Fluctuations and light scattering in thin smectic films

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An analysis of the layer-displacement fluctuations in thin smectic-A liquid-crystal films is presented. Explicit formulas for the displacement-displacement correlation function, taking into account the surface tension and the anchoring strength, are derived. It is shown that the boundary conditions essentially affect the fluctuations not only on the surface but in the interior of the film as well. The light scattering on the displacement-associated director fluctuations is considered. This consideration is carried out on the basis of the Green's function of the electromagnetic field for an optically anisotropic film. The calculations include multiple-reflection effects for incident and scattered light beams. It is shown that the scattered light intensity differs essentially from the intensity for the same volume in an infinite smectic medium. There is no forward scattering in the case of normal incidence and an undulation behavior of the intensity as a function of scattering angle takes place. There are maxima in the angular distribution. The magnitudes of the maxima are sensitive to the surface-tension coefficient. The possibility of measuring the surface-tension coefficient by means of an optical experiment is discussed.

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I. INTRODUCTION

Light-scattering methods are widely used in liquid-crystal studies. But due to thermodynamical and optical differences it is necessary to carry out the theoretical analysis for each liquid-crystal type separately. This paper is devoted to smectic liquid crystals. Smectic liquid crystals are characterized by the long-range orientational order of the elongated molecules, which in addition are partially segregated into layers. A smectic liquid crystal may be described as a system with one-dimensional translational ordering. Long-range translational order in such a system is destroyed by thermal fluctuations [1]. Strong thermal fluctuations lead to characteristic features of the light scattering, which were first detected by Ribotta, Salin, and Durand [2]. Physical properties of smectic finite-size systems, thin films in particular, arouse a lot of interest. In this case surface phenomena such as surface tension [3] and anchoring strength [4] must be taken into account. The influence of the boundary conditions on the displacement-displacement correlation function and the x-ray scattering process was analyzed by Holyst, Tweet, and Sorenson [5]. The analysis was carried out numerically on the basis of a discrete version of the free energy. An analytical solution of this problem seems to be desirable. Another problem connected with the light scattering exists in the case of thin films. First, there are ordinary and extraordinary electromagnetic waves in film, because the smectic-A phase is optically uniaxial medium, so scattering of waves of one type into waves of another takes place. Secondly, an incident and the scattered beams are multiply reflected from interfaces. These optical phenomena must be taken into account in the theory because they are able to affect essentially experimental data.

The outline of this paper is as follows. In Sec. II we discuss the fluctuations and correlation function and their dependence on smectic elastic constants, the surface tension and the anchoring strength; explicit expression for the displacement-displacement correlation function in the case of the smectic-A film is presented. Section III is devoted to the light scattering. In Sec. III A the fluctuations of the dielectric tensor are considered and the problem of light scattering in smectic films is formulated. In Sec. III B the incident field in the interior of the film in the case of normal incidence is calculated. Section III C is devoted to the Green's function for an optically anisotropic film; multiple-reflection effect is analyzed. In Sec. III D we derive an expression for the intensity for light scattering in smectic-A film and the angular distribution of the intensity is analyzed. In Sec. IV we summarize the results obtained, discuss briefly a case of oblique incidence, and consider the possibility of measuring the surface tension coefficient by means of optical experiment.

II. THE FREE ENERGY AND THE FLUCTUATIONS IN A SMECTIC-A FILM

We consider a homeotropic aligned smectic film with thickness L. Let the film be confined between the planes $z=\pm L/2$ in the Cartesian coordinate system. The parameter which describes a deformation of a smectic liquid crystal is the layer displacement $u(\mathbf{r})$ along the z axis. It is known that this parameter is a hydrodynamical one [6] and its fluctuations disperse slowly. Apart from the layer structure there is a direction of the preferred molecular alignment $\mathbf{n}$ (the director) in the smectic liquid crystal. There are different types of smectic phases which are classified by the director orientation with respect to the layers. We will consider the smectic-A phase, i.e., assume the director to be normal to the layers. In this case the displacement gradients of the director fluctuation $\delta\mathbf{n}(\mathbf{r})$ are equal to $-\nabla_\perp u(\mathbf{r})$, where $\nabla_\perp$ is the gradient in x,y variables. To describe the fluctuations we start from the expression for the free energy F, which is a sum of the bulk F_{bulk} and the surface E_{surf} terms. The bulk contribution is equal to [7]

$$F_{\text{bulk}}=\tfrac{1}{2}\int d^3r\{B[\partial_z u(\mathbf{r}_\perp,z)]^2+K[\Delta_\perp u(\mathbf{r}_\perp,z)]^2\} \ . \tag{1}$$

Here B is the smectic elastic constant associated with the layer compressions, K is the elastic constant associated with the layer undulations, and $\Delta_\perp$ is the Laplacian in x,y variables. The integration is carried out over the volume V of the sample. The surface term depends on the sample's environment. First, there is a contribution induced by the surface tension [5]

$$F_{\text{surf}}=\tfrac{1}{2}\int d^2r_\perp\gamma[\nabla_\perp u(\mathbf{r})]^2 \ , \tag{2}$$

where the integration is carried out over the sample surface and γ is the surface tension coefficient. This term describes the additional energy cost associated with increasing of the surface area. Secondly, if the smectic film is not freely suspended, there may be anchoring on the surface. Usually the surface anchoring is described by the Rapini potential [4] $\frac{1}{2}W_0\sin^2(\phi)$, where W_0 is the anchoring strength and ϕ is the angle between the direction $\mathbf{n}$ and the surface normal. For small ϕ the potential is equal to $\frac{1}{2}W_0[\nabla_\perp u(\mathbf{r})]^2$. It is easy to notice that the anchoring term only changes the surface tension coefficient γ. So we will not introduce the additional term, connected with anchoring. Taking into account that the opposite sides of the film may be set under different conditions in a real experiment, we assume the surface $z=L/2$ to be characterized by coefficient γ_+ and the surface $z=-L/2$ to be characterized by γ_-. The surface contribution to the free energy thus is given by

$$F_{\text{surf}}=\tfrac{1}{2}\int d^2r_\perp\{\gamma_+[\nabla_\perp u(\mathbf{r}_\perp,L/2)]^2+\gamma_-[\nabla_\perp u(\mathbf{r}_\perp,-L/2)]^2\} \ . \tag{3}$$

Our aim in this section is to calculate the layer-displacement correlation function $G(\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')=\langle u(\mathbf{r}_\perp,z)u(\mathbf{r}'_\perp,z')\rangle$. Here the statistical average $\langle\ \rangle$ is taken with respect to $u(\mathbf{r})$, i.e.,

$$\langle\cdots\rangle=\int Du\cdots\exp\left[-\frac{F}{k_BT}\right]\Big/\int Du\exp\left[-\frac{F}{k_BT}\right] , \tag{4}$$

where the free energy F is equal to a sum of expressions (1) and (3).

The considered system is homogeneous in the xy plane, so it is appropriate to take the Fourier transform with respect to these variables,

$$u(\boldsymbol{\kappa},z)=\int d^2r_\perp e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}_\perp}u(\mathbf{r}_\perp,z) \ , \tag{5}$$

where $\boldsymbol{\kappa}$ is the wave vector in the xy plane. The free energy can be written as follows:

$$F=\frac{1}{(2\pi)^2}\int d^2\kappa F_\kappa \ , \tag{6}$$

where F_κ is the contribution of fluctuations with wave vector $\boldsymbol{\kappa}$,

$$F_\kappa=\tfrac{1}{2}\left\{\int_{-L/2}^{L/2}dz[B|\partial_z u(\boldsymbol{\kappa},z)|^2+K\kappa^4|u(\boldsymbol{\kappa},z)|^2]+\kappa^2[\gamma_+|u(\boldsymbol{\kappa},+L/2)|^2+\gamma_-|u(\boldsymbol{\kappa},-L/2)|^2]\right\} . \tag{7}$$

If the fluctuation $u(\boldsymbol{\kappa},z)$ satisfies the boundary conditions

$$a_\pm u(\boldsymbol{\kappa},\pm L/2)\pm\partial_z u(\boldsymbol{\kappa},\pm L/2)=0 \ , \tag{8}$$

where

$$a_\pm=\frac{\gamma_\pm}{B}\kappa^2 \ , \tag{9}$$

then the contribution of the surface terms vanishes and expression (7) transforms into a quadratic form

$$F_\kappa=\tfrac{1}{2}B\int_{-L/2}^{L/2}dz\, u^*(\boldsymbol{\kappa},z)\hat{A}u(\boldsymbol{\kappa},z) \ , \tag{10}$$

with the operator

$$\hat{A}=-\partial_z^2+\frac{K}{B}\kappa^4 \ . \tag{11}$$

One can prove that the operator $\hat{A}$ acting on the functions which satisfy the boundary conditions (8) is a self-adjoint one [8]. Thus there are eigenvalues $\lambda_m(\kappa)$ and eigenfunctions $f_m(\kappa,z)$ (fluctuation modes) which compose a basis. It should be noted that utilization of the boundary conditions (8) does not imply any restrictions on our consideration of fluctuations, because any function $u(\mathbf{r})$ can be expanded in terms of $f_m(\kappa)$.

