# Light scattering on director fluctuations in smectic- A liquid crystals

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Theoretical analysis of peculiarities of light propagating and scattering in smectic-A liquid crystals is carried out. The optical anisotropy of the medium is incorporated in the analysis. The model takes into account distortion of the layer structure as well as of the field of the directors. The intensity of the light scattered on the director fluctuations is calculated for arbitrary directions of the incident and scattered light. Peculiarities of the angle dependence of intensity are analyzed in detail. All geometries of the optical experiment, which permit separate observation of the two fluctuation modes of the director, are obtained. The extinction coefficients of the ordinary and extraordinary rays far from the smectic-A-nematic phase transition as well as its vicinity are calculated. As a result, we obtain that, contrary to nematic liquid crystals, outside the nearst vicinity of the phase transition point, the extinction coefficients of the ordinary and extraordinary rays possess a similar angle dependence and are of the same order.

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#### I. INTRODUCTION

At the present time light-scattering methods are extensively used for studies of physical properties of various systems. An application of these methods to liquid crystals is extremely interesting, because there are highly strong fluctuations in these systems. Due to this property, light scattering in liquid crystals is strong. At the same time, the experimental data interpretation has been complicated by their intricate optical structure and, in particular, by their optical anisotropy. For nematic liquid crystals, a description taking into consideration this optical anisotropy has been carried out [1-4]. It allows one to develop methods of finding the thermodynamic and kinetic parameters from the light-scattering data [1-3,5].

For smectic-A liquid crystals, the anisotropic description has still not been carried out. It is known, however, that there are some interesting features of light scattering in these media. In particular, the prevailing part of light scattered in a smectic-A liquid crystal is concentrated in the region of the wave vectors cone whose axis is directed along the normal to the smectic layers. This effect was predicted by de Gennes [6] and experimentally confirmed by Ribotta, Salin, and Durand [7].

Recently, a special interest in studying the smectic-A-nematic phase transition has occurred. A peculiarity of this transition is that there are two correlation lengths possessing in the general case different critical indices. A main method of investigation of this transition is to study the light scattering intensity. Since the problem of measuring these indices exactly is rather complicated, it becomes important to give the correct theoretical description of the corresponding optical experiment. First and foremost, this involves rigorous consideration

of the optical anisotropy.

The second problem is to define those geometries of an optical experiment, for which the parameters of the smectic-A liquid crystal are contained in the relationships for the intensity of scattering in a simple way. It is common to use for these purposes the geometries, for which the contribution to scattering is given by one of the two fluctuation modes of the director [8-11]. However, the problem of finding all such geometries has not been regarded yet. Meanwhile, a knowledge of these geometries extends the capabilities of experiments.

The third problem is to calculate the extinction coefficient. It is well known [12,13] that extinction measurements near a phase-transition point are a rather important source of the information about the critical behavior of a system. Up to now, this problem for smectic liquid crystals has not been analyzed either theoretically or experimentally.

In the present paper, we calculate the angle distribution of the light-scattering intensity and the extinction coefficient for ordinary and extraordinary waves in smectic-A liquid crystals, taking properly into account their optical anisotropy. We supposed the medium to be unlimited. In Sec. II different types of fluctuations are analyzed in the Gaussian approximation. The modes describing the fluctuations of the director which give the main contribution into light scattering are regarded comprehensively. We take into account the fluctuations of the director itself as well as fluctuations of the director being caused by fluctuations of the layer structure. In Sec. III the intensity of the scattered light on the two modes connected with the fluctuations of the director is treated. All conditions of separately observing these modes in a light-scattering experiment are examined. We obtain relationships taking into account the optical anisotropy. In Sec. IV calculations of the extinction coefficient are carried out. A comprehensive analysis of its behavior near the point of the smectic-A-nematic phase transition is given.

The results obtained may be used for determining the parameters of smectic-A liquid crystals from light-scattering measurements.

#### II. FLUCTUATIONS

Being examined as an optical system, smectic-A liquid crystals display properties of a uniaxial medium with the following equilibrium value of the dielectric tensor:

$$\varepsilon_{\alpha\beta}^{0} = \varepsilon_{\perp} \delta_{\alpha\beta} + \varepsilon_{\alpha} n_{\alpha}^{0} n_{\beta}^{0} , \qquad (2.1)$$

where the unit vector  $\mathbf{n}^0$  directed along the optical axis of the system is called the "director." Here  $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$  and  $\varepsilon_{\parallel}$  and  $\varepsilon_{\perp}$  are the dielectric permeabilities parallel and perpendicular to  $\mathbf{n}^0$ . In smectic-A liquid crystals, as in nematic ones, the vector  $\mathbf{n}^0$  is directed along the prevailing direction of the long molecular axes.

From the microscopic point of view, smectic liquid crystals are layered systems with one-dimensionally period structures along the z axis. The value of the period length is of the order of the intermolecular distance. For smectic-A liquid crystals, z is parallel to  $\mathbf{n}^0$ , i.e., the layers are perpendicular to the director in the equilibrium state.

If we are interested in light-scattering problems, we can confine our examination to studying fluctuations of the dielectric tensor  $\delta \hat{\epsilon}$ . There are six fluctuation modes for any symmetrical tensor of the second rank, such as δε. There are one scalar mode (fluctuations of Trε), one longitudinal mode (fluctuations of the anisotropy  $\varepsilon_a$ ), two uniaxial transversal modes (fluctuations of the director), and two biaxial transversal modes (a local breaking of the uniaxiality of ê). Scalar, biaxial, and longitudinal fluctuations in smectic-A liquid crystals are comparatively small and of the order of those in ordinary organic liquids. (The exceptions may be connected with the biaxial fluctuations near the point of the phase transitions  $A \rightarrow C$ , and with the longitudinal fluctuations near the point of the phase transitions  $A \rightarrow I$ .) That is why our examination can be confined to the director fluctuations only. In this case in the first-order approximation, the fluctuations of the dielectric tensor are given by

$$\delta \varepsilon_{\alpha\beta}(\mathbf{r}) = \varepsilon_a \left[ \delta n_{\alpha}(\mathbf{r}) n_{\beta}^{0} + \delta n_{\beta}(\mathbf{r}) n_{\alpha}^{0} \right], \qquad (2.2)$$

where

$$\delta \mathbf{n}(\mathbf{r}) = \mathbf{n}(\mathbf{r}) - \mathbf{n}^0 \tag{2.3}$$

is the quantity of the fluctuation deflection of the local director  $\mathbf{n}(\mathbf{r})$  from its equilibrium value  $\mathbf{n}^0$ .

Note that above we have dealt with spontaneous thermal fluctuations. Aside from them in systems with degenerated symmetry, there may be contributions to  $\delta \hat{\epsilon}$  from other physical quantities. These contributions are connected with the so-called module conservation principle [14]. One of the possible approaches to calculate this kind of contribution to scalar fluctuations of  $\hat{\epsilon}$  from shift fluctuations of the smectic layers has been proposed in

Ref. [15].

For smectic-A liquid crystals, the director fluctuations can result from two causes. First, there are purely spontaneous thermal fluctuations of the long molecular axes. Contrary to nematic liquid crystals, such fluctuations require rather large energy, because they connect with the local deflection of the director from the normal to the layer (so-called tilt mode). Second, the director fluctuations can be produced by a local shift of the layers  $\mathbf{u}(\mathbf{r})$  (dilation mode). The only component of the shift vector  $\mathbf{u}$  fluctuating strongly is the component that is parallel to the z axis. Further, we shall designate it as u.

From this point of view the density of the elastic energy of a smectic-A deformation contains three types of contributions. The first contribution is the Frank energy,

$$F_N = \frac{1}{2} [K_{11} (\text{div}\mathbf{n})^2 + K_{22} (\mathbf{n} \cdot \text{rot}\mathbf{n})^2 + K_{33} (\mathbf{n} \times \text{rot}\mathbf{n})^2],$$
(2.4)

where  $K_{11}$ ,  $K_{22}$ , and  $K_{33}$  are the Frank moduli. This energy is connected with fluctuation deflections of the local directors  $\mathbf{n}(\mathbf{r})$  from the homogeneous state. The second contribution is the elastic energy  $F_L$  of the layered structure itself, and the third contribution is the energy  $F_{NL}$  connected with deflections of the local directors from a normal to the layers. For the sum of the second and the third contributions, we use the representation of de Gennes [16,17], adding the terms of the fourth order in  $\nabla$ .

$$F_{L} + F_{NL} = \frac{1}{2} a |\Psi|^{2} + \frac{1}{4} b |\Psi|^{4} + \frac{1}{2} C_{\parallel} |\nabla_{\parallel} \Psi|^{2} + \frac{1}{2} d_{\perp} |\nabla_{\perp}^{2} \Psi|^{2}$$

$$+ \frac{1}{2} d' |\nabla_{\parallel}^{2} \Psi|^{2} + \frac{1}{4} d'' |\nabla_{\perp} \nabla_{\parallel} \Psi|^{2}$$

$$+ \frac{1}{2} C_{\perp} |(\nabla_{\perp} + iq_{0} \delta \mathbf{n}_{\perp}) \Psi|^{2} , \qquad (2.5)$$

where

$$\Psi(\mathbf{r}) = |\Psi(\mathbf{r})| \exp\{iq_0 u(\mathbf{r})\}$$
 (2.6)

is the smectic's order parameter,  $|\Psi(\mathbf{r})|$  is proportional to the amplitude of the smectic density wave,  $2\pi/q_0$  is the period of the smectic structure,  $a=a'(T-T_{NA})$ ,  $T_{NA}$  is the temperature of the phase transition  $N\to A$ , and a', b,  $C_{\parallel}$ ,  $C_{\perp}$ ,  $d_{\perp}$ , d', and d'' are constants (further, we suppose them to be positive values, except d''; in addition, the inequality  $d_{\perp}d'>d''^{2}$  is supposed to be valid). In the Gaussian approximation over small deflections from the equilibrium value  $\xi(\mathbf{r})=|\Psi(\mathbf{r})|-|\Psi_{0}|$  and  $u(\mathbf{r})$ , we obtain from Eq. (2.5) with an accuracy of the surface terms,

$$\begin{split} F_L + F_{NL} &= F(|\Psi_0|) + \frac{1}{2} A \xi^2 + \frac{1}{2} C_{\parallel} (\nabla_{\parallel} \xi)^2 + \frac{1}{2} C_{\perp} (\nabla_{\perp} \xi)^2 \\ &+ \frac{1}{2} B (\nabla_{\parallel} u)^2 + \frac{1}{2} D (\nabla_{\perp} u + \delta \mathbf{n}_{\perp})^2 + \frac{1}{2} K_L (\nabla_{\perp}^2 u)^2 \\ &+ \frac{1}{2} K_L' (\nabla_{\parallel}^2 u)^2 + \frac{1}{2} K_L'' \nabla_{\parallel}^2 u \nabla_{\parallel}^2 u , \end{split}$$
(2.7)

where

$$A = -2a'(T - T_{NA}) = 2b |\Psi_0|^2, \quad B = C_{\parallel} q_0^2 |\Psi_0|^2,$$

$$D = C_{\perp} q_0^2 |\Psi_0|^2, \quad K_L = d_1 q_0^4 |\Psi_0|^2,$$

$$K_L' = d' q_0^4 |\Psi_0|^2, \quad K_L'' = d'' q_0^4 |\Psi_0|^2.$$
(2.8)

Here  $|\Psi_0|$  is the equilibrium value of the order parameter

which has the form  $|\Psi_0|^2 = -a'(T - T_{NA})/b$  for the mean-field approximation. In the expansion over  $\xi(\mathbf{r})$ , we have neglected the terms of the fourth order in  $\nabla$  in Eq. (2.7). It is a consequence of Eq. (2.7) that the mode  $\xi(\mathbf{r})$  does not interact with fluctuations of  $u(\mathbf{r})$  and  $\delta \mathbf{n}_1(\mathbf{r})$ .

