NANOSYSTEMS: PHYSICS, CHEMISTRY, MATHEMATICS, 2015, 6 (4), P. 489–500

The interaction of polarization charges in freely suspended smectic-C* films

D. A. Murich[1], V. P. Romanov[1], S. V. Ul'yanov[1,2]

[1]Saint Petersburg State University, Ul'yanovskaya, 1, Petrodvoretz, Saint Petersburg 198504, Russia

[2]Saint Petersburg State University of Commerce and Economics, Saint Petersburg 194021, Russia

murichdim@mail.ru, ulyanov_sv@mail.ru

PACS 61.30.Dk, 61.30.Gd, 42.79.Hp, 42.70.Df

DOI 10.17586/2220-8054-2015-6-4-489-500

The fluctuations of the director orientation in a freely suspended smectic-C* film were theoretically investigated. In the free energy expression of the film, not only were the elastic energy and the weak external electric field interaction considered, the interaction of polarization charges arising from fluctuations of the polarization vector were also included. The correlation function of the director fluctuations was obtained for a film of finite thickness. Calculations of light scattering intensity were provided. It has been found that due to the interaction of polarization charges, the angular dependence of the scattering intensity significantly depends on the magnitude of spontaneous polarization.

Keywords: smectic-C*, freely suspended films, polarization charges, correlation function, light scattering intensity.

Received: 20 June 2015

1. Introduction

Smectic-C* liquid crystals have attracted wide attention from researchers for several decades. This is primarily due to the unique physical properties inherent to these substances [1]. Smectics-C* (Sm-C*) are well-documented as layered systems consisting of monomolecular layers composed of elongated molecules which are inclined relative to the normal to the layers. This leads to the creation of a director vector, **n**, which gives the average direction of preferred molecule orientation, and is inclined at an angle θ relative to the normal **N** to the smectic layers. At constant temperature, the angle θ can be considered constant throughout the liquid crystal. Each layer in Sm-C* can be considered as a two-dimensional liquid. In addition, the Sm-C* possesses spontaneous polarization, and the polarization vector **P** in each point of the liquid crystal is perpendicular to both the director **n**, and the normal **N**. In bulk Sm-C* samples, when passing from layer to layer, the polarization vector **P** rotates around the normal **N** by a certain angle that is the same for all layers. The number of layers over which the vector **P** makes a full rotation may vary from five or six to thousands [1-3]. As a result, the director **n** also uniformly rotates about the normal **N** when passing from one smectic layer to the next, forming a helical structure. In the bulk samples of Sm-C*, the vector **P** can experience several full rotations, and therefore, throughout the entire sample, the polarization is zero.

In free-standing Sm-C* films, an average a constant direction of the polarization vector **P** can be achieved by an external electric field, or by a small film thickness. Due to fluctuations of the director orientation $\mathbf{n}$, fluctuations of the polarization $\mathbf{P}$ may also arise. The occurrence of spatial inhomogeneities for spontaneous polarization leads to the appearance of polarization charges with density ρ =-div$\mathbf{P}$. The electrostatic interaction of the polarization charges changes the spatial correlation function of the director fluctuations and it can be manifested in light scattering experiments. It is usually assumed that the interaction of the polarization charges is completely or partially screened by impurity charges [1]. At the same time, the contribution of the unscreened Coulomb interaction was observed in the light scattering experiments which were performed on highly-pure Sm-C* samples [4-8].

The Coulomb interaction of polarization charges in Sm-C* is usually assumed to be isotropic [1]. As was shown in [9], screened Coulomb interaction in thin Sm-C* films may lead to renormalization of the bend elastic modulus. For thin freely suspended Sm-C* films in [4-8] the correlation function of orientation fluctuations was calculated and experiments were performed on highly-purified Sm-C* samples which revealed the Coulomb interaction contribution to the angular dependence of the light scattering. For a plane Sm-C* cell with bookshelf geometry, the calculations of the correlation function for orientation fluctuations were carried out in [10], taking into account the finite thickness of the cell. The angular dependence of light scattering intensity for Sm-C* was calculated for different spontaneous polarization values. Anisotropy of the Coulomb interaction in the bulk samples of Sm-C* was considered in the theoretical description performed in [11].

In the present work, the correlation function of orientation fluctuations is calculated for a freely suspended Sm-C* film, taking into account the finite film thickness and the Coulomb interaction of polarization charges. The results of the calculations are used to find the angular dependence of scattered light intensity.

2. Basic equations

Let's assume that in a freely suspended Sm-C* film the helical structure of director $\mathbf{n}$ is unwound. This can be achieved by an external electric field, $\mathbf{E}$, which is directed along the smectic layers. We assume that the field applied is not too large, so that it was possible to consider only the interaction of a field with the polarization vector $\mathbf{P}$. In this case, the free energy of distortion in the film Sm-C* can be represented as the sum of three terms:

$$F = F_{Fr} + F_P + F_C. \tag{2.1}$$

Here,F_{Fr} – the elastic free energy of distortion of the director field, which, in the unwound Sm-C*, can be represented in the following form:

$$F_{Fr} = \frac{1}{2}\int d\mathbf{r}\left[K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\cdot\operatorname{rot}\mathbf{n})^2 + K_{33}(\mathbf{n}\times\operatorname{rot}\mathbf{n})^2\right]. \tag{2.2}$$

