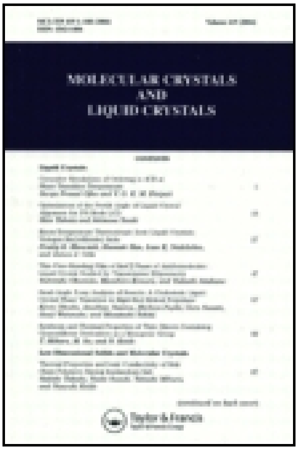

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Longitudinal Fluctuations of the Order Parameter in Discotic Liquid Crystals

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The role of the singular longitudinal fluctuations in discotic liquid crystals is considered. The expression for the extinction coefficient of the light scattering due to these fluctuations is calculated.

Several years ago, Pokrovskii and Patashinskii[1] drew attention to one general property of so-called degenerate systems, i.e., those in which the appearance of the order parameter breaks the continuous symmetry of the high temperature phase. In particular it was shown that large transverse fluctuations of the order parameter are necessarily accompanied by strong longitudinal fluctuations. Pokrovskii and Patashinskii studied isotropic magnetic systems and the terminology of transverse and longitudinal fluctuations refers to the direction of the spontaneous magnetic moment M. The magnitude of the moment M^2 is determined by the exchange forces. The degeneracy of the system implies that the free energy does not depend on the direction of $\mathbf{M}$.

The transverse and longitudinal fluctuations of M are constrained by the requirement of a constant modulus (the principle of the constancy of the modulus is in the terminology of the authors[1]:

$$2M\delta M_{\parallel} = -\delta M_{\perp}^2 \qquad (1)$$

where M is the magnitude of the spontaneous moment, and $\delta M_{\perp}$ is the transverse fluctuation.

In order to observe the longitudinal fluctuations, it was suggested[1] that the corresponding susceptibilities should be measured. The degeneracy of the system implies a singularity of the transverse susceptibility in zero magnetic field:

$$\chi_\perp \sim H^{-1}$$

As a consequence of Eq. (1), the longitudinal suceptibility also has a singularity

$$\chi_\parallel \sim H^{-1/2}$$

However, the existence of anisotropy interferes with the experimental observation of these laws, and to our knowledge there has been no experimental observation of these longitudinal fluctuations.

Nematic liquid crystals are also degenerate in this sense, but, in the absence of anisotropy they are more suitable for the experimental observation of longitudinal fluctuations. In particular, through light scattering studies, the effect of the longitudinal fluctuations can be isolated.[2,3]

In this paper we consider the longitudinal fluctuations in a system with the translational symmetry broken in two directions. The existence of such systems was predicted by Peirls and Landau, recently they have been obtained in the laboratory,[4] and today they are investigated intensively.

These systems (so called discotic liquid crystals) consist of molecules in the form of discs forming columns which form a two-dimensional lattice. Along the columns, however, the system is liquid-like. The role of the director is played by the normal to the prevailing orientation of the planes of the discs, and the fluctuation of the director is connected with the bending of the columns. This means that in the expression for the elastic energy of the Frank type, the terms with div **n** and **n** curl **n** are missing. The role of the transverse coordinate is played by the displacements of the two-dimensional lattice.

We shall assume for simplicity that the lattice is square. Then the density can be written in the form

$$\rho = \rho_0 + m[\cos(p_0 x + p_0 u_x) + \cos(p_0 y + p_0 u_y)] \tag{2}$$

where ρ_0 is the average density of the system, m is the amplitude of modulation of the two-dimensional lattice, p_0 is the wave vector corresponding to the given lattice, the quantities u_x and u_y are displacements of the lattice.

We denote the corresponding transverse fluctuation by

$$\delta\varphi_\alpha = p_0 u_\alpha, \quad \alpha = x, y \tag{3}$$

By means of the well-known expression derived by Landau and Peierls[5] for the displacement u_α of the three-dimensional system with a two-dimensional translational ordering[5] we have:

$$\langle\delta\varphi_\alpha\delta\varphi_\beta\rangle = p_0^2\langle u_\alpha u_\beta\rangle = \frac{p_0^2 T}{K}\frac{\delta_{\alpha\beta} - \dfrac{q_\alpha q_\beta}{q^2}}{q_z^4 + p_0^2 q_\perp^2} \tag{4}$$

where K is a Frank constant.

