Fluctuations of the tensor order-parameter modes in a cholesteric liquid crystal

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We use a Landau–de Gennes free energy to calculate the fluctuations of the five independent modes of the tensor order parameter for a cholesteric liquid crystal. Our results include, as a limiting case, the two classical director modes, known as the twist mode and the "umbrella" mode. We find, however, in contrast to the classical director model, that there can be substantial temperature dependence to the umbrella mode, as well as three additional modes near the transition to the isotropic phase. We comment on a recent experiment that suggests that two of these additional modes may have already been detected.

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INTRODUCTION

The first calculations of director fluctuations in a cholesteric liquid crystal were done quite some time ago [1–3]. It was shown theoretically that there are two principal types of director fluctuation modes. When a fluctuation $\delta\boldsymbol{n}$ of the director $\boldsymbol{n}$ is perpendicular to the helical wave vector $\boldsymbol{q}_0$, the deformation is known as a twist mode and is illustrated in Fig. 1(b). This mode may be thought of as a modulation of the phase of the helicoidal structure. The second case, where $\delta\boldsymbol{n}$ is parallel to $\boldsymbol{q}_0$, is referred to as either a conical mode or an "umbrella" mode and is shown in Fig. 1(c). This mode involves a modulation of the tilt of the director away from its preferred direction perpendicular to the helical axis. The two modes illustrated in Fig. 1 both have their wave vectors along the helical axis. One can also have modes whose wave vectors have components perpendicular to $\boldsymbol{q}_0$, but it turns out that the analysis of these modes is much more complicated and has only been done for the case where the wave vector is much less than $\boldsymbol{q}_0$. All other studies have treated only the situation where the wave vector is along the helix and the discussion in this paper will be limited to this case as well.

The average square fluctuations for the two director modes were found by applying the equipartition theorem to the Frank free energy for director distortions with the result [1–4]

$$\langle|\delta n_\perp(k)|^2\rangle = \frac{k_B T}{K_2 k^2}, \quad k = q \pm 2q_0 \quad \text{(twist)}, \tag{1a}$$

$$\langle|\delta n_\parallel(k)|^2\rangle = \frac{k_B T}{K_3 q_0^2 + K_1 k^2}, \quad k = q \pm q_0 \quad \text{(umbrella)}. \tag{1b}$$

Here the subscripts "⊥" and "∥" represent, respectively, perpendicular and parallel to the helical axis. The scattering wave vector is denoted by q and that of the helix by q_0. Thus, the umbrella mode reaches its maximum at points q_0 and $-q_0$ along the q_z axis in reciprocal space, or in other words when the tilt perturbation is commensurate with the helix periodicity, as in Fig. 1(c). In this case the orientational order is essentially the same as in a smectic-C^* phase, although the magnitude of the "tilt angle" is quite different in the two cases.

The twist mode fluctuations diverge at points $2q_0$ and $-2q_0$ on the q_z axis. In other words a twist modulation whose periodicity is half that of the helix is a Goldstone mode, since such a mode would constitute a uniform rotation or translation of the helical structure. In practice finite size effects limit the observation of the full divergence.

The director mode model is a two-component description of the fluctuations. In actuality the total number of modes capable of showing fluctuations is five, the number of independent components of the tensor orientational order parameter. There is often important information to be learned by looking at the "extra" three modes, even if the strength of their fluctuations is not particularly large. In a previous theoretical paper [5] it was shown how the five modes fluctuate in nematic liquid crystals in a wide variety of circumstances, including in the presence of external fields. Here we propose to do the same thing for cholesteric liquid crystals of various

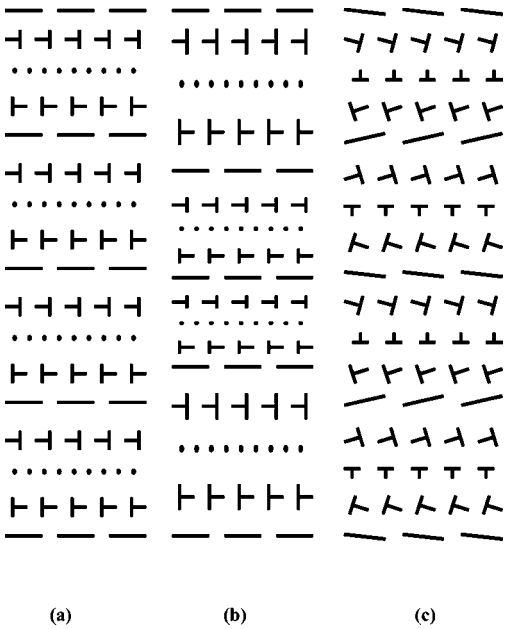

FIG. 1. Schematic representations of (a) a uniform cholesteric, (b) a twist mode, and (c) a conical or "umbrella" mode.

chiralities. The case of cholesteric liquid crystals in external fields will be covered separately.

