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Anisotropy of electrostatic interaction in smectic-C^* liquid crystals

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The contribution to the free energy of distortion of the ferroelectric smectic-C^* due to the electrostatic interaction of polarization charges is calculated. These calculations are performed by accounting for the anisotropy of the permittivity, which is essential for smectic-C^*. Fluctuations of the **c** director in an external electric field are considered. It is shown that the anisotropy of the permittivity strongly affects the interaction of the polarization charges, the spectrum orientation fluctuations, and the angular dependence of the light scattering intensity.

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I. INTRODUCTION

For many years smectic-C^* (Sm-C^*) has attracted considerable attention due to its unusual physical properties and its various practical applications [1]. These unique properties are caused by the fact that Sm-C^* is formed by chiral molecules that lead to the formation of the spontaneous polarization **P** in the plane of the smectic layers. In each smectic layer the polarization vector **P** is constant, but it can rotate from layer to layer, preserving its absolute value. It leads to the existence of various types of Sm-C^* such as ferroelectrics, antiferroelectrics, and ferrielectrics [2]. When the temperature changes, phase transitions may occur between different types of Sm-C^*, for example, between ferroelectric and antiferroelectric [3,4]. The period of the orientational structure in Sm-C^* can vary from five or six to thousands of smectic layers [5–8]. The uniform orientational structure can be organized by the bounding surfaces or by the external electric field.

The presence of spontaneous polarization **P** allows one to operate the orientational structure of Sm-C^*. Weak external fields can rotate the vector director **n** around the normal to the smectic layers. In smectic-A^* the strong electric field can cause deviations of the optical axis from the normal [9], and in Sm-C^* it can cause the transition from the helical to the planar smectic structure [10,11].

The presence of bounding surfaces and thermal fluctuations in systems with spontaneous polarization leads to the emergence of polarization charges with density $-\operatorname{div}\mathbf{P}$. The effect of the electrostatic interaction of these charges is essential both for the equilibrium structure of Sm-C^* and for orientation fluctuations in the external fields. Most often the long-range electrostatic interactions are considered to be totally [1] or partially [12] screened by impurity charges. The experimental data obtained for the texture of islands [12] and for periodic stripe patterns [13] in free-standing Sm-C^* films are consistent with the theoretical conclusions proposed in Ref. [12], that the screened Coulomb interaction in two-dimensional systems leads to renormalization of the bend elastic modulus.

The interaction between islands on freely suspended Sm-C^* films was experimentally studied [14] by using optical tweezers. The results were compared with the numerical calculations [15] where only the elastic energy was taken into account. It was pointed out that it is necessary to consider the spontaneous polarization in order to account, in detail, for the interactions of chiral islands. The effect of spontaneous polarization on the structural defects and orientation dynamics was investigated in Ref. [16]. In this work it was shown that spontaneous polarization results in an increase of the orientation rigidity and orientation viscosity. Also, the relaxation dynamics in Sm-C^* was studied in Ref. [17], where the multistage nature of the orientation relaxation process was found.

Sm-C^* can be uniaxial or biaxial and the dielectric anisotropy can be positive or negative [10,18]. The properties of Sm-C^* are dependent on the anisotropy of the permittivity since it is not small. In particular, the description of the unusual electro-optic response in systems with essential molecular biaxiality and a large slope of the molecular axes, up to 45°, become possible only when the biaxial permittivity tensor is taken into account [19].

The contribution of unscreened electrostatic interactions of the polarization charges to the intensity of the scattered light was observed in experiments with well cleaned samples [20–24]. At small wave numbers, i.e., for long-wavelength fluctuations, the role of the electrostatic interaction due to spontaneous polarization increases. As a rule, when calculating the spectrum of the fluctuation orientation, the interactions of polarization charges are considered by introducing an averaged permittivity [1], i.e., without accounting for the dielectric anisotropy of the medium. However, the anisotropy of the permittivity is essential for a quantitative comparison between theory and experiment [19]. It is well established from the description of the light scattering in liquid crystals, studies of the Freedericksz effect, investigation of the flexoelectric effect, etc.

In this paper the free energy of the distortion of Sm-C^* in the external electric field is obtained by taking into account the Coulomb interactions of polarization charges in an anisotropic medium. The spectra of the **c**-director thermal fluctuations and of the angular dependence of the scattered light intensity are calculated. It is shown that the anisotropy of the permittivity has a significant influence on the obtained results.