According to the principle of the constancy of the modulus Eq. (1), which in our case is of the form:

$$2m\,\delta m = -(\delta\varphi_\alpha)^2$$

we have for the correlation function of the longitudinal fluctuation δm the following expression

$$G_\parallel(\mathbf{p}) \equiv \langle \delta m(\mathbf{p})\delta m(-\mathbf{p})\rangle = \frac{T^2m^2p_0^4}{16\pi^3K^2}$$

$$\times\int \frac{d^3q}{(q_z^4 + p_0^2q_\perp^2)[(q_z + p_z)^4 + p_0^2(\mathbf{q}_\perp + \mathbf{p}_\perp)^2]} \quad (5)$$

Integrating over the angles and the transverse coordinate we obtain

$$G_\parallel(\mathbf{p}) = \frac{T^2m^2p_0^2}{16\pi^2K^2} I(p_z, p_\perp, p_0) \quad (6)$$

where

$$I(p_z, p_\perp, p_0) = \int_{-\infty}^{\infty}\frac{dq}{A}\ln\frac{(A + p_0^2p_\perp^2)^2 + B^2}{(A - p_0^2p_\perp^2)^2 - B^2}\,|p_\perp \neq 0|$$

$$A = \{[(q + p_z)^4 + p_0^2p_\perp^2 - q^4]^2 + 4p_0^2p_\perp^2q^4\}^{1/2}$$

$$B = (q + p_z)^4 - q^4 \quad (7)$$

In the general case, the integration cannot be fulfilled analytically. However, it can be obtained in a closed form in two special cases. For example, when $p_z = 0$

$$G_\parallel = \frac{T^2m^2}{K^2}\cdot\frac{p_0^{1/2}}{p_\perp^{3/2}}C_1 \quad (8)$$

where C_1 is the number.

$$C_1 = \frac{\Gamma^2\left(\frac{1}{4}\right)}{16\sqrt{2\pi}}$$

In the other case, when $p_\perp = 0$

$$G_\parallel = \frac{T^2m^2p_0^2}{8K^2p_z^3} \quad (9)$$

It is possible to obtain an interpolation formula by expanding Eq. (7) in powers of the parameter

$$p^4 = \frac{p_z^4}{p_0^2 p_\perp^2} \tag{10}$$

and subsequent matching of the resulting expressions. The expressions for I when $\delta \ll 1$ or $\delta \gg 1$ can be obtained analytically.

We give the final formula for the longitudinal correlation function

$$G_\parallel = \frac{T^2 m^2 p_0^2}{8K^2} \frac{1}{p_z^3 + A(p_\perp p_0)^{3/2}} \tag{11}$$

where

$$A = \frac{(2\pi)^{3/2}}{\left[\Gamma\left(\frac{1}{4}\right)\right]^2}$$

We now turn to light scattering on the longitudinal fluctuations. The differential coefficient of extinction can be written in the following form

$$dh = \frac{\omega^4}{32\pi^2 c^4} \langle \delta\epsilon(\mathbf{q})\delta\epsilon(-\mathbf{q})\rangle(1 + \cos^2\theta)d0$$

where

$$q^2 = 4\frac{\omega^2}{c^2}\sin^2\frac{\theta}{2},$$

where θ is the scattering angle, and $\delta\epsilon$ is the fluctuation of the dielectric permeability.

According to Eq. (2) we obtain

$$\delta\epsilon = \frac{\partial\epsilon}{\partial\rho}\Delta m[\cos(p_0 x + \varphi_x) + \cos(p_0 y + \varphi_y)]$$

$$+ \frac{1}{4}\frac{\partial^2\epsilon}{\partial\rho^2}(\delta m)^2[1 + \cos 2(p_0 x + \varphi_x) + \ldots] \tag{12}$$

Since $p_0 \gg \omega/c$ the contribution of the first term can be neglected because of the fast oscillations. In that case, the main contribution to the scattering comes from the second order scattering. It is important to note that in the case of broken rotational symmetry, the light scattering is of the first order.

