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Arcadi N. Shalaginov^a

^a Marine Technical University of St. Petersburg, Department of
Physics, 101 Leninski pr., St. Petersburg, 198262, Russia.

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FINITE-SIZE EFFECTS IN FLUCTUATIONS AND LIGHT SCATTERING IN LIQUID CRYSTALS (LC)

ARCADI N. SHALAGINOV

Marine Technical University of St.Petersburg,
Department of Physics, 101 Leninski pr.,
St.Petersburg, 198262, Russia.

Abstract. The angular distribution of the intensity of the light scattered by director fluctuations in thin LC layers is theoretically considered. The analysis is carried out for a freely suspended smectic-A film and a homeotropically aligned nematic LC cell. It is shown that surface tension, anchoring strength and the splay-bend surface elastic constant (in the case of nematic LC) affect the angular distribution of the scattered light intensity. The correlation function of the fluctuations is written down in a closed explicit form rather than infinite series of natural modes.

It is known that light scattering in LC is much stronger than in liquids due to local optical anisotropy and well-developed fluctuations of the director. An advanced theory¹ of such a process allows one to investigate LC by means of light-scattering experiments. LC layers, that are of interest from the practical as well as fundamental point of view, could be investigated by the same method if two difficulties were overcome. First, there is either an interaction between the confining surfaces and the director or the surface tension, that gives rise to a surface contribution to the free energy. Saddle-splay (K_{24}) and splay-bend (K_{13}) elastic constants of nematic LC also lead to surface contributions^{2,3}. These affect the director fluctuations in the layers. Second, incident and scattered light multiple reflects at the interfaces between optically anisotropic and isotropic media. It is the aim of the paper to fill the gaps in the theory for both the homeotropically aligned cell and the thin smectic-A film.

Let us consider a homeotropically aligned nematic layer confined between two planes situated at $z=\pm L/2$ in a Cartesian coordinate system. The free energy is given by

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

where \vec{n}_{\perp} and \vec{r}_{\perp} are the in-plane components of \vec{n} and \vec{r} . The last term, proposed by Rapini-Papoular⁴, accounts for the surface anchoring. We assume the equilibrium director \vec{n}^0 to be normal to the surfaces. We shall consider only small fluctuations of the director $\delta \vec{n}(\vec{r}) = \vec{n}(\vec{r}) - \vec{n}^0$. Since \vec{n} is a unit vector, only two components, namely δn_x and δn_y , are independent. The correlation function $G_{\alpha\beta}(\vec{r}_{\perp} - \vec{r}'_{\perp}, z, z') = \langle \delta n_{\alpha}(\vec{r}) \delta n_{\beta}(\vec{r}') \rangle$ in (\vec{r}, z, z') - representation, where \vec{r} is an in-plane wave vector, can be expressed in the explicit form^{4,5}. Without any restriction we can assume \vec{r} to be normal to the y-axis and get⁵

$$G_{11}(\kappa, z, z') = \frac{k_B T}{2\alpha_1 K_{33} \Delta_1} \left\{ (\alpha_1^2 - w^2) \cosh(\alpha_1(z+z')) + \left[(\alpha_1^2 + w^2) \cosh(\alpha_1 L) + 2\alpha_1 w \sinh(\alpha_1 L) \right] \cosh(\alpha_1(z-z')) - \Delta_1 \sinh(\alpha_1 |z-z'|) \right\}, \quad (2)$$

where

$$\Delta_1 = (\alpha_1^2 + w^2) \sinh(\alpha_1 L) + 2w\alpha_1 \cosh(\alpha_1 L), \\ w = W_0 / (K_{33} - K_{13}), \quad \alpha_1 = (K_{11} / K_{33})^{1/2} \kappa \quad (3)$$

$i=1$ and $i=2$ correspond to δn_x and δn_y , respectively.

To analyze light-scattering process one should consider the director-associated fluctuations $\delta \epsilon_{\alpha\beta}(\vec{r})$ of the dielectric tensor

$$\delta \epsilon_{\alpha\beta}(\vec{r}) = \epsilon_a (n_{\alpha}^0 \delta n_{\beta}(\vec{r}) + n_{\beta}^0 \delta n_{\alpha}(\vec{r})), \quad (4)$$

where $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$, ε_{\parallel} and ε_{\perp} being the permittivities along and transverse to \vec{n} . The intensity and polarization properties of the scattered light can be expressed in terms of the function $\langle E_{\alpha}(\vec{r}) E_{\beta}^*(\vec{r}) \rangle$, which in the Born approximation is given by

$$\langle E_{\alpha}(\vec{r}) E_{\beta}^*(\vec{r}) \rangle = \frac{\omega^4}{c^4} \int d^3 r' d^3 r'' T_{\alpha\gamma}(\vec{r}, \vec{r}') T_{\beta\lambda}^*(\vec{r}, \vec{r}'') \langle \delta\varepsilon_{\gamma\mu}(\vec{r}') \delta\varepsilon_{\lambda\nu}(\vec{r}'') \rangle | E_{\mu}^0 E_{\nu}^0 \exp(i \vec{k}_1(\vec{r}' - \vec{r}'')) \rangle. \quad (5)$$

Here E^0 and $\vec{k}_{(1)}$ are the amplitude and wave vector of the incident light, $T_{\alpha\beta}(\vec{r}, \vec{r}')$ is the Green's function for an optically anisotropic medium with an account for the boundaries. The detailed analysis of the multi-reflection and the optical anisotropy effects in the case of a homeotropically aligned liquid crystal cell is presented elsewhere. The integration in (5) can be carried out analytically. The curves in Figure 1 show the angular dependence of the intensity of extraordinary scattered waves as a function of the angle between the normal of the homeotropically aligned cell and the scattering direction.