According to the equipartition theorem the correlation function $G(\kappa,z,z')$ is given by

$$G(\kappa,z,z')=\frac{k_BT}{B}\sum_m\lambda_m^{-1}(\kappa)f_m(\kappa,z)f_m^*(\kappa,z') \ . \tag{12}$$

The eigenvalues $\lambda_m(\kappa)$ can be calculated only numerically and it is very difficult to analyze this expression analytically. A more effective approach is to use the discrete version for free energy [5]. This method reduces the problem to a simple $N\times N$ matrix inversion, where N is the number of layers in the smectic film. But this problem can be solved analytically.

As one can see from Eq. (12) the correlation function in (κ,z,z') representation must satisfy the equation

$$(g^2-\partial_z^2)G(\kappa,z,z')=\frac{k_BT}{B}\delta(z-z') \ , \tag{13a}$$

where

$$g=\sqrt{K/B}\,\kappa^2 \; . \tag{13b}$$

The associated boundary conditions are

$$a_\pm G(\kappa,\pm L/2,z')\pm\partial_z G(\kappa,\pm L/2,z')=0 \; . \tag{14}$$

When z is not equal to z' we get zero in the right part of Eq. (13a) with the well-known solutions. The solution of this main problem can be expressed through the solutions $u_+(z)$ and $u_-(z)$ of Eq. (13a) with zero in the right part [9],

$$G(\kappa,z,z')=\frac{k_B T}{B[u_+\partial_z u_- - u_-\partial_z u_+]}\times\begin{cases}u_+(z)u_-(z') & \text{if } z>z'\\ u_-(z)u_+(z') & \text{if } z<z' \; .\end{cases} \tag{15}$$

To satisfy the conditions (14) we must choose $u_\pm(z)$ which satisfy similar boundary conditions,

$$a_\pm u_\pm\left[\pm\frac{L}{2}\right]\pm\partial_z u_\pm\left[\pm\frac{L}{2}\right]=0 \; . \tag{16}$$

The required functions are

$$u_\pm(z)=(g\mp a_\pm)\exp\left[g\left[z\mp\frac{L}{2}\right]\right]+(g\pm a_\pm)\exp\left[-g\left[z\mp\frac{L}{2}\right]\right] \; . \tag{17}$$

Using Eqs. (17) and (15), we have

$$G(\kappa,z,z')=\frac{k_B T}{2Bg\Delta}\{(g^2-a_+a_-)\cosh[g(z+z')]+g(a_- - a_+)\sinh[g(z+z')]+[(g^2+a_+a_-)\cosh(gL)+g(a_-+a_+)\sinh(gL)]\cosh[g(z-z')]-\Delta\sinh(g|z-z'|)\} \; , \tag{18a}$$

where

$$\Delta=(g^2+a_+a_-)\sinh(gL)+g(a_++a_-)\cosh(gL) \; . \tag{18b}$$

It should be mentioned that the expansion of G given by Eq. (18) as a meromorphic function of g to a sum of simple fractions yields a formula like Eq. (12). The function G has singularities at the points where $\Delta=0$. The points can be found only numerically in general, but if the surface tension coefficient is equal to zero or infinity, the expansion can be carried out analytically. For example, when $\gamma_+=\gamma_-=0$ (film with negligibly small surface tension) the singularity points are $g_m=i\pi m/L$ and we get

$$G(\kappa,z,z')=\frac{k_B T}{BL}\left\{\frac{1}{g^2}+2L^2\sum_{m>0}\frac{1}{(gL)^2+(\pi m)^2}\times\cos\left[\frac{\pi m z}{L}\right]\times\cos\left[\frac{\pi m z'}{L}\right]\right\} . \tag{19}$$

It is easy to notice that expressions (18) reduce to those for the infinite smectic film when thickness L is large,

$$G(\kappa,z,z')=\frac{k_B T}{2gB}\exp(-g|z-z'|) \; . \tag{20}$$

Continuous Fourier transform of Eq. (20) with respect to $z-z'$ leads us to the well-known expression for the correlation function in $\mathbf{q}$ representation [7]

$$G(\mathbf{q})=\frac{k_B T}{K\kappa^4+Bq_z^2} \; , \tag{21}$$

where $\mathbf{q}=(\boldsymbol{\kappa},q_z)$.

The distinction of this kind of smectic film is that the displacement fluctuation goes Goldstone type for any finite anchoring strength and surface tension. Indeed, expression (18) has a singularity at the point $\boldsymbol{\kappa}=0$ if $\gamma_\pm$ are finite. But the director fluctuations are not of the same type due to the coefficient $\boldsymbol{\kappa}$ in the relation $\delta\mathbf{n}(\boldsymbol{\kappa},z)=-i\boldsymbol{\kappa}u(\boldsymbol{\kappa},z)$.

A mean-square fluctuation at the point with coordinate z can be calculated using the expression

$$\langle u(\mathbf{r}_\perp,z)u(\mathbf{r}_\perp,z')\rangle=\frac{1}{(2\pi)^2}\int d^2\kappa G(\kappa,z,z) \; . \tag{22}$$

The integral in Eq. (22) diverges at the upper and lower limits of κ. The divergence at the short-wavelength limit arises since the theory is not valid for distances less than a molecular diameter a_0, so one must introduce the cutoff parameter $2\pi/a_0$. The divergence at the long-wavelength limit is connected with the Landau-Peierls instability, hence another cutoff parameter $2\pi/l$ appears with l being the transverse size of the film. Thus the limits in this integral are $2\pi/l<\kappa<2\pi/a_0$. For example, the mean-square displacement in the case of a freely suspended film with $\gamma_-=\gamma_+=\gamma$ can be written as

$$\langle|u(\mathbf{r}_\perp,z)|^2\rangle=\frac{k_B T}{8\pi\sqrt{KB}}\int_{\xi_1}^{\xi_2}d\xi\frac{2(1-\eta^2)e^{-\xi}\cosh(\alpha\xi)+(1+\eta)^2+(1-\eta)^2e^{-2\xi}}{\xi[(1+\eta)^2-(1-\eta)^2e^{-2\xi}]} \; , \tag{23a}$$

and in a case of one hard boundary ($\gamma_+=\gamma,\gamma_-=\infty$) as

$$\langle|u(\mathbf{r}_\perp,z)|^2\rangle=\frac{k_B T}{8\pi\sqrt{KB}}\int_{\xi_1}^{\xi_2}d\xi\frac{(1-\eta)(e^{\xi(\alpha-1)}-e^{-2\xi})+(1+\eta)(1-e^{-\xi(\alpha+1)})}{\xi[1+\eta+(1-\eta)e^{-2\xi}]} \; , \tag{23b}$$

where the dimensionless variables used are

$$\eta=\frac{\gamma}{\sqrt{KB}}, \quad \alpha=\frac{2z}{L}, \quad \xi_1=L\left[\frac{K}{B}\right]^{1/2}\left[\frac{2\pi}{l}\right]^2,$$

$$\xi_2=L\left[\frac{K}{B}\right]^{1/2}\left[\frac{2\pi}{a_0}\right]^2.$$

The variation of the mean-square fluctuation with the z coordinate is presented in Fig. 1. The calculation has been carried out on the basis of Eqs. (23). Figure 1(a) corresponds to the case of a freely suspended film with the same surface tension coefficient γ on both sides. Figure 1(b) corresponds to the case when the fluctuations at one boundary are fully frozen (hard boundary) but the other boundary is characterized by coefficient γ. We chose a film containing 61 layers and the cutoff parameters a_0 and l being equal to 4 and 4×10^4 Å, respectively, in order to compare the results to those obtained through the numerical routine in [5]. The calculations were performed assuming for $k_BT=4.0\times10^{-14}$ ergs and typical smectic parameters $K=10^{-6}$ dyn, $B=2.5\times10^7$ dyn/cm^2, and several values of γ. The comparison shows that the expressions presented above and the model of the discrete layers give similar results. It is seen from Fig. 1(a) that in systems with small surface tension the fluctuations on the surface are in fact larger than those in the interior of the system. Then the particular value of γ can be estimated from Eq. (23a). The sign of the first term in the integral depends on the correlation between 1 and τ^2. When $\gamma<\sqrt{KB}$ the fluctuations at the surface are larger than in the interior. If $\gamma=\sqrt{KB}$ then the mean-square fluctuation does not depend on z.