II. THE DISTORTION ENERGY OF Sm-C^* IN EXTERNAL ELECTRIC FIELD

Let us consider ferroelectric Sm-C^* in the case when the helix of the director rotation is unwound. We assume that the

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electric field is directed parallel to the smectic layers and that the field is not too strong, so that in the free energy we can take into account the interaction of the field with the spontaneous polarization **P** only. The distortion free energy can be presented as the sum of three contributions,

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

The first contribution, F_{Fr}, describes the energy of the elastic distortion of the director field **n**, and in the unwound Sm-C^* it has the form

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

Here K_{11}, K_{22}, and K_{33} are Frank modules.

The second term, F_P, describes the interaction of the external electric field **E** with the spontaneous polarization **P**,

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

The third contribution, F_C, describes the Coulomb interaction of polarization charges, which in inhomogeneous ferroelectric Sm-C^* has a density

$$\rho = -\operatorname{div}\mathbf{P}. \tag{2.4}$$

It is possible to get an explicit form for this contribution by using the expression for the field of the pointlike charge e in a homogeneous anisotropic medium. The induction **D** obeys the equation

$$\operatorname{div}\mathbf{D} = 4\pi e\delta(\mathbf{r}), \tag{2.5}$$

where

$$D_i = \varepsilon_{ik}E_k = -\varepsilon_{ik}\frac{\partial\phi}{\partial x_k}, \tag{2.6}$$

and ε_{ik} is the tensor of permittivity. For the potential ϕ we have the equation

$$\varepsilon_{ik}\frac{\partial^2\phi}{\partial x_i\partial x_k} = -4\pi e\delta(\mathbf{r}). \tag{2.7}$$

This equation is reduced to the Poisson equation for the pointlike charge $e/\sqrt{\det\hat{\varepsilon}}$ by the linear transformation of the coordinates. Here $\det\hat{\varepsilon}$ is the determinant of the permittivity tensor. The solution of Eq. (2.7) for the potential has the form [25]

$$\phi = \frac{e}{\sqrt{\det\hat{\varepsilon}\,\varepsilon^{-1}_{ik}x_i x_k}}, \tag{2.8}$$

where $\hat{\varepsilon}^{-1}$ is the inverse of the permittivity tensor. Thus the contribution of the Coulomb interaction of polarization charges to the free energy can be presented in the form

$$F_C = \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\frac{\operatorname{div}\mathbf{P}(\mathbf{r})\operatorname{div}'\mathbf{P}(\mathbf{r}')}{\sqrt{\det\hat{\varepsilon}\,\varepsilon^{-1}_{ik}(\mathbf{r}-\mathbf{r}')_i(\mathbf{r}-\mathbf{r}')_k}}. \tag{2.9}$$

When writing F we mean the thermodynamic potential which should be minimized in a fixed external field [1].

In equilibrium the polarization vector **P** is directed along the external electric field **E** and the vector director **n** is in the plane perpendicular to the field. Let us introduce the coordinate frame with the z axis perpendicular to the smectic layers which are supposed to be plane and parallel. The external electric field is directed along the y axis and the projection of the vector director **n** on the plane xy is directed along the x axis in the equilibrium. The temperature of the Sm-C^* liquid crystal is supposed to be constant. Therefore the angle θ between the director **n** and the normal to the smectic layers $\mathbf{N} = (0,0,1)$ is kept constant.

By introducing the notations $\varepsilon_1,\varepsilon_2,\varepsilon_3$ for the permittivities along the principal axes it is possible to parametrize the permittivity tensor in the form

$$\varepsilon_{ik} = \varepsilon_1\delta_{ik} + (\varepsilon_3-\varepsilon_1)n_{0i}n_{0k} + (\varepsilon_2-\varepsilon_1)p_{0i}p_{0k}. \tag{2.10}$$

Here we used the notations

$$\mathbf{n}_0 = (\sin\theta, 0, \cos\theta), \quad \mathbf{c}_0 = (1,0,0), \quad \mathbf{p}_0 = [\mathbf{N}\times\mathbf{c}_0] = (0,1,0), \tag{2.11}$$

where $\mathbf{n}_0$ is the equilibrium vector director, $\mathbf{c}_0$ is the equilibrium **c** director, and $\mathbf{p}_0$ is the unit vector directed in the equilibrium along the vector of the spontaneous polarization **P**.