In the limit  $D \rightarrow \infty$ , we can set identically

$$\delta \mathbf{n}_{1}(\mathbf{r}) = -\nabla_{1} u(\mathbf{r}) \tag{2.9}$$

in Eq. (2.7). This approximation corresponds to the situation when the local director is strictly perpendicular to the layers and its fluctuations are only determined by the fluctuations of the layer shift  $u(\mathbf{r})$ . In this case, the total contribution into the elastic energy density,

$$F_{A} = F_{N} + F_{L} + F_{NL} , \qquad (2.10)$$

connected with the layer shift fluctuations has the usual form (cf. Refs. 18,19])

$$F_{A}(u) = \frac{1}{2}B(\nabla_{\parallel}u)^{2} + \frac{1}{2}K_{1}(\nabla_{\perp}^{2}u)^{2} + \frac{1}{2}K'(\nabla_{\parallel}^{2}u)^{2} + \frac{1}{2}K''\nabla_{\perp}^{2}u\nabla_{\parallel}^{2}u , \qquad (2.11)$$

where

$$K_1 = K_{11} + K_L, \quad K' = K'_L, \quad K'' = K_{33} + K''_L$$
 (2.12)

The term  $K_{22}(\mathbf{n}\cdot\mathbf{rotn})^2$  from Eq. (2.4) does not give any contribution to  $F_A(u)$  in this case. For a finite value of D, deflections of the local director from a normal to the layers become possible. To take them into account it is necessary to use Eqs. (2.4) and (2.7) rather than (2.11).

Going over to the Fourier components, it is convenient to represent  $\delta n_{1a}$  in the form

$$\delta \mathbf{n}_{1q} = \delta n_{1q} \mathbf{e}_1 + \delta n_{2q} \mathbf{e}_2 , \qquad (2.13)$$

where the unit vectors  $\mathbf{e}_1$ ,  $\mathbf{e}_2$ , and  $\mathbf{n}^0$  constitute a right-hand orthogonal coordinate system, and  $\mathbf{e}_1$  lays in the plane defined by the vectors  $\mathbf{q}$  and  $\mathbf{n}^0$ . Thus for the given vector  $\mathbf{q}$ , the contribution to the elastic energy density has the form

$$F(\xi_{\mathbf{q}}, u_{\mathbf{q}}, \delta \mathbf{n}_{\mathbf{q}}) = \frac{1}{2} [(A + C_{\parallel} q_{\parallel}^{2} + C_{\perp} q_{\perp}^{2}) |\xi_{\mathbf{q}}|^{2} + (D + K_{33} q_{\parallel}^{2} + K_{11} q_{\perp}^{2}) |\delta n_{1\mathbf{q}}|^{2} + (D + K_{33} q_{\parallel}^{2} + K_{22} q_{\perp}^{2}) |\delta n_{2\mathbf{q}}|^{2} + (Bq_{\parallel}^{2} + Dq_{\perp}^{2} + K_{L} q_{\parallel}^{4} + K_{L}' q_{\parallel}^{2} q_{\perp}^{2}) |u_{\mathbf{q}}|^{2} + iDq_{\perp} (u_{\mathbf{q}} \delta n_{\perp \mathbf{q}}^{*} - u_{\mathbf{q}}^{*} \delta n_{\perp \mathbf{q}})],$$
(2.14)

where  $q_{\parallel}$  and  $q_{\perp}$  are the components of the vector  ${\bf q}$  parallel and perpendicular to  ${\bf n}^0$ . Only one mode  $\delta n_1$  from the two modes of the director fluctuations interacts with the layer shift fluctuations u. The quadratic form (2.14) allows us to obtain correlators of all fluctuation modes. In particular, for the director fluctuations, we can write (cf. Ref. [20])

$$\langle |\delta n_{jq}|^2 \rangle = \frac{k_B T}{\Delta_j + K_{jj} q_{\perp}^2 + K_{33} q_{\parallel}^2},$$
 (2.15)

$$\langle \delta n_{1q} \delta n_{2q}^* \rangle = 0 , \qquad (2.16)$$

where i = 1, 2,

$$\Delta_{1} = D \frac{Bq_{\parallel}^{2} + K_{L}q_{\perp}^{4} + K_{L}''q_{\parallel}^{2}q_{\perp}^{2} + K_{L}'q_{\parallel}^{4}}{Dq_{\perp}^{2} + Bq_{\parallel}^{2} + K_{L}q_{\perp}^{4} + K_{L}''q_{\parallel}^{2}q_{\perp}^{2} + K_{L}'q_{\parallel}^{4}}, \qquad (2.17)$$

$$\Delta_2 = D . (2.18)$$

Note that, contrary to nematic liquid crystals in smectic-A liquid crystals, the director fluctuations are not Goldstone fluctuations.

Far from the point  $T=T_{NA}$  the typical values of B and  $K_{11}$  are  $B\sim 2\times 10^7$  g/(cm sec<sup>2</sup>),  $K_{11}\sim 10^{-6}$  g cm/sec<sup>2</sup> (see Ref. [10]),  $K_{33}\sim 10^2K_{11}$  (see Ref. [11]). For estimations, we can consider  $D\sim B$ ,  $K_{22}\sim K_{33}$ ,  $K_L\sim K_{11}$ ,  $K_L\sim K_{11}\sim K_{12}\sim K_{33}$ ,  $q_{\parallel}\sim q_{\perp}\sim 10^5$  cm<sup>-1</sup>. Thus the contributions to the elastic energy connected with the changing of the interlayer distance (the coefficient B) and the deflection of the director from the normal to the layers (the coefficient D) is far greater than the contributions connected with the deflections of the director from the homogeneous state (the coefficients  $K_{11}$ ,  $K_{22}$ , and  $K_{33}$ ) and the layer bending (the coefficients  $K_L$ ,  $K'_L$ , and  $K''_L$ ):

$$B, D \gg K_{ii} q_{\perp}^2 + K_{33} q_{\parallel}^2 \tag{2.19}$$

and

$$Bq_{\parallel}^{2} + Dq_{\perp}^{2} \gg K_{L}q_{\perp}^{4} + K_{L}''q_{\perp}^{2}q_{\parallel}^{2} + K_{L}'q_{\parallel}^{4} . \qquad (2.20)$$

In the lowest order over B and D, we obtain

$$\langle |\delta n_{1q}|^2 \rangle = k_B T [B^{-1} q_{\perp}^2 q_{\parallel}^{-2} + D^{-1}],$$
  
 $\langle |\delta n_{2q}|^2 \rangle = k_B T D^{-1}.$  (2.21)

For  $q_{\parallel} \rightarrow 0$ , the first relationship in Eq. (2.21) becomes invalid. In this case, it is necessary to take into account the correction terms  $K_L q_{\perp}^4$  and  $K_{11} q_{\perp}^2$  in Eqs. (2.15) and (2.17). Thus we obtain

$$\langle |\delta n_{1q}|^2 \rangle = k_B T [Bq_{\parallel}^2 q_{\perp}^{-2} + K_1 q_{\perp}^2]^{-1} .$$
 (2.22)

Equation (2.22) coincides with the usual relationship for the director fluctuations in smectic-A liquid crystals [18,19]. The only difference is that  $K_1$  is the coefficient defined by Eq. (2.12) rather than the Frank modulus  $K_{11}$ .

Near the point of the phase transition  $T=T_{NA}$  the values  $B,D\to 0$ . In this case, the inequalities (2.19) are invalid and now we cannot neglect the terms  $K_{jj}q_{\perp}^2+K_{33}q_{\parallel}^2$  in the denominators of Eqs. (2.15), as we do in Eqs. (2.21). If the inequality (2.20) remains valid as before [for example, for the mean-field theory in accordance with Eqs. (2.8), the ratios between values  $B,D,K_L$ ,  $K_L'$ , and  $K_L''$  do not depend on a temperature], we obtain

$$\Delta_{1}(q_{\parallel},q_{\perp}) = \frac{BDq_{\parallel}^{2}}{Bq_{\parallel}^{2} + Dq_{\perp}^{2}}, \qquad (2.23)$$

and the temperature dependence of  $\Delta_1$  is defined by the coefficients B and D. In particular,

$$\Delta_{1}(q_{\parallel}, q_{\perp}) = \begin{cases} Bq_{\parallel}^{2}q_{\perp}^{-2} & \text{if } q_{\parallel}^{2} \ll q_{\perp}^{2} \\ D & \text{if } q_{\parallel}^{2} \gg q_{\perp}^{2} \end{cases}$$
 (2.24)

If inequality (2.20) becomes invalid for  $T \rightarrow T_{NA}$  because of the fluctuations interaction, the temperature dependence of  $\Delta_1$  is defined by all sets of coefficients B, D,  $K_L$ ,  $K'_L$ , and  $K''_L$  in Eq. (2.17).

Thus Eqs. (2.21) and (2.22) give us that far from  $T=T_{NA}$  the modes  $\langle |\delta n_{1q}|^2 \rangle$  and  $\langle |\delta n_{2q}|^2 \rangle$  are of the same order if  $q_{\parallel} \sim q_{\perp}$ . However, for  $q_{\parallel}/q_{\perp} \rightarrow 0$ , the mode  $\langle |\delta n_{2q}|^2 \rangle$  is practically invariable, and the mode  $\langle |\delta n_{1q}|^2 \rangle$  rises sharply. Near  $T=T_{NA}$  for  $q_{\parallel} \neq 0$ , the modes  $\langle |\delta n_{jq}|^2 \rangle$  rise critically. If  $q_{\parallel} = 0$ , the behavior of  $\langle |\delta n_{1q}|^2 \rangle$  is Goldstone's one:  $\langle |\delta n_{1q}|^2 \rangle \sim q_{\perp}^{-2}$ .