Here, K_{11}, K_{22}, K_{33} are Frank modules. The second term in Eq. (2.1) arises from the interaction of the spontaneous polarization $\mathbf{P}$ with external electric field $\mathbf{E}$:

$$F_P = -\int d\mathbf{r}(\mathbf{P}\cdot\mathbf{E}). \tag{2.3}$$

The last term in Eq. (2.1) considers Coulomb interaction between the polarization charges arising in the inhomogeneous ferroelectric Sm-C*. Neglecting anisotropy of the dielectric constant, and taking it as equal to the average value of ε, for this contribution, we have:

$$F_C = \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\frac{\operatorname{div}\mathbf{P}(\mathbf{r})\ \operatorname{div}'\mathbf{P}(\mathbf{r}')}{\varepsilon\ |\mathbf{r}-\mathbf{r}'|}. \tag{2.4}$$

The layers are flat in a free standing Sm-C* film at equilibrium. In the description of the director **n**, fluctuation deformations of the layered structure are usually neglected. This is due to the fact that the possibility of free rotation of the director **n** around the normal **N** to the smectic layers, leads to much greater distortions of the director **n** field than small change the direction of the normal **N**. In a Sm-C* film in which the helical structure is unwound by an external electric field, the orientation of the director **n** is uniform.

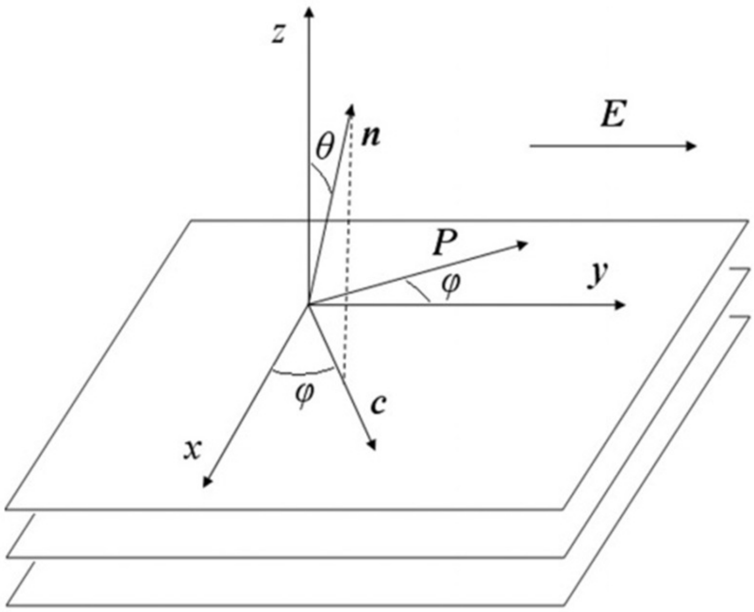

FIG. 1. The system coordinates and vectors **n**,**c**, **P**. The electric field is directed along the y axis

For the calculations, it is convenient to use a coordinate system in which the xy plane is parallel to the smectic layers and is located in the middle of the film. The direction of the y-axis is chosen along the external field, **E**, and the z-axis is normal to the layers. The orientation structure in Sm-C* conveniently described by **c**-director, which is a unit vector codirectional with the projection of the director **n** onto the plane of the smectic layer. Director **n** is defined by angles θ and φ as shown in Fig. 1. The angle θ determines the slope of director **n** with respect to the normal to the layers. This angle depends on the temperature, which we assumed to be constant. The angle φ describes the deviation of **c**-director from the equilibrium direction along the x-axis. The polarization vector **P** is given by:

$$\mathbf{P} = P\,[\mathbf{N} \times \mathbf{c}] \tag{2.5}$$

where P - the polarization of Sm-C*. The vectors **n**, **c**, **P** have the coordinates:

$$\begin{gathered} \mathbf{n} = (\sin\theta\cos\varphi,\ \sin\theta\sin\varphi,\ \cos\theta) \\ \mathbf{c} = (\cos\varphi,\ \sin\varphi,\ 0) \\ \mathbf{P} = P(-\sin\varphi,\ \cos\varphi,\ 0). \end{gathered} \tag{2.6}$$

Believing the **c**-director fluctuations and hence the angle φ fluctuations are small, we have:

$$\begin{gathered} \mathbf{n} \approx \left[\left(1 - \frac{\varphi^2}{2}\right)\sin\theta,\ \varphi\sin\theta,\ \cos\theta\right], \\ \mathbf{c} \approx \left(1 - \frac{\varphi^2}{2},\ \varphi,\ 0\right), \end{gathered} \tag{2.7}$$

$$\mathbf{P} \approx P(-\varphi,\ 1-\frac{\varphi^2}{2},\ 0).$$

The contribution of the **c**-director fluctuations to the free energy in the Gaussian approximation can be represented as follows:

$$\begin{aligned}\delta F = \tfrac{1}{2}\int d\mathbf{r}\Big[B_1\left(\tfrac{\partial\varphi}{\partial x}\right)^2 + B_2\left(\tfrac{\partial\varphi}{\partial y}\right)^2 + B_3\left(\tfrac{\partial\varphi}{\partial z}\right)^2 + 2B_{13}\left(\tfrac{\partial\varphi}{\partial x}\right)\left(\tfrac{\partial\varphi}{\partial z}\right) + \\ +PE\varphi^2 + \tfrac{P^2}{\varepsilon}\int d\mathbf{r}'\frac{\frac{\partial\varphi(\mathbf{r})}{\partial x}\frac{\partial\varphi(\mathbf{r}')}{\partial x'}}{|\mathbf{r}-\mathbf{r}'|}\Big].\end{aligned} \tag{2.8}$$