It is an interesting fact that the first experimental attempt to verify the results of Eqs. (1) did not take place until nearly three decades after their theoretical prediction. Borsali *et al.* [6] reported the results of light scattering measured at different scattering angles but a fixed temperature that are claimed to be consistent with the predictions of Eqs. (1). We believe, however, that there is a major problem in their interpretation since the data that they identify with the umbrella mode require that the bend elastic constant be more than three orders of magnitude larger than the splay elastic constant, which is totally unexpected on physical grounds. We suggest in a later section that these data are more likely due to one of the other "extra" tensor modes. Furthermore, Borsali *et al.* say that they have unreported data associated with an unidentified "fast" mode, which we believe to be yet another one of the five tensor modes. Clearly the multiplicity of observed modes calls for a description that goes beyond the two-mode director description.

One particularly interesting feature of the tensor order parameter description of the cholesteric liquid crystal is that the predicted *temperature* dependence of the modes is in general much more dramatic than when the director mode picture is employed, particularly when large chiralities are involved. Considering, for example, the results of Eqs. (1), we see that the main temperature dependence of the director modes stems from that of the elastic constants appearing in the denominators. In mean field theory it is expected that the elastic constants will all be proportional to the square of the order parameter. Now the intensities of scattered light will also be proportional to the square of the order parameter multiplied by the appropriate $\langle \delta n^2 \rangle$. Thus the intensity of light scattered from the director modes is expected to be more or less *independent of temperature* as far as the director mode picture is concerned. Our results obtained via the tensor mode description show that while the twist mode is essentially temperature independent, the other modes have temperature dependences that can be substantial, particularly in the vicinity of the transition to the isotropic phase. The reason for these differences is that the director picture is essentially a uniaxial model requiring only one parameter to describe the degree of order, whereas the tensor description is a biaxial model requiring two parameters to describe the order. It is the interaction of the two order parameters in the tensor model that accounts for the temperature dependence.

In the next section we review what is already known about the tensor model for the cholesteric liquid crystal and its transition to the isotropic phase. The main conclusions to be shown are the temperature dependences of the order parameters and the temperature-chirality phase diagram for the cholesteric phase. Brazovskii and Dmetriev [7] were the first to present these results, which by now have been elaborated upon by many others. The section following that gives additional results, namely, our predictions for the fluctuations of the five order parameter components or modes. We compare and contrast these predictions with those of the director model throughout, and at the end with the experimental data that presently exist.

FREE ENERGY, ORDER PARAMETER, AND TEMPERATURE-CHIRALITY PHASE DIAGRAM: REVIEW OF KNOWN RESULTS

We take, as usual, the order parameter $Q_{\alpha\beta}$ to be the anisotropic part of the dielectric tensor, which we represent as [5]

$$Q_{\alpha\beta}(\vec{r}) = \begin{pmatrix} -\frac{1}{\sqrt{6}}Q(\vec{r}) + \frac{1}{\sqrt{2}}P_1(\vec{r}) & \frac{1}{\sqrt{2}}P_2(\vec{r}) & \frac{1}{\sqrt{2}}R_x(\vec{r}) \\ \frac{1}{\sqrt{2}}P_2(\vec{r}) & -\frac{1}{\sqrt{6}}Q(\vec{r}) - \frac{1}{\sqrt{2}}P_1(\vec{r}) & \frac{1}{\sqrt{2}}R_y(\vec{r}) \\ \frac{1}{\sqrt{2}}R_x(\vec{r}) & \frac{1}{\sqrt{2}}R_y(\vec{r}) & \frac{2}{\sqrt{6}}Q(\vec{r}) \end{pmatrix}. \tag{2}$$

In Landau theory the free energy density is expanded as a power series in this order parameter, which to fourth order, and in the one-elastic-constant approximation, looks like

$$f = \frac{1}{2}AQ_{\alpha\beta}Q_{\beta\alpha} + \frac{1}{2}LQ_{\alpha\beta,\gamma}Q_{\beta\alpha,\gamma} - 2Lq_0\varepsilon_{\alpha\beta\gamma}Q_{\alpha\mu}Q_{\mu\beta,\gamma} - \sqrt{\frac{2}{3}}BQ_{\alpha\beta}Q_{\beta\gamma}Q_{\gamma\alpha} + \frac{1}{4}C(Q_{\alpha\beta}Q_{\beta\alpha})^2. \tag{3}$$