In general, the free energy of deformation in smectics contains the contributions due to the displacement of smectic layers from the equilibrium position:

$$F_{\rm Sm} = \frac{1}{2}\int d\mathbf{r}\left\{B\left(\frac{\partial u}{\partial z}\right)^2 + K(\Delta_\perp u)^2\right\}. \tag{2.12}$$

Here u is the displacement of the smectic layers along the z axis, and B and K are the layer compression and layer bend elastic constant, respectively. In free-standing Sm-C^* films the free energy of deformation also contains the surface term

$$F_{Sf} = \frac{\gamma}{2}\int d\mathbf{r}_\perp\{(\nabla_\perp u_1)^2 + (\nabla_\perp u_N)^2\}. \tag{2.13}$$

Here γ is the surface tension, and the displacements of the two free surfaces of the film are denoted by u_1 and u_N. In what follows we will be interested in the distortions of the **c**-director field, and in this case we will neglect deviations of the normal to the smectic layers from the z axis, as it is usually done [12,16,20–24]. So, the fluctuations of the director orientation are considered in the film consisting of flat layers. Due to the thermal fluctuations, the director can rotate around the z axis and then transit from one layer to another while maintaining the angle θ. In this approximation, the free energy of orientation deformation can be written as (2.1).

III. C-DIRECTOR FLUCTUATIONS

Random deviations of the **c** director from the x axis can emerge due to thermal fluctuations. In this case,

$$\mathbf{n} = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta), \quad \mathbf{c} = (\cos\varphi, \sin\varphi, 0), \quad \mathbf{p} = (-\sin\varphi, \cos\varphi, 0) = [\mathbf{N}\times\mathbf{c}], \quad \mathbf{P} = P\mathbf{p}, \tag{3.1}$$

where P is the spontaneous polarization, and φ is the angle between the **c** director and the x axis. The directions of vectors **P**, **n**, **c** are shown in Fig. 1. Supposing that the fluctuations are

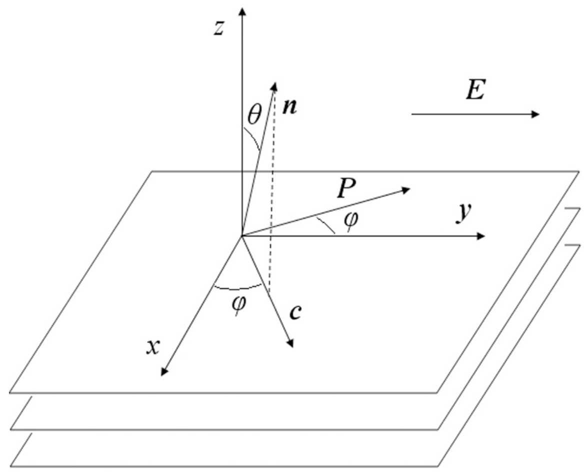

FIG. 1. Positions of the vectors **n**, **c**, and **P** in smectic-C^*. The external electric field **E** is directed along the y axis.

small, we have

$$\mathbf{n} \approx \left[\left(1-\frac{\varphi^2}{2}\right)\sin\theta, \varphi\sin\theta, \cos\theta\right], \quad \mathbf{c} \approx \left(1-\frac{\varphi^2}{2}, \varphi, 0\right), \tag{3.2}$$
$$\mathbf{P} \approx P\left(-\varphi, 1-\frac{\varphi^2}{2}, 0\right).$$

The contribution of the **c**-director fluctuations to the distortion of the free energy has the following form in a Gaussian approximation:

$$\delta F = \frac{1}{2}\int d\mathbf{r}\Bigg[K_{11}\sin^2\theta\left(\frac{\partial\varphi}{\partial y}\right)^2 + K_{22}\sin^2\theta\left(\cos\theta\frac{\partial\varphi}{\partial x} - \sin\theta\frac{\partial\varphi}{\partial z}\right)^2 + K_{33}\sin^2\theta\left(\sin\theta_0\frac{\partial\varphi}{\partial x} + \cos\theta\frac{\partial\varphi}{\partial z}\right)^2 + PE\varphi^2 + P^2\int d\mathbf{r}'\frac{\frac{\partial\varphi(\mathbf{r}')}{\partial x'}\frac{\partial\varphi(\mathbf{r})}{\partial x}}{\sqrt{\det\hat{\varepsilon}(\hat{\varepsilon}^{-1})_{ik}(\mathbf{r}-\mathbf{r}')_i(\mathbf{r}-\mathbf{r}')_k}}\Bigg]. \tag{3.3}$$

