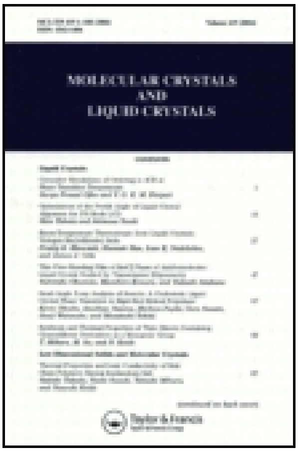

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Surface Effects in Dynamics of Thermal Director Fluctuations

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SURFACE EFFECTS IN DYNAMICS OF THERMAL DIRECTOR FLUCTUATIONS

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Abstract The two-time correlation function of the director fluctuations is calculated for a homeotropically aligned nematic cell in an external magnetic field. The anchoring strength, the splay-bend elastic constant are taken into account. An influence of the surface parameters on light -scattering data is discusses. It is shown, the ratio K_{13}/K_{33} affect the frequency spectrum of the scattered light when the in-plane component of the scattering vector is equal to zero. A geometry of an experiment, that gives the frequency spectrum different from single Lorentzian, is offered.

INTRODUCTION

Physical properties of liquid crystals (LC) are of a great interest from both fundamental and applied points of view. The light-scattering method [1], well developed for infinite media, provides a lot of information on LC systems. In order to apply this method to bounded LC one must know the correlation function of thermal director fluctuations. In view of the fact that according to the fluctuation dissipation theorem [2] this function also describes a response of a system to an external action, it is also useful in applications. The aim of the present work is to find out how surface parameters such as the anchoring strength and the surface like constants K_{13}, K_{24} affect dynamics of the fluctuations in the bulk and to calculate the two-time correlation function in the case of homeotropically aligned cell. K_{13} and K_{24} are under extensive experimental [3, 4] as well as theoretical study [5, 6]. The most studies are based on the fact, that an equilibrium director field depends on these coefficients if a size of a nematic LC is small. Apart from that dependence, the surface contribution to the free energy also changes random thermal deviations of the director from an equilibrium field in the bulk. It turns out [7], that K_{24} is irrelevant in the case of homeotropical nematic cell and the correlation function of the director fluctuations, which can be studied experimentally by means of light scattering, depends on some combination of anchoring strength W_0, Frank elastic constant K_{33} and K_{13}. Additional information on the surface parameters can be extracted from the frequency distribution of the scattered light. It is usually assumed [8, 9] the frequency spectrum to be a single Lorentzian. Though in a case of bounded LC the fluctuations with various wave vectors [10] and, hence, various decay times are responsible for the scattering on one angle a deviation from a single Lorentzian has not been noticed experimentally. It turns out the deviation can be found if the in-plane component of the scattering vector is equal to zero.

This paper is arranged as follows: in the next section an expression for the two-time correlation function is derived with taking into account the applied magnetic field, which is assumed to be less than critical one, and the surface contribution to the free energy. Then, the dependance of the frequency distribution of the scattered light upon the magnetic field and the ratio K_{13}/K_{33} is analyzed. Corresponding graphs are presented. In the last section the results are summarized and discussed.

CORRELATION FUNCTION OF DIRECTOR FLUCTUATIONS

Let the homeotropically aligned nematic cell be confined between the two plates situated at $z = \pm L/2$ in the Cortesian coordinate system. The starting point of the description is the free energy expression including the surface like elastic terms [6]

$$F = \frac{1}{2}\int d^3r[K_{11}(div\mathbf{n})^2 + K_{22}(\mathbf{n}\cdot curl\mathbf{n})^2 + K_{33}(\mathbf{n}\times curl\mathbf{n})^2 \qquad (1)$$
$$-2(K_{22}+K_{24})div(\mathbf{n}div\mathbf{n}+\mathbf{n}\times curl\mathbf{n}) + 2K_{13}div(\mathbf{n}div\mathbf{n})^2]$$
$$+\frac{W_0}{2}\int d^2r[\mathbf{n}_\perp^2(\mathbf{r}_\perp, L/2) + \mathbf{n}_\perp^2(\mathbf{r}_\perp, -L/2)],$$