From Eqs. (2.2) and (2.13), it is easy to find the correlation function of the fluctuations of the dielectric tensor  $G_{\mu\alpha\nu\beta}(\mathbf{r}_1,\mathbf{r}_2) = \langle \delta \varepsilon_{\mu\alpha}(\mathbf{r}_1) \delta \varepsilon_{\nu\beta}^*(\mathbf{r}_2) \rangle$ . In (q) representation, it takes the form

$$G_{\mu\alpha\nu\beta}(\mathbf{q}) = \varepsilon_a^2 \sum_{j=1,2} \langle |\delta n_{jq}|^2 \rangle (e_{j\mu} n_{\alpha}^0 + e_{j\alpha} n_{\mu}^0) \times (e_{j\nu} n_{\beta}^0 + e_{j\beta} n_{\nu}^0) . \tag{2.25}$$

# III. INTENSITY

## A. General relations

The intensity of a single scattering inside an anisotropic medium may be written in the form [21,4]:

$$I_{(s)}^{(i)} = I_0^{(i)} \frac{V}{R^2} \left[ \frac{\omega^2}{4\pi c^2} \right]^2 \frac{n_{(s)}}{n_{(i)} \cos \delta_{(i)} \cos^3 \delta_{(s)}}$$

$$\times f_{(s)}^2 e_{\mu}^{(i)} e_{\nu}^{(i)} G_{\mu\alpha\nu\beta}(\mathbf{q}) e_{\alpha}^{(s)} e_{\beta}^{(s)} .$$
(3.1)

Here and below the index (i) is related to the incident wave; the index (s) is related to the scattering wave; i,s=1 for the ordinary wave; i,s=2 for the extraordinary wave;  $I_0^{(i)}$  is the intensity of the incident light; V is the scattering volume; R is the distance between the scattering point and the observation point; the condition  $V^{1/3} \ll R$  is supposed to be valid;  $\omega$  is the circular frequency; c is the light velocity in a vacuum;  $\mathbf{q} = \mathbf{k}_{(s)} - \mathbf{k}_{(i)}$ ;  $\mathbf{k}_{(i)}$  and  $\mathbf{k}_{(s)}$  are the wave vectors;  $n_{(i)}$  and  $n_{(s)}$  are the refraction indexes;  $e^{(i)}$  and  $e^{(s)}$  are the polarization vectors; and  $\delta_{(i)}$  and  $\delta_{(s)}$  are the angles between the vectors of the electric field intensity and the vectors of the electric field induction, related to the corresponding waves. The factors  $f_{(s)}$  are connected with the wave surface Gaussian curvatures. In uniaxial media for the waves with the wave vectors  $\mathbf{k}_{(j)} = k_{(j)} \mathbf{m}$  (j = 1, 2), we obtain [4]

$$n_{(1)} = \varepsilon_{1}^{1/2}, \quad n_{(2)} = \left(\frac{\varepsilon_{\parallel}\varepsilon_{\perp}}{\beta(\vartheta)}\right)^{1/2}, \quad \mathbf{e}^{(1)} = \frac{[\mathbf{m} \times \mathbf{n}^{0}]}{\sin\vartheta},$$

$$\mathbf{e}^{(2)} = \frac{\mathbf{m}\varepsilon_{\parallel}\cos\vartheta - \mathbf{n}^{0}\beta(\vartheta)}{\sin(\vartheta)\alpha(\vartheta)}, \quad \cos\delta_{(1)} = 1,$$

$$\cos\delta_{(2)} = \frac{\beta(\vartheta)}{\alpha(\vartheta)}, \quad f_{(1)} = 1, \quad f_{(2)} = \frac{\alpha(\vartheta)\beta^{1/2}(\vartheta)}{\varepsilon_{\parallel}^{1/2}\varepsilon_{\parallel}}.$$
(3.2)

Here and below we use the notation

$$\alpha(\vartheta) = (\varepsilon_{\parallel}^2 \cos^2 \vartheta + \varepsilon_{\perp}^2 \sin^2 \vartheta)^{1/2} ,$$
  

$$\beta(\vartheta) = \varepsilon_{\parallel} \cos^2 \vartheta + \varepsilon_{\perp} \sin^2 \vartheta ,$$
(3.3)

and  $\vartheta$  is the angle between the vectors **m** and  $\mathbf{n}^0$ .

Equation (3.1) corresponds to the case where the incident and scattering waves propagate inside the medium. Since the real measurements are carried out outside the medium it is necessary to take into account the influence of the boundaries. This problem is rather difficult; it was regarded in detail by Lax and Nelson [21]. The corrections connected with the influence of the boundaries result in additional angle factors in Eq. (3.1), which depend on the angle  $\vartheta$  as well as on the geometry of the experiment.

Note that the real direction of the wave propagation is the Poynting vector S. Out of the medium its direction coincides with the direction of the wave vector k, and inside the anisotropic medium S is not parallel to k. However, the laws of refraction and reflection are formulated in more convenient form for k. That is why all equations for the scattering inside media are represented here in terms of the wave vectors k rather than S.

Calculating the convolution in Eq. (3.1) for the correlation function given by Eq. (2.25), we obtain

$$e_{\mu}^{(i)} e_{\nu}^{(i)} G_{\mu\alpha\nu\beta}(\mathbf{q}) e_{\alpha}^{(s)} e_{\beta}^{(s)}$$

$$= \varepsilon_{a}^{2} \sum_{j=1,2} \langle |\delta n_{j\mathbf{q}}|^{2} \rangle Q_{j}(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}, \mathbf{q}) , \quad (3.4)$$

where angle factors

$$Q_{j}(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}, \mathbf{q}) = [(\mathbf{e}^{(i)} \cdot \mathbf{e}_{j})(\mathbf{e}^{(s)} \cdot \mathbf{n}^{0}) + (\mathbf{e}^{(s)} \cdot \mathbf{e}_{j})(\mathbf{e}^{(i)} \cdot \mathbf{n}^{0})]^{2}.$$
(3.5)

# B. Conditions for the separate observation of the modes $\delta n_1$ and $\delta n_2$ in scattering

For investigation of smectic-A properties by light-scattering methods, it is convenient to use the experimental geometries for which the scattering contribution is given separately by modes  $\delta n_1$  and  $\delta n_2$  [8–11]. These geometries are determined by turning into zero one of the factors  $Q_j(\mathbf{e}^{(i)},\mathbf{e}^{(s)},\mathbf{q})$ . The detailed calculations of all these geometries are given in the Appendix. It turns out that there are seven such geometries for  $(o) \rightarrow (o)$ ,  $(o) \rightarrow (e)$ ,  $(e) \rightarrow (o)$ , and  $(e) \rightarrow (e)$  types of scattering [here the indices (o) and (e) correspond to the ordinary and extraordinary waves]. We designate them as G0–G6.

First and foremost, the intensity of  $(o) \rightarrow (o)$  scattering turns into zero identically for both modes  $\delta n_1$  and  $\delta n_2$  and for every direction of  $\mathbf{k}_{(i)}$  and  $\mathbf{k}_{(s)}$ —the geometry G0.

Let us describe the other geometries G1-G6. We consider the vectors  $\mathbf{n}^0$  and  $\mathbf{k}_{(i)}$  to be fixed. Then every such geometry is defined by totality of the vectors  $\mathbf{k}_{(s)}$ . It is convenient to use the spherical coordinate system with the polar axis being parallel to  $\mathbf{n}^0$ . The polar angles of the vectors  $\mathbf{k}_{(i)}$  and  $\mathbf{k}_{(s)}$  we shall designate as  $\vartheta_i$  and  $\vartheta_s$ , the azimuthal angle of the vector  $\mathbf{k}_{(i)}$  is chosen to be

equal to zero, and that of the vector  $\mathbf{k}_{(s)}$  is designated as  $\phi$ . Every geometry is characterized by its own relation among the angles  $\vartheta_i$ ,  $\vartheta_s$ , and  $\phi$  which defines the corresponding bold line in Fig. 1. The components  $q_{\parallel}$  and  $q_{\perp}$ , which are necessary for the calculations of  $\langle |\delta n_{jq}|^2 \rangle$ , are represented as functions of  $\vartheta_i$ ,  $\vartheta_s$ , and  $\phi$  for every geometry. (The relationships for  $\langle |\delta n_{jq}|^2 \rangle$  are given in Sec. II.) In the following, we use the notation

$$A_0 = I_0^{(i)} \frac{V \varepsilon_a^2}{R^2} \left[ \frac{\omega^2}{4\pi c^2} \right]^2. \tag{3.6}$$

# 1. Geometry G1

The scattering involves the mode  $\langle |\delta n_{2q}|^2 \rangle$ . The type of scattering is  $(o) \rightarrow (e)$ . The equation of the line characterizing the scattering for this mode only is  $\phi = 0$  [Fig. 1(a)]. The intensity of the scattering for this geometry is

$$I_{(2)}^{(1)} = A_0 \frac{\sin^2 \vartheta_s \alpha^3(\vartheta_s)}{\varepsilon_{\parallel}^{1/2} \beta^{5/2}(\vartheta_s)} \langle |\delta n_{2q}|^2 \rangle , \qquad (3.7)$$

$$q_{\parallel} = (\omega/c)[n_{(2)}(\vartheta_s)\cos\vartheta_s - n_{(1)}\cos\vartheta_i],$$

$$q_{\perp} = (\omega/c)[n_{(2)}(\vartheta_s)\sin\vartheta_s - n_{(1)}\sin\vartheta_i].$$
(3.8)

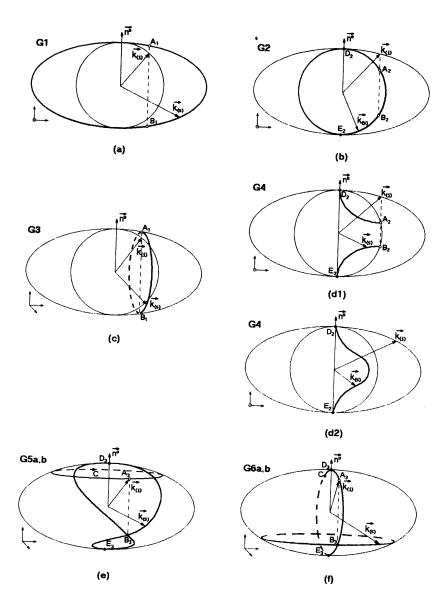


FIG. 1. The arrangement of the scattering wave vectors  $\mathbf{k}_{(s)}$  for which the angle factor  $Q_j$  turns into zero (the case of  $\varepsilon_a > 0$ ). The thick lines exhibit the points on the wave surface defined by this kind of  $\mathbf{k}_{(s)}$  for the preliminary given incident wave vector  $\mathbf{k}_{(i)}$ . The thin lined circle and ellipse exhibit the wave surface for a uniaxial medium. (a) i=1, s=2, j=1. (b) i=2, s=1, j=1. (c) i=1, s=2, j=2. (d1) i=2, s=1, j=2,  $k_{(i)_1} < k_{(s)}$ . (d2) i=2, s=1, j=2,  $k_{(i)_1} > k_{(s)}$ . (e) i=s=2, j=1. (f) i=s=2, j=2. The corresponded coordinate system is shown in the bottom left-hand corner of every figure.