Here:

$$\begin{aligned} B_1 &= K_{22}\sin^2\theta\ \cos^2\theta + K_{33}\sin^4\theta, \\ B_2 &= K_{11}\sin^2\theta, \\ B_3 &= K_{22}\sin^4\theta\ + K_{33}\sin^2\theta\cos^2\theta, \\ B_{13} &= \sin^2\theta\ \cos\theta\,(K_{33}-K_{22}).\end{aligned} \tag{2.9}$$

We will consider a film of finite thickness, located in the area [-$L/2$; $L/2$], where L is the film thickness. In the xy plane, the film has a macroscopic size and therefore it is convenient to use the following Fourier representation:

$$\begin{aligned}\varphi_{\mathbf{q}_\perp}(z) &= \int d\mathbf{r}_\perp e^{-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\varphi(\mathbf{r}_\perp, z), \\ \varphi(\mathbf{r}_\perp, z) &= \tfrac{1}{(2\pi)^2}\int d\mathbf{q}_\perp e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\varphi_{\mathbf{q}_\perp}(z).\end{aligned} \tag{2.10}$$

In expression (2.8), for the free energy, we turn to the Fourier representation and take into account the ratio:

$$\int d\mathbf{r}_\perp\frac{e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}}{\sqrt{r_\perp^2+(z-z')^2}} = \frac{2\pi}{q_\perp}e^{-q_\perp|z-z'|}. \tag{2.11}$$

The result is as follows:

$$\begin{aligned}\delta F = \tfrac{1}{2(2\pi)^2}\int d\mathbf{q}_\perp\Big[\int_{-\frac{L}{2}}^{\frac{L}{2}}dz\,\varphi^*_{\mathbf{q}_\perp}(z)\left(B_1q_x^2+B_2q_y^2+PE-B_3\tfrac{\partial^2}{\partial z^2}-2B_{13}iq_x\tfrac{\partial}{\partial z}\right)\varphi_{\mathbf{q}_\perp}(z)+ \\ +\tfrac{2\pi P^2}{q_\perp\varepsilon}q_x^2\int_{-\frac{L}{2}}^{\frac{L}{2}}dz\int_{-\frac{L}{2}}^{\frac{L}{2}}dz'\,e^{-q_\perp|z-z'|}\varphi^*_{\mathbf{q}_\perp}(z')\varphi_{\mathbf{q}_\perp}(z)+ \\ +\left(B_3\varphi^*_{\mathbf{q}_\perp}(z)\tfrac{\partial\varphi_{\mathbf{q}_\perp}(z)}{\partial z}+2B_{13}iq_x\varphi^*_{\mathbf{q}_\perp}(z)\varphi_{\mathbf{q}_\perp}(z)\right)\Big|_{-\frac{L}{2}}^{\frac{L}{2}}\Big].\end{aligned} \tag{2.12}$$

The free energy (2.12) is a quadratic form, $\frac{1}{2}\left(\varphi, \hat{M}\varphi\right)$. We select by square brackets kernel of the operator explicitly:

$$\begin{aligned}\delta F = \tfrac{1}{2(2\pi)^2}\int d\mathbf{q}_\perp\int_{-\frac{L}{2}}^{\frac{L}{2}}dz\,\varphi^*_{\mathbf{q}_\perp}(z)\int_{-\frac{L}{2}}^{\frac{L}{2}}dz'\ \Big[\delta(z-z')\left(B_1q_x^2+B_2q_y^2+PE- \right.\\ \left. -B_3\tfrac{\partial^2}{\partial z'^2}-2B_{13}iq_x\tfrac{\partial}{\partial z'}\right)+\tfrac{2\pi P^2}{q_\perp\varepsilon}q_x^2\,e^{-q_\perp|z-z'|} \\ +\delta(z-z')\left(\delta\left(z'-\tfrac{L}{2}\right)-\delta\left(z'+\tfrac{L}{2}\right)\right)\left(B_3\tfrac{\partial}{\partial z'}+2B_{13}iq_x\right)\Big]\varphi_{\mathbf{q}_\perp}(z').\end{aligned} \tag{2.13}$$

The resulting expression for the free energy enables us to find the correlation function for the angle φ, ie, **c**-director fluctuations:

$$g_{\mathbf{q}_\perp}(z,\ z') = \int d\mathbf{r}_\perp\ e^{-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\left\langle\varphi(\mathbf{r}_\perp, z)\,\varphi(\mathbf{0}, z')\right\rangle. \tag{2.14}$$

Here $\langle ...\rangle$ represents the statistical averaging over all configurations of the angle φ field. Correlation function $g_{\mathbf{q}_\perp}(z,\ z')$ is the core of inverse operator $\hat{M}^{-1}$, multiplying by