where $\mathbf{n}_\perp$ and $\mathbf{r}_\perp$ are the in-plane components of $\mathbf{n}$ and $\mathbf{r}$. The last term proposed by Rapini and Papoular [11] accounts for the surface anchoring. Let us consider only small thermal deviations $\delta\mathbf{n}(\mathbf{r}) = \mathbf{n}(\mathbf{r}) - \mathbf{n}^0$ of the director from the normal to the interfaces $\mathbf{n}^0$. Since $\mathbf{n}$ is unit vector, only two components, namely δn_x and δn_y, are independent. If the sample is in the magnetic field $\mathbf{H} = (H, 0, 0)$, the additional therm must be included into the free energy [12]. Full expression for a free energy deviation from equilibrium magnitude can be written as a sum of bulk and surface contributions

$$\Delta F = \frac{1}{2}\int d^2\mathbf{r}_\perp \int_{-L/2}^{L/2} dz[K_{11}(div\delta\mathbf{n}_\perp)^2 + K_{22}(\partial_x\delta n_y - \partial_y\delta n_x)^2 - K_{33}\delta\mathbf{n}_\perp \cdot \partial_z^2\delta\mathbf{n}_\perp - \chi_a H^2 n_x^2] + \frac{1}{2}\int d^2\mathbf{r}_\perp\{W_0[\delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, L/2) + \delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, -L/2)] + \frac{1}{2}(K_{33} - 2K_{13})\partial_z \cdot [\delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, L/2) - \delta\mathbf{n}_\perp^2(\mathbf{r}_\perp, -L/2)]\} \tag{2}$$

In order to study the dynamics of the fluctuations one has to consider corresponding set of hydrodynamic equations [13, 14, 15] accounting for coupling of the director field with the flow. To distinguish an effect of the surface parameters we neglect coupling and restrict ourselves to the simplest model assuming the dissipative function to be of the form

$$R = \frac{1}{2}\gamma_1 \int d^3r(\frac{\partial\delta\mathbf{n}}{\partial t})^2, \tag{3}$$

where γ_1 is a constant with the dimension of a viscosity. In order to take into account dissipation at the boundaries, the surface viscosity coefficient γ_{surf} has been introduced [17, 16]

$$R_{surf} = \frac{1}{2}\gamma_{surf}\int d^2r_\perp[(\partial_t\delta\mathbf{n}(\mathbf{r}_\perp, z = L/2, t))^2 + (\partial_t\delta\mathbf{n}(\mathbf{r}_\perp, z = -L/2, t))^2] \tag{4}$$

According to 2, 3 the director field in the bulk must satisfy the equation

$$\gamma_1\frac{\partial\delta\mathbf{n}}{\partial t} = -\Lambda\delta\mathbf{n}, \tag{5}$$

Λ being a self-adjoint matrix with elements

$$\Lambda_{11} = -(K_{11}\partial_x^2 + K_{22}\partial_y^2 + \chi_a H^2)$$

$$\Lambda_{12} = \Lambda_{21} = (K_{22} - K_{11})\partial_x\partial_y$$

$$\Lambda_{22} = -(K_{22}\partial_x^2 + K_{11}\partial_y^2 + K_{33}\partial_z^2)$$

According to [17] the supplementary boundary conditions for the case $K_{13} = 0$ are given by

$$\gamma_{surf}\frac{\partial\delta\mathbf{n}(\mathbf{r}_\perp, \pm L/2, t)}{\partial t} = -(\frac{W_0}{K_{33}} \pm \partial_z)\delta\mathbf{n}(\mathbf{r}_\perp, \pm L/2, t), \tag{6}$$

and γ_{surf} should be taken into account only if high a frequency behavior of the director field near the interfaces is of an interest. Keeping in mind that a contribution of the surface layers to the light -scattering intensity is much less than that of the bulk, we assume γ_{surf} to be zero. This assumption also implies that characteristic time of director alignment at the surfaces is much less than the time of alignment in the bulk, so in the bulk alignment time -scale one can apply equilibrium boundary conditions. It has been shown [6, 7] that in the case of homeotropical cell K_{13} can be taken into account by replacing W_0 with $W_0K_{33}/(K_{33} - 2K_{13})$, hence, for the boundary conditions we have