Determining the Fourier transformation as

$$\varphi_{\mathbf{q}} = \int d\mathbf{r}\,\varphi(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}}, \quad \varphi(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{q}}\varphi_{\mathbf{q}}e^{i\mathbf{q}\cdot\mathbf{r}}, \tag{3.4}$$

where V is the system volume, we get

$$\delta F = \frac{1}{2V}\sum_{\mathbf{q}}\left[K_{11}q_y^2\sin^2\theta + K_{22}\sin^2\theta(q_x\cos\theta - q_z\sin\theta)^2 + K_{33}\sin^2\theta(q_x\cos\theta + q_z\sin\theta)^2 + PE + P^2q_x^2G_{-\mathbf{q}}\right]\times|\varphi_{\mathbf{q}}|^2, \tag{3.5}$$

where

$$G_{-\mathbf{q}} = \int d\mathbf{r}e^{i\mathbf{q}\cdot\mathbf{r}}\frac{1}{\sqrt{\det\hat{\varepsilon}(\hat{\varepsilon}^{-1})_{ik}r_ir_k}}. \tag{3.6}$$

The radicand in (3.6) has the form

$$\det\hat{\varepsilon}\,(\hat{\varepsilon}^{-1})_{ik}r_ir_k = \frac{\varepsilon_1\varepsilon_2\varepsilon_3\left[(\alpha r_x - \beta r_z)^2 + \gamma^2r_y^2 + r_z^2\right]}{\varepsilon(\theta)}, \tag{3.7}$$

where

$$\varepsilon(\theta) = \varepsilon_1\sin^2\theta + \varepsilon_3\cos^2\theta, \quad \alpha = \frac{\varepsilon(\theta)}{\sqrt{\varepsilon_1\varepsilon_3}}, \quad \beta = \frac{(\varepsilon_3-\varepsilon_1)\sin\theta\cos\theta}{\sqrt{\varepsilon_1\varepsilon_3}}, \quad \gamma = \sqrt{\frac{\varepsilon(\theta)}{\varepsilon_2}}. \tag{3.8}$$

To provide integration in Eq. (3.6) it is convenient to introduce the new variables R_x, R_y, R_z:

$$r_x = \frac{1}{\alpha}R_x + \frac{\beta}{\alpha}R_z, \quad r_y = \frac{1}{\gamma}R_y, \quad r_z = R_z. \tag{3.9}$$

For the function $G_{-\mathbf{q}}$ we get

$$G_{-\mathbf{q}} = \frac{1}{\varepsilon(\theta)}\int d\mathbf{R}\frac{e^{i\mathbf{Q}\cdot\mathbf{R}}}{R} = \frac{1}{\varepsilon(\theta)}\frac{4\pi}{Q^2}, \tag{3.10}$$

where

$$\mathbf{Q} = \left(\frac{q_x}{\alpha}, \frac{q_y}{\gamma}, q_z + \frac{\beta}{\alpha}q_x\right). \tag{3.11}$$

Thus we have

$$G_{-\mathbf{q}} = \frac{4\pi\varepsilon(\theta)}{\varepsilon_1\varepsilon_3q_x^2 + \varepsilon_2\varepsilon(\theta)q_y^2 + [q_x(\varepsilon_3-\varepsilon_1)\sin\theta\cos\theta + q_z\varepsilon(\theta)]^2}. \tag{3.12}$$

Substituting Eq. (3.12) into Eq. (3.5) we obtain the following expression for the distortion free energy of the Sm-C^*:

$$\delta F = \frac{1}{2V}\sum_{\mathbf{q}}M(\mathbf{q})|\varphi_{\mathbf{q}}|^2. \tag{3.13}$$

Here

$$M(\mathbf{q}) = \Bigg\{K_{11}q_y^2\sin^2\theta + K_{22}\sin^2\theta(q_x\cos\theta - q_z\sin\theta)^2 + K_{33}\sin^2\theta(q_x\cos\theta + q_z\sin\theta)^2 + PE + \frac{4\pi P^2q_x^2\varepsilon(\theta)}{\varepsilon_1\varepsilon_3q_x^2 + \varepsilon_2\varepsilon(\theta)q_y^2 + [q_x(\varepsilon_3-\varepsilon_1)\sin\theta\cos\theta + q_z\varepsilon(\theta)]^2}\Bigg\}. \tag{3.14}$$

Note that the last term in the curly brackets is positive for any relation between the principal values of the permittivity tensor.