Here, as usual, the coefficient A is assumed to have a linear temperature dependence, $A=A_0(T-T^*)$, while A_0, T^*, L, q_0, B, and C are assumed to be constant. The free energy for the helicoidal structure is obtained by substituting $Q(\vec{r})=Q_0$, $P_1=P_0\cos(2q_0z+\phi)$, and $P_2=P_0\sin(2q_0z+\phi)$ into Eqs. (2) and (3), where Q_0 and P_0 are (temperature dependent) constants and ϕ is an arbitrary phase angle. P_0 may be seen to be a measure of how well the molecules are aligned along the local director that is perpendicular to the helix and whose direction changes as one moves along the z direction. Q_0, which is invariably negative, is a measure of how well the molecules point *away* from the helical axis. If the cho-

lesteric structure were locally uniaxial, then the parameters P_0 and Q_0 would be related by $P_0^2=3Q_0^2$. When there is twist, however, there is local *biaxiality* and this relationship does not hold [see Eq. (6) below].

It is useful to cast Eq. (3) into dimensionless form by scaling out a factor of B/C from $Q_{\alpha\beta}$ and a factor of B^4/C^3 from f. In this manner the dimensionless free energy density for the helicoidal structure in equilibrium is found to be

$$f_{\rm eq}=\frac{1}{2}t(Q_0^2+P_0^2)+\frac{1}{4}(Q_0^2+P_0^2)^2-\frac{1}{3}Q_0^3+P_0^2Q_0-\frac{1}{2}\kappa^2P_0^2. \tag{4}$$

In the above a reduced temperature $t\equiv AC/B^2$ and a reduced chirality parameter $\kappa\equiv 2\ell q_0$ are defined, where $\ell\equiv\sqrt{LC}/B$. Minimizing this expression with respect to its two order parameters yields

$$tQ_0+Q_0^3+P_0^2Q_0-Q_0^2+P_0^2=0 \tag{5a}$$

and

$$P_0(t+P_0^2+Q_0^2+2Q_0-\kappa^2)=0. \tag{5b}$$

Eliminating the temperature between these two equations shows that the relationship

$$P_0^2=Q_0(3Q_0-\kappa^2) \tag{6}$$

is always valid. Equation (6) may in turn be substituted back into Eq. (5) to obtain an expression that may be solved to give the temperature dependence of the order parameter Q_0. In this manner we find

$$Q_0(t,\kappa)=\frac{1}{8}(-2+\kappa^2-\sqrt{\kappa^4+12\kappa^2-16t+4}). \tag{7}$$

Then Eq. (7) may be used with Eq. (6) to obtain the temperature dependence of P_0.

As was first noticed by Brazovskii and Dmetriev [7], a careful examination of the free energy expression (4) reveals that the transition from the helicoidal cholesteric phase to the isotropic phase becomes second order for $\kappa\geqslant 2$ and takes place at a temperature

$$t_c=\kappa^2 \quad (\kappa^2\geqslant 2). \tag{8}$$

If $\kappa^2<2$, the transition is first order and takes place at a temperature

$$t_c=\frac{1}{9}+\frac{1}{2}\kappa^2+\frac{\sqrt{2}}{36}(2+3\kappa^2)^{3/2} \quad (\kappa^2<2). \tag{9}$$

Figure 2 shows the phase diagram for the isotropic-cholesteric transition, including the tricritical point where this changeover takes place.

Defining $\varepsilon=t_c-t$, the distance in temperature from the transition, gives more useful expressions for the temperature variations of the order parameter Q_0:

$$Q_0=\frac{1}{8}[\kappa^2-2-\sqrt{(\kappa^2-2)^2+16\varepsilon}] \quad (\kappa^2\geqslant 2), \tag{10}$$

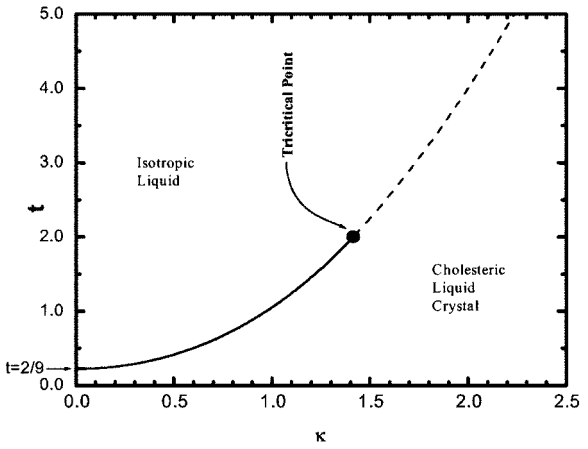

FIG. 2. Temperature vs chirality phase diagram for the cholesteric-isotropic transition. The solid (dashed) line represents a first (second) order transition.