## 2. Geometry G2

The scattering involves the mode  $\langle |\delta n_{2q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (o)$ . The equation of the line characterizing the scattering for this mode only is  $\phi = 0$  [Fig. 1(b)]. The intensity of the scattering for this geometry is

$$I_{(1)}^{(2)} = A_0 \frac{\varepsilon_1^2 \sin^2 \vartheta_i}{\varepsilon_{\parallel}^{1/2} \alpha(\vartheta_i) \beta^{1/2}(\vartheta_i)} \langle |\delta n_{2q}|^2 \rangle , \qquad (3.9)$$

$$q_{\parallel} = (\omega/c) [n_{(1)}\cos\vartheta_s - n_{(2)}(\vartheta_i)\cos\vartheta_i] ,$$
  

$$q_{\perp} = (\omega/c) [n_{(1)}\sin\vartheta_s - n_{(2)}(\vartheta_i)\sin\vartheta_i] .$$
(3.10)

### 3. Geometry G3

The scattering involves the mode  $\langle |\delta n_{1q}|^2 \rangle$ . The type of scattering is  $(o) \rightarrow (e)$ . The equation of the line characterizing the scattering for this mode only is  $n_{(1)} \sin \vartheta_i = n_{(2)} (\vartheta_s) \sin \vartheta_s \cos \phi$  [Fig. 1(c)]. The intensity of the scattering for this geometry is

$$I_{(2)}^{(1)} = A_0 \frac{\sin^2 \vartheta_s \alpha^3(\vartheta_s)}{\varepsilon_{\parallel}^{1/2} \beta^{5/2}(\vartheta_s)} \langle |\delta n_{1q}|^2 \rangle , \qquad (3.11)$$

$$q_{\parallel} = (\omega/c) [n_{(2)}(\vartheta_s) \cos \vartheta_s - n_{(1)} \cos \vartheta_i],$$
  

$$q_{\perp} = (\omega/c) [n_{(2)}^2(\vartheta_s) \sin^2 \vartheta_s - n_{(1)}^2 \sin^2 \vartheta_i]^{1/2}.$$
(3.12)

## 4. Geometry G4

The scattering involves the mode  $\langle |\delta n_{1q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (o)$ . The equation of the line characterizing the scattering for this mode only is  $n_{(1)} \sin \vartheta_s = n_{(2)} (\vartheta_i) \sin \vartheta_i \cos \phi$  [Fig. 1(d)]. The intensity of the scattering for this geometry is

$$I_{(1)}^{(2)} = A_0 \frac{\varepsilon_1^2 \sin^2 \vartheta_i}{\varepsilon_1^{1/2} \alpha(\vartheta_i) \beta^{1/2}(\vartheta_i)} \langle |\delta n_{1q}|^2 \rangle , \qquad (3.13)$$

$$q_{\parallel} = (\omega/c) [n_{(1)}\cos\vartheta_s - n_{(2)}(\vartheta_i)\cos\vartheta_i],$$
  

$$q_{\perp} = (\omega/c) [n_{(2)}^2(\vartheta_i)\sin^2\vartheta_i - n_{(1)}^2\sin^2\vartheta_s]^{1/2}.$$
(3.14)

#### 5. Geometry G5a

The scattering involves the mode  $\langle |\delta n_{2q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (e)$ . The equation of the line characterizing the scattering for this mode only is  $\vartheta_i = \vartheta_s$  [Fig. 1(e)]. The intensity of the scattering for this geometry is

$$I_{(2)}^{(2)} = A_0 \frac{\varepsilon_{\parallel} \alpha^2(\vartheta_i)}{\beta^3(\vartheta_i)} \sin^2(2\vartheta_i) \cos^2(\phi/2) \langle |\delta n_{2q}|^2 \rangle , \quad (3.15)$$

$$q_{\parallel} = 0$$
,  
 $q_{\perp} = 2(\omega/c)n_{(2)}(\vartheta_i)\sin\vartheta_i\sin(\phi/2)$ . (3.16)

#### 6. Geometry G5b

The scattering involves the mode  $\langle |\delta n_{2q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (e)$ . The equation of the line characterizing the scattering for this mode only is

$$\cos\phi = \frac{[n_{(2)}(\vartheta_s)\cos\vartheta_s - n_{(2)}(\vartheta_i)\cos\vartheta_i]\sin\vartheta_i\sin\vartheta_s}{n_{(2)}(\vartheta_i)\cos\vartheta_s\sin^2\vartheta_i - n_{(2)}(\vartheta_s)\cos\vartheta_i\sin^2\vartheta_s}$$

[Fig. 1(e)]. The intensity of the scattering for this geometry is

$$I_{(2)}^{(2)} = A_0 \frac{\varepsilon_{\parallel} \alpha^3(\vartheta_s)}{\alpha(\vartheta_i) \beta^{1/2}(\vartheta_i) \beta^{5/2}(\vartheta_s)} \sin(\vartheta_i - \vartheta_s) \sin(\vartheta_i + \vartheta_s) \frac{n_{(2)}(\vartheta_i) \cos\vartheta_s \sin^2\vartheta_i + n_{(2)}(\vartheta_s) \cos\vartheta_i \sin^2\vartheta_s}{n_{(2)}(\vartheta_i) \cos\vartheta_s \sin^2\vartheta_i - n_{(2)}(\vartheta_s) \cos\vartheta_i \sin^2\vartheta_s} \langle |\delta n_{2q}|^2 \rangle .$$

$$(3.17)$$

$$q_{\parallel} = (\omega/c) \left[ n_{(2)}(\vartheta_s) \cos\vartheta_s - n_{(2)}(\vartheta_i) \cos\vartheta_i \right],$$

$$q_{\perp} = (\omega/c) \left| \frac{n_{(2)}(\vartheta_i) \cos\vartheta_s \sin^2\vartheta_i + n_{(2)}(\vartheta_s) \cos\vartheta_i \sin^2\vartheta_s}{n_{(2)}(\vartheta_i) \cos\vartheta_s \sin^2\vartheta_i - n_{(2)}(\vartheta_s) \cos\vartheta_i \sin^2\vartheta_s} \right|^{1/2} \left| n_{(2)}^2(\vartheta_i) \sin^2\vartheta_i - n_{(2)}^2(\vartheta_s) \sin^2\vartheta_s \right|^{1/2}.$$
(3.18)

(3.19)

# 7. Geometry G6a

The scattering involves the mode  $\langle |\delta n_{1q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (e)$ . The equation of the line characterizing the scattering on this mode is  $\phi = 0$  [Fig. 1(f)]. The intensity of the scattering for this geometry is

$$I_{(2)}^{(2)} = A_0 \frac{\varepsilon_{\parallel} \alpha^3(\vartheta_s)}{\alpha(\vartheta_i) \beta^{1/2}(\vartheta_i) \beta^{5/2}(\vartheta_s)} \sin^2(\vartheta_i + \vartheta_s) \langle |\delta n_{1q}|^2 \rangle ,$$

8. Geometry G6b

The scattering involves the mode  $\langle |\delta n_{1q}|^2 \rangle$ . The type of scattering is  $(e) \rightarrow (e)$ . The equation of the line characterizing the scattering for this mode is  $\vartheta_i = \vartheta_s$  [Fig. 1(f)]. The intensity of the scattering for this geometry is

$$q_{\parallel} = (\omega/c) [n_{(2)}(\vartheta_s) \cos \vartheta_s - n_{(2)}(\vartheta_i) \cos \vartheta_i] ,$$
  

$$q_{\perp} = (\omega/c) [n_{(2)}(\vartheta_s) \sin \vartheta_s - n_{(2)}(\vartheta_i) \sin \vartheta_i] .$$
(3.20)

$$I_{(2)}^{(2)} = A_0 \frac{\varepsilon_{\parallel} \alpha^2(\vartheta_i)}{\beta^3(\vartheta_i)} \sin^2(2\vartheta_i) \sin^2(\phi/2) \langle |\delta n_{1q}|^2 \rangle , \qquad (3.21)$$

$$q_{\parallel} = -2(\omega/c) n_{(2)}(\vartheta_i) \cos \vartheta_i ,$$

$$q_{\perp} = 2(\omega/c) n_{(2)}(\vartheta_i) \sin \vartheta_i \sin(\phi/2) .$$
(3.22)

Note that the definition of the vectors  $\mathbf{e}_1$  and  $\mathbf{e}_2$  given in Eq. (2.13) becomes impossible for  $\mathbf{q}_1 = \mathbf{0}$ . That is why partitioning  $\delta \mathbf{n}_q$  into two modes  $\delta n_{jq}$  also becomes impossible in this case. In Fig. 1 the points  $A_m$  and  $B_m$ , m=1,2,3, corresponds to this condition, where  $\mathbf{q}_1 = \mathbf{0}$ . In fact, all these points are the points of the intersection of the lines on which  $Q_1$  and  $Q_2$  turn to zero. Then one would expect that the intensity of scattering at these points is equal to zero. However, it is not true in general. In this case it is necessary to regard the complete intensity. In the limit  $\mathbf{q}_1 \rightarrow \mathbf{0}$ , Eqs. (2.15) and (2.17) give

$$\langle |\delta n_{1q}|^2 \rangle = \langle |\delta n_{2q}|^2 \rangle = \frac{k_B T}{D + K_{33} q_{\parallel}^2} . \qquad (3.23)$$