For the **c**-director fluctuations we have

$$\langle|\varphi_{\mathbf{q}}|^2\rangle = \frac{kTV}{M(\mathbf{q})}, \tag{3.15}$$

where T is the temperature and k is the Boltzmann constant.

IV. LIGHT SCATTERING BY C-DIRECTOR FLUCTUATIONS

It has been shown that accounting for the anisotropy of the permittivity in the Coulomb interaction of polarization charges alters the correlation function of the **c**-director fluctuations. It is of interest to find out how it affects the angular dependence of the light scattering intensity caused by the **c**-director fluctuations. The intensity of the scattered light can be expressed as [26–28]

$$I = \frac{VI_0k_0^4}{(4\pi R)^2}e_\alpha^{(s)}e_\beta^{(s)}W_{\alpha\nu\beta\mu}(\mathbf{q}_{\rm sc})e_\nu^{(i)}e_\mu^{(i)}, \tag{4.1}$$

where I is the intensity of the scattered light, I_0 is the intensity of the incident radiation, k_0 is the wave number of the incident and scattered light, R is the distance between the scattered volume and the observation point, $\mathbf{e}^{(\mathbf{i})}$ and $\mathbf{e}^{(\mathbf{s})}$ are the polarization vectors of the incident and the scattered light, respectively, $\mathbf{q}_{\rm sc} = \mathbf{k}_s - \mathbf{k}_i$ is the scattering vector, and $\mathbf{k}_i$ and $\mathbf{k}_s$ are the wave vectors of the incident and the scattered light, respectively. The description of the light scattering is provided in the Born approximation for the isotropic medium. The function $W_{\alpha\nu\beta\mu}(\mathbf{q}_{\rm sc})$ in Eq. (4.1) is the Fourier image of the correlation function of the permittivity fluctuations $\tilde{\varepsilon}$ at the optical frequency. The notation $\tilde{\varepsilon}$ is introduced in order to distinguish it from the static permittivity which was included in the previous sections. In the coordinate representation $W_{\alpha\nu\beta\mu}$ has the form

$$W_{\alpha\nu\beta\mu}(\mathbf{r}_1,\mathbf{r}_2) = \langle\delta\tilde{\varepsilon}_{\alpha\nu}(\mathbf{r}_1)\delta\tilde{\varepsilon}_{\beta\mu}(\mathbf{r}_2)\rangle, \tag{4.2}$$

and in the homogeneous medium it depends on the difference $\mathbf{r}_1 - \mathbf{r}_2$. The brackets $\langle\cdots\rangle$ correspond to the statistical averaging. The fluctuation of the permittivity tensor $\tilde{\varepsilon}_{\alpha\beta} = \tilde{\varepsilon}_1\delta_{\alpha\beta} + (\tilde{\varepsilon}_3 - \tilde{\varepsilon}_1)n_\alpha n_\beta + (\tilde{\varepsilon}_2 - \tilde{\varepsilon}_1)p_\alpha p_\beta$ in the case of small deviation angles φ has the form

$$\delta\tilde{\varepsilon}_{\alpha\beta} = \left[\Delta\tilde{\varepsilon}\left(\frac{\partial n_\alpha}{\partial\varphi}n_\beta + n_\alpha\frac{\partial n_\beta}{\partial\varphi}\right) + \delta\tilde{\varepsilon}\left(\frac{\partial p_\alpha}{\partial\varphi}p_\beta + p_\alpha\frac{\partial p_\beta}{\partial\varphi}\right)\right]\varphi, \tag{4.3}$$

where $\Delta\tilde{\varepsilon} = \tilde{\varepsilon}_3 - \tilde{\varepsilon}_1$ and $\delta\tilde{\varepsilon} = \tilde{\varepsilon}_2 - \tilde{\varepsilon}_1$. Variables in the expression in square brackets are calculated for $\varphi = 0$. In explicit form the fluctuation contribution to the permittivity tensor is presented as

$$\delta\tilde{\varepsilon} = \begin{pmatrix} 0 & \Delta\tilde{\varepsilon}\sin^2\theta - \delta\tilde{\varepsilon} & 0 \\ \Delta\tilde{\varepsilon}\sin^2\theta - \delta\tilde{\varepsilon} & 0 & \Delta\tilde{\varepsilon}\sin\theta\cos\theta \\ 0 & \Delta\tilde{\varepsilon}\sin\theta\cos\theta & 0 \end{pmatrix}\varphi. \tag{4.4}$$