$$(w \pm \partial_z)\delta\mathbf{n}(\mathbf{r}_\perp, \pm L/2, t) = 0, \tag{7}$$

where

$$w = \frac{W_0}{K_{33} - 2K_{13}}. \tag{8}$$

It is worth mention that the consideration is valid only for positive w, because negative w corresponds to non-stable homeotropical cell. The correlation function of thermal director fluctuations

$$G_{mn}(\mathbf{r}_\perp - \mathbf{r}'_\perp, z, z'; t) = \langle \delta n_m(\mathbf{r}, t)\delta n_n(\mathbf{r}', 0)\rangle. \tag{9}$$

must satisfy the following equation

$$\gamma_1 \partial_t G_{mn}(\mathbf{r}_\perp, z, z'; t) = -\Lambda_{mk} G_{kn}(\mathbf{r}_\perp, z, z'; t) \tag{10}$$

with boundary conditions

$$(w \pm \partial_z) G_{mn}(\mathbf{r}_\perp, \pm L/2, z', t) = 0. \tag{11}$$

At $t = 0$ this function coincides with the one-time correlation function, which can be derived from [7]

$$\Lambda_{mk} G_{kn}(\mathbf{r}_\perp - \mathbf{r}'_\perp, z, z', 0) = k_B T \delta_{mn} \delta(\mathbf{r} - \mathbf{r}'). \tag{12}$$

It is convenient to take Fourier transform with respect to $\mathbf{r}_\perp$ and t:

$$G^+_{mn}(\mathbf{q}_\perp, z, z'; \omega) = \int d^2 r_\perp \int_0^\infty dt \exp[-\imath \mathbf{q}_\perp \cdot \mathbf{r}_\perp + \imath\omega t] G_{mn}(\mathbf{r}_\perp, z, z'; t).$$

Rotation around the z-axis on angle ψ [8]

$$\tan(2\psi) = \frac{(K_{11} - K_{22})q_\perp^2 \sin(2\phi)}{(K_{11} - K_{22})q_\perp^2 \cos(2\phi) - \chi_a H^2},$$

where ϕ is an angle between the two-dimensional wave vector $\mathbf{q}_\perp = q_\perp(\cos\phi, \sin\phi, 0)$ and $\mathbf{H}$ gives the diagonal system of linear equations:

$$\tilde{G}_{12}^+ = \tilde{G}_{21}^+ = 0,$$

$$[\lambda_j(\omega) - \partial_z^2]\tilde{G}_{jj}^+(\mathbf{q}_\perp, z, z'; \omega) = \frac{\gamma_1}{K_{33}}\tilde{G}_{jj}(\mathbf{q}_\perp, z, z'; t = 0), \tag{13}$$

where

$$\lambda_j(\omega) = \mathcal{K}_j(\mathbf{q}_\perp) - \frac{\imath\omega\gamma_1}{K_{33}} \tag{14}$$

$$\mathcal{K}_j(\mathbf{q}_\perp) = \frac{1}{2K_{33}}[(K_{11} + K_{22})q_\perp^2 - \chi_\perp^2 H^2$$
$$\pm\sqrt{[(K_{11} - K_{22})\mathbf{q}_\perp^2 - \chi_\perp^2 H^2]^2 + 2\chi_a H^2 (K_{11} - K_{22})\mathbf{q}_\perp^2(1 - \cos(2\phi))}].$$