Taking into account the identity

$$e_{1\alpha}e_{1\beta} + e_{2\alpha}e_{2\beta} + n_{\alpha}^{0}n_{\beta}^{0} = \delta_{\alpha\beta}$$
, (3.24)

we can rewrite the correlation function (2.25) in the form

$$G_{\mu\alpha\nu\beta}(q_{\parallel},q_{\perp}=0) = \frac{\varepsilon_{\alpha}^{2}k_{B}T}{D+K_{33}q_{\parallel}^{2}} (\delta_{\mu\nu}n_{\alpha}^{0}n_{\beta}^{0} + \delta_{\mu\beta}n_{\alpha}^{0}n_{\nu}^{0} + \delta_{\alpha\beta}n_{\mu}^{0}n_{\nu}^{0} + \delta_{\alpha\nu}n_{\mu}^{0}n_{\beta}^{0} - 4n_{\mu}^{0}n_{\alpha}^{0}n_{\nu}^{0}n_{\beta}^{0}) .$$
(3.25)

Then from Eqs. (3.25) and (3.1) we have

$$I_{(s)}^{(i)} \sim (\mathbf{n}^{0} \cdot \mathbf{e}^{(i)})^{2} + (\mathbf{n}^{0} \cdot \mathbf{e}^{(s)})^{2}$$

$$+ 2(\mathbf{e}^{(i)} \cdot \mathbf{e}^{(s)})(\mathbf{n}^{0} \cdot \mathbf{e}^{(i)})(\mathbf{n}^{0} \cdot \mathbf{e}^{(s)})$$

$$- 4(\mathbf{n}^{0} \cdot \mathbf{e}^{(i)})^{2}(\mathbf{n}^{0} \cdot \mathbf{e}^{(s)})^{2}$$
(3.26)

Thus for geometries G1 and G3, or G2 and G4, the intensities of the scattering at the points  $A_1$  and  $B_1$  or  $A_2$  and  $B_2$ , correspondingly (see Fig. 1),

$$I_{(s)}^{(i)} \sim (\mathbf{n}^0 \cdot \mathbf{e}^{(2)})^2$$
, (3.27)

differs from zero for  $\vartheta_i \neq 0$ ,  $\vartheta_i \neq \pi/2$ . For the geometries G5 and G6 at the point  $A_3$  (forward scattering),

$$I_{(2)}^{(2)} \sim 4(\mathbf{n}^0 \cdot \mathbf{e}^{(2)})^2 [1 - (\mathbf{n}^0 \cdot \mathbf{e}^{(2)})^2],$$
 (3.28)

also differs from zero for  $\vartheta_i \neq 0$ ,  $\vartheta_i \neq \pi/2$ ; and, at the point  $B_3$ ,

$$I_{(2)}^{(2)} \equiv 0$$
 (3.29)

Thus it is necessary to exclude the points  $A_1$ ,  $B_1$ ,  $A_2$ ,  $B_2$ , and  $A_3$  from the lines of the intensities equal to zero (in Fig. 1 they are designated by unfilled circles). On the contrary, at the point  $B_3$ , the intensity is actually equal to zero.

The calculations of intensities with the correlation function given by Eq. (3.25) show that one can make use of Eq. (3.7) or (3.11) for the points  $A_1$  and  $B_1$ . For  $q_{\parallel}$ , we

have for these points

$$q_{\parallel} = (\omega/c) \varepsilon_{\perp}^{1/2} \sin \vartheta_{i} (\cot \vartheta_{s} - \cot \vartheta_{i}) . \tag{3.30}$$

Analogously, for the points  $A_2$  and  $B_2$ , one can make use of Eq. (3.9) or (3.13). For  $q_{\parallel}$ , we have for these points

$$q_{\parallel} = (\omega/c) \varepsilon_1^{1/2} \sin \vartheta_s (\cot \vartheta_s - \cot \vartheta_i) . \tag{3.31}$$

Note that for the points  $A_1$ ,  $B_1$ ,  $A_2$ , and  $B_2$  the angles  $\vartheta_i$  and  $\vartheta_s$  are related by

$$\cot^2\vartheta_2 - \cot^2\vartheta_1 = \varepsilon_a / \varepsilon_{\parallel} . \tag{3.32}$$

For the point  $A_3$  (forward scattering), one can make use of Eqs. (3.15) or (3.19), taking into consideration the conditions  $\phi=0$  or  $\vartheta_i=\vartheta_s$ , correspondingly. In this case  $\mathbf{q}=\mathbf{0}$ . For all these special points, it is implied that Eq. (3.23) is used for  $\langle |\delta n_{1\mathbf{q}}|^2 \rangle$  and  $\langle |\delta n_{2\mathbf{q}}|^2 \rangle$ .

Note that the simultaneous turning of  $Q_1$  and  $Q_2$  to zero corresponds to the absence of the contribution of the director fluctuations to the scattering. For the geometry G0, the condition  $Q_1 = Q_2 = 0$  is obeyed identically. For the geometries G1 and G3, the simultaneous turning of  $Q_1$  and  $Q_2$  to zero is impossible because the corresponding curved lines have no points of intersection. For the geometries G2 and G4, the points  $D_2$  and  $E_2$  are common but the directions of the polarization vectors  $e^{(s)}$ defined by these geometries are orthogonal to each other for each of these points. Thus it is impossible to satisfy the conditions  $Q_1 = 0$  and  $Q_2 = 0$  at each of the points  $D_2$ and  $E_2$  simultaneously. The same conclusion is true for the points  $D_3$  and  $E_3$  for the geometries G5 and G6. At the common points for the last geometries, points C and  $B_3$ , the conditions of the simultaneous turning of  $Q_1$  and  $Q_2$  to zero are noncontradictory. Moreover, for G5 and G6, for  $\vartheta_i = \pi/2$ , there is a whole line ("equator") of common points, where  $Q_1 = Q_2 = 0$ . Note that this analysis of the turning  $Q_1$  and  $Q_2$  to zero is also valid for nematic liquid crystals. Particularly, the condition  $Q_1 = Q_2 = 0$  for them had been analyzed [4,22]. It is easy to see that the results of these two examinations are in complete agreement.

# C. Features of the scattering far from the phase-transition point $T_{NA}$

Far from the point  $T_{NA}$ , the mode  $\langle |\delta n_{1q}|^2 \rangle$  increases rapidly in the region of  $q_{\parallel}/q_{\perp} \rightarrow 0$ . This fact was mentioned at the end of Sec. II. In this region, the mode  $\langle |\delta n_{2q}|^2 \rangle$  can be ignored. Taking into account that the quantities  $q_{\parallel}$  and  $q_{\perp}$  in optics are of the order of  $\omega/c$ , we have from Eq. (2.22) under the condition  $(\omega/c)^2 K_1/B \ll 1$ ,

$$\langle |\delta n_{1q}|^2 \rangle \approx \frac{\pi k_B T \delta(q_{\parallel})}{(K_1 B)^{1/2}} . \tag{3.33}$$

Here we use the well-known representation for the  $\delta$  function,

$$\delta(x) = \pi^{-1} \lim_{\varepsilon \to 0} \frac{\varepsilon}{\varepsilon^2 + x^2} .$$

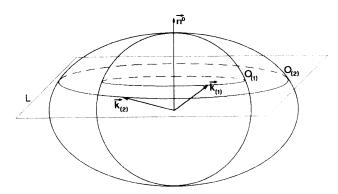


FIG. 2. Mutual arrangement of the incident  $(\mathbf{k}_{(i)})$  and scattered  $(\mathbf{k}_{(s)})$  wave vectors on the wave surface for the intensive scattering. (The case of  $\varepsilon_a > 0$ .) The cases i = 1, s = 2 and i = 2, s = 1 are possible.

One can see from Eqs. (2.25), (3.1), and (3.33) that the scattered light is concentrated in the region of those directions of the wave vectors  $\mathbf{k}_{(s)}$ , for which  $q_{\parallel} = 0$ , i.e.,  $k_{(s)_{\parallel}} = k_{(i)_{\parallel}}$ . For uniaxial media, this equality means that  $\mathbf{k}_{(s)}$  must be situated on the surfaces of two circular cones, with  $\mathbf{n}_0$  as the axis of symmetry. This effect was predicted by de Gennes [6] and experimentally confirmed

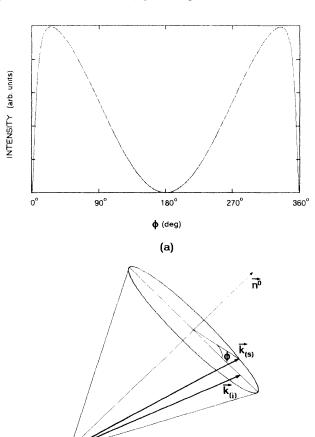


FIG. 3. (a) The angle dependence of the intensity of the scattering along the ring calculated by use of Eq. (3.35). We set  $\varepsilon_{\parallel} = 3.5$ ,  $\varepsilon_{\perp} = 2.5$ . (b) The corresponding geometry of scattering (i=1, s=2).

(b)

by Ribotta, Salin, and Durand [7].

Figure 2 shows the  $\mathbf{k}_{(i)}$  and  $\mathbf{k}_{(s)}$  wave-vector arrangement for the scattering in a smectic-A liquid crystal. The plane L which is perpendicular to  $\mathbf{n}^0$  selects on the wave surfaces two circles  $O_{(1)}$  and  $O_{(2)}$ . These circles are related to the ordinary and extraordinary waves. Let the vector  $\mathbf{k}_{(i)}$  lie on one of them. Then the condition  $k_{(s)_{\parallel}} = k_{(i)_{\parallel}}$  gives us that  $\mathbf{k}_{(s)}$  must lie on one of them too. The values of the polar angles  $\vartheta_1$  (between  $\mathbf{k}_{(1)}$  and  $\mathbf{n}^0$ ) and  $\vartheta_2$  (between  $\mathbf{k}_{(2)}$  and  $\mathbf{n}^0$ ), which are determined from the circles  $O_{(1)}$  and  $O_{(2)}$  in Fig. 2, are connected by

$$\tan \vartheta_2 = (\varepsilon_{\parallel}/\varepsilon_{\perp})^{1/2} \tan \vartheta_1 \ . \tag{3.34}$$

It is the consequence of the geometries G0 and G5a (Sec. III B) that the intensities  $I_{(1)}^{(1)}$  and  $I_{(2)}^{(2)}$  are equal to zero for these directions. This fact was mentioned in Refs. [7,23].

