$.

Expanding the correlation function into a full series of natural modes, Eq. (29) becomes

$$g(\mathbf{q},\omega) = \frac{4k_BT}{\gamma L^2}\left\{\frac{2}{\omega^2+\Gamma^{(0)2}}\frac{\sin^2[(q_z+b_{13}q_x)L/2]}{(q_z+b_{13}q_x)^2} + \sum_{n=1}^{\infty}\frac{1}{\omega^2+\Gamma^{(n)2}}\left[\frac{\sin[(\pi n/L - q_z - b_{13}q_x)L/2]}{\pi n/L - q_z - b_{13}q_x} + (-1)^n\frac{\sin[(\pi n/L + q_z + b_{13}q_x)L/2]}{\pi n/L + q_z + b_{13}q_x}\right]^2\right\}. \quad (30)$$

Integration over z and z' in Eq. (29) can be carried out using Eq. (25). We thus obtain the more compact expression

$$g(\mathbf{q},\omega) = \frac{2k_BT}{B_3L\omega}\mathrm{Im}\frac{1}{\sqrt{-\lambda}\sinh(\sqrt{-\lambda}L)\,[(q_z+b_{13}q_x)^2-\lambda]} \times\left\{\sqrt{-\lambda}L\sinh(\sqrt{-\lambda}L) + \frac{2(q_z+b_{13}q_x)^2}{(q_z+b_{13}q_x)^2-\lambda} \times\left[\cosh(\sqrt{-\lambda}L) - \cos[(q_z+b_{13}q_x)L]\right]\right\}, \quad (31)$$

where $\lambda = \lambda(\mathbf{q}_\perp,\omega)$ is defined by Eq. (18).

Equation (31) allows one to take the limits $L \to \infty$ and $L \to 0$

$$\lim_{L\to\infty} g(\mathbf{q},\omega) = \frac{2k_BT}{\gamma}\frac{1}{\omega^2 + \left[B_1q_x^2 + B_2q_y^2 + B_3q_z^2 + 2B_{13}q_xq_z\right]^2/\gamma^2}, \quad (32)$$

$$\lim_{L\to 0} g(\mathbf{q},\omega) = \frac{2k_BT}{\gamma}\frac{1}{\omega^2 + \left[(B_1 - B_{13}^2/B_3)q_x^2 + B_2q_y^2\right]^2/\gamma^2}. \quad (33)$$

The first limit (see also Ref. [20]) corresponds to the statement that in an infinite sample the intensity is proportional to the correlation function in the $(\mathbf{q},\omega)$ representation. The latter can be derived directly from Eq. (1) by taking the appropriate Fourier transform and using the equipartition theorem. A comparison of these two limits shows that the half-widths of the curves differ from each other considerably. It should be emphasized that in order to get the half-width in the case of a very thin film it is not sufficient simply to set $q_z = 0$ in the expression for an infinite sample. Rather, the corresponding expression for a thin film should be obtained by choosing q_z such that the denominator of Eq. (32) is minimized. One sees that $B_1q_x^2 + B_2q_y^2 + B_3q_z^2 + 2B_{13}q_xq_z$ for fixed q_x and q_y is minimal if $q_z = -b_{13}q_x$, the minimum being equal to $(B_1 - B_{13}^2/B_3)q_x^2 + B_2q_y^2$. This value also can be written as $\gamma\Gamma^{(0)}$, where $\Gamma^{(0)}$, defined be Eq. (9), is the relaxation rate of the main mode. For thin films, this mode is the only one that needs to be taken into account.

Thin films can be considered on the basis of the two-dimensional (2D) formulation [21] according to which φ is uniform across the film and the elastic energy per unit area is given by $\frac{1}{2}[K^B(\partial_x\varphi)^2 + K^S(\partial_y\varphi)^2]$, where K^B and K^S are the bend and splay elastic constants of the c director. The relaxation rate Γ measured is given by $(K^Bq_x^2 + K^Sq_y^2)/\eta$ [21], where η is the 2D viscosity. These two formulations give the same relaxation rate provided that $\eta = L\gamma$ and

$$K^S = LB_2, \qquad K^B = L(B_1 - B_{13}^2/B_3). \quad (34)$$

In fact, these 2D effective constants depend on bulk elastic constants as well as boundary conditions. In the case of the free-standing film, they are given by (34). Equations (3) allow one to express them in terms of θ and the Frank constants.

A useful quantity is the anisotropy K^S/K^B. It does not depend on an absolute measurements of the elasticity and can be obtained from light scattering experiments. Using Eqs. (3) in Eqs. (34) we obtain

$$\frac{K^S}{K^B} = \frac{K_{11}}{K_{22}}\cos^2\theta + \frac{K_{11}}{K_{33}}\sin^2\theta. \quad (35)$$

IV. CONCLUSION

The fluctuations of the c director in free-standing smectic films have been studied theoretically. The films have been modeled using elastic theory for smectic-C liquid crystals. In studying the dynamical behavior of the fluctuations, we have chosen the simplest feasible hydrodynamical model. The correlation analysis has been carried out both in terms of the natural modes of the system and in terms of the response function. The latter gives compact analytical expressions and can be easily extended to the case of a film on a substrate or a cell. It has been shown that the correlation and response functions for a film in the $(\mathbf{q}_\perp, z, z', \omega)$ representation obey the same relation as those for an infinite sample in the $(\mathbf{q},\omega)$ representation. In fact, this statement can be generalized to the case of arbitrary boundary conditions. The expressions derived show how the three-dimensional system evolves into a two-dimensional one with diminishing film thickness. We have established that the two-dimensional elastic model cannot be derived simply by assuming that the fluctuations in a thin film are uniform across the film. Instead, the two-dimensional model should be properly derived from the three-dimensional one by minimizing the free energy over all possible dependence across the

film. A signature of this result is that B_{13} features in the effective two-dimensional bend elastic coefficient (34).

The elastic anisotropy is given in Eq. (35). Such a dependence of the elastic anisotropy on the tilt angle can be checked in light scattering experiments. Section III presents a complete set of expressions requisite for interpretation of experimental data. Equation (31) takes into account contributions from all the natural modes and allows one to analyze the frequency spectrum of the inelastic scattering. By adding a term $-\chi_a H^2 \cos^2(\theta)/B_3$ to the right-hand side of Eq. (18), the effect of an aligning magnetic field H applied along the x axis could be accommodated.

The dynamical model chosen in this paper does not take into account the influence of mass flow on the reorientation of the c director. There is evidence from numerical calculations [22] that this effect [23] is not negligible. In the case of hard boundary conditions it gives rise to the so-called backflow speeding up switching effect. A similar effect can be found in homeotropically aligned cells [24]. It has been pointed out [24] that the coupling of the director with mass flow prevents one from describing the dynamical processes in terms of the stationary natural modes of the director. This is because the mass flow field and the director must satisfy different boundary conditions. One suspects such coupling will depend strongly on the boundary conditions. A precise analysis of these dynamical effects in the case of a free-standing film is beyond the scope of this paper. We defer it as the possible subject of a separate study.

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