$$Q_0=\frac{1}{8}\left[\kappa^2-2-\sqrt{\kappa^4+4\kappa^2+\frac{20}{9}+16\varepsilon-\frac{2}{9}(6\kappa^2+4)^{3/2}}\right] \quad (\kappa^2<2). \tag{11}$$

The temperature behavior of P_0 can then be obtained through use of (6). From these equations we can see that at the tricritical point $(\kappa^2=2)Q_0=-\frac{1}{2}\varepsilon^{1/2}$ and $P_0\approx\varepsilon^{1/4}$. Thus P_0 is seen to be the primary order parameter with the usual mean field tricritical exponent $\beta=\frac{1}{4}$, while Q_0 is seen to be a secondary order parameter. Likewise, along the line of critical points $(\kappa^2>2)P_0$ has the usual mean field critical exponent $\beta=\frac{1}{2}$ whereas Q_0 varies linearly with ε.

After further consideration Brazovskii and Dmetriev [7] realized that this tricritical point and line of second order transitions would not actually be reached because at chiralities of $\kappa\sim 1$ and beyond multiply periodic structures, which today are called the "blue phases" [4], would intervene. That is, although the cholesteric liquid crystal may act as though it is approaching a second order transition to the isotropic phase, it will undergo a first order transition to one of the blue phases before it reaches the hypothetical second order transition. The calculations of the fluctuations presented here are still useful up to the point that a blue phase is encountered; indeed there are reasons for believing that these fluctuations may actually play a dynamic role in the creation of the blue phases. Also, as we shall show in a separate paper, in certain circumstances the application of a large electric field can quench the blue phases and a true second order transition can be reached. Therefore, for all these reasons, the calculation of the fluctuations at the approach to the cholesteric-isotropic transition are of more than just academic interest and so we proceed.

ORDER-PARAMETER FLUCTUATIONS: PRESENT RESULTS

Now that the local minimum of the free energy has been found we need to express the free energy to second order in fluctuations about this minimum. This analysis is facilitated

by switching representations from one where the coordinate axis are fixed in space, as in Eq. (2), to one in which the reference axes rotate in the same way as does the helix, so that the unperturbed structure always appears uniform. Accordingly we define

$$Q(\mathbf{r}) = Q_0 + \delta Q(\mathbf{r}),$$

$$P_{\parallel}(\mathbf{r}) = P_0 + \delta P_{\parallel}(\mathbf{r}),$$

$$P_{\perp}(\mathbf{r}) = 0 + \delta P_{\perp}(\mathbf{r}),$$

$$R_1(\mathbf{r}) = 0 + \delta R_1(\mathbf{r}),$$

$$R_2(\mathbf{r}) = 0 + \delta R_2(\mathbf{r}). \tag{12}$$

Here we see that δQ and $\delta P_{\parallel}$ are fluctuations in the *magnitudes* of the two measures of alignment, Q and P, respectively. The quantity $\delta P_{\perp}$ is a local fluctuation in the *phase* ϕ of the helix or, equivalently, a small rotation of the local director perpendicular to the helix; it is, in other words, a *twist* mode. Finally, δR_1 and δR_2 are *both* umbrella modes. There are two umbrella modes because the local biaxiality implies that there are two directors: R_1, the primary one pointing along the alignment direction of the long axes of the molecules, and a secondary one, R_2, perpendicular to the first and also to the helical axis. Each of these directors can tilt, or develop a conical structure, as shown in Fig. 3, and so there are two independent modes.

As mentioned earlier, we will only be considering the case where the scattering wave vector $\boldsymbol{q}$ is along the z direction. Accordingly, $\delta Q_{\alpha\beta}$ will be considered to be a function of z only and can then be expressed in terms of its Fourier coefficients as

$$\delta Q_{\alpha\beta}(z) = \sum_q \delta\tilde{Q}_{\alpha\beta}(k) e^{ikz}. \tag{13}$$

When Eq. (13) is entered into the free energy density (3) and then this is integrated over all space, the free energy expression that results to second order in $\delta\tilde{Q}_{\alpha\beta}$ is