Thus the intensive scattering is concentrated in the narrow ring of  $\mathbf{k}_{(s)}$  in the plane which is perpendicular to  $\mathbf{n}^0$ . Its center is defined by the director, and the angle radius is equal to  $\vartheta_s$  and can be found as a function of  $\vartheta_i$  from Eq. (3.34). The angle distribution of the intensity along this ring can be found from Eq. (3.1),

$$I_{(s)}^{(i)} = A_{(s)}^{(i)}(\vartheta_i) \frac{\varepsilon_{\parallel} \sin^2 \phi}{\varepsilon_{\perp} + \varepsilon_{\parallel} - 2(\varepsilon_{\perp} \varepsilon_{\parallel})^{1/2} \cos \phi} , \qquad (3.35)$$

where  $\phi$  is the azimuthal angle measured with respect to the direction of  $\mathbf{k}_{(i)_1}$ . Here we use Eqs. (3.4), (3.5), and (3.33). One can see that if  $\phi = 0$  and  $\phi = \pi$ , the intensity of the scatteribution becomes equal to zero. Figure 3 shows the angle distribution of the intensity along the ring. The factors  $A_{(s)}^{(i)}(\vartheta_i)$  defining the dependence of the intensities on the polar angles  $\vartheta_i$  are calculated in Sec. IV. The distribution (3.35) coincides at least qualitatively with experimental data [23,24].

### IV. EXTINCTION COEFFICIENT

The relationship for the extinction coefficient in anisotropic media has the form [4] (cf. Ref. [1])

$$\tau_{(i)} = \frac{(\omega/c)^4}{(4\pi)^2} \frac{e_{\mu}^{(i)} e_{\nu}^{(i)}}{n_{(i)} \cos^2 \delta_{(i)}} \sum_{s=1}^2 \int d\Omega_{(s)} \frac{n_{(s)} e_{\alpha}^{(s)} e_{\beta}^{(s)}}{\cos^2 \delta_{(s)}} \times G_{\mu\alpha\nu\beta}(\mathbf{k}_{(s)} - \mathbf{k}_{(i)}) ,$$
(4.1)

where  $d\Omega_s$  denotes the integration over all orientations of the unit vector  $\mathbf{m}_{(s)} = \mathbf{k}_{(s)}/k_{(s)}$  and  $G_{\mu\alpha\nu\beta}$  is defined by Eq. (2.25).

Since for  $\tau_{(i)}$  calculations one must carry out the summation over the two types of the scattered waves, s=1,2, in Eq. (4.1) as well as over the two types of fluctuation modes, j=1,2, in Eq. (2.25), it is convenient to use the designation  $\tau_{j(i,s)}$  for the contribution to the extinction of the wave of the type (s) which arises from the scattering of the wave of the type (i) on the fluctuation mode  $\delta n_j$ , i.e.,

$$\tau_{(i)} = \sum_{j,s=1,2} \tau_{j(i,s)} . \tag{4.2}$$

Note that  $\tau_{j(1,1)}=0$  by virtue of the geometry G0 from Sec. III.

The direct calculation of the two-dimensional integral (4.1) is rather cumbersome. However, it is possible to obtain more simple approximate relationships for  $\tau_{(i)}$ . Let us regard two different cases—the system far from its critical point, when we can make use of the relationships (2.21) and (2.22), and the system in the nearest vicinity of this point, when it is necessary to use Eqs. (2.15), (2.17), (2.18), and (2.23).

In the first case, we can use the fact that the parameters  $\zeta_D = D/(K_{mm}k_0^2)$  and  $\zeta_B = B/(K_{mm}k_0^2)$  [here  $m=1,2,3, k_0=(\omega/c)^{-1}$ ] satisfy the inequalities  $\zeta_D \gg 1$ and  $\zeta_B >> 1$ . Let us search for the asymptotical approximation of  $\tau_{(i)}$  in Eq. (4.1) under the conditions  $\zeta_D \to \infty$ and  $\zeta_B \rightarrow \infty$ . Making use of Eqs. (2.21), it is easy to see from Eqs. (4.1) and (4.2) that  $\tau_{1(i,s)} \sim \alpha_{(i,s)} \xi_B^{-1} + \beta_{(i,s)} \xi_D^{-1}$ and  $\tau_{2(i,s)} \sim \gamma_{(i,s)} \zeta_D^{-1}$ , where  $\alpha_{(i,s)}$ ,  $\beta_{(i,s)}$ , and  $\gamma_{(i,s)}$  are some constants. One can obtain, however, that the twodimensional integrals defining the values of the coefficients  $\alpha_{(i,s)}$  are diverging for  $q_{\parallel} \rightarrow 0$ ,  $i \neq s$ . (The value of  $\alpha_{(2,2)}$  is finite by virtue of the geometry G5a.) One of the consequences of this divergence is the fact that the main contribution in the integral (4.1) gives the vicinity of  $q_{\parallel} = 0$  only, i.e., the vectors  $\mathbf{k}_{(s)}$  situated in the vicinity of the circles  $O_{(1)}$  and  $O_{(2)}$  (see Fig. 2). Thus the contribution to the intensity in the vicinity of these circles prevails not by its absolute value only, as mentioned in Sec. III, but also by its contribution to the integral.

From the mathematical point of view the equality of  $\alpha_{(i,s)}$  to infinity means that the asymptotic behavior of  $\tau_{1(i,s)}$  is slower than  $\zeta_B^{-1}$ . For its determination it is sufficient to set  $q_{\parallel}=0$  (i.e., to replace  $k_{(s)_{\parallel}}$  by  $k_{(i)_{\parallel}}$ ) in all factors in the integral (4.1), except  $\langle |\delta n_{1q}|^2 \rangle$ , the divergence with which it is connected. After this replacement,  $\langle |\delta n_{1q}|^2 \rangle$  is the only factor integrating over  $k_{(s)_{\parallel}}$ . Since the main contribution in the integration contributes only in the small vicinity of  $k_{(s)_{\parallel}} = k_{(i)_{\parallel}}$ , we can use Eq. (2.22) for it and extend the limits of this integration to  $-\infty$  and  $+\infty$ . Carrying out this procedure is equivalent to making use of  $\langle |\delta n_{1q}|^2 \rangle$  in the form (3.33) in the integral (4.1). This gives the main term of the asymptotic expansion being proportional to  $B^{-1/2}$ . The proper asymptotic form for  $\zeta_D \to \infty$  and  $\zeta_B \to \infty$  is

$$\tau_{(i)} = a \xi_B^{-1/2} + b \xi_B^{-1} + c \xi_D^{-1} + \cdots , \qquad (4.3)$$

where a, b, and c are finite constants. Under the assumption that B and D are values of the same order, it is sufficient to take into account only the terms  $\tau^B_{(i)}$  proportional to  $\xi^{-1/2}_B$ , neglecting the terms proportional to  $\xi^{-1}_B$  and  $\xi^{-1}_D$  in Eq. (4.3).

Equation (3.33) and the relationship

$$\int f(x)\delta(g(x))dx = \sum_{x^*} |g'(x^*)|^{-1} f(x^*), \qquad (4.4)$$

where  $x^*$  are the roots of the equation g(x)=0, allow us to carry out one integration in Eq. (4.1). Parametrizing

the vector  $\mathbf{k}_{(s)}$  on the circle of the rest integration  $O_{(s)}$  as  $k_{(s)\perp}(\cos\phi,\sin\phi)$ ,  $0<\phi\leq 2\pi$ , and taking into account Eqs. (3.4) and (3.5), we have

$$\tau_{(i)}^{B} = \frac{(\omega/c)^{3}}{16\pi} \frac{\varepsilon_{a}^{2} k_{B} T}{(K_{1}B)^{1/2}} \frac{n_{(s)}}{n_{(i)} \cos^{2} \delta_{(i)} \cos^{2} \delta_{(s)} J_{(s)}} \times (\mathbf{e}^{(2)} \cdot \mathbf{n}^{0})^{2} \int_{0}^{2\pi} d\phi (\mathbf{e}^{(1)} \cdot \mathbf{e}_{1})^{2} , \qquad (4.5)$$

where  $s \neq i$  and

$$J_{(s)} = n_{(s)} - \frac{\partial n_{(s)}}{\partial \vartheta_s} \cot \vartheta_s . \tag{4.6}$$

Here we make use of the relationship  $(\mathbf{e}^{(1)} \cdot \mathbf{n}^0) = 0$  and get the values  $n_{(s)}$ ,  $\cos \delta_{(s)}$ ,  $J_{(s)}$ , and  $(\mathbf{e}^{(2)} \cdot \mathbf{n}^0)$  out from the integration, because they do not depend on the azimuthal angle  $\phi$ . It is easy to obtain the values of  $n_{(i)}(\vartheta_i)$ ,  $n_{(s)}(\vartheta_s)$ ,  $\cos \delta_{(i)}(\vartheta_i)$ ,  $\cos \delta_{(s)}(\vartheta_s)$ ,  $(\mathbf{e}^{(2)} \cdot \mathbf{n}^0)$ , and  $J_{(s)}$  from Eqs. (3.2). In particular,

$$J_{(1)} = n_{(1)}(\vartheta_1) = \varepsilon_1^{1/2} ,$$

$$J_{(2)} = n_{(2)}(\vartheta_2) \frac{\varepsilon_1}{\beta(\vartheta_2)} .$$
(4.7)

The integration over  $\phi$  in Eq. (4.5) for both cases i=1, s=2 and i=2, s=1 yields

$$\int_0^{2\pi} d\phi (\mathbf{e}^{(1)} \cdot \mathbf{e}_1)^2 = \frac{\pi \varepsilon_{\parallel}}{\max(\varepsilon_{\perp}, \varepsilon_{\parallel})} . \tag{4.8}$$

Thus we obtain from Eqs. (4.5)-(4.8),

$$\tau_{(i)}^{B}(\vartheta_{i}) = \frac{(\omega/c)^{3}}{16} \frac{k_{B}T}{(K_{1}B)^{1/2}} \frac{\varepsilon_{a}^{2} \varepsilon_{\parallel}}{\varepsilon_{\perp}^{1/2} \max(\varepsilon_{\perp}, \varepsilon_{\parallel})} g_{(i)}(\vartheta_{i}) ,$$
(4.9)

where

$$g_{(1)}(\vartheta_1) = \sin^2 \vartheta_1 ,$$

$$g_{(2)}(\vartheta_2) = \frac{\varepsilon_1^2 \sin^2 \vartheta_2}{\varepsilon_1^{1/2} (\varepsilon_1 \sin^2 \vartheta_2 + \varepsilon_1 \cos^2 \vartheta_2)^{3/2}} .$$
(4.10)

For typical smectic-A liquid crystals, where  $B \sim 2 \times 10^7$  g/(cm sec<sup>2</sup>),  $K_{11} \sim 10^{-6}$  dyn,  $\varepsilon_a \sim 1$ , and  $\varepsilon \sim 3$ , it is easy to obtain the following estimation for the extinction coefficient far from the point of the phase transition:

$$\tau_{(1)} \sim \tau_{(2)} \sim \frac{(\omega/c)^3}{16} \frac{k_B T}{(K_1 B)^{1/2}} \frac{\epsilon_a^2}{\epsilon^{1/2}}$$
  
 $\sim 0.4 \text{ cm}^{-1}$ . (4.11)

It is interesting to pay attention to the fact that the value  $\tau_{(1)}$  is one order less, and  $\tau_{(2)}$  is two orders less, than the corresponding values in nematic liquid crystals [1,3,4]. Meanwhile, the extinction coefficient in smectic-A liquid crystals is several orders more than that in ordinary organic liquids [25].