Let us consider the intensity of scattered light in the geometry shown in Fig. 2 when the incident light is directed along the normal to the smectic layers, i.e., $\mathbf{k}_i = k_0(0,0,1)$.

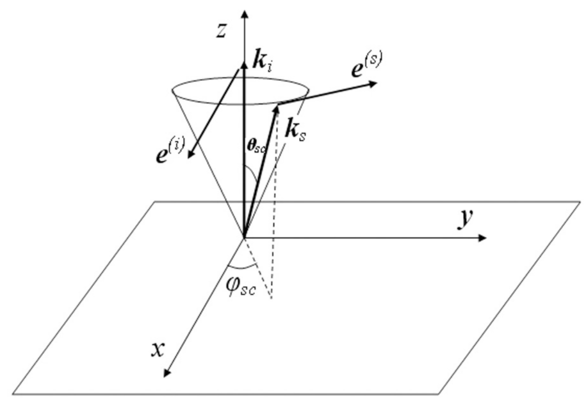

FIG. 2. Positions of the vectors **n**, **c**, and **P** in smectic-C^*. External electric field **E** is directed along the y axis.

For definiteness we assume that the incident light polarization vector is directed perpendicular both to the normal of the smectic layers and to the external electric field, i.e., along the x axis, $\mathbf{e}^{(i)} = (1,0,0)$. The scattered radiation is determined by the wave vector $\mathbf{k}_s = k_0(\sin\theta_{\rm sc}\cos\varphi_{\rm sc}, \sin\theta_{\rm sc}\sin\varphi_{\rm sc}, \cos\theta_{\rm sc})$, where $\theta_{\rm sc}$ and $\varphi_{\rm sc}$ are the polar and the azimuthal angles, respectively. We consider the case when the polarization vector $\mathbf{e}^{(s)}$ is parallel to the smectic layers, i.e., $\mathbf{e}^{(s)} = (-\sin\varphi_{\rm sc}, \cos\varphi_{\rm sc}, 0)$.

The angular dependence of the scattered light intensity in this case is given by

$$I \sim \cos^2\varphi_{\rm sc}(\Delta\tilde{\varepsilon}\sin^2\theta - \delta\tilde{\varepsilon})^2\langle|\varphi_{\mathbf{q}}|^2\rangle. \tag{4.5}$$

The calculations are performed according to Eq. (4.5) for two geometries. In the first case the intensity of light scattering in the xz plane was analyzed. In another geometry the dependence of the scattering intensity on the azimuthal angle $\varphi_{\rm sc}$ was studied. In the calculations for Sm-C^*, the following values of the parameters were used: $K_{11} = K_{33} = 1.2\times10^{-11}$ N, $K_{22} = 0.6\times10^{-11}$ N, $\tilde{\varepsilon}_1 = 2.25$, $\tilde{\varepsilon}_2 = 2.56$, $\tilde{\varepsilon}_3 = 2.89$, $\theta = 15^\circ$, $k_0 = 10^7$ m^{-1}. We assumed that the system is in a uniform external electric field $E = 0.3$ statvolt/cm = 8994 V/m directed along the y axis. The results of the calculations in these geometries are presented in Fig. 3. As far as, according to Eqs. (4.1)–(4.3), the scattering intensity I for any geometry contains the quantity $\langle|\varphi_{\mathbf{q}}|^2\rangle$, we also present its angular dependence, which is shown in Fig. 4. The calculations were performed with the same parameters of the liquid crystal as in Fig. 3.