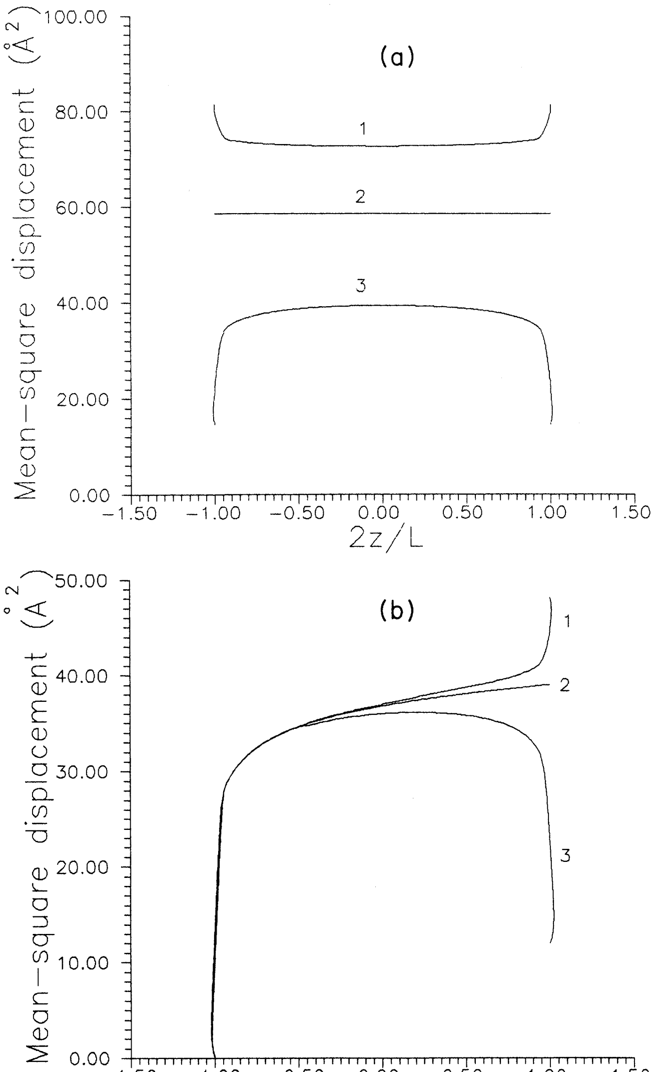

FIG. 1. The mean-square displacement fluctuation profile vs coordinate z for a 61-layer-thick film; $k_BT=4.0\times10^{-14}$ ergs; $B=2.5\times10^7$ dyn/cm^2, $K=1.0\times10^{-6}$ dyn are the smectic elastic constants, and γ is the surface tension coefficient; (1) $\gamma=3$ dyn/cm, (2) $\gamma=5$ dyn/cm, and (3) $\gamma=30$ dyn/cm. Graph (a) corresponds to the case of a freely suspended film with the same surface tension coefficient γ on the both sides. Graph (b) corresponds to the case when the fluctuations on one boundary are fully frozen but another boundary is characterized by coefficient γ.

III. LIGHT SCATTERING

A. Fluctuations of the dielectric tensor and light-scattering process

In order to analyze the influence of the surface phenomena on a light-scattering process let us consider the dielectric tensor $\epsilon_{\alpha\beta}(\mathbf{r})$. Due to an optical anisotropy it can be written in the following form:

$$\epsilon_{\alpha\beta}(\mathbf{r})=\epsilon_{\parallel}n_\alpha(\mathbf{r})n_\beta(\mathbf{r})+\epsilon_{\perp}[\delta_{\alpha\beta}-n_\alpha(\mathbf{r})n_\beta(\mathbf{r})]\ , \tag{24}$$

where $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the permittivities along and transverse to $\mathbf{n}$. Displacement $u(\mathbf{r})$ leads, first, to a fluctuation of the director $\delta\mathbf{n}(\mathbf{r})=-\nabla_{\perp}u(\mathbf{r}_{\perp},z)$ and second to a fluctuation of $\epsilon_{\parallel}$ and $\epsilon_{\perp}$, because they depend on the density. The contribution to the light-scattering intensity from the second type of fluctuations is negligible compared with the first one [10]. So we assume the fluctuation of the dielectric tensor is given by

$$\delta\epsilon_{\alpha\beta}(\mathbf{r})=\epsilon_a[n_\alpha^0\delta n_\beta(\mathbf{r})+n_\beta^0\delta n_\alpha(\mathbf{r})]\ , \tag{25}$$

where $\epsilon_a=\epsilon_{\parallel}-\epsilon_{\perp}$.

Since characteristic frequency of the fluctuations is much less than the angular frequency ω of light, we assume that the incident and scattered waves have the same frequency and vary with time as $e^{-i\omega t}$. An incident wave with amplitude $\mathbf{E}^{\text{inc}}$ creates electric field with amplitude $\mathbf{E}^0(\mathbf{r})$ in the interior of the film. Due to multiple-reflection effect this field in general consists of four waves (two ordinary and two extraordinary), which propagate in different directions. These four coherent waves are scattered by the random inhomogeneities $\delta\hat{\epsilon}(\mathbf{r})$. The scattered light after multiple reflections at the boundaries $z=\pm L/2$ and refraction at one boundary produces the field with complex amplitude $\mathbf{E}'(\mathbf{r})$ outside the film. The intensity and the polarization of the scattered light are completely described in terms of correlation function

$\langle \mathbf{E}'_\alpha(\mathbf{r})\mathbf{E}'^*_\beta(\mathbf{r})\rangle$, which can be calculated in the Born approximation [11]

$$\langle \mathbf{E}'_\alpha(\mathbf{r})\mathbf{E}'^*_\beta(\mathbf{r})\rangle=\frac{\omega^4}{c^4}\int d^3r'd^3r''T_{\alpha\gamma}(\omega,\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z') \times T^*_{\beta\lambda}(\omega,\mathbf{r}_\perp-\mathbf{r}''_\perp,z,z'') \times\langle\delta\epsilon_{\gamma\mu}(\mathbf{r}')\delta\epsilon_{\lambda\nu}(\mathbf{r}'')\rangle \times E^0_\mu(\mathbf{r}')\mathbf{E}^{0*}_\nu(\mathbf{r}'') , \quad (26)$$

where c is the velocity of light and the integration is carried out over illuminated volume V, and $\hat{T}$ is the Green's function. Thus in order to study the light-scattering process we should calculate the incident electric field in the interior of the film and the Green's function.