$k_B T$, where T is the temperature and k_B is the Boltzmann constant. Hence, the correlation function $g_{\mathbf{q}_\perp}(z,\, z')$ must satisfy the equation:

$$\hat{M} g = k_B T\, \delta\left(z - z'\right), \tag{2.15}$$

or in explicit form:

$$\begin{array}{l} \left(B_1 q_x^2 + B_2 q_y^2 + PE - B_3 \frac{\partial^2}{\partial z^2} - 2B_{13} i q_x \frac{\partial}{\partial z}\right) g_{\mathbf{q}_\perp}(z,\, z') + \\ + \int_{-\frac{L}{2}}^{\frac{L}{2}} dz'' \, \frac{2\pi P^2}{q_\perp \varepsilon}\, q_x^2\, e^{-q_\perp |z - z''|}\, g_{\mathbf{q}_\perp}(z'',\, z') + \\ + \left(\delta\left(z - \frac{L}{2}\right) - \delta\left(z + \frac{L}{2}\right)\right)\left(B_3 \frac{\partial}{\partial z} + 2B_{13} i q_x\right) g_{\mathbf{q}_\perp}(z,\, z') = k_B T\, \delta(z - z') \quad . \end{array} \tag{2.16}$$

To solve the integral-differential equation, it is convenient to enterthe following function $v_{\mathbf{q}_\perp}(z,\, z')$, defined as:

$$v_{\mathbf{q}_\perp}(z,\, z') = q_\perp \int_{-\frac{L}{2}}^{\frac{L}{2}} dz''\, e^{-q_\perp |z - z''|}\, g_{\mathbf{q}_\perp}(z'',\, z'). \tag{2.17}$$

This allows us to solve the following system of differential equations instead of the integral-differential equation:

$$\begin{cases} \left(-\partial_z^2 - 2b_{13} i q_x \partial_z + b_1 q_x^2 + b_2 q_y^2 + \frac{PE}{B_3}\right) g_{\mathbf{q}_\perp}(z,\, z') + \\ + \frac{2\pi P^2}{B_3\, \mathbf{q}_\perp \varepsilon} q_x^2\, v_{\mathbf{q}_\perp}(z,\, z') = \frac{k_B T}{B_3} \delta(z - z') \\ \left(-\partial_z^2 + q_\perp^2\right) v_{\mathbf{q}_\perp}(z,\, z') - 2q_\perp^2 g_{\mathbf{q}_\perp}(z,\, z') = 0, \end{cases} \tag{2.18}$$

Here, we use the notations:

$$\begin{array}{lll} \partial_z = \frac{\partial}{\partial z}, & \partial_z^2 = \frac{\partial^2}{\partial z^2}, & \\ b_1 = \frac{B_1}{B_3}, & b_2 = \frac{B_2}{B_3}, & b_{13} = \frac{B_{13}}{B_3}. \end{array}$$

The boundary conditions for the system (2.18) have the form:

$$\begin{cases} \pm \partial_z v_{\mathbf{q}_\perp}\left(z = \pm\frac{L}{2},\, z'\right) + q_\perp v_{\mathbf{q}_\perp}\left(z = \pm\frac{L}{2},\, z'\right) = 0 \\ \partial_z g_{\mathbf{q}_\perp}\left(z = \pm\frac{L}{2},\, z'\right) + 2 i q_x b_{13} g_{\mathbf{q}_\perp}\left(z = \pm\frac{L}{2},\, z'\right) = 0. \end{cases} \tag{2.19}$$

The first of these conditions permits the removal of terms with δ-functions from the left-hand side of Eq. (2.16), and the second follows from the rule for differentiating of the function $v_{\mathbf{q}_\perp}(z,\, z')$ at the boundary of the film.

For the solution of the system (2.18) with boundary conditions (2.19), it is convenient to enter two four-dimensional vectors in the same way as it was done in [10]:

$$\begin{array}{l} \mathbf{W} = \left(g\,,\ v\,,\ \partial_z g\,,\ \partial_z v\right)^T, \\ \mathbf{D} = \frac{k_B T}{B_3}\left(0\,,\ 0\,,\ 1\,,\ 0\right)^T. \end{array} \tag{2.20}$$

Here, the upper index "T" denotes the transposition. In the given notations, the system (2.18) has the form:

$$(\partial_z - \hat{H})\mathbf{W} = -\mathbf{D}\delta(z - z'), \tag{2.21}$$

where:

$$\hat{H} = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ a & b & -f & 0 \\ -2q_\perp^2 & q_\perp^2 & 0 & 0 \end{pmatrix}. \tag{2.22}$$

We have introduced here the following notation:

$$a = b_1 q_x^2 + b_2 q_y^2 + \tfrac{PE}{B_3},$$
$$b = \tfrac{2\pi\, P^2 q_x^2}{B_3\, q_\perp^2\, \varepsilon},$$
$$f = 2b_{13} q_x\, i.$$

The boundary conditions for the Eq. (2.21), as it follows from (2.19), take the following form:

$$\hat{\Gamma}_\sigma \mathbf{W}_{\mathbf{q}_\perp}\left(z = \sigma\frac{L}{2},\ z'\right) = 0, \tag{2.23}$$

where $\sigma = \pm$ corresponds to $z = \pm L/2$, and the matrix $\hat{\Gamma}_\sigma$ has the form:

$$\hat{\Gamma}_\sigma = \begin{pmatrix} f & 0 & 1 & 0 \\ 0 & q_\perp & 0 & \sigma 1 \end{pmatrix}. \tag{2.24}$$

The boundary conditions (2.23) are satisfied by the vectors proportional to the following linearly independent vectors:

$$\mathbf{w}^{(1)} = (1\,,\ 0\,,\ -f\,,\ 0)^T\,,$$
$$\mathbf{w}^{(\sigma)} = (0\,,\ \sigma 1\,,\ 0\,,\ -q_\perp)^T \quad ,$$