When  $T \rightarrow T_{NA}$  the coefficients B and D decrease. As long as the parameters  $\zeta_B$  and  $\zeta_D$  are still rather big, there is no reason to take into account for  $\tau_{(i)}$  the correc-

tion term proportional to  $\xi_B^{-1}$  in Eq. (4.3). But this statement is not, in general, valid for the term proportional to  $\xi_D^{-1}$ . The fact is that [26]

$$B \sim |T - T_{NA}|^{\phi}$$
,  
 $D \sim |T - T_{NA}|^{\phi'}$  (4.12)

for  $T \rightarrow T_{NA}$ . There are different theoretical predictions as well as experimental results for the values of  $\phi$  and  $\phi'$  [26], but all theories predict  $\phi' \geq \phi$ . The usual experimental data are [27]  $\phi \approx 0.3$  and  $\phi' \approx 0.5$ . In this case, the increasing of the term  $c\,\xi_D^{-1}$  from Eq. (4.3) is quicker than that of the term  $a\,\xi_B^{-1/2}$  (and quicker than  $b\,\xi_B^{-1}$ ). As a result, it may be that the values  $c\,\xi_D^{-1}$  and  $a\,\xi_B^{-1/2}$  are of the same order for the temperature region, where Eqs. (2.21) and (2.22) are still valid. For the calculation of the contribution of the order of  $\xi_D^{-1}$  into extinction, note that this contribution, in contrast to the term in Eq. (4.3) of the order of  $\xi_B^{-1/2}$ , is defined by all possible directions of the unit vectors  $\mathbf{m}_{(s)}$ , rather than the vicinity of  $q_{\parallel} = 0$ . In this case one can use Eq. (2.21) and, by analogy with Eqs. (3.23) and (3.25), the corresponding contribution into  $G_{\mu\alpha\nu\beta}(\mathbf{q})$  being proportional to  $\xi_D^{-1}$  has the form

$$\frac{\varepsilon_{\alpha}^{2}k_{B}T}{D} \left(\delta_{\mu\nu}n_{\alpha}^{0}n_{\beta}^{0} + \delta_{\mu\beta}n_{\alpha}^{0}n_{\nu}^{0} + \delta_{\alpha\beta}n_{\mu}^{0}n_{\nu}^{0} + \delta_{\alpha\gamma}n_{\mu}^{0}n_{\nu}^{0} + \delta_{\alpha\gamma}n_{\mu}^{0}n_{\beta}^{0} - 4n_{\mu}^{0}n_{\alpha}^{0}n_{\nu}^{0}n_{\beta}^{0}\right).$$
(4.13)

Substituting (4.13) into Eq. (4.1) and integrating over  $\phi$  and  $\vartheta_s$ , we obtain the corresponding contributions into  $\tau_{(i)}$ ,

$$\tau_{(1)}^{D} = \frac{(\omega/c)^4}{6\pi} \frac{k_B T}{D} \varepsilon_a^2 , \qquad (4.14)$$

$$\tau_{(2)}^{D}(\vartheta_{i}) = \frac{(\omega/c)^{4}}{12\pi} \frac{k_{B}T}{D} \varepsilon_{a}^{2}$$

$$\times \frac{\varepsilon_{\perp}(3\varepsilon_{\perp} + \varepsilon_{\parallel})\sin^{2}\vartheta_{i} + 2\varepsilon_{\parallel}^{2}\cos^{2}\vartheta_{i}}{\varepsilon_{\parallel}^{1/2}\beta^{3/2}(\vartheta_{i})}, \qquad (4.15)$$

where  $\beta(\vartheta)$  is defined by Eq. (3.3). Note that  $\tau_{(1)}^D$  does not depend on the direction of the incident ray.

depend on the direction of the incident ray. For  $\vartheta_i = 0$ , the value  $\tau_{(i)}^B$  being calculated from Eq. (4.9) turns out to be equal to zero. In this case, it is necessary to take into consideration the next term of the asymptotic expansion over  $\zeta_B$ . In our case,  $\tau_{(i)}^B(0) \sim \zeta_B^{-1}$  and this contribution is defined by the mode  $\delta n_1$  only. For its calculation, we can make use of Eq. (2.21) taking into account only the term being proportional to  $\zeta_B^{-1}$ . The divergence for  $q_{\parallel} = 0$  cancels because of the angle factor. After integration over  $\phi$  and  $\vartheta_s$  we have

$$\tau_{(i)}^{B}(0) = \frac{(\omega/c)^4}{6\pi} \frac{k_B T}{B} \frac{\varepsilon_a^2 \varepsilon_{\parallel}}{\varepsilon_{\perp}} . \tag{4.16}$$

For the same values of the parameters of the smectic-A liquid crystal as before, one can obtain the following estimation:  $\tau_{(i)}(0) = \tau_{(i)}^B(0) + \tau_{(i)}^D(0) \sim 10^{-2} \text{ cm}^{-1}$ .

Thus the extinction coefficient far from  $T_{NA}$  is

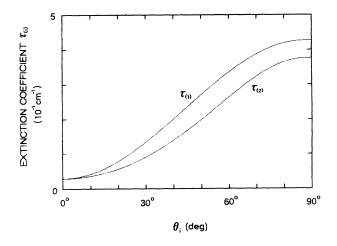
$$\tau_{(i)}(\vartheta_i) = \tau_{(i)}^B(\vartheta_i) + \tau_{(i)}^D(\vartheta_i) , \qquad (4.17)$$

where  $\tau_{(i)}^{B}(\vartheta_i)$  and  $\tau_{(i)}^{D}(\vartheta_i)$  are defined by Eqs. (4.9), (4.14), and (4.15).

Figure 4 illustrates the angle dependence of  $\tau_{(1)}$  and  $\tau_{(2)}$  far from the point of the phase transition. In the case of  $\epsilon_{\parallel} > \epsilon_{1}$ , we have  $\tau_{(1)} > \tau_{(2)}$ , and if  $\epsilon_{\parallel} < \epsilon_{1}$ , the situation is reverse.

Let us now regard the case of the vicinity of  $T_{NA}$ , where  $\xi_B \ll 1$  and  $\xi_D \ll 1$ . In this case for  $\langle |\delta n_{jq}|^2 \rangle$ , we must use Eq. (2.15) regarding  $\Delta_j$  as small parameters. In the limit  $\Delta_j \rightarrow 0$ , the director fluctuations for smectic-A liquid crystals are very similar to that for nematic liquid crystals in the external field  $\mathbf{H}$ . The only difference is that, for a nematic liquid crystal,  $\Delta_1$  and  $\Delta_2$  are equal to each other and do not depend on  $\mathbf{q}$ ,  $\Delta_1 = \Delta_2 = \chi_a H^2$ .

The limit transition  $\zeta_B, \zeta_D \to 0$  is possible for  $\tau_{(i)}(\vartheta_i)$ , if the incident wave is ordinary, i.e., i=1. In this case, the expression for the extinction coefficient coincides completely with that for the ordinary wave in nematic liquid crystals [4],



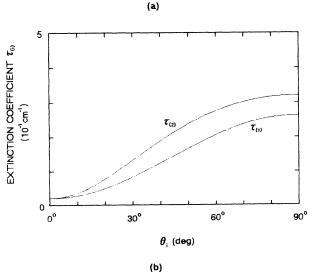


FIG. 4. The angle dependence of the extinction coefficients for the ordinary and extraordinary incident rays. Here we use the following values of the parameters:  $(\omega/c)=10^5$  cm $^{-1}$ , T=300 K,  $B=D=2\times10^7$  g/(cm sec $^2$ ) and  $K_{11}=10^{-6}$  dyn. (a) The case of  $\varepsilon_a>0$ :  $\varepsilon_\parallel=3.5$ ,  $\varepsilon_\perp=2.5$ . (b) The case of  $\varepsilon_a<0$ :  $\varepsilon_\parallel=2.5$ ,  $\varepsilon_\perp=3.5$ .

$$\tau_{(1)}(\vartheta_{i}) = \frac{(\omega/c)^{2}}{8\pi} \frac{k_{B}T}{K_{33}} \frac{\varepsilon_{a}^{2} \varepsilon_{1}}{\varepsilon_{\parallel}^{1/2}} \int_{0}^{\pi} d\vartheta \frac{\sin^{3}\vartheta}{\beta^{3/2}(\vartheta)} \left[ \sin^{2}\vartheta \frac{t_{2} - t_{1}}{\Phi_{1}t_{2} + \Phi_{2}t_{1}} \left[ \frac{t_{1}}{M + At_{1} + \Phi_{1}} + \frac{t_{2}}{M + At_{2} + \Phi_{2}} \right] + \Phi_{2}^{-1} \right], \quad (4.18)$$

where

$$A = \frac{\beta(\vartheta)}{\varepsilon_{\parallel}} \sin^2 \vartheta_i + \sin^2 \vartheta, \quad M = \left[ \frac{\beta^{1/2}(\vartheta)}{\varepsilon_{\parallel}^{1/2}} \cos \vartheta_i - \cos \vartheta \right]^2,$$

$$\Phi_m = \left[ M^2 + 2t_m AM + t_m^2 (A - 2\sin^2 \vartheta)^2 \right]^{1/2}, \quad t_m = \frac{K_{mm}}{K_{33}}, \quad m = 1, 2.$$

$$(4.19)$$

In the case of the extraordinary incident wave (i=2),  $\tau_{(2)}(\vartheta_i)$  diverges logarithmically for  $\zeta_B, \zeta_D \to 0$ . Further, we confine our examination by the logarithmical approximation over  $\zeta_B, \zeta_D$ . Since only the small vectors **q** contribute to the integral, we can use Eq. (2.23) instead of Eq. (2.17) regardless of the validity of inequality (2.20). Then we can obtain for the extinction coefficient (cf. Ref. [4] for nematic liquid crystals)

$$\tau_{(2)}(\vartheta_i) = \frac{(\omega/c)^2}{8\pi} \frac{k_B T}{K_{33}} \frac{\varepsilon_a^2 \varepsilon_1}{\beta^2(\vartheta_i)} \sin^2(2\vartheta_i) \left[ \frac{t_1 \cos \vartheta_i}{F_1(F_1 + t_1 \cos \vartheta_i)} \ln(\xi_D^{-1} + \xi_B^{-1} \sin^{-2}\vartheta_i) + \frac{1}{F_2 + t_2 \cos \vartheta_i} \ln(\xi_D^{-1}) \right], \tag{4.20}$$

where

$$F_{m} = \left[ t_{m}^{2} \cos^{2}\vartheta_{i} + t_{m} \frac{\varepsilon_{\perp}^{2}}{\varepsilon_{\parallel}^{2}} \sin^{2}\vartheta_{i} \right]^{1/2}, \tag{4.21}$$

 $t_m$  is defined by Eqs. (4.19), and m=1,2. Note that in the limit  $T \rightarrow T_{NA}$ , the condition  $\phi' > \phi$  gives that both logarithms in Eq. (4.20) are defined by the parameter  $\zeta_D^{-1}$ . The expression for  $\tau_{(2)}/\ln(T-T_{NA})$  contains the index  $\phi'$  as a factor. We emphasize that, contrary to Eq. (4.9), the Frank moduli  $K_{11}$  and  $K_{33}$  appear in Eqs. (4.18) and (4.20) rather than the effective moduli  $K_1$  and K'', from Eq. (2.15).