B. The incident field in the interior of the film

To simplify our calculation we restrict ourselves to the case of normal incidence and assume the incident wave to be polarized in the xz plane, i.e., $\mathbf{E}^{\rm inc}=E^{\rm inc}\mathbf{e}_x\exp[ik_0(z+L/2)]$, where $k_0=\omega\sqrt{\epsilon_0}/c$ ($\epsilon_0=1$ for air). Only two ordinary waves with polarization vector $\mathbf{e}_x$ and wave number $k_{\rm tr}=\omega\sqrt{\epsilon_\perp}/c$, but passing in opposite directions, are produced in this case. The complex amplitude of electric field in all the regions can be written as follows:

$$E_\uparrow\exp\left[ik_0\left[z-\frac{L}{2}\right]\right] \quad \text{if } z<\frac{L}{2} ,$$

$$E^{\rm inc}[d_\uparrow\exp(ik_{\rm tr}z)+d_\downarrow\exp(-ik_{\rm tr}z)] \text{ if } |z|<\frac{L}{2} ,$$

$$E^{\rm inc}\exp\left[ik_0\left[z+\frac{L}{2}\right]\right]+E_\downarrow\exp\left[-ik_0\left[z+\frac{L}{2}\right]\right] \quad \text{if } z<-\frac{L}{2} ,$$

where subscripts ↑ and ↓ indicate the propagation directions of the waves. The coefficients $d_\uparrow$ and $d_\downarrow$ have to be determined. By utilizing standard electromagnetic conditions at $z=\pm L/2$, we get four equations,

$$E_\uparrow=E^{\rm inc}\left[d_\uparrow\exp\left[ik_{\rm tr}\frac{L}{2}\right]+d_\downarrow\exp\left[-ik_{\rm tr}\frac{L}{2}\right]\right] ,$$

$$E^0+E_\downarrow=E^{\rm inc}\left[d_\uparrow\exp\left[-ik_{\rm tr}\frac{L}{2}\right]+d_\downarrow\exp\left[ik_{\rm tr}\frac{L}{2}\right]\right] ,$$

$$k_0E_\uparrow=k_{\rm tr}E^{\rm inc}\left[d_\uparrow\exp\left[ik_{\rm tr}\frac{L}{2}\right]-d_\downarrow\exp\left[-ik_{\rm tr}\frac{L}{2}\right]\right] ,$$

$$k_0(E^{\rm inc}-E_\downarrow)=k_{\rm tr}E^{\rm inc}\left[d_\uparrow\exp\left[-ik_{\rm tr}\frac{L}{2}\right]-d_\downarrow\exp\left[ik_{\rm tr}\frac{L}{2}\right]\right] .$$

The first pair of equations provides for the continuity of the tangential with respect to the surface components of vector $\mathbf{E}$, the other one provides for the continuity of the same components of magnetic field. These equations give

$$d_\uparrow=\frac{2k_0(k_{\rm tr}+k_0)\exp(-ik_{\rm tr}L/2)}{(k_{\rm tr}+k_0)^2\exp(-ik_{\rm tr}L)-(k_{\rm tr}-k_0)^2\exp(ik_{\rm tr}L)} , \quad (27a)$$

$$d_\downarrow=\frac{2k_0(k_{\rm tr}-k_0)\exp(ik_{\rm tr}L/2)}{(k_{\rm tr}+k_0)^2\exp(-ik_{\rm tr}L)-(k_{\rm tr}-k_0)^2\exp(ik_{\rm tr}L)} . \quad (27b)$$

Thus the amplitude of the electric field in the interior of the film in the case of normal incidence is

$$\mathbf{E}^0(\mathbf{r})=E^{\rm inc}\mathbf{e}_x[d_\uparrow\exp(ik_{\rm tr}z)+d_\downarrow\exp(-ik_{\rm tr}z)] . \quad (28)$$

C. Green's function

The system has translational-invariance symmetry parallel to the surfaces, so the Green's function depends on $t-t'$, $\mathbf{r}_\perp-\mathbf{r}'_\perp$, z, and z' and satisfies the equation

$$\left[\nabla\times\nabla\times+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\hat{\epsilon}(\mathbf{r})\right]\hat{T}(t-t',\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')=\hat{I}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') , \quad (29)$$

where $\hat{I}$ is the identity matrix and tensor $\hat{\epsilon}$ is given by

$$\hat{\epsilon}_{\alpha\beta}(\mathbf{r})=\begin{cases}\epsilon_0\delta_{\alpha\beta} & \text{if } |z|>L/2\\ \epsilon_\parallel n^0_\alpha n^0_\beta+\epsilon_\perp(\delta_{\alpha\beta}-n^0_\alpha n^0_\beta) & \text{if } |z|<L/2 .\end{cases} \quad (30)$$

Due to the causality principle this function is equal to zero when t is less than t'. According to Eq. (26) we must calculate the Green's function in $(\omega,\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')$ representation for $|z'|<L/2$. It is convenient to calculate it in $(\omega,\boldsymbol{\kappa},z,z')$ representation

$$\hat{T}(\omega,\boldsymbol{\kappa},z,z')=\int d^2r_\perp\int dt\, e^{i\omega t-i\boldsymbol{\kappa}\cdot\mathbf{r}}\hat{T}(t,\mathbf{r}_\perp,z,z') \quad (31)$$

and then to complete inverse Fourier transform in the xy plane. It should be noted that due to the causality principle one can take the limit $\omega=\omega+i0$ in Eq. (31), which allows one to carry our integration if singularities take place.

We will find the Green's function $\hat{T}(\omega,\boldsymbol{\kappa},z,z')$ in the following form:

$$T_{\alpha\beta}(\omega,\boldsymbol{\kappa},z,z')=\sum_{j=1}^{2}A^{(\sigma j)}e'^{(\sigma j)}_\alpha e^{(\sigma j)}_\beta \times\exp[i(k_0^2-\kappa^2)^{1/2}(\sigma z-L/2)] \quad (32a)$$

if $|z|>L/2$, and

$$T_{\alpha\beta}(\omega,\boldsymbol{\kappa},z,z')=T^{(0)}_{\alpha\beta}(\omega,\boldsymbol{\kappa},z,z')+\sum_{j=-2}^{2}{}'B^{(j)}e^{(j)}_\alpha e^{(j)}_\beta\exp[ik_z^{(j)}z] \quad (32b)$$

if $|z|>L/2$. Here $\hat{T}^{(0)}$ is the Green's function for an infinite optically uniaxial medium, while the other terms being multiplied by $\exp(i\kappa+\mathbf{r}_\perp)$ represent amplitudes of plane waves with polarization $e'^{(j)}_\alpha$ in the exterior and with $e^{(j)}_\alpha$ in the interior of the film, $j=-2,-1,1,2$. The positive numbers j correspond to waves propagating in the positive direction with respect to the z axis, the negative ones correspond to waves that propagate in the negative direction, and σ is equal to $\mathrm{sgn}(z-z')$. The indices $j=1,-1$ are referred to amplitudes, wave vectors, and polarization vectors of waves that polarized in the xy plane and $j=2,-2$ are referred to characteristics of waves with polarization in the $\boldsymbol{\kappa}z$ plane. Wave vectors $\mathbf{k}^{(j)}$ of the ordinary and extraordinary waves in the film can be written as

$$\mathbf{k}^{(\pm1)}=(\boldsymbol{\kappa},\pm(k_{\mathrm{tr}}^2-\kappa^2)^{1/2}) , \quad (33a)$$

$$\mathbf{k}^{(\pm2)}=(\boldsymbol{\kappa},\pm(k_{\mathrm{tr}}/k_{\mathrm{al}})(k_{\mathrm{al}}^2-\kappa^2)^{1/2}) , \quad (33b)$$

where $k_{\mathrm{al}}=\omega\epsilon_\parallel^{1/2}/c$. The corresponding unit polarization vectors $\mathbf{e}^{(j)}$ are given by [12]

$$\mathbf{e}^{(\pm1)}=\frac{[\mathbf{k}^{(\pm1)},\mathbf{n}^0]}{|[\mathbf{k}^{(\pm1)},\mathbf{n}^0]|} , \quad (34a)$$

$$\mathbf{e}^{(\pm2)}=\frac{[\hat{\epsilon}\mathbf{k}^{(\pm2)},[\mathbf{n}^0,\mathbf{k}^{(\pm2)}]]}{|[\mathbf{n}^0,\mathbf{k}^{(\pm2)}]|(\mathbf{k}^{(\pm2)},\hat{\epsilon}^2\mathbf{k}^{(\pm2)})^{1/2}} . \quad (34b)$$

The expressions for wave vectors and for polarization vectors $\mathbf{e}'^{(j)}$ in the case of an isotropic surrounding medium with permittivity ϵ_0 are given by similar formulas but with $\epsilon_{\alpha\beta}$ replaced by $\epsilon_0\delta_{\alpha\beta}$ and with $k_{\mathrm{al}},k_{\mathrm{tr}}$ replaced by k_0.