Therefore, the solution of Eq. (2.21) satisfying the boundary condition (2.23) can be found in the form:

$$\mathbf{W}^\sigma_{\mathbf{q}_\perp}(z,\ z') = e^{\left(z-\sigma\frac{L}{2}\right)\hat{H}}\left(\mathbf{w}^{(1)} C^{(1)}_\sigma(z') + \mathbf{w}^{(\sigma)} C^{(2)}_\sigma(z')\right). \tag{2.25}$$

Here, $\mathbf{W}^+_{\mathbf{q}_\perp}(z,\ z')$ corresponds to $\mathbf{W}_{\mathbf{q}_\perp}(z,\ z')$ at $z > z'$ and $\mathbf{W}^-_{\mathbf{q}_\perp}(z,\ z')$ corresponds to $\mathbf{W}_{\mathbf{q}_\perp}(z,\ z')$ at $z < z'$. Unknown functions $C^1_\sigma(z')$ and $C^2_\sigma(z')$ can be found by substituting of Eq. (2.25) into Eq. (2.21) and integrating the resulting equation with respect to z over the interval $[z' - \varepsilon,\ z' + \varepsilon]$ where $\varepsilon \to +0$. As a result, we obtain the following algebraic vector equation for the functions $C^1_\sigma(z')$ and $C^2_\sigma(z')$:

$$\mathbf{W}^+_{\mathbf{q}_\perp}(z',\ z') - \mathbf{W}^-_{\mathbf{q}_\perp}(z',\ z') = -\mathbf{D}. \tag{2.26}$$

The solution of this equation can easily be found in the following form:

$$\left(C^{(1)}_+,\ C^{(2)}_+,\ C^{(1)}_-,\ C^{(2)}_-\right)^T = -\hat{S}^{-1} e^{\left(\frac{L}{2}-z'\right)\hat{H}}\mathbf{D}. \tag{2.27}$$

Here, $\hat{S}^{-1}$ is the matrix inverse to the matrix $\hat{S}$ consisting of the following columns:

$$\hat{S} = \left(\mathbf{w}^{(1)},\quad \mathbf{w}^{(+)},\quad -e^{-L\hat{H}}\mathbf{w}^{(1)},\quad -e^{-L\hat{H}}\mathbf{w}^{(-)}\right). \tag{2.28}$$

The functions $C^1_\sigma(z')$ and $C^2_\sigma(z')$, found in Eq. (2.27), can be substituted into Eq. (2.25). The desired the correlation function $g_{\mathbf{q}_\perp}(z,\ z')$ is given by the first component of the found four-dimensional vector$\mathbf{W}_{\mathbf{q}_\perp}(z,\ z')$.

3. Light scattering by c-director fluctuations

Light scattering in Sm-C* occurs with fluctuations of the permittivity, which are connected with the fluctuations of **c**-director. In the lowest approximation with respect to the angle φ, we have:

$$\delta\tilde{\varepsilon}_{\alpha\beta} = \tilde{\varepsilon}_a\left(\frac{\partial n_\alpha}{\partial\varphi} n_\beta + n_\alpha \frac{\partial n_\beta}{\partial\varphi}\right)\varphi. \tag{3.1}$$

Here, $\tilde{\varepsilon}_a$ is the anisotropy of the permittivity at optical frequencies. In the Born approximation, the intensity of the scattered light can be represented as [12-14]:

$$I = \frac{VI_0 k_0^4}{(4\pi R)^2} e_\alpha^{(s)} e_\beta^{(s)} W_{\alpha\nu\beta\mu}(\mathbf{q}_{sc}) e_\nu^{(i)} e_\mu^{(i)}, \tag{3.2}$$

where I is the intensity of the scattered light, I_0 is the intensity of the incident beam, V is the scattering volume, k_0 is the wave number of the incident and scattered beams, R is the distance from the film to the observation point, $\mathbf{e}^{(i)}$ and $\mathbf{e}^{(s)}$ are the polarization vectors in the incident and scattered rays, $\mathbf{q}_{sc} = \mathbf{k}_s - \mathbf{k}_i$ is the scattering vector, $\mathbf{k}_i$ and $\mathbf{k}_s$ are the wave vectors of the incident and scattered rays. The function $W_{\alpha\nu\beta\mu}(\mathbf{q}_{sc})$ is the Fourier transform of the correlation function for permittivity fluctuations at optical frequencies, namely:

$$W_{\alpha\nu\beta\mu}(\mathbf{q}) = \frac{1}{L} \int_{-\frac{L}{2}}^{\frac{L}{2}} dz \int_{-\frac{L}{2}}^{\frac{L}{2}} dz' \, e^{-iq_z(z-z')} \int d\mathbf{r}_\perp e^{-i\mathbf{q}_\perp \cdot \mathbf{r}_\perp} \left\langle \delta\tilde{\varepsilon}_{\alpha\nu}(\mathbf{r}_\perp, z) \delta\tilde{\varepsilon}_{\beta\mu}(\mathbf{0}, z') \right\rangle . \tag{3.3}$$

Here, for the wave vector used, the notation $\mathbf{q} = (\mathbf{q}_\perp, q_z)$. After substituting Eqs. (3.1) and (3.3) into Eq. (3.2), and taking into account the definition (2.14), we obtain the following expression for the intensity of light scattering:

$$I \sim \tilde{\varepsilon}_a^2 \left[e_\alpha^{(s)} \left(\frac{\partial n_\alpha}{\partial \varphi} n_\beta + n_\alpha \frac{\partial n_\beta}{\partial \varphi} \right) e_\beta^{(i)} \right]^2 G_{\mathbf{q}_{sc}}, \tag{3.4}$$

Where:

$$G_{\mathbf{q}} = \frac{1}{L} \int_{-\frac{L}{2}}^{\frac{L}{2}} dz \int_{-\frac{L}{2}}^{\frac{L}{2}} dz' \, e^{-iq_z(z-z')} g_{\mathbf{q}_\perp}(z, z'). \tag{3.5}$$

The numerical calculations were performed for the beam incident on the film surface at the angle θ_i relative to the z axis in the xz plane. Fig. 2 shows the geometry of the optical experiment for the case of incidence of the beam normal to the Sm-C* film.