Note that all panels of Fig. 3 illustrate the angular dependence of the scattered light intensity for three systems with the same mean value of the permittivity $\varepsilon = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ but with different principal values of the permittivity tensor. The systems considered, isotropic, uniaxial and biaxial, differ from each other only by the term describing the Coulomb interaction of the polarization charges. From this figure one can see that the maximum difference in the angular dependence of the scattered light intensity is observed in the region of small angles for $\theta_{\rm sc}$ and for small angles and angles close to 180° for $\varphi_{\rm sc}$. The calculations are completed for not too large values of polarization P. Nevertheless, in the region of small $\theta_{\rm sc}$ and $\varphi_{\rm sc}$ the calculated scattered light intensity for the anisotropic

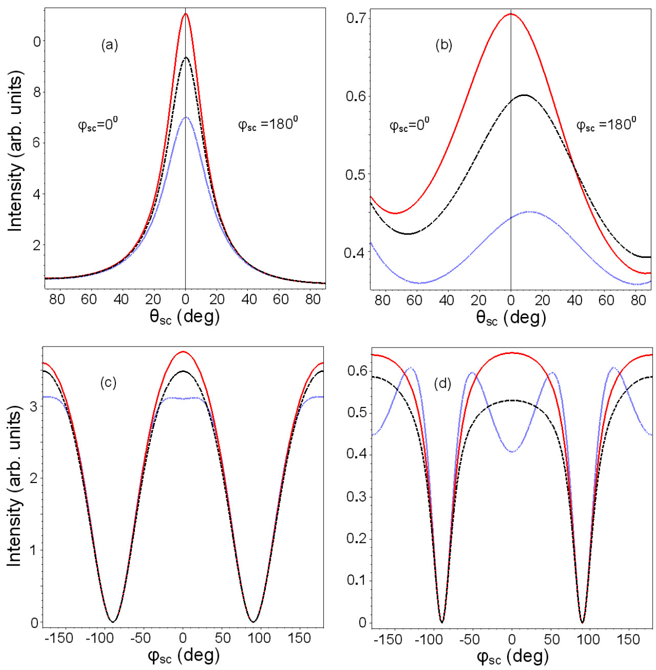

FIG. 3. (Color online) Angular dependence of the light scattering intensity. (a) and (b) show the dependencies of the scattered light intensity on the polar angle θ_{sc} for fixed azimuth angles ϕ_{sc}. (c) and (d) show the dependence of the scattered light intensity on the azimuth angle ϕ_{sc} for the fixed polar angle $\theta_{sc} = 20^\circ$. In (a) and (c) the polarization $P = 5$ statcoulomb/cm$^2 = 1.67 \times 10^4$ nC/m^2. In (b) and (d) the polarization $P = 20$ statcoulomb/cm$^2 = 6.67 \times 10^4$ nC/m^2. In all panels the scattering intensity is presented for three different combinations of the principal values of the static permittivity tensor. The solid red lines correspond to $\varepsilon_1 = \varepsilon_2 = \varepsilon_3 = 5$, the dashed black lines are for the uniaxial system with $\varepsilon_1 = \varepsilon_2 = 4$ and $\varepsilon_3 = 7$, and the dotted blue lines are for the biaxial system with $\varepsilon_1 = 3$, $\varepsilon_2 = 7$, and $\varepsilon_3 = 5$.

model systems may differ by more than half as much compared with the isotropic system. This is due to the difference in the form of the correlation function of the orientation fluctuations, whose angular dependencies are shown in Fig. 4.

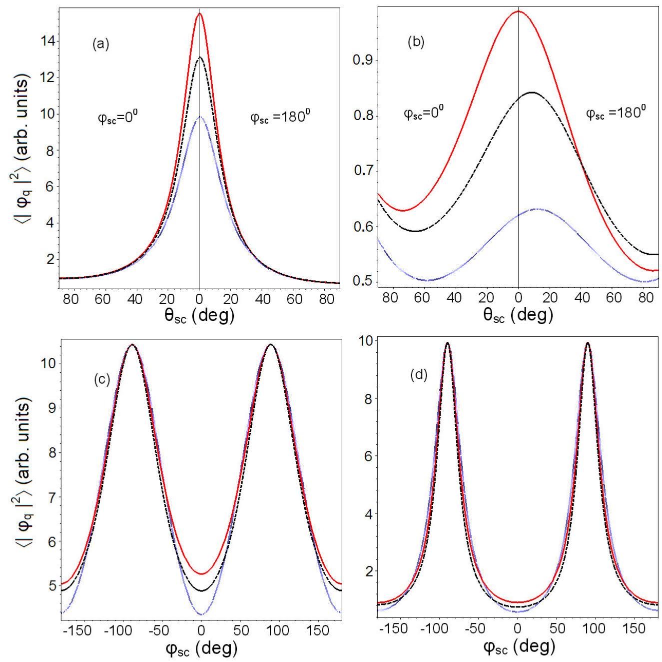

FIG. 4. (Color online) Angular dependence of the Fourier image of the **c**-director fluctuation correlation function. The parameters used are the same as in Fig. 3.