One can see that the function in form (32) with arbitrary coefficients $A^{(j)}$ and $B^{(j)}$ satisfies Eq. (29) everywhere except boundaries due to the special choice of $\hat{T}^{(0)}$. The Green's function for an infinite optically uniaxial medium is discussed in detail in Ref. [12]. In order to obtain $\hat{T}^{(0)}(\omega,\boldsymbol{\kappa},z-z')$ it is convenient to take inverse Fourier transform of the Green's function in $(\omega,\mathbf{k})$ representation over k_z. Applying the results of [12] and taking into account the causality principle one can get

$$T^{(0)}_{\alpha\beta}(\omega\boldsymbol{\kappa}z-z')=\sum_{j=1,2} h^{(j)}\exp[i|z-z'|k_z^{(j)}]e_\alpha^{(\sigma j)}e_\beta^{(\sigma j)} -\frac{1}{k_{\mathrm{al}}^2}\delta_{\alpha3}\delta_{\beta3}\delta(z-z') , \quad (35)$$

where

$$h^{(1)}=-\frac{1}{2i(k_{\mathrm{tr}}^2-\kappa^2)^{1/2}} ,\qquad h^{(2)}=-\frac{\kappa^2(k_{\mathrm{tr}}^2-k_{\mathrm{al}}^2)+k_{\mathrm{al}}^4}{2ik_{\mathrm{al}}^3k_{\mathrm{tr}}(k_{\mathrm{al}}^2-\kappa^2)^{1/2}} , \quad (36)$$

and $\sigma=\mathrm{sgn}(z-z')$.

We must find such coefficients $A^{(j)}$ and $B^{(j)}$, which satisfy boundary conditions at $z=\pm L/2$. It should be noted that there is no wave type mixing, because the director is normal to the boundaries, i.e., reflected and refracted waves are polarized in the same plane as the incident one. According to the Fresnel formulas, the reflected and refracted wave amplitudes are proportional to the incident one. Let $\tau^{(j)}$ and $\rho^{(j)}$ denote Fresnel coefficients for refracted and reflected waves, respectively. In the case of incidence out of an optically uniaxial medium they are given by [11]

$$\rho^{(\pm1)}=\frac{k_z-k_{\uparrow z}}{k_{\uparrow z}-k_{\downarrow z}},\quad \tau^{(\pm1)}=\frac{2k_z}{k_{\uparrow z}-k_{\downarrow z}} , \quad (37a)$$

$$\rho^{(\pm2)}=\frac{\epsilon_0k_z-\epsilon_{\mathrm{tr}}k_{\uparrow z}}{\epsilon_0k_z+\epsilon_{\mathrm{tr}}k_{\uparrow z}} ,\qquad \tau^{(\pm2)}=\frac{2\epsilon_\parallel k_0}{(\mathbf{k},\hat{\epsilon}\mathbf{k})}\frac{\epsilon_{\mathrm{tr}}k_z}{\epsilon_0k_z+\epsilon_{\mathrm{tr}}k_{\uparrow z}} , \quad (37b)$$

where $\mathbf{k}$, $\mathbf{k}_\uparrow$, and $\mathbf{k}_\downarrow$ are the incident, refracted, and reflected wave vectors, respectively. Their x,y components are the same, while z components depend on the type and propagation direction of the waves. Thus the boundary conditions that determine the coefficients $A^{(j)}$ and $B^{(j)}$ $(j=-2,-1,1,2)$ can be expressed in terms of $\rho^{(j)}$ and $\tau^{(j)}$ as follows:

$$A^{(m)}=\tau^{(m)}\left\{h^{(m)}\exp\left[ik_z^{(m)}\left[\frac{L}{2}-z'\right]\right] +B^{(m)}\exp\left[ik_z^{(m)}\frac{L}{2}\right]\right\} , \quad (38a)$$

$$B^{(-m)}=\rho^{(m)}\{h^{(m)}\exp[ik_z^{(m)}(L-z')] +B^{(m)}\exp[ik_z^{(m)}L]\} , \quad (38b)$$

$$A^{(-m)}=\tau^{(m)}\left\{h^{(m)}\exp\left[ik_z^{(m)}\left[\frac{L}{2}+z'\right]\right] +B^{(-m)}\exp\left[ik_z^{(m)}\frac{L}{2}\right]\right\} , \quad (38c)$$

$$B^{(m)}=\rho^{(m)}\{h^{(m)}\exp[ik_z^{(m)}(L+z')] +B^{(-m)}\exp[ik_z^{(m)}L]\} , \quad (38d)$$

where $m=1,2$. This system of equations leads to the expressions

$$B^{(m)}=\rho^{(m)}h^{(m)}\frac{\exp[ik_z^{(m)}z']+\rho^{(m)}\exp[ik_z^{(m)}(L-z')]}{\exp[-ik_z^{(m)}L]-\rho^{(m)2}\exp[ik_z^{(m)}L]} , \quad (39a)$$

$$A^{(m)}=\tau^{(m)}h^{(m)}\exp\left[ik_z^{(m)}\frac{L}{2}\right] \times\frac{\exp[-ik_z^{(m)}(L+z')]+\rho^{(m)}\exp[ik_z^{(m)}z']}{\exp[-ik_z^{(m)}L]-\rho^{(m)2}\exp[ik_z^{(m)}L]} . \quad (39b)$$

The corresponding coefficients $A^{(j)}$ and $B^{(j)}$ with $j=-m$ are given by similar expressions but with z' replaced by $-z'$.

We are interested in the Green's function in real-space representation when the point $\mathbf{r}'$ is in the interior of the film, while $\mathbf{r}$ is outside of it. According to Eqs. (31) and (32a) this function can be written as

$$T_{\alpha\beta}(\omega,\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')=\frac{1}{(2\pi)^2}\int d^2\kappa \exp\left[i\boldsymbol{\kappa}\cdot(\mathbf{r}_\perp-\mathbf{r}'_\perp)+i(k_0^2-\kappa^2)^{1/2}\left[\sigma z-\frac{L}{2}\right]\right]\sum_{j=1}^{2}A^{(\sigma j)}(\boldsymbol{\kappa},z')e'^{(\sigma j)}_\alpha e^{(\sigma j)}_\beta \ . \tag{40}$$

Since the light-scattering process takes place in the interior of the film but the intensity is detected outside of it at the large distances (compared with the wavelength of light), we can use in Eq. (26) the asymptotic expression for $\hat{T}$ instead of the precise one. We assume that $\mathbf{r}=\mathbf{s}R$, where $\mathbf{s}$ is a unit vector, and $R \gg |\mathbf{r}'|$. Carrying out the integration over $\boldsymbol{\kappa}$ in Eq. (40) by means of the stationary phase method, we get

$$T_{\alpha\beta}(\omega,\mathbf{r}_\perp-\mathbf{r}'_\perp,z,z')=\frac{k_0|s_z|}{2i\pi R}\exp\left[ik_0R-ik_0\left[\mathbf{s}_\perp\cdot\mathbf{r}'_\perp+|s_z|\frac{L}{2}\right]\right]\sum_{j=1}^{2}A^{(\sigma j)}(k_0\mathbf{s}_\perp,z')e'^{(\sigma j)}_\alpha e^{(\sigma j)}_\beta \ , \tag{41}$$

where $\sigma=\mathrm{sgn}(z-z')$, s_z and $\mathbf{s}_\perp$ are the components of unit vector $\mathbf{s}$, and $\mathbf{e}'^{(j)}$ and $\mathbf{e}^{(j)}$ are the polarization vectors (in surrounding space and in the film, respectively) of waves of type j with wave vector $(\mathbf{s}_\perp k_0, k_z^{(j)})$. This Green's function takes into account the air-film interface effects and can be utilized when the optical axis in the optically uniaxial film is normal to its surfaces.