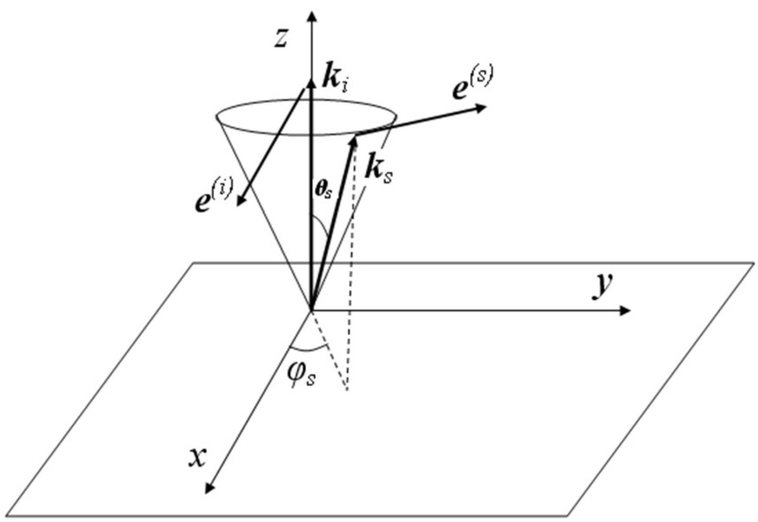

FIG. 2. Direction of the wave vectors of the incident and scattered rays in an optical experiment, $\mathbf{k}_i$ and $\mathbf{k}_s$, respectively. Also shown are the direction of the polarization vectors in the incident and scattered rays: $\mathbf{e}^{(i)}$ and $\mathbf{e}^{(s)}$

The wave vectors of the incident and scattered rays and the polarization vectors in this geometry are given by:

$$\begin{aligned} \mathbf{k}_i &= k_0(\sin\theta_i,\ 0, \cos\theta_i)\ , \\ \mathbf{k}_s &= k_0(\sin\theta_s\cos\varphi_s,\ \sin\theta_s\sin\varphi_s,\ \cos\theta_s)\ , \\ \mathbf{e}^{(i)} &= (1,\ 0,\ 0)\ , \\ \mathbf{e}^{(s)} &= (-\sin\varphi_s,\ \cos\varphi_s,\ 0). \end{aligned} \tag{3.6}$$

Here, θ_i and θ_s polar angles of the incident and scattered rays, φ_s is the azimuth angle of the scattered beam. In this case, we obtain from Eq. (3.4) the following angular dependence of the scattered light intensity:

$$I\sim\tilde{\varepsilon}_a^2\sin^4\theta\ \cos^2\varphi_s\ G_{\mathbf{q}_{sc}}. \tag{3.7}$$

It should be noted that the angular dependence of the intensity is determined entirely by the last two factors in Eq. (3.7), while the first two factors are material constants.

The numerical calculations were performed via Eq. (3.7) for the free standing Sm-C* film with the following parameters: $L = 10^{-4}$ cm, $\theta = 15°$, $\tilde{\varepsilon}_a = 5$, $k_0 = 10^5$ cm^{-1}, E=0.3 statvolt/cm = 8994 V/m, $K_{11}=0.7\cdot10^{-6}$ dyn=$0.7\cdot10^{-11}$ N, $K_{22}=0.4\cdot10^{-6}$ dyn=$0.4\cdot10^{-11}$ N, $K_{33} = 1.7\cdot10^{-6}$ dyn=$1.7\cdot10^{-11}$ N. For spontaneous polarization, different values were used, namely, P = 10, 15, 20 statcoulomb/cm^2 = $3.34\cdot10^4$, $5.01\cdot10^4$, $6.67\cdot10^4$ nC/m^2.

In Figs. 3 and 4 are shown the angular dependences for the scattered light intensity in the case where the xz plane is the scattering plane. Since in this case $\varphi_s = 0$, it follows from the Eq. (3.7) that precisely the same angular dependence has the correlation function of the **c**-director fluctuations $G_{\mathbf{q}_{sc}}$.