The one-time correlation function has been obtained in [7] and can be written in the form

$$\tilde{G}_{jj}(\mathbf{q}_\perp, z, z', 0) = \frac{k_B T}{K_{33}}\mathcal{R}_{\lambda_j(0)}(z, z'), \tag{15}$$

where $\mathcal{R}$ is the resolving function, corresponding to the boundary conditions 11, of the operator $-\partial_z^2$ [18]

$$\mathcal{R}_\lambda(z, z') = \frac{1}{2\sqrt{\lambda}\Delta}[(\lambda - w^2)\cosh(\sqrt{\lambda}(z + z')) \tag{16}$$
$$+((\lambda + w^2)\cosh(\sqrt{\lambda}L) + 2\sqrt{\lambda}w\sinh(\sqrt{\lambda}L))\cosh(\sqrt{\lambda}(z - z'))$$
$$-\Delta\sinh(\sqrt{\lambda}|z - z'|],$$

$$\Delta = (\lambda + w^2)\sinh(\sqrt{\lambda}L) + 2w\sqrt{\lambda}\cosh(\sqrt{\lambda}L) \tag{17}$$

The spectrum of the operator is discrete and essentially negative. The eigenvalues are defined by equation $\Delta = 0$. The real part of $\lambda_j(\omega)$ depends on H and is larger than maximal eigenvalue if H is less than the critical magnitude H_c of the Freèderickzs transition. For example, in the case of strong anchoring ($W_0 = \infty$) the maximal eigenvalue is equal to $-(\frac{\pi}{L})^2$ and gives $H_{c\infty} = \frac{\pi}{L}\sqrt{\frac{K_{33}}{\chi_a}}$. This function also describes a response of the system to an external action and the response function differs from $\mathcal{R}_{\lambda_j(\omega)}(z,z')$ only by some coefficient [2].

The first formula for resolving function of self–adjoint operator yields

$$\tilde{G}^{+}_{jj}(\mathbf{q}_\perp, z, z'; \omega) = \frac{\gamma_1 k_B T}{K^2_{33}\cdot[\lambda_j(\omega) - \lambda_j(0)]}[\mathcal{R}_{\lambda_j(0)}(z,z') - \mathcal{R}_{\lambda_j(\omega)}(z,z')]. \qquad (18)$$

For the full Fourier product one gets

$$\tilde{G}_{jj}(\mathbf{q}_\perp, z, z'; \omega) = \frac{2k_B T}{K_{33}\omega} Im\mathcal{R}_{\lambda_j(\omega)}(z,z'). \qquad (19)$$

Needed correlation function of the director fluctuations is given by

$$G_{ij}(\mathbf{r}_\perp, z, z'; \omega) = \frac{1}{(2\pi)^2}\int d^2\, q_\perp \exp(\imath \mathbf{q}_\perp \cdot \mathbf{r}_\perp) G_{ij}(\mathbf{q}_\perp, z, z'; \omega), \qquad (20)$$

where

$$G_{11}(\mathbf{q}_\perp, z, z'; \omega) = \cos^2(\psi)\tilde{G}_{11} + \sin^2(\psi)\tilde{G}_{22}$$

$$G_{22}(\mathbf{q}_\perp, z, z'; \omega) = \cos^2(\psi)\tilde{G}_{22} + \sin^2(\psi)\tilde{G}_{11}$$

$$G_{12}(\mathbf{q}_\perp, z, z'; \omega) = G_{21}(\mathbf{q}_\perp, z, z'; \omega) = \sin(\psi)\cos(\psi)(\tilde{G}_{11} - \tilde{G}_{22}).$$

LIGHT-SCATTRING INTENSITY

To analyze an influence of the boundary conditions on a light scattering process let us consider the director associated fluctuations $\delta\varepsilon_{\alpha\beta}(\mathbf{r},t)$ of the dielectric tensor

$$\delta\varepsilon_{\alpha\beta}(\mathbf{r},t) = \varepsilon_a(n^0_\alpha \delta n_\beta(\mathbf{r},t) + n^0_\beta \delta n_\alpha(\mathbf{r},t)), \qquad (21)$$

where ε_a is the optical anisotropy of the nematic LC. The intensity of scattered light in the Born approximation is given by [1]