It is easy to estimate the order of the values of  $\tau_{(1)}$  and  $\tau_{(2)}$  in Eqs. (4.18) and (4.20) making use of the corresponding values in nematic liquid crystals [4]:  $\tau_{(1)N} \sim 10$  c...<sup>-1</sup>,  $\tau_{(2)N} \sim 50$  cm<sup>-1</sup>. Taking into account the experimental data for the coefficient  $K_{33}$  behavior near  $T_{NA}$ 

1] (the value of  $K_{33}$  for  $T=T_{NA}$  is one order more than its alue in the nematic phase), it may be concluded that the corresponding values for smectic-A liquid crystals are one order less, i.e.,  $\tau_{(1)A} \sim 1$  cm<sup>-1</sup>,  $\tau_{(2)A} \sim 5$  cm<sup>-1</sup>. However, it is necessary to note that measurements of  $\tau_{(2)}$  in this case are rather difficult because of the strong forward scattering. This difficulty was mentioned for nematic liquid crystals [4,28].

## **APPENDIX**

Let us regard those geometrical conditions which give the angle factors  $Q_j(\mathbf{e}^{(i)},\mathbf{e}^{(s)},\mathbf{q})$  turning to zero. Let us examine the cases of different polarizations.

(1) i=1, s=1. It is easy to obtain from Eqs. (3.2) that  $(\mathbf{e}^{(1)} \cdot \mathbf{n}^0) = 0$ . Using this fact one can see that  $Q_1 \equiv 0$  and  $Q_2 \equiv 0$ . It gives us the geometry G0.

(2) i=1, s=2; i=2, s=1. The equality  $Q_1=0$  gives the condition  $(\mathbf{e}_1 \cdot \mathbf{e}^{(1)})=0$  because of the relationships

 $(\mathbf{e}^{(1)} \cdot \mathbf{n}^0) = 0$ ,  $(\mathbf{e}^{(2)} \cdot \mathbf{n}^0) \neq 0$ . [The case of  $(\mathbf{e}^{(2)} \cdot \mathbf{n}^0) = 0$  corresponds to  $\mathbf{k}_{(2)} \| \mathbf{n}^0$  is, in fact, related to case (1) above.] From the equations  $(\mathbf{e}_1 \cdot \mathbf{n}^0) = 0$ ,  $(\mathbf{e}^{(1)} \cdot \mathbf{n}^0) = 0$ ,  $(\mathbf{e}_1 \cdot \mathbf{e}^{(1)}) = 0$  we have  $\mathbf{e}_2 \| \mathbf{e}^{(1)}$ . Because of  $\mathbf{k}_{(1)} \perp \mathbf{e}^{(1)}$ , the vector  $\mathbf{k}_{(1)}$  lays in the plane defined by the vectors  $\mathbf{e}_1$  and  $\mathbf{n}^0$ . The vector  $\mathbf{q} = \mathbf{k}_{(s)} - \mathbf{k}_{(i)}$  also lays in this plane. That is why the vector  $\mathbf{k}_{(2)}$  lays in this plane too. Thus we have that, in the general case, the condition  $Q_1 = 0$  means that the vector  $\mathbf{k}_{(s)}$  lays in the plane defined by the vectors  $\mathbf{n}^0$  and  $\mathbf{k}_{(i)}$  [see Figs. 1(a) and 1(b)]—the geometries G1 and G2. In the degenerate situations for  $\mathbf{k}_{(2)} \| \mathbf{n}^0$ , case (1) is realized, and, if  $\mathbf{k}_{(1)} \| \mathbf{n}^0$ , an additional condition is necessary: the vector  $\mathbf{e}^{(1)}$  must be perpendicular to the plane of the vectors  $\mathbf{k}_{(2)}$  and  $\mathbf{n}^0$ .

Analogously for the condition  $Q_2=0$ , we have  $(\mathbf{e}_2 \cdot \mathbf{e}^{(1)})=0$ . The equations  $(\mathbf{e}_2 \cdot \mathbf{n}^0)=0$ ,  $(\mathbf{e}^{(1)} \cdot \mathbf{n}^0)=0$  gives  $\mathbf{e}_1 \| \mathbf{e}^{(1)}$ . Because of the fact that  $\mathbf{k}_{(1)}$  is perpendicular to  $\mathbf{e}^{(1)}$ , the vector  $\mathbf{k}_{(1)}$  lays in the plane defined by the vectors  $\mathbf{e}_2$  and  $\mathbf{n}^0$ . From the conditions  $(\mathbf{q} \cdot \mathbf{e}_2)=0$ ,  $\mathbf{k}_{(s)}=\mathbf{q}+\mathbf{k}_{(i)}$  we have  $(\mathbf{k}_{(s)}\cdot\mathbf{e}_2)=(\mathbf{k}_{(i)}\cdot\mathbf{e}_2)$ . Note that in this case,  $\mathbf{e}_2$  is perpendicular to the plane defined by the vectors  $\mathbf{e}^{(1)}$  and  $\mathbf{n}^0$ . Thus for  $Q_2=0$ , we have the following situations. If the vectors  $\mathbf{n}^0$  and  $\mathbf{k}_{(1)}$  are fixed, the end of the vector  $\mathbf{k}_{(2)}$  must be situated on the curved line defined by the intersection of the wave surface and the plane which is parallel to the vectors  $\mathbf{n}^0$  and  $\mathbf{e}^{(1)}$ , and passes through the end of the vector  $\mathbf{k}_{(1)}$ . (If  $\mathbf{k}_{(1)} \| \mathbf{n}^0$ , then an additional condition is necessary: the vector  $\mathbf{e}^{(1)}$  must be situated in the plane of the vectors  $\mathbf{k}_{(2)}$  and  $\mathbf{n}^0$ .)

In the case of i=1, s=2, the construction carried out above defines the geometry of scattering, i.e., the arrangement of  $\mathbf{k}_{(s)}$  for which  $Q_2=0$ . In this case, the ends of the seeking vectors  $\mathbf{k}_{(s)}$  are situated on the ellipse [Fig. 1(c)]—the geometry G3. For  $\varepsilon_a > 0$ , such geometries exist for every  $\mathbf{k}_{(i)}$ , and, for  $\varepsilon_a < 0$ , only if the condition  $\varepsilon_1 \sin^2 \vartheta_i \le \varepsilon_1$  is valid.

For further analysis, it is convenient to use the spheri-

cal coordinate system with the polar axis  $z || \mathbf{n}^0$  and the vector  $\mathbf{k}_{(i)}$  lying in the plane of the axes x and z:

$$\mathbf{k}_{(i)} = (\omega/c) n_{(i)}(\vartheta_i) (\sin \vartheta_i, 0, \cos \vartheta_i) ,$$

$$\mathbf{k}_{(s)} = (\omega/c) n_{(s)}(\vartheta_s) (\sin \vartheta_s \cos \varphi_s, \sin \vartheta_s \sin \varphi_s, \cos \vartheta_s) .$$
(A1)

In the case of i = 2, s = 1, the condition of turning of  $Q_2$  to zero described above yields

$$n_{(1)}\sin\vartheta_s = n_{(2)}(\vartheta_i)\sin\vartheta_i\cos\varphi_s . \tag{A2}$$

The line defined by this condition on the wave surface is shown in Figs. 1(d1) and 1(d2)—the geometry G4. For  $\varepsilon_a > 0$ , both cases (d1) and (d2) can be realized, and, for  $\varepsilon_a < 0$ , only case (d1) can.

(3) i=s=2. In the coordinate system described above, the condition  $Q_1=0$  takes the form

$$[n_{(2)}(\vartheta_i)\sin\vartheta_i\tan\vartheta_i - n_{(2)}(\vartheta_s)\sin\vartheta_s\tan\vartheta_s]\cos\varphi_s + [n_{(2)}(\vartheta_i)\cos\vartheta_i - n_{(2)}(\vartheta_s)\cos\vartheta_s]\tan\vartheta_i\tan\vartheta_s = 0.$$
(A3)

The following two cases are possible. First, Eq. (A3) is true identically for every  $\varphi_s$  if  $\vartheta_i = \vartheta_s$  [see the bold circle in Fig. 1(e)]—the geometry G5a. Secondly, for  $\vartheta_i \neq \vartheta_s$ , there is only the value of  $\cos \varphi_s$  defined by Eq. (A3) for every couple  $\vartheta_i, \vartheta_s$ . It is easy to show that this value of  $\cos \varphi_s$  satisfy the condition  $-1 \leq \cos \varphi_s \leq 1$ . That is why there is another curved line on the wave surface besides the circle mentioned above for which  $Q_1 = 0$  [Fig. 1(e)]—the geometry G5b. It passes through the poles of the ellipsoid (i.e., the points  $\vartheta_i = 0$  and  $\vartheta_i = \pi$ ) and has one point of self-intersection.

The condition  $Q_2=0$  in our coordinate system has the form

$$[n_{(2)}(\vartheta_i)\sin\vartheta_i\tan\vartheta_i + n_{(2)}(\vartheta_s)\sin\vartheta_s\tan\vartheta_s]\sin\varphi_s = 0.$$
(A4)

Here two cases are also possible. First, for every  $\vartheta_i, \vartheta_s$ , we have  $\sin \varphi_s = 0$  [Fig. 1(f)]—the geometry G6a. Secondly, the other factor in Eq. (A4) turns to zero, if  $\vartheta_s = \pi - \vartheta_i$  [Fig. 1(f)]—the geometry G6b.

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