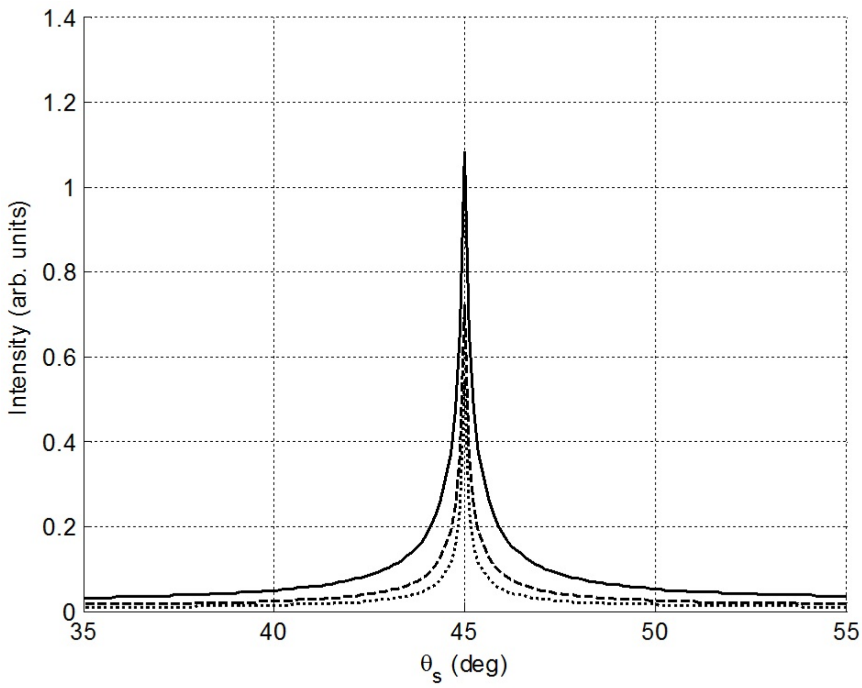

FIG. 3. The angular dependence of the scattered light intensity is shown for different spontaneous polarization values. For solid, dashed and dotted lines, the spontaneous polarization values are respectively: 10, 15, 20 statcoulomb/cm^2. The beam of light falls on the film at an angle of 45° to the normal. The scattering plane coincides with the xz plane

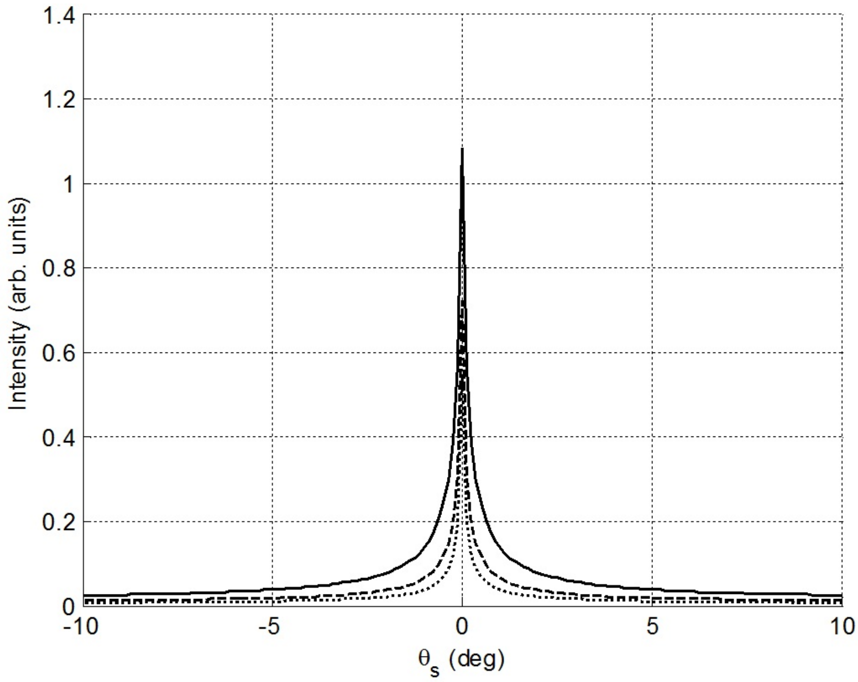

FIG. 4. The same as in Fig. 3, but in this instance, the light beam's angle of incidence is perpendicular to the film's surface. Positive values of θ_s correspond to $\varphi_s = 0°$, and negative to $\varphi_s = 180°$

As can be seen from Figs. 3, 4, the correlation function of **c**-director fluctuations $G_{\mathbf{q}}$ and the scattered light intensity are significantly dependent on the spontaneous polarization P. These dependences are mainly determined by the electrostatic interaction of the polarization charges. With increased spontaneous polarization P, the correlation function $G_{\mathbf{q}_{sc}}$ and the scattered light intensity are reduced. Hence, the **c**-director fluctuations are suppressed with increasing P; i.e., the system becomes more rigid in orientation, as was noted in [4-9, 15].

In Figs. 5 and 6 are shown respectively the dependences of the scattered light intensity and the correlation function $G_{\mathbf{q}_{sc}}$ on the azimuthal angle φ_s at normal incidence of light on the film. The incident beam is polarized in the direction of the x axis, perpendicular to the external electric field. The scattered beam propagates at an angle of 10° relative to the normal to the smectic layers and is polarized in the xy plane. Fig. 5 shows that the correlation function $G_{\mathbf{q}_{sc}}$ significantly depends on the value of the spontaneous polarization P and the azimuthal angle φ_s. The function $G_{\mathbf{q}_{sc}}$ reaches a maximum when $\mathbf{q}_{sc}$ is within the xy plane, and minimal, if $\mathbf{q}_{sc} \in xz$. Fig. 6 shows that in accordance with Eq. (3.7), the effects associated with the geometry of the experiment are added to the above relationships in the intensity of the scattered light. If the direction of the wave vector and the polarization vector in the incident and scattered rays are given by Eq. (3.6), the maximum intensity should be observed in the scattering plane xz, and the minimum occurs in the xy plane.