$$I(bfe^{(i)}, \mathbf{e}^{(s)}, \omega) = C\int_{V_s} d^3\mathbf{r} d^3\mathbf{r}' e^{(s)}_\alpha e^{(s)}_\beta T_{\alpha\gamma}(\mathbf{r}, \mathbf{r}_1; \omega + \omega_0) \qquad (22)$$
$$\times T^*_{\beta\delta}(\mathbf{r}, \mathbf{r}_2; \omega + \omega_0)\mathcal{G}_{\gamma\mu\delta\nu}(\mathbf{r}_1, \mathbf{r}_2; \omega) E^{(i)}_0(\mathbf{r}_1) E^{(i)}_0(\mathbf{r}_2) e^{(i)}_\mu e^{(i)}_\nu,$$

where V_s is the illuminated volume, C is a constant connected with a definition of the intensity, $\mathbf{e}^{(i)}$ and $\mathbf{e}^{(s)}$ are unit polarization vectors of incident and scattered light, ω_0

and $E_0^{(i)}$ are the frequency and the amplitude of the incident light, ω is a frequency shift. $\mathcal{G}_{\gamma\mu\delta\nu}(\mathbf{r}_1, \mathbf{r}_2; \omega)$ is dielectric tensor - dielectric tensor correlation function

$$\mathcal{G}_{\gamma\mu\delta\nu}(\mathbf{r}_1, \mathbf{r}_2; \omega) = \int_{-\infty}^{\infty} dt \exp(-\imath\omega t)\langle\delta\varepsilon_{\gamma\mu}(\mathbf{r}_1, t)\delta\varepsilon_{\delta\nu}(\mathbf{r}_2, 0)\rangle$$

The Green's function $T_{\alpha\gamma}$ that takes into account optical anisotropy of the medium, multiple reflection at the interfaces has been presented in [10]. To simplify the consideration we assume here the Green's function to be one for a far zone in an infinite medium with permittivity $\varepsilon = (\varepsilon_\perp + \varepsilon_{||})/2$. Assuming the incident and scattered light to be plane waves with the wave vectors $\mathbf{k}^{(i)}$, $\mathbf{k}^{(s)}$ and carrying out the integration with respect to the in-plane coordinates in refe19 one gets

$$I(\mathbf{e}^{(i)}, \mathbf{e}^{(s)}, \omega) \sim \int_{-L/2}^{L/2} dz \int_{-L/2}^{L/2} dz' \exp(-\imath q_z(z - z')) \times e_\alpha^{(s)} e_\beta^{(s)} \mathcal{G}_{\alpha\mu\beta\nu}(\mathbf{q}_\perp, z, z'; \omega) e_\mu^{(i)} e_\nu^{(i)}, \quad (23)$$

where $\mathbf{q} = \mathbf{k}^{(s)} - \mathbf{k}^{(i)}$ is the scattering vector with components $(\mathbf{q}_\perp, q_z)$, the function

$$\mathcal{G}_{\alpha\mu\beta\nu}(\mathbf{q}_\perp, z, z'; \omega) = \int d^2(r_{1\perp} - r_{2\perp})\mathcal{G}_{\alpha\mu\beta\nu}(\mathbf{r}_1, \mathbf{r}_2; \omega)$$

can be easy expressed in terms of the director correlation function in $(\mathbf{q}_\perp, z, z'; \omega)$–representation 19. It seen from the expression for the resolving function, that the boundary conditions affect final results when a real part of $\lambda_j(\omega)$ is small. It means, that to distinguish effects of the boundaries one should consider geometries with which $\mathbf{q}_\perp = 0$. Keeping in mind that ordinary waves are not scattered into ordinary ones by the director fluctuations and there is no scattering of extraordinary—extraordinary type when $\mathbf{q}_\perp = 0$, let us consider scattering of extraordinary waves into ordinary ones. Setting the polarization and wave vectors of the incident and scattered light

$$\mathbf{k}^{(i)} = k_0(0, \sin\theta_i, \cos\theta_i); \mathbf{k}^{(s)} = k_0(0, \sin\theta_s, \cos\theta_s);$$

$$\mathbf{e}^{(i)} = (0, -\cos\theta_i, \sin\theta_i); \mathbf{e}^{(s)} = (1, 0, 0)$$

and carrying out integration in 23 with respect to z and z' yield

$$I \sim \sin^2(\theta_i)\frac{1}{\omega} Im\{(w + g)[2q_z^2 - 2wg + L(w + g)(q_z^2 + g^2)] \quad (24)$$