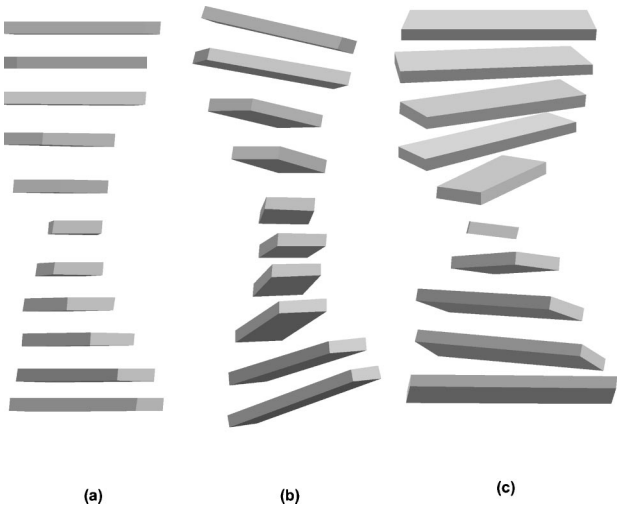

FIG. 3. Schematic representation of (a) the unperturbed cholesteric, (b) a primary umbrella mode R_1, and (c) a secondary umbrella mode R_2. The primary director is parallel to the long axis of the parallelepiped; the secondary director is parallel to the intermediate axis.

$$\begin{aligned}\Delta F_q = {} & \frac{1}{2}(t + 3Q_0^2 + P_0^2 - 2Q_0 + \ell^2 k^2)|\delta\tilde{Q}(k)|^2 + \frac{1}{2}(t + Q_0^2 \\ & + 3P_0^2 + 2Q_0 - \kappa^2 + \ell^2 k^2)|\delta\tilde{P}_{\parallel}(k)|^2 + \frac{1}{2}(t + Q_0^2 + P_0^2 \\ & + 2Q_0 - \kappa^2 + \ell^2 k^2)|\delta\tilde{P}_{\perp}(k)|^2 + \frac{1}{2}\left(t + Q_0^2 + P_0^2 - Q_0 \right. \\ & \left. - \sqrt{3}P_0 - \frac{1}{4}\kappa^2 + \ell^2 k^2\right)|\delta\tilde{R}_1(k)|^2 + \frac{1}{2}\left(t + Q_0^2 + P_0^2 - Q_0 \right. \\ & \left. + \sqrt{3}P_0 - \frac{1}{4}\kappa^2 + \ell^2 k^2\right)|\delta\tilde{R}_2(k)|^2 + \frac{1}{2}(2Q_0P_0 + 2P_0) \\ & \times[\delta\tilde{Q}(k)\delta\tilde{P}_{\parallel}^*(k) + \delta\tilde{Q}^*(k)\delta\tilde{P}_{\parallel}(k)]. \end{aligned} \tag{14}$$

The equipartition theorem may then be employed to find the thermal averages of the fluctuations. The coefficients appearing in Eq. (14) may be reexpressed using Eqs. (5) and (6) previously developed and thus we obtain

$$\langle|\delta\tilde{P}_{\perp}(k)|^2\rangle = \frac{k_B T}{\ell^2 k^2}, \tag{15}$$

$$\langle|\delta\tilde{R}_{1,2}(k)|^2\rangle = \frac{k_B T}{-3Q_0 \mp \sqrt{3Q_0(3Q_0 - \kappa^2)} + \frac{3}{4}\kappa^2 + \ell^2 k^2}, \tag{16}$$

$$\langle|\delta\tilde{Q}(k)|^2\rangle = \frac{k_B T[2Q_0(3Q_0 - \kappa^2) + \ell^2 k^2]}{2Q_0(3Q_0 - \kappa^2)(\kappa^2 - 8Q_0 - 2) + [8Q_0^2 - 2(\kappa^2 + 2)Q_0 + \kappa^2]\ell^2 k^2 + \ell^4 k^4}, \tag{17}$$

$$\langle|\delta\tilde{P}_{\parallel}(k)|^2\rangle = \frac{k_BT[2Q_0(Q_0-2)+\kappa^2+\ell^2k^2]}{2Q_0(3Q_0-\kappa^2)(\kappa^2-8Q_0-2)+[8Q_0^2-2(\kappa^2+2)Q_0+\kappa^2]\ell^2k^2+\ell^4k^4}. \tag{18}$$

