Fluctuation absorption of sound in compensated cholesterics

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A novel fluctuation effect in compensated cholesterics is predicted. It is shown that near the compensation line fluctuations of the director lead to the appearance of an anomalous contribution to the sound damping. This damping diverges in the low frequency region in proportion to the pitch of the cholesteric helix.

1. Nonlinear and fluctuation phenomena in the hydrodynamics of liquid crystals have been attracting much interest of late.[1,2] The fluctuations are quite substantial in liquid crystals with partial translational ordering (smectics and discotics), owing to the special softness of these systems, i.e., to the absence of a shear modulus in one or two dimensions. In nematics, on the other hand, fluctuations in the long-wave and low-frequency limit make only a small contribution to the kinetic coefficients ($\propto \omega^{1/2}$, where ω is the frequency) and in this sense nematics do not differ from ordinary isotropic liquids.

The situation is somewhat different in cholesteric liquid crystals. The cholesteric phase is known to be realized in substances consisting of chiral molecules. As a result, the director n in a cholesteric is "twisted" into a helix.[3] The pitch of this helix is usually of the same order as the wavelength of visible light, so that the wave vector q_0 of the helix determines the position of the Bragg maxima when light is diffracted by a cholesteric.

Its helical structure makes a cholesteric the equivalent of a smectic over scales much larger than the helix pitch.[4] As shown in Refs. 1 and 2, in smectics the contributions of the thermal fluctuations to the bulk-viscosity coefficients diverge in proportion to ω^{-1}. However, the coefficient in this dependence, which is indicative of the strength of the fluctuations, is determined by the compression modulus B of the smectic layer, and in cholesterics this modulus is equal to $K_2 q_0^2$ (K_2 is the elastic torsion modulus) and is considerably smaller than the compression modulus in real smectics. Fluctuations can therefore manifest themselves in cholesterics only at wavelengths too large and at frequencies too low to be realized in real experiments.

A very important property of cholesterics, which makes them highly useful in applications, is the substantial dependence of q_0 on the pressure P, on the temperature T, and (in the case of a mixture) on the concentration c of the components. Of particular interest, and frequently used in applications, are binary mixtures of chiral substances with different signs and magnitudes of the component chirality. The phase diagram of such a mixture, plotted in coordinates T and c, usually contains a solid line along which $q_0 = 0$. It is called the compensation line, and the binary mixture with $q_0 = 0$ is called a compensated or racemic cholesteric.

The director distribution in compensated cholesterics is homogeneous, just as in nematics. However (see below), the dynamics of a compensated cholesteric differs radically from that of a nematic. The reason is that any action that leads to deviation from the compensation line deforms substantially the homogeneous distribution of the director. The role of the fluctuations in the dynamics of compensated cholesterics turns out therefore to be very important.

2. We are interested in fluctuation effects due to the smallness of the parameter q_0 near the compensation line. It is easy to verify that the contribution of long-wave thermal fluctuations to the static characteristics of the cholesteric is not substantial also if q_0 is small. To investigate the contribution of the fluctuations to the dynamic characteristics it is necessary to use the diagram technique developed in Ref. 5, where a perturbation theory was developed in terms of the nonlinearities in the hydrodynamic equation. The corresponding diagram technique is generated by an effective action that is determined directly from these dynamic equations.

The nonlinear equations of the hydrodynamics of cholesterics are given in Ref. 4. Analysis shows that the only substantial nonlinear term in these equation is due to the following contribution to the pressure:

$$P = K_2 q_* (\mathbf{n}\,\mathrm{rot}\,\mathbf{n} - q_0). \tag{1}$$

Here

$$q_* = -\rho \partial q_0 / \partial \rho. \tag{2}$$

The derivative with respect to ρ is taken in Eq. (2) at a constant value of the specific entropy. The quantity q_* is of the order of the wave vector q_0 far from the compensation line.

The contribution (1) to the pressure corresponds to the following term, of third order in density, of the Lagrange function that determines the aforementioned effective action:

$$\mathscr{L}^{(3)} = K_2 q_* \nabla_i p_i \delta\mathbf{n}\,\mathrm{rot}\,\delta\mathbf{n}. \tag{3}$$

Here p_i is the auxiliary Bose field conjugate to the momentum density j_i, and $\delta\mathbf{n}$ is the director fluctuation.