$$+4g\exp(-gL)[(w^2-q_z^2)\cos(q_zL)-2wq_z\sin(q_zL)]$$
$$+(g-w)\exp(-2gL)[2wg+2q_z^2+L(w-g)(g^2+q_z^2)\}$$
$$\times\{(g^2+q_z^2)^2[(w+g)^2-\exp(-2gL)(w-g)^2]\}^{-1},$$

where

$$q_\perp = k_0|\sin\theta_s - \sin\theta_i|;\, q_z = k_0(\cos\theta_s - \cos\theta_i);$$
$$g = \sqrt{(K_{11}\mathbf{q}_\perp^2 - \chi_a H^2 - \imath\omega\gamma)/K_{33}}$$

A real part of the root must be positive. The character $\sim$ in equation 24 means that some coefficient independent of the frequency and the surface constants has been dropped for a brevity. This expression allows one to calculate the scattering intensity and compare it to data obtained in an inelastic light scattering experiment. Fig. 1 shows the inverse intensity I^{-1} as function of ω^2 for various ratios K_{13}/K_{33}. If the frequency distribution were of a single Lorentzian type, we would get strait lines. One can find the deviation from the linear low at the range of small frequencies, hence, the dependence of the intensity upon time is not exponential one. Characteristic frequency at which the linear low beaks and the shape of the lines depend on the thickness of the cell and the combination $W_0/(K_{33}-2K_{13})$. This allows one to study experimentally surface parameters of the nematic LC analyzing spectral properties of the scattered light by means of correlation spectroscopy. The deviation from strait lines disappears when the magnetic field is applied. It happens because the mode with maximal eigenvalue, which is responsible for the Freèderickzs transition, dominates and leads to a single Lorentzian. Fig. 2 shows linear behavior for $H/H_c = 0.5$. The slopes of the lines differ from each other because the intensity also depends on an effective anchoring strength w, which is a function of K_{13}.

DISCUSSION

While the dynamics in a case of nematic cell is studied, the director is usually assumed to be fixed at the boundaries (strong anchoring) [15, 19]. An attempt to take into account finite anchoring strength and the splay-bend elastic constant K_{13} is made here. As for K_{24}, it is irrelevant in the geometry considered. The

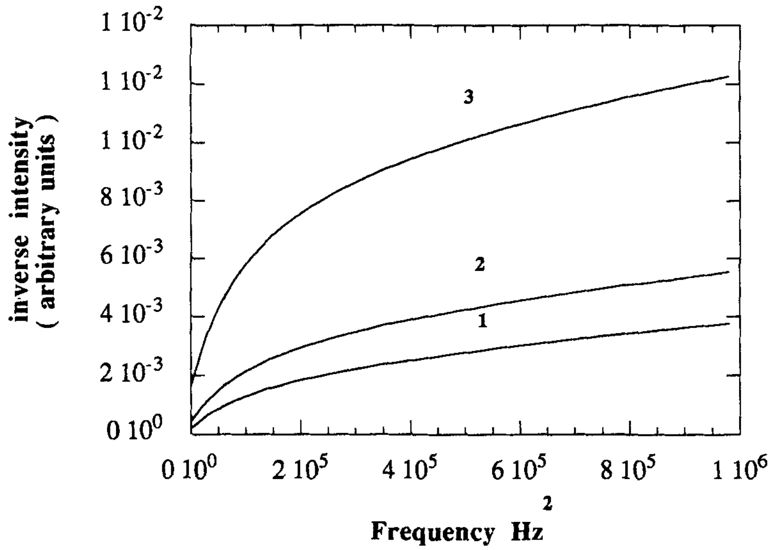

FIGURE 1: Inverse intensity I^{-1} of extraordinary—ordinary type light scattering vs. ω^2. The calculation was carried out on the basis of 24 for $K_{11}/K_{33} = K_{22}/K_{33} = 0.7$, $K_{33} = 1. \times 10^{-6} dyne$, $W_0 = 1. \times 10^{-2} \frac{erg}{cm^2}$, $L = 1. \times 10^{-3} cm$, $H/H_c = 0.$, $\gamma_1 = 1. \times 10^{-2}\ g/c \cdot cm$, $\theta_i = \pi/4$, $\theta_s = 3\pi/4$ and $k_0 = 1. \times 10^5\ cm^{-1}$, (1) $K_{13}/K_{33} = -0.25$, (2) $K_{13}/K_{33} = 0.$, (3) $K_{13}/K_{33} = 0.25$.