The fluctuations of $P_\perp$ are, as in the director-mode picture, inverse proportional to k^2 and relatively insensitive to temperature. The fluctuations in R_1, the primary umbrella mode, are likewise as given by the director model if one is far from the cholesteric-isotropic transition and/or the chirality κ is small. That is, the expansion of Eq. (16) gives

$$\langle|\delta\tilde{R}_1(k)|^2\rangle \approx \frac{k_BT}{\ell^2q_0^2+\ell^2k^2} \quad (-3Q_0 \gg \kappa^2), \tag{19}$$

which is equivalent to Eq. (1b) in the one-elastic-constant case. If, however, the cholesteric-isotropic transition is nearby and/or the chirality κ is large, then the situation is quite different: the primary umbrella fluctuations *decrease* substantially as the temperature is increased. The secondary umbrella mode has just the opposite behavior: it is normally very weak but becomes much larger when cholesteric-isotropic transition is approached and/or the chirality κ is large. We show these behaviors, as well as that for all of the other modes, in Fig. 4 for a chirality of $\kappa=0.7$, which has a first order phase transition and in Fig. 5 for a chirality of $\kappa=1.8$, which has a second order phase transition. Here we plot k_BT divided by each average squared fluctuation *evaluated at its peak* ($k=0$).

Although they are not the focus of the present study, the results for the isotropic phase are also included in Figs. 4 and 5 for the sake of completeness. These are easily obtained by applying the equipartition theorem to Eq. (14) after first setting Q_0 and P_0 equal to zero. Calculations of this sort were first done by de Gennes [8] and then elaborated on by many others in the tensor fomulation including especially Hornreich and Shtrikman [9], who also discussed scattering from the blue phases, but not from the cholesteric phase itself. Recently Longa *et al.* [10] have considered what happens in the isotropic phase when the harmonic approximation breaks down and coupling between the five, formerly independent, modes ensues; this is their model for the transition from the ordinary isotropic phase to the amorphous phase known as BPIII. It would be interesting to see if mode coupling would also modify the results presented here for the cholesteric phase.

From Eq. (18) it may easily be seen that $P_\parallel$ plays the role of the "soft mode" in that its fluctuations diverge when the second transition is approached: as $k_BT/4\varepsilon$ at the tricritical point ($\kappa^2=2$) and as $k_BT/2\varepsilon$ along the line of critical points ($\kappa^2>2$). That the *amplitude as well as the phase* of the primary order parameter should diverge suggests that defects may eventually play an important role as the transition is approached. The defects most commonly seen in cholesterics are the λ and τ singularities, whose cores resemble the double-twisted structures found in the blue phases. Perhaps then there is a dynamical aspect to the formation of the blue phases in addition to the energetics of static structures that has been considered up to now.

The Q mode fluctuations [Eq. (17)] also grow as the transition is approached, as $k_BT/[16\varepsilon+(\kappa^2-2)^2]^{1/2}$, but as may be seen from this expression it diverges at the tricritical point only ($k^2=2$). The critical exponent of $\frac{1}{2}$ that it has at the tricritical point is another indication that Q is a secondary order parameter.

It is important to realize that although the fluctuations of all five modes peak at $k=0$, these peaks are actually separated in reciprocal space. Thus the $P_\perp$ and $P_\parallel$ modes peak at $q=\pm 2q_0$, the umbrella modes R_1 and R_2 at $q=\pm q_0$, and the Q mode at $q=0$. This separation can help somewhat in the experimental detection of the different modes, although there is still considerable overlap between some of the modes. This is

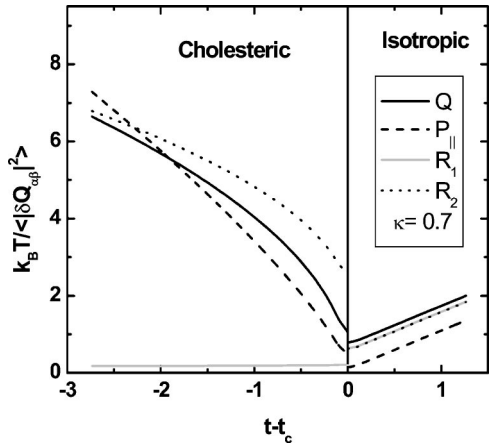

FIG. 4. Inverse of the average squared fluctuations of the order parameter components for a chirality ($\kappa=0.7$) having a first order transition.

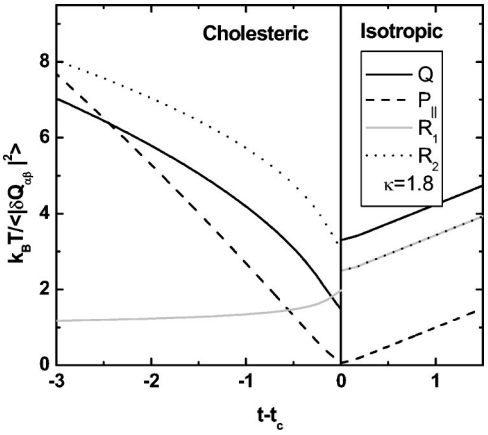

FIG. 5. Inverse of the average squared fluctuations of the order parameter components for a chirality ($\kappa=1.8$) having a second order transition.