D. The intensity of the scattered light

According to Eq. (35) the scattered field in the far zone is a spherical wave but because R is much longer than the wavelength, the field can be considered locally as a plane wave with wave vector $k_0\mathbf{s}_\perp$. Thus the intensity I at the point $\mathbf{r}=R\,\mathbf{s}$ of the scattered waves at the point $\mathbf{r}=R\,\mathbf{s}$ is given by

$$I=\frac{c\epsilon_0}{8\pi}\mathrm{Tr}\langle \mathbf{E}'(\mathbf{r})\otimes\mathbf{E}'^*(\mathbf{r})\rangle \ . \tag{42}$$

We will calculate separately the intensity $I^{(2)}$ of waves polarized in the scattering plane and intensity $I^{(1)}$ of waves polarized in the xy plane. Applying Eqs. (42), (26), and (41) we get

$$I^{(m)}=I_0\frac{\omega^4k_0^2s_z^2}{c^4(2\pi R)^2}e^{(\sigma m)}_\gamma e^{(\sigma m)}_\lambda\int d^3r'd^3r''\exp[-ik_0\mathbf{s}_\perp\cdot(\mathbf{r}'_\perp-\mathbf{r}''_\perp)]A^{(\sigma m)}(k_0\mathbf{s}_\perp,z')A^{(\sigma m)*}(k_0\mathbf{s}_\perp,z'')$$
$$\times\langle\delta\epsilon_{\gamma 1}(\mathbf{r}')\delta\epsilon_{\lambda 1}(\mathbf{r}'')\rangle[d_\uparrow\exp(ik_{\mathrm{tr}}z')+d_\downarrow\exp(-ik_{\mathrm{tr}}z')][d_\uparrow^*\exp(-ik_{\mathrm{tr}}z'')+d_\downarrow^*\exp(ik_{\mathrm{tr}}z'')] \ , \tag{43}$$

where I_0 is the intensity of the incident light, $m=1,2$. The integration over $\mathbf{r}'_\perp-\mathbf{r}''_\perp$ is Fourier transform of the correlation function but integration over $\mathbf{r}''_\perp$ is reduced to multiplication by illuminated surface S. Using expressions (40) and (39b) in Eq. (43) we get in the case of normal incidence

$$I^{(1)}=0 \ , \tag{44}$$
$$I^{(2)}=I_0Cs_z^2s_x^2s_\perp^2\frac{\epsilon_\perp^2k_0(k_0^2s_\perp^2(k_{\mathrm{tr}}^2-k_{\mathrm{al}}^2)+k_{\mathrm{al}}^2)^2}{k_{\mathrm{al}}^6k_{\mathrm{tr}}^2(k_{\mathrm{al}}^2-k_0^2s_\perp^2)(\mathbf{k},\hat{\epsilon}\mathbf{k})}\left|\frac{\tau^{(2)}}{1-[\rho^{(2)}\exp(ik_z^{(2)}L)]^2}\right|^2\frac{1}{L}$$
$$\times\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''G(k_0\mathbf{s},z',z'')\{\exp[-ik_z^{(2)}(L+\sigma z')]+\rho^{(2)}\exp[ik_z^{(2)}\sigma z']\}$$
$$\times[d_\uparrow\exp(ik_{\mathrm{tr}}z')+d_\downarrow\exp(-ik_{\mathrm{tr}}z')]\{\exp[ik_z^{(2)}(L+\sigma z'')]+\rho^{(2)}\exp[-ik_z^{(2)}\sigma z'']\}$$
$$\times\{d_\uparrow^*\exp(ik_{\mathrm{tr}}z')+d_\downarrow^*\exp(-ik_{\mathrm{tr}}z')\} \ ,$$

where

$$C=\frac{\omega^4k_0V\epsilon_a^2}{c^4(4\pi R)^2} \ .$$

We also used the following relations:

$$e_z^{(2)}=\frac{\epsilon_{\mathrm{tr}}k_0s_\perp}{(\mathbf{k},\hat{\epsilon}^2\mathbf{k})},\quad V=SL \ ,$$

$$k_z^{(2)}=\frac{k_{\mathrm{tr}}}{k_{\mathrm{al}}}(k_{\mathrm{al}}^2-s_\perp^2k_0^2)^{1/2} \ .$$

All integrals in Eq. (44) can be calculated analytically due to exponential dependence of the correlation function G on z and z'. We note that there is no scattering of the ordinary wave to the ordinary one in the smectic liquid crystal. The same situation takes place in an aligned nematic liquid crystal [7].

The theory presented above takes into account the multiple-reflection effects for the incident and the scattered beams. Expression (44) together with Eq. (18) allows one to calculate scattered light intensity measured experimentally for arbitrary surface tensions $\gamma_\pm$.

We restrict ourselves to symmetric boundary conditions ($\gamma_+=\gamma_-=\gamma$). Under such circumstances the following equalities are valid:

$$G(\boldsymbol{\kappa},z',z'')=G(\boldsymbol{\kappa},z'',z')=G(\boldsymbol{\kappa},-z',-z'')\ . \tag{45}$$

Intensity of scattered light $I^{(2)}$ can be written as

$$I^{(2)}=I_0 C s_z^2 s_x^2 s_\perp^2 \left| \frac{\tau^{(2)}}{1-[\rho^{(2)}\exp(ik_z^{(2)}L)]^2} \right|^2 \frac{\epsilon_\perp^2 k_0^4 [s_\perp^2 k_0^2 (k_{\rm tr}^2-k_{\rm al}^2)+k_{\rm al}^4]^2}{k_{\rm tr}^6 (k_{\rm al}^2 - s_\perp^2 k_0^2) k_{\rm tr}^2}$$
$$\times\{ W(k_{\rm tr}-\sigma k_z^{(2)},-k_{\rm tr}+\sigma k_z^{(2)})[|d_\uparrow|^2+|\rho^{(2)}d_\downarrow|^2]+W(k_{\rm tr}+\sigma k_z^{(2)},-k_{\rm tr}-\sigma k_z^{(2)})[|d_\downarrow|^2+|\rho^{(2)}d_\uparrow|^2]$$
$$+W(k_{\rm tr}+\sigma k_z^{(2)},k_{\rm tr}+\sigma k_z^{(2)})[\rho^{(2)}\exp(ik_z^{(2)}L)d_\uparrow d_\downarrow^* +\text{c.c.}]$$
$$+W(k_{\rm tr}-\sigma k_z^{(2)},k_{\rm tr}-\sigma k_z^{(2)})[\rho^{(2)}\exp(ik_z^{(2)}L)d_\downarrow d_\uparrow^* +\text{c.c.}]$$
$$+W(k_{\rm tr}-\sigma k_z^{(2)},k_{\rm tr}+\sigma k_z^{(2)})[d_\uparrow d_\downarrow^* +\rho^{(2)^2}d_\downarrow d_\uparrow^* +\text{c.c.}]$$
$$+W(k_{\rm tr}+\sigma k_z^{(2)},-k_{\rm tr}+\sigma k_z^{(2)})[|d_\uparrow|^2+|d_\downarrow|^2][\rho^{(2)}\exp(ik_z^{(2)}L)+\text{c.c.}]\}\ , \tag{46}$$

where the function $W(q',q'')$ is defined by

$$W(q',q'')=\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''e^{iq'z'+iq''z''}G(sk_0,z',z'')\ . \tag{47}$$

This function represents the fluctuation properties of smectic-A film that manifest themselves in the light-scattering process. This function possesses, similar to (45), symmetry properties

$$W(q',q'')=W(q'',q')=W(-q',-q'')\ ,$$

which were used in Eq. (46). Utilizing the expression for correlation function (18) and expressions for integrals

$$\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''e^{iq'z'+iq''z''}\cosh(gz'+gz'')$$
$$=\frac{2}{(q'^2+g^2)(q''^2+g^2)}\left\{(q'q''-g^2)\left[\cos\left[(q'-q'')\frac{L}{2}\right]-\cos\left[(q'+q'')\frac{L}{2}\right]\cosh(gL)\right]\right.$$
$$\left.+g(q'+q'')\sin\left[(q'+q'')\frac{L}{2}\right]\sinh(gL)\right\}\ , \tag{48}$$

$$\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''e^{iq'z'+iq''z''}\sinh(g|z'-z''|)$$
$$=\frac{2}{(q'^2+g^2)(q''^2+g^2)}\left\{-g\frac{q'^2+q''^2+2g^2}{q'+q''}\sin\left[(q'+q'')\frac{L}{2}\right]\right.$$
$$\left.+g(q'-q'')\sin\left[(q'-q'')\frac{L}{2}\right]\cosh(gL)+(q'q''+g^2)\cos[(q'-q'')]\sinh(gL)\right\}\ , \tag{49}$$

one can get the explicit formula for W,

$$W(q',q'')=\frac{k_BT}{BL}\left\{2(a+g)\left[-(q'+q'')(ag+q'q'')\cos\left[(q'+q'')\frac{L}{2}\right]\right.\right.$$
$$\left.+[a(g^2-q'q'')+g(g^2+q'^2+q''^2+q'q'')]\sin\left[(q'+q'')\frac{L}{2}\right]\right]$$
$$+4g(q'+q'')e^{-gL}\left[(a^2+q'q'')\cos\left[(q'-q'')\frac{L}{2}\right]-a(q'-q'')\sin\left[(q'-q'')\frac{L}{2}\right]\right]$$
$$-2(a-g)e^{-2gL}\left[(q'+q'')(ag-q'q'')\cos\left[(q'+q'')\frac{L}{2}\right]\right.$$
$$\left.\left.+[a(g^2-q'q'')-g(g^2+q'^2+q''^2+q'q'')]\sin\left[(q'+q'')\frac{L}{2}\right]\right]\right\}$$
$$\times\{(q'+q'')(q'^2+g^2)(q''^2+g^2)[(a+g)^2-e^{-2gL}(a-g)^2]\}^{-1}\ , \tag{50}$$

where $g(K/B)^{1/2}s_\perp^2k_0^2$, $a=(\gamma/B)s_\perp^2k_0^2$.