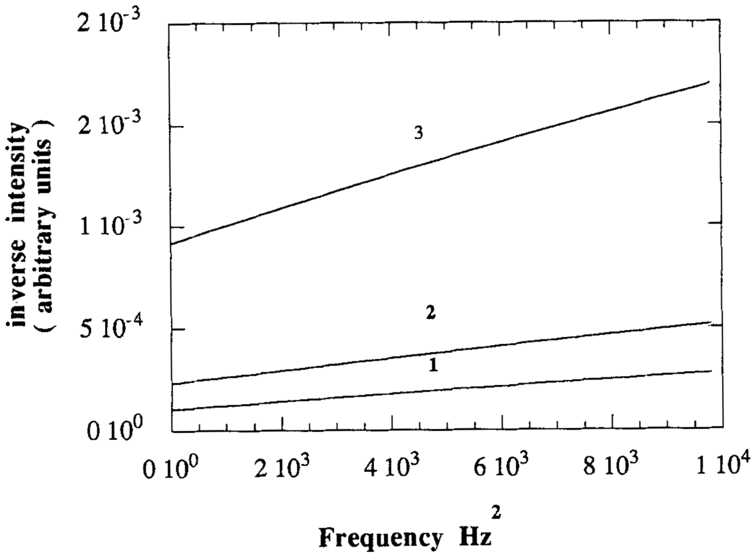

FIGURE 2: Inverse intensity I^{-1} of extraordinary—ordinary type light scattering vs. ω^2 for $H/H_c = 0.5$, (1) $K_{13}/K_{33} = -0.25$, (2) $K_{13}/K_{33} = 0.$, (3) $K_{13}/K_{33} = 0.25$. The other parameters are the same as for Fig. 1

designed model is based on the statement made in [17] that the director orients at the interfaces much faster than in the bulk. This assumption implies surface viscosity γ_{surf} to be zero. To the author's knowledge, there is no experimental determination of γ_{surf} up to now, so it is impossible to verify the statement using existing data. The other important point, that must be mentioned, concerns hydrodynamic boundary conditions. These conditions in the simplest situation of ordinary fluid in a presence of solid walls are under study now [20, 21]. The case of LC where the flow interacts with the director is much more complicated and requires special analyzes. Another problem concerning the flow is that the director field can not be expressed in terms of stationary modes with corresponding exponential decays if the director coupls with the flow [14]. It happens because the director field and the velocity must satisfy different boundary conditions. All the problems lay out of the frame of the paper and will be the subject of separate consideration. The offered model does not account for these effects and the reason for this is that the flow only slightly changes light-scattering data.

The results obtained can be summarized as follows. An explicit formula for the two-time correlation function of the director fluctuations for homeotropical nematic cell with an applied external magnetic field is presented. Useful in applications the response function can be easy expressed in terms of obtained resolving function 16. The expressions have compact form, take automatically into account all natural modes and allow one to analyze the dependence on the anchoring strength and the splay-bend elastic coefficient. The dependence can also be studied by means of correlation spectroscopy. The frequency distribution of light scattered by the director fluctuations is sensitive to the combination $W_0/(K_{33} - 2K_{13})$, which can be obtained by fitting experimental data. The distribution in most geometries is of a single Lorentzian type, but if the in-plane component of the scattering vector is equal to zero, considerable deviation from this low can be found in the range of small frequencies. The deviation is easy to study experimentally in a case of scattering of extraordinary waves into ordinary waves. The deviation disappears with increasing the magnetic field, because the mode, which is responsible for the

Freedericksz transition, becomes dominating.

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