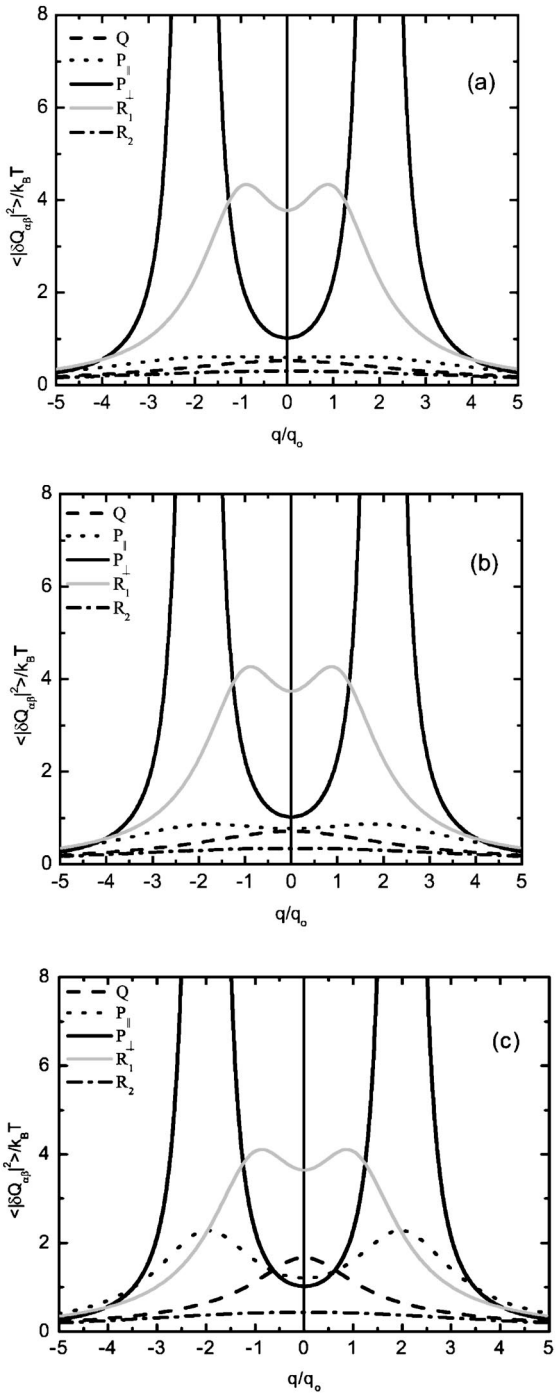

FIG. 6. The average squared fluctuations of the order parameter components for a chirality ($\kappa=0.7$). The temperature from the transition is $\varepsilon=$ (a) 0.2, (b) 0.1, and (c) 0.

demonstrated in Fig. 6 in which we plot the mean-squared fluctuations vs q for a chirality of $\kappa=0.7$ and three different temperatures. Note that the R_2 modes at $q=\pm q_0$ overlap to such an extent that they are never resolved as two separate peaks.

THE LOW CHIRALITY LIMIT: COMPARISON TO EXPERIMENT

The experiment of Borsali *et al.* [6] was performed on a nematic doped with a small amount of chiral material, giving a cholesteric with a pitch of about 10 μm. Unfortunately, a cholesteric with a pitch this large does not really display many of the characteristic features that would be found in a more typical material having a pitch comparable to the wavelength of light. The separation of the mode peaks in reciprocal space, for example, is not at all evident; the smallest q^2 in the experiment is about three orders of magnitude larger than q_0^2, so the distinction between q and k is irrelevant. Nonetheless, the analysis we have presented here should still apply to the experiment; we have only to take the limit where κ or q_0 is zero. In this limit, and to first order in k^2, we have for k_BT *divided by* each average squared fluctuation

$$\frac{k_BT}{\langle|\delta\tilde{R}_1(k)|^2\rangle}\approx \ell^2k^2, \tag{20}$$

$$\frac{k_BT}{\langle|\delta\tilde{R}_2(k)|^2\rangle}\approx -6Q_0+\ell^2k^2, \tag{21}$$

$$\frac{k_BT}{\langle|\delta\tilde{Q}(k)|^2\rangle}\approx -2(4Q_0+1)+\frac{(4Q_0^2+2Q_0+1)}{3Q_0^2}\ell^2k^2, \tag{22}$$

$$\frac{k_BT}{\langle|\delta\tilde{P}_\parallel(k)|^2\rangle}\approx \frac{-6Q_0(4Q_0+1)}{Q_0-2}+\frac{(4Q_0^2+2Q_0+7)}{(Q_0-2)^2}\ell^2k^2. \tag{23}$$