Thus, the cross-section for the scattering is determined by the correlator

$$\langle\delta\epsilon(\mathbf{q})\delta\epsilon(-\mathbf{q})\rangle = \frac{1}{(4\pi)^3}\left(\frac{\partial^2\epsilon}{\partial\rho^2}\right)^2 \int d^3p\, G_{\parallel}(\mathbf{p}+\mathbf{q})G_{\parallel}(\mathbf{p}) \tag{13}$$

Performing the necessary calculations with the help of Eq. (11) we obtain for the extinction coefficient the following expressions

$$dh \sim \left(\frac{\partial^2\epsilon}{\partial\rho^2}\right)^2\left(\frac{\omega Tmp_0}{cK}\right)^4 \frac{1}{q_z p_0^2}, \quad q_\perp = 0$$

$$dh \sim \left(\frac{\partial^2\epsilon}{\partial\rho^2}\right)^2\left(\frac{\omega Tmp_0}{cK}\right)^4 \frac{1}{p_0^2(p_0 q_\perp)^{1/2}}, \quad q_z = 0 \tag{14}$$

The longitudinal scattering can be observed if the fluctuations associated with the bending of the liquid columns are suppressed. The polarization factor entering the expression for light scattering on the bends is equal to zero if one or both polarization vectors are perpendicular to the director.

There is also present the "ordinary" scattering associated with the density fluctuations

$$\langle\delta\rho(\mathbf{q})\delta\rho(-\mathbf{q})\rangle \sim \frac{T\rho_0}{s^2} \tag{15}$$

where s is the sound velocity.

It is clear that at small scattering angles, in the long wavelength limit, the longitudinal scattering dominates. A rough estimate from the expressions (14) and (15) with

$$\frac{\partial^2\epsilon}{\partial\rho^2} \sim \rho^{-1}\frac{\partial\epsilon}{\partial\rho}, \quad m \sim \frac{Kp_0^2 s^2}{\rho},$$

gives $K \sim 10^{-7}$ dyne, $s \sim 10^5$ cm s^{-1}, $\rho \sim 1$ g cm^{-3}, $p_0 \sim 10^7$ cm^{-1} and shows that at the wave length $\lambda \sim 6000$ Å the longitudinal scattering is essentially at scattering angles of several degrees.

We would like to point out some qualitative physical feature of the longitudinal scattering.

First, the frequency dependence is not ω^4 (as is the case in ordinary liquids) but rather ranges between ω^3 and $\omega^{7/2}$. Secondly, the line width is weakly dependent on the scattering wave vector

$$\sim q_z^{1/2}, \quad q_\perp = 0$$

$$\sim q_\perp^{1/4}, \quad q_z = 0$$

instead of the q^2 low in nematics.

It is interesting to note that longitudinal scattering of this type is not distinguished by any singular features in systems with broken three-dimensional translational symmetry, i.e., in real crystals. For this reason it cannot be observed on the background of other scattering mechanisms. The reason for this is that the correlation function of the transverse fluctuations, i.e., the elastic displacements of the crystal atoms, has the form

$$\langle \delta_u^2 \rangle \sim \frac{T}{q^2}$$

For the longitudinal correlator in this case we have

$$G_{\parallel} \sim \int d^3q \, \langle \delta_u^2 \rangle^2 \sim \frac{T^2}{q}$$

However, as mentioned above, in systems with broken translational symmetry, second order scattering takes place. Therefore the scattering cross-section is

$$dh \sim \int G_{\parallel}^2 d^3q$$

This integral converges at $q \to 0$ and consequently there is no singular longitudinal scattering. It can be asserted that discotic crystals are systems with the maximal translational group still allowing singular longitudinal scattering.

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