Formulas (50) and (46) describe the light-scattering process in smectic-A film. In order to understand it we will consider separately each term of Eq. (50). Assume σ to be equal to 1. It means that the point $\mathbf{r}$, where the intensity $I^{(2)}$ is studied, is above the film $(z>0)$. The term with coefficient $|d_\uparrow|^2$ in the first term in braces is the contribution to the intensity of scattering of wave with wave vector $\mathbf{e}_z k_{\rm tr}$ into the extraordinary one with wave vector $(\mathbf{s}k_0, k_z^{(2)})$. The term with coefficient $|\rho^{p(2)}d_\downarrow|^2$ is the contribution of wave with wave vector $-\mathbf{e}_z k_{\rm tr}$ into one with wave vector $(\mathbf{s}k_0, -k_z^{(2)})$ and then reflected from the bottom boundary $(z=L/2)$. The term with coefficient $|d_\downarrow|^2$ in the second term of the expression in braces is the contribution of scattering of wave with wave vector $\mathbf{e}_z k_{\rm tr}$ into one with $(\mathbf{s}k_0, k_z^{(2)})$. The term with coefficient $|\rho^{(2)}d_\uparrow|^2$ in the same square brackets is the contribution of scattering of wave with wave vector $\mathbf{e}_z k_{\rm tr}$ into one with $(\mathbf{s}k_0, -k_z^{(2)})$ and then reflected from the bottom boundary. The other terms of Eq. (50) describe interference of the scattered waves with different wave vectors. For example, the term with coefficient $\rho^{(2)}d_\uparrow d_\downarrow^*$ corresponds to interference of two scattered waves, one of which reflected from a boundary, with wave vectors $(\mathbf{s}k_0, k_z^{(2)})$ and $(\mathbf{s}k_0, -k_z^{(2)})$. The main contribution to the intensity is related with scattering of that wave which propagates upward because $|d_\downarrow|$ is less than $|d_\uparrow|$. According to Eq. (27) the ratio $|d_\downarrow|/|d_\uparrow|$ for typical permittivities $(\epsilon_0=1, \epsilon_{\rm tr}=3)$ is about 0.25.

To analyze the effects connected with the fluctuation features we neglect now interface and anisotropy optical effects. Assume Green's function $\hat{T}$ to be one for the far zone in an infinite isotropic medium with permittivity ϵ_0 and neglect the reflected incident wave in the interior $(d_\uparrow=1, d_\downarrow=0)$. We get in this case

$$I^{(2)}=I_0Cs_x^2s_\perp^2W(q_z,-q_z)\;, \tag{51}$$

where $\mathbf{q}=k_0(\mathbf{s}_\perp, s_z-1)$ is the scattering vector. The function W is reduced to the simpler expression

$$W(q_z,-q_z)=\frac{k_BT}{BL}\{(a+g)[2q_z^2-2ag+L(a+g)(q_z^2+g^2)]+4ge^{-gL}[(a^2-q_z^2)\cos(q_zL)-2aq_z\sin(q_zL)]$$
$$+(g-a)e^{-2gL}[2ag+2q_z^2+L(a-g)(g^2+q_z^2)]\}$$
$$\times\{(g^2+q_z^2)^2[(a+g)^2-e^{-2gL}(a-g)^2]\}^{-1}\;, \tag{52}$$

where a and g are the same as in Eq. (50). Considering the scattering process in the xz plane we also assume $s_x=s_\perp=\sin\theta$, where θ is a scattering angle. So we can set

$$q_z=-2k_0\sin^2(\theta/2),\quad g=(K/B)^{1/2}k_0^2\sin^2\theta\;,$$
$$a=(\gamma/B)k_0^2\sin^2\theta\;. \tag{53}$$

It is seen that function W has singularity at the point $\theta=0$. Applying expansions $e^{-gL}=1-gL+O(\theta^4)$, $\sin(q_zL)=q_zL+O(\theta^6)$, and $\cos(q_zL)=1+O(\theta^4)$, one can find that W increases as θ^{-2} if L is finite. Taking into account the dependence of $s_\perp$ on the scattering angle, according to Eq. (51), we get that the intensity $I^{(2)}$ decreases as θ^2 when angle θ vanishes. Rather different angular dependence takes place if the thickness L is infinite. Expression (52) for infinite sample evolves to

$$W(q_z,-q_z)=\frac{k_BT}{B(g^2+q_z^2)}\;, \tag{54}$$

so we get the well-known [7] expression for the intensity $I^{(2)}$,

$$I^{(2)}=I_0Cs_\perp^2s_x^2\frac{k_BT}{Kq_\perp^4+Bq_z^2} \tag{55a}$$

per the same illuminated volume, where $\mathbf{q}=(\mathbf{q}_\perp, q_z)$ is the scattering vector. As distinct from the intensity in the film case this value remains finite when the scattering angle vanishes. Thus the fact that there is no forward scattering in a film is a pure finite-size effect.

To analyze the angular dependence let us consider separately the case of $\gamma_+=\gamma_-=0$ in more detail. We can apply expansion (19) of the correlation function to the sum of modes. The use of Eq. (19) in Eq. (47) gives the expression

$$I^{(2)}=I_0C\frac{2k_BT}{BL^2}s_\perp^2s_x^2\left\{\frac{2}{q_z^{(2)2}g^2}\sin^2\left[q_z^{(2)}\frac{L}{2}\right]\right.$$
$$\left.+L^4\sum_{m>0}\frac{1}{(gL)^2+(\pi m)^2}\left[\frac{\sin[(\pi m-q_z^{(2)}L)/2]}{\pi m-q_z^{(2)}L}+\frac{\sin[(\pi m+q_z^{(2)}L)/2]}{\pi m+q_z^{(2)}L}\right]^2\right\}\;, \tag{55b}$$

where $q_z^{(2)}$ and g are the same as in Eq. (35). A main contribution for thin film comes from the first term in the braces. Due to the coefficient $\sin^2(q_zL/2)$ this contribution oscillates with the increasing of the scattering angle. According to Eq. (55b) extrema must be at the points θ_m, which satisfy the equation

$$\pi m-Lk_0(1-\cos\theta_m)\;. \tag{56}$$

Odd numbers m correspond to maxima and even numbers correspond to minima. So, there are oscillations in angular dependence and the positions of the extrema are defined by correlation of the thickness and the wavelength. It is seen that the frequency of the oscillations increases with the thickness of the film. A similar situation takes place when two waves, reflected from different interfaces of a film, are interfering. Here we have no reflections but only scattering in the film takes place.

We now apply the presented theory to consider a dependence of the intensity on the thermodynamical and optical parameters of the film. Curves, calculated for films with various thickness (10^{-4} cm, 10^{-3} cm, ∞) and with the other parameters similar ($B=2.5\times10^7$ dyn/cm, $K=10^{-6}$ dyn, $\gamma=10$ dyn/cm) and $k_0=10^5$ cm^{-1} are presented in Fig. 2 to show dependence on the thickness L. This calculation has been carried out on the basis of Green's function for an isotropic infinite medium, so it does not take into account multiple-reflection and anisotropical effects. The dashed line corresponds to intensity in infinite sample per the same volume. It can be seen that there is a gap at zero scattering angle, if L is a finite value. A width of the gap decreases with increasing of L. So, there is no gap in the case of an infinite sample.