V. DISCUSSION

The importance of the Coulomb interaction of polarization charges in Sm-C^* manifests itself in many studies. This interaction affects the equilibrium texture and dynamics in Sm-C^* films [12–14,16,17], as well as fluctuations in the orientation and the scattering of light on them [20–24]. It was found that the presence of a large spontaneous polarization leads to an increase of the effective rigidity of the orientation of the director field [12,16,20–24] and to an increase in the effective viscosity of the orientation [16,20–24].

The relaxation of the director fields around a vortex of strength +1 in free-standing Sm-C^* films was studied experimentally in Ref. [17] by means of polarizing microscopy. The relaxation of the initial twisted configuration to equilibrium was also studied. It was found that the relaxation dynamics is highly nonlinear and contains two stages: a slow one when the phase is pinned in the center and on the boundary of the film, and a fast one when the **c** director flips by 180° in the center of the film, resulting in unwinding of the spiral pattern. This phenomenon is the result of differences in the elastic moduli K_s and K_b. In the theoretical analysis of the spontaneous polarization this is completely neglected, since in these systems it was small. At the same time it was pointed out that in systems with a large polarization, an account of the electrostatic interaction is needed.

In Ref. [13] the effect of spontaneous polarization on elastic properties of free-standing Sm-C^* films was studied experimentally. It was pointed out that the influence of polarization charges on elasticity had not been completely understood. For a qualitative description of the elastic properties, the one-constant approximation is sometimes used. It was pointed out that this approximation simplifies the analysis, but it is generally not justified by experiments in both polar and nonpolar smectics [16,29]: K_b may essentially differ from K_s and the elastic anisotropy K_b/K_s depends strongly on polarity. The magnitude of the bare ratio K_b/K_s was determined in the racemic mixture from the structure of periodic stripes and 2π walls in a magnetic field. Periodic stripe structures were used to study the influence of polarization on elasticity. Polarization dependent contributions to the elastic anisotropy and to the bend elastic constant show a P^2 behavior, in agreement with theoretical predictions [12].

The proposed theoretical models which consider the Coulomb interaction in the presence of polarization charges screening ionic impurities [12,23] show that a combination of large spontaneous polarization and a low concentration of screening ions produces the largest increase in the effective bend elastic constant.

Obviously, the anisotropy of the material plays a crucial role in the description of Sm-C^* cells and films. The one-constant approximation in the elastic contributions to the free energy allows one to only qualitatively describe the observed effects, and not for all cases. It would be natural to expect that for a quantitative description of the experimental data it is necessary to consider the interaction of the polarization charges by taking into account the anisotropic nature of the medium in which these charges interact. In particular, it is of interest to carry out the generalization of the theoretical models with an averaged permittivity which was used to describe the Coulomb interaction in two-dimensional films [1,12,20,21], or in three-dimensional systems [23] using the real anisotropic permittivity. In this paper we have shown that the inclusion of the anisotropy of permittivity in the description of the Coulomb interaction of the polarization charges in Sm-C^* is not a difficult problem, and at the same time it leads to noticeable effects. Our description was provided for the simplest, but interesting, case where the helix of the director rotation is unwound, because for this case we can obtain the analytical expression for the correlation function of the **c**-director orientation fluctuations in particular. As an illustration, we have considered the angular dependence of a scattered light intensity. From Figs. 3 and 4, which illustrate our calculations, we can see a sufficient difference between the angular dependencies of the light scattering intensity with and without accounting for the anisotropy of the permittivity.

It will be interesting to provide a light scattering experiment for Sm-C^* films with sufficient and different anisotropies of the permittivity, but with similar parameters. It is of interest to take into account the anisotropy of permittivity in the Coulomb interaction of polarization charges to describe the textures in films of Sm-C^* with spatial structural distortions arising from the presence of impurities, islands, special boundary conditions, external electric or magnetic fields, etc. But it seems that for these cases appropriate calculations can be provided only numerically.

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