The term $\mathscr{L}^{(3)}$ specifies the structure of the interaction vertices in the diagram technique. The unrenormalized correlators of the hydrodynamic quantities are determined by the linear equations of the dynamics of cholesterics.[5] We shall need hereafter only the director correlator $D_{ij} = \langle \delta n_i \delta n_j \rangle$, determined completely in the short-wave region by two functions D_l and D_t (the longitudinal and transverse components relative to the wave vector, which take the following form in the Fourier representation (ω is the frequency and $\mathbf{q}$ is the wave vector):

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$$D_l=\frac{2T\eta_l}{\eta_l{}^2\omega^2+K_l{}^2q^4},\quad D_t=\frac{2T\eta_t}{\eta_t{}^2\omega^2+K_t{}^2q^4}. \tag{4}$$

Here η_l and η_t are certain combinations of the coefficients, and K_l and K_t are combinations of Frank constants. All these combinations have a complicated dependence on the angle betwen the wave vector **q** and the director. Their explicit forms can be found in Ref. 4. Note that the fluctuation corrections to (4) are negligible.

That part of the Lagrangian $\mathscr{L}^{(3)}$ which describes the interaction generates the following fluctuation contribution to the dissipative part of the density of the Lagrangian:

$$\mathscr{L}^{(d)}={}^1/_2ip_i\Pi_{ik}p_k. \tag{5}$$

In the short-wave region, the polarization operator Π_{ik} is of the form

$$\Pi_{ik}(\omega,\mathbf{k})=2K_2{}^2q_*{}^2k_ik_k\varepsilon_{\alpha\beta}\varepsilon_{\gamma\delta}\times\int\frac{d\nu\, d^3q}{(2\pi)^4}(\mathbf{qn})^2D_{\alpha\gamma}(\nu,\mathbf{q})D_{\beta\delta}(\omega+\nu,\mathbf{k}+\mathbf{q}). \tag{6}$$

In this equation, the Greek subscripts denote components in a plane perpendicular to the director, and $\varepsilon_{\alpha\beta}$ is a two-dimensional antisymmetric tensor. Expression (6) corresponds to the diagram shown in Fig. 1, in which the dashed lines denote the correlators $\hat{D}$, and the vertices are specified by Eq. (3) for $\mathscr{L}^{(3)}$.

The presence of the term (5) in $\mathscr{L}$ is equivalent to the onset of sound damping with a decrement

$$\gamma_{fl}=\eta_{fl}\rho^{-1}q^2. \tag{7}$$

The specific expression for η_{fl} depends on the sound frequency ω. We consider first frequencies $\omega\ll Kq_0^2/\eta$, where K and η are respectively quantities of the order of the Frank moduli and of the unrenormalized (i.e., without allowance for the fluctuations) viscosity coefficients. In this case it follows from (5) and (6) that

$$\eta_{fl}=\frac{K_2q_*{}^2}{T}\int\frac{d\nu\, d^3q}{(2\pi)^4}(\mathbf{qn})^2D_t(\nu,\mathbf{q})D_t(\omega+\nu,\mathbf{q}). \tag{8}$$

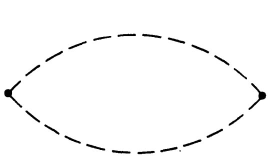

FIG. 1.

The integral in (8) can be determined only numerically, but by using Eqs. (4) it is easy to obtain for it the estimate

$$\eta_{fl}\sim10^{-2}q_*{}^2T\eta^{1/2}K^{-1/2}\omega^{-1/2}. \tag{9}$$

Expression (9) is incorrect for frequencies $\omega\ll Kq_0^2/\eta$, since the characteristic wave vectors q in the integral become smaller than q_0, meaning that Eqs. (4) cannot be used (recall that Eqs. (4) pertain to small scales, over which a cholesteric is similar to a nematic, whereas at large scales it is similar to a smectic). Equation (8) can nevertheless be used to estimate the fluctuation contribution to the sound damping also at low frequencies. To this end it is necessary to put $\omega=0$ in (8) and cut off the integral over the wave vector at a value on the order of q_0. As a result we get

$$\eta_{fl}\sim10^{-2}q_*{}^2T\eta K^{-1}q_0{}^{-1}. \tag{10}$$

3. The result of our investigation is the following conclusion, which can be directly verified in experiment. In the high-frequency region, the fluctuation damping of sound has an unusual frequency dependence ($\propto\omega^{3/2}$). In the low-frequency region the sound damping is, as usual, proportional to ω^2, but the coefficient of ω^2 diverges like q_0^{-1} near the compensation line. In particular, on "moving" to the compensation line as the temperature is varied, this coefficient diverges like $|T-T_*|^{-1}$, where T_* is the temperature at which $q_0=0$.

The fluctuation contribution to sound absorption can thus be experimentally identified, in both large and small scales, with the aid of the specific frequency dependence and the proximity to the compensation line.

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