4. Conclusion

A number of works have suggested that the electrostatic interaction between the polarization charges may be very important in the study of Sm-C* [4-8, 16-19]. Systems with a large spontaneous polarization have been shown to increase the effective rigidity of the orientation and increase the effective orientation viscosity [4-9, 15]. We have studied

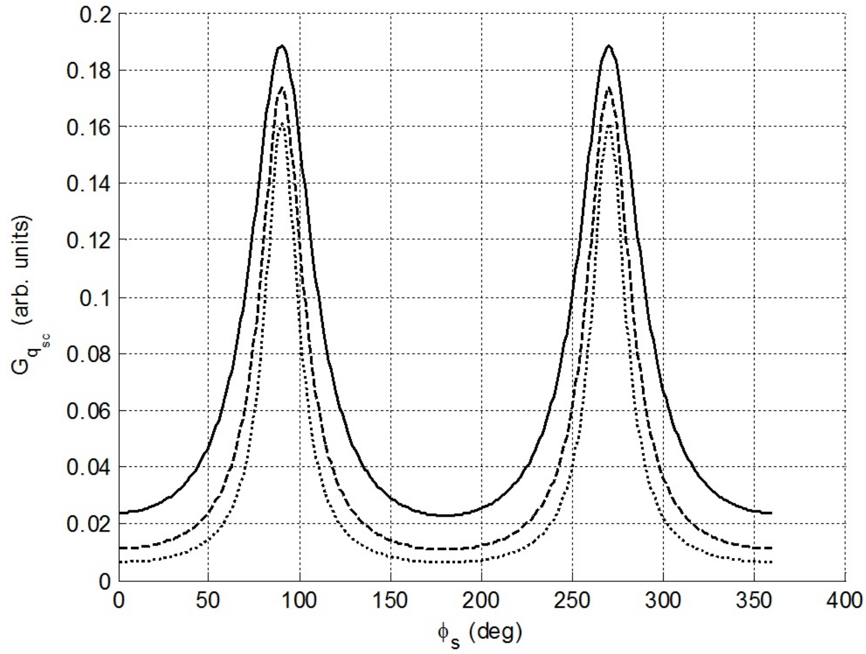

FIG. 5. The dependence of the correlation function $G_{\mathbf{q}_{sc}}$ on the azimuthal angle φ_s is shown for the same spontaneous polarization values as in Fig. 3. A beam of light falls perpendicular to the surface of the film. The value $\theta_s = 10°$ remains constant

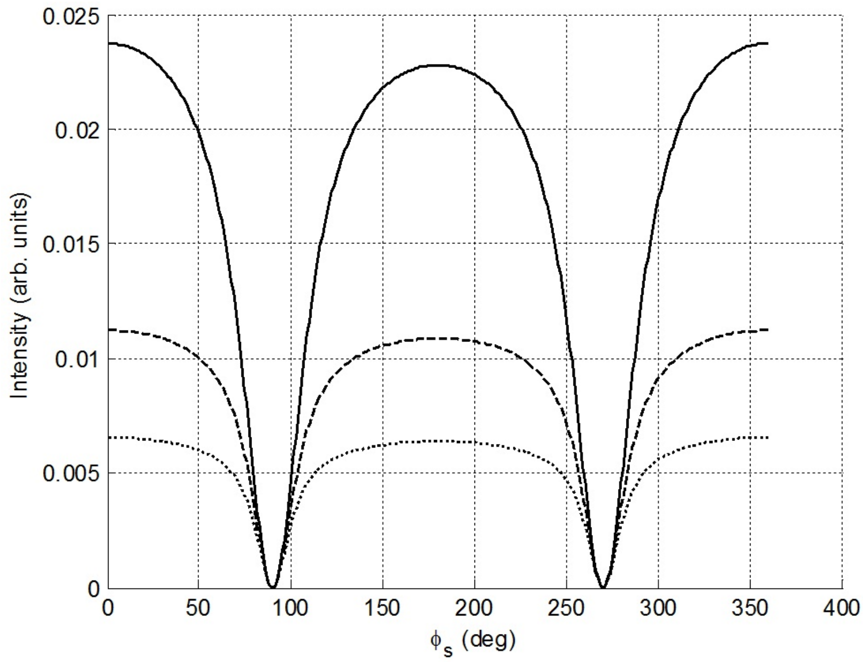

FIG. 6. The dependence of the scattered light intensity on the azimuthal angle φ_s is shown for the same conditions used in Fig. 5

the role of the Coulomb interaction between the polarization charges in the formation of the correlation function of director fluctuations in freely standing Sm-C* films of finite thickness. The film considered was in an external electric field directed along the layers. At equilibrium, the **c**-director helicoid was unwound. The obtained correlation function permitted the calculation of the angular dependence for the scattered light intensity. As a result of the calculations, the correlation function for the orientation fluctuations and the scattered light intensity were found to significantly depend on the magnitude of the spontaneous polarization. These relationships are caused by the Coulomb interaction between the polarization charges occurring due to orientation fluctuations. Increasing the magnitude of polarization was shown to significantly reduce the correlation function of the orientation fluctuations as well as the scattered light intensity, indicating that the suppression of the director fluctuations occurs as a result of the electrostatic interaction between the polarization charges. This can be regarded as a definite increase in the orientation rigidity of the system.

Acknowledgments

The authors acknowledge Saint Petersburg State University for research Grants No. 11.37.145.2014 and No. 11.37.161.2014, and the Russian Foundation for Basic Research for Grant No. 13-02-01255-a.

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