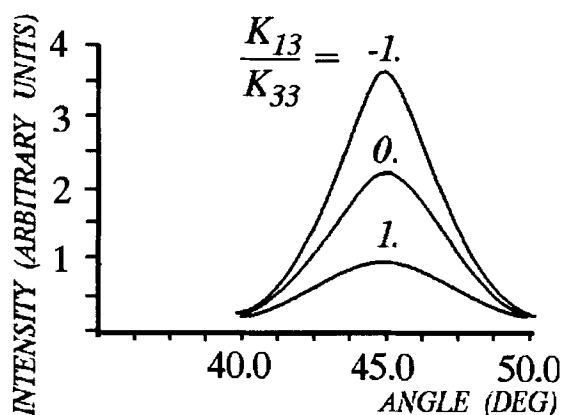


FIGURE 1 The light scattering intensity for the case of homeotropically aligned nematic cell.

In order to distinguish the influence of the surface parameters W_0 and K_{13} the interface optical effects were

omitted. The calculation was carried out for an oblique incident extraordinary wave (the tilt angle is equal to $\pi/4$), $W_0 = 5.0 \times 10^{-3} \text{ dyn/cm}$, $K_{11}/K_{33} = 0.7$, $K_{33} = 10^{-6} \text{ dyn}$, $k = 10^5 \text{ cm}^{-1}$, $L = 10^{-3} \text{ cm}$.

A smectic-A freely suspended film with thickness L can be treated by a similar manner⁷. The parameter, which describes a deformation of a smectic liquid crystal is the layer displacement $u(\vec{r})$ along the z axis. We assume the director fluctuation $\delta \vec{n}(\vec{r})$ is equal to $-\nabla_{\perp} u(\vec{r})$, where ∇_{\perp} is the gradient in the x, y variables. The expression for the free energy F a sum of the bulk and the surface terms

$$F = \frac{1}{2} \left\{ \int d^3 r \left[B (\partial_z u(\vec{r}_{\perp}, z))^2 + K (\Delta_{\perp} u(\vec{r}_{\perp}, z))^2 \right] + \int d^2 r_{\perp} \gamma (\nabla_{\perp} u(\vec{r}))^2 \right\}. \quad (6)$$

Here B is the smectic elastic constant associated with the layer compressions, K is the elastic constant associated with the layer undulations and Δ_{\perp} is the Laplacian in the x, y variables. The correlation function $G(\vec{k}, z, z') = \langle u(\vec{k}, z) u(-\vec{k}, z') \rangle$ is given by expressions⁷ (2), (3), where one must set $\alpha_1 = \alpha_2 = g = \sqrt{K/B} k^2$, $w = \gamma k^2 / B$, and B instead of K_{33} . The relation between $\delta \varepsilon_{\alpha\beta}$ and $\delta \vec{n}$ is the same as for nematic LC.

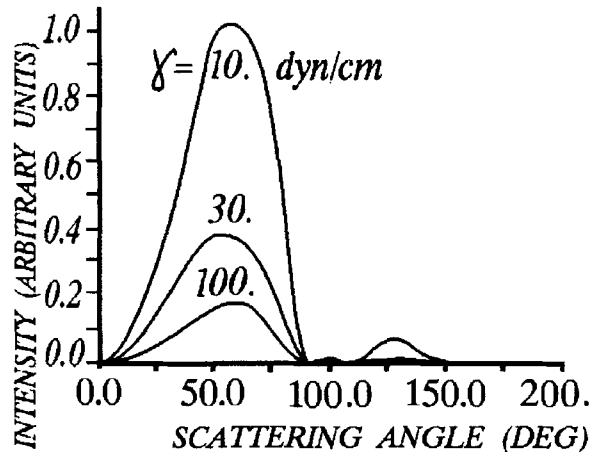


FIGURE 2 The intensity of the scattered light vs the scattering angle for the case of the normal incidence on the freely suspended film; $\varepsilon_{\parallel} = 3.3$, $\varepsilon_{\perp} = 3.0$, $k_0 = 10^5 \text{ cm}^{-1}$, $L = 10^{-4} \text{ cm}$, $B = 2.5 \times 10^7 \text{ dyn/cm}^2$, $K = 10^{-6} \text{ dyn}$.

The curves in Figure 2 show the angular dependence taking into account all the optical interface effects, namely optical anisotropy and multiple-reflection effects for the incident and scattered beams. In order to show the dependence on surface tension γ three values have been chosen. One can see that there are peaks in the variation and the magnitudes of the peaks depend strongly on the surface tension. The origin of this peaks is connected with the thermodynamical properties of smectic liquid crystals. Indeed, smectic layers are practically incompressible, hence the random displacements of different layers are well-correlated in the direction normal to the layers. This leads to the undulation behaviour of the angular dependence.

The main result of this calculation is that the surface parameters of liquid crystals affect the angular distribution, so can be investigated by means of light-scattering experiment.

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