So in this limit we see from Eq. (20) that the primary umbrella mode is indistinguishable from the twist mode, which is still given by Eq. (15). The second mode identified by Borsali *et al.* must therefore be one of the other modes (as must be the third "fast mode" discussed briefly by them). The only alternative, the one actually considered by Borsali *et al.*, is that the product $K_3q_0^2$ stays *finite* as q_0 becomes negligible, which would require the bend elastic constant to become anomalously large for no apparent reason.

Most likely the second mode that Borsali *et al.* have discovered is the $P_\parallel$ mode. We base this upon the fact that it is this mode that lies closest to the twist and primary umbrella modes for the most likely ranges of parameters. In the range of $-1<Q_0<-1/3$ (the smallest negative value allowed for $\kappa=0$) the $P_\parallel$ mode is always the mode that is closest to the twist and primary umbrella modes. For an order parameter of $Q_0=-1$ the Q, $P_\parallel$, and R_2 modes are coincident; they have the same slopes and the same intercept of 6. This intercept places these modes considerably farther away from the twist and primary umbrella modes than what is found in the experiment. In Fig. 7 we show a plot of the inverses of the various modes versus ℓ^2k^2 for a Q_0 value of -0.4, which seems to correspond much more to the experimental situation. It is not possible to give an exact comparison between our results and the experimental data, however, because what

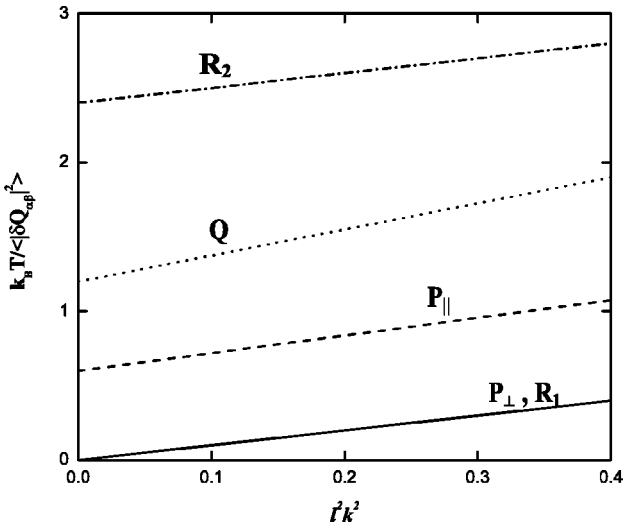

FIG. 7. Inverse of the average squared fluctuations of the order parameter components vs ℓ^2k^2 for zero chirality and $Q_0=-0.4$ ($\varepsilon=0.3597$).

we calculate is related to the intensities of light scattering whereas the experiment has measured relaxation rates, which are related to the intensity autocorrelation function. The latter are scaled to the former by combinations of (unknown) viscosity coefficients, but this probably should not change the relative importance of the different modes by very much. Also, the value of the length ℓ is not known, but if we assume that it is 200 Å then the range of the ℓ^2k^2 shown in the figure corresponds to the experiment.

If Fig. 7 is an accurate guide, then the third "fast mode" detected by Borsali *et al.* is probably the *Q* mode. More measurements of the modes and their k^2 slopes at different temperatures should provide significant new information to enable the more positive identification of each mode.

CONCLUSION

The tensor order-parameter description of fluctuations in a cholesteric liquid crystal complements and extends the director-model theory. It shows that far from the transition to the isotropic phase the most dominant modes by far are the twist and umbrella modes and that their temperature dependences are approximately as given by the director model. As the isotropic phase is approached, however, the temperature dependences of the twist and umbrella modes begin to differ substantially from the predictions of the director model. At the same time the strengths of the other three fluctuation modes, not included in the director model, grow substantially. There may be some interesting physics to be learned by measuring all five of these modes, not just at the transition to the isotropic phase but also at transitions to the blue phases or even at transitions to smectic liquid crystals or other lower temperature phases. To some extent the five modes may be singled out experimentally by virtue of their different angular dependences, although this may prove difficult because they are so broad. In principle, polarization selection of incoming and scattered light, using both linear and circular polarizers and analyzers, can also be used to separate the modes to some extent at least, but multiple scattering may complicate this method. Finally, as was done in [6], the modes may be resolved in the time domain especially if they have very different relaxation rates. A combination of all these methods may be needed for an unambiguous elucidation.

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