To estimate the positions of the extrema let us apply Eq. (55a), which is true only if the surface tension is equal to zero. According to Eq. (56) we get $\theta_1=14.4°$, $\theta_3=25.1°$ for maxima and $\theta_2=20.4°$, $\theta_4=29.0°$ for minima, when $L=10^{-3}$ cm. The same results show the curve Fig. 2, which is plotted for $\gamma=10$ dyn/cm. Thus the positions of the extrema depend slightly on γ and are primarily determined by L and wavelength.

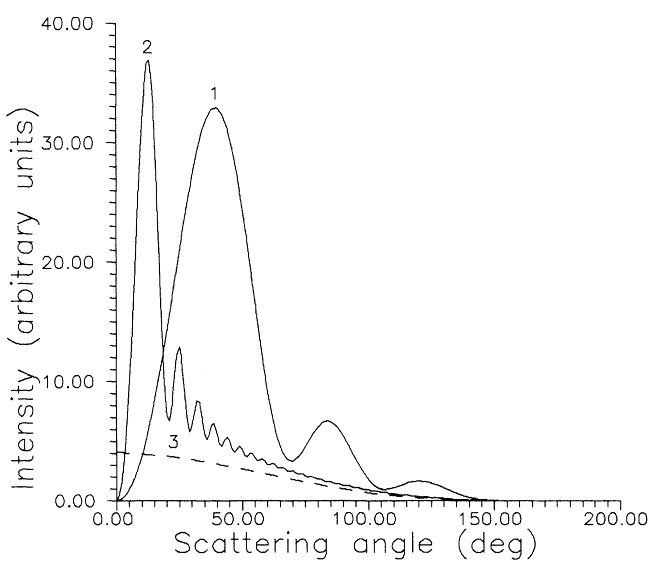

FIG. 2. The intensity of the scattered light $I^{(2)}$ vs the scattering angle θ. The calculation was carried out for the case of scattering in the xz plane without taking into account interface effects; $k_0=10^5$ cm^{-1}, $\gamma=10$ dyn/cm, $B=2.5\times10^7$ dyn/cm^2, $K=10^{-6}$ dyn; (1) $L=10^{-4}$ cm, (2) $L=10^{-3}$ cm, and (3) the dashed curve corresponds to the intensity per the same volume in an infinite sample.

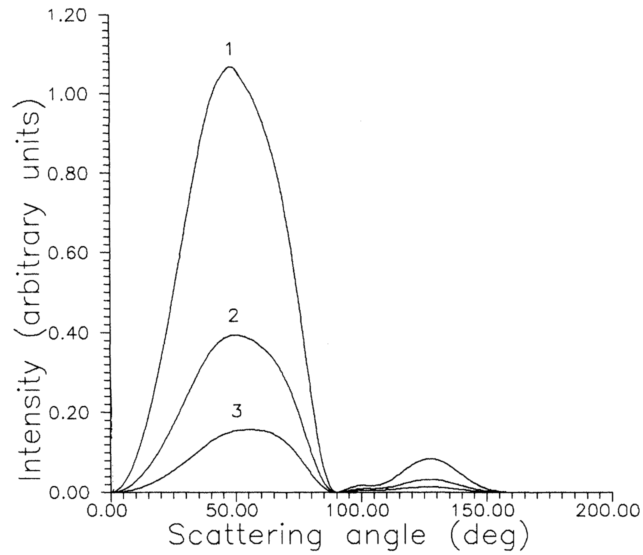

FIG. 3. The intensity of the scattered light $I^{(2)}$ vs the scattering angle θ. The calculation was carried out for the case of scattering in the xz plane, taking into account interface effects and optical anisotropy of the film; $\epsilon_\parallel=3.3$, $\epsilon_\perp=3.0$, $k_0=10^5$ cm^{-1}, $L=10^{-4}$ cm, $B=2.5\times10^7$ dyn/cm^2, $K=10^{-6}$ dyn; (1) $\gamma=10$ dyn/cm, (2) $\gamma=30$ dyn/cm, and (3) $\gamma=100$ dyn/cm.

The curves in Fig. 3 show angular dependence when interface effects take place. The calculation has been carried out taking into account optical anisotropy and multiple-reflection effects for the incident and scattered beams. Film parameters have been assumed to be $L=10^{-4}$ cm, $\epsilon_\parallel=3.3$, and $\epsilon_\perp=3.0$, elastic constants K and B are the same as for Fig. 2. In order to show the dependence on surface tension γ three values have been chosen ($\gamma=10$, 30, and 100 dyn/cm). It can be seen, first, that the positions of the extrema differ from that of the corresponding curve in Fig. 2. It occurs due to refraction at the boundaries. There is undulation behavior of the intensity as a function of θ when $\theta>\pi/2$ (backward scattering). The origin of this behavior is connected with reflection of forward scattered light from the interface $z=L/2$ and scattering of the reflected incident wave with amplitude $\mathbf{E}_0 d_\downarrow$ on small angles. It also can be seen that the magnitudes of the peaks depend strongly on the surface tension.

IV. DISCUSSION

The analysis presented involves a calculation of the displacement-displacement correlation function for smectic-A films including surface phenomena. The expressions for the case of unequal surface tension coefficients describing the opposite sides of a film can be applied to systems studied experimentally. It is shown that the boundary conditions strongly affect the fluctuations not only at the surface but in the interior of the film as well. The explicit expressions presented for the displacement-displacement correlation function can also be used for an analysis of x-ray scattering in smectic liquid crystals.

The developed theory of the light-scattering process in smectic-A films allows one to calculate the intensity measured experimentally. The interface and anisotropy optical effects are taken into account. The theory elucidates two finite-size effects. First, there is no forward scattering in the case of normal incidence, whereas in the infinite sample the scattering with scattering angle equal to zero is stronger than that with any others. The width of the gap at $\theta=0$ in the variation of the intensity with θ tends to zero when the thickness of the film increases. Second, there is the undulation behavior of this variation. This behavior keeps on when interface optical effects are not taken into account, so it is not caused by reflections from the boundaries. The origin of this behavior is connected with the thermodynamical properties of smectic liquid crystals. Indeed, smectic layers are practically incompressible, hence the random displacements of different layers are well correlated in the direction normal to layers. This gives rise to the interference of waves scattered at points with the same xy but various z coordinates. The increasing of the film thickness leads to an increase in the frequency of the oscillations. The positions of the extrema hardly depend on surface tension, but the magnitudes of the peaks do.

Since the angular distribution of the scattered light is sensitive to the surface tension coefficient, the latter can be measured by means of the optical experiment. It is an important result because smectic liquid crystals, because of their elastic properties and extremely large viscosity, are similar to solids, so the surface tension coefficient cannot be measured by means of the traditional methods available for liquids.

Although the formulas presented for the intensity are valid for normal incidence, they allow one to understand what happens in the case of oblique incidence. The description can be carried out in analogous fashion to the one presented for normal incidence with the same Green's function and W, but with another dependence of wave and polarization vectors on the scattering angle. For example, if the incident wave outside the film is polarized in the plane normal to the incident plane, two ordinary waves are produced in the interior of the film. Only extraordinary scattered waves are produced in this case, so $I^{(1)}=0$. Since the ordinary and extraordinary wave numbers differ from each other, the scattering vector is not equal to zero for any scattering angle. Thus W as a function of the scattering angle has no singularities. A quite different situation takes place when the incident wave outside the film is polarized in the incidence plane. Two incident extraordinary waves are produced inside the film. They are scattered into extraordinary as well as ordinary waves, so both intensities $I^{(1)}$ and $I^{(2)}$ are not equal to zero in general. The function W for extraordinary-ordinary light scattering goes finite for the same reason, while for extraordinary-extraordinary light scattering it is infinite when the scattering angle is equal to zero. This function increases as θ^{-2} as θ decreases. Due to other angular-dependent coefficients in the expression for the intensity the last goes finite. The particular case when $I^{(2)}$ tends to zero is the case of the normal incidence considered in detail above.

We also note that the expression presented here for the Green's function can be applied to studies of arbitrary films with optical axis normal to surfaces when multiple-reflection effect and optical anisotropy are essential. It can be modified easily for various interface conditions depending on experimental setup by replacing Fresnel coefficients $\tau^{(j)}$ and $\rho^{(j)}$ with appropriate ones in Eq. (38).

We hope that the analysis presented will be useful for further experimental and theoretical studies of thin smectic systems.

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