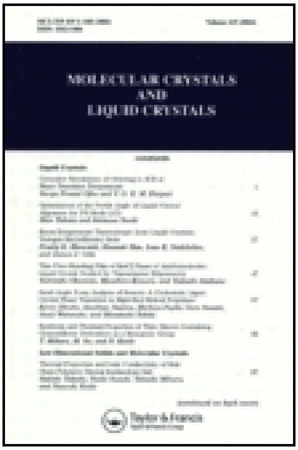

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FLUCTUATION SPECTRUM OF DIRECTOR IN CHOLESTERIC PHASE WITH EXTERNAL FIELD

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Fluctuation spectrum of molecular orientation in the chloesteric phase exposed to the external field with positive anisotropy is calculated for the general case of Frank's elastic constants. The stability of the phase is certified in the meaning of peierls-Landau theory.

Keywords: cholesteric phase; fluctuation spectrum; Lame equation; Peierls-Landau instability; transverse external field

INTRODUCTION

The cholesteric phase is known to be unstable for the fluctuation of helix angle with long wave number in the meaning of Peierls-Landau analysis. This instability comes from the one-dimensional structure together with the continuous rotational symmetry around the cholesteric axis [1]. In the transverse magnetic (or, electric) field violating this rotational symmetry, the helicoidal structure is distorted and so-called soliton lattice is achieved. Then, the system is shown to be stabilized in spite of the one dimensional structure, where the position of soliton lattice is degenerate continuously in the direction of the helical axis [2,3]. The chiral smectic C phase has also helicoidal structure, and fluctuation of azimuthal angle called a phason is similar variable to the helix angle in cholesterics. Experimentally, the phason mode has been observed, where the gap of dispersion relation at the Brillouin zone boundary is stressed [4,5].

In the previous study the fluctuation spectrum in the cholesteric phase is calculated under the one constant approximation for Frank's elastic coefficients, ($k_1 = k_2 = k_3 = k$), from which the stability is proved [3]. In view of the correspondence of theory with the practical experiments, the one

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constant approximation is insufficient. Here, the calculation of the fluctuation spectrum for the general case with individual Frank's coefficients is reported.

GENERAL FORMALISM

Soliton Lattice State

Calculations are carried out after the theory in the literature of reference 3, and here an outline of the formalism is sketched. The Frank's free energy for the director field ***n(r)*** is given by

$$F = \frac{1}{2V}\int d^3r[k_1(\mathrm{div}\,\boldsymbol{n})^2 + k_2(\boldsymbol{n}\cdot\mathrm{rot}\,\boldsymbol{n} + q_0)^2 + k_3\{\boldsymbol{n}\times(\mathrm{rot}\,\boldsymbol{n})\}^2 - \chi_a(\boldsymbol{H}\cdot\boldsymbol{n})^2], \quad (1)$$

where V is the volume of the system, $\chi_a > 0$ and q_0 is 2π times of the inverse of pitch p_0 of the cholesteric phase. Here, we chose the cholesteric axis in z-axis and the magnetic field is in y-axis. Then, the director is expressed in terms of the twist angle $\theta(z)$ as

$$(n_x(z), n_y(z), n_z(z)) = (\cos\theta(z), \sin\theta(z), 0). \quad (2)$$

The Euler-Lagrange equation for $\theta(z)$ is obtained with the magnetic coherence length $\xi = (k_2/\chi_a)^{1/2}H^{-1}$ as

$$\frac{d^2(2\theta)}{dz^2} + \xi^{-2}\sin(2\theta) = 0. \quad (3)$$

The equilibrium solution to Eq. (3) is given by

$$\sin\theta(z) = \mathrm{sn}\; z/\kappa\xi, \quad (4)$$

where sn x is Jacobi's sn-function and a modulus κ is determined from

$$E(\kappa)/\kappa = \pi q_0\xi/2, \quad (5)$$

in which $E(\kappa)$ is the complete elliptic integral of second kind.

Fluctuation Spectrum

Fluctuations of the director from $\theta(z)$ have two component, $\varphi(\boldsymbol{r})$ and $\psi(\boldsymbol{r})$, where the former is in the twist plane and the latter is perpendicular to that plane, respectively. The excess free energy due to fluctuations relative to the soliton lattice state (5) is expressed as

$$\Delta F = \frac{1}{2V}\int d^3r\ \Phi^+(\boldsymbol{r})A\ \Phi(\boldsymbol{r}), \tag{6}$$

where $\boldsymbol{A}$ is a 2×2 matrix and the hermit conjugate of $\Phi(\boldsymbol{r})$ is expressed as

$$\Phi^+(\boldsymbol{r}) = (\varphi(\boldsymbol{r})^*, \Psi(\boldsymbol{r})^*). \tag{7}$$

The free energy ΔF is calculated by the minimum eigenvalue of matrix $\boldsymbol{A}$, which is sufficient at the present purpose up to the quadratic terms of wave number. The system is uniform in x-y plane and the fluctuations are assumed to be proportional to $\exp\{i(k_x x + k_y y)\}$. Then, components of $\boldsymbol{A}$ are expressed using notations, $k_\pm = k_x \pm ik_y$, as,

$$\begin{aligned} A_{11} &= (k_1\sin^2\theta + k_3\cos^2\theta)k_x^2 + (k_1\cos^2\theta + k_3\sin^2\theta)k_y^2 - k_2\frac{\mathrm{d}^2}{\mathrm{d}z^2} \\ &\quad - 2(k_1 - k_3)k_x k_y \sin\theta\cos\theta + k_2\xi^{-2}(\sin^2\theta - \cos^2\theta), \end{aligned} \tag{8}$$

$$\begin{aligned} A_{22} &= (k_2\sin^2\theta + k_3\cos^2\theta)k_x^2 + (k_2\cos^2\theta + k_3\sin^2\theta)k_y^2 - k_1\frac{\mathrm{d}^2}{\mathrm{d}z^2} \\ &\quad - 2(k_2 - k_3)k_x k_y \sin\theta\cos\theta + k_3\left(\frac{\mathrm{d}\theta}{\mathrm{d}z}\right)^2 \\ &\quad + k_2\left\{2q_0\frac{\mathrm{d}\theta}{\partial z} - 2\left(\frac{\mathrm{d}\theta}{\partial z}\right)^2 + \xi^{-2}\sin^2\theta\right\}, \end{aligned} \tag{9}$$

$$\begin{aligned} A_{12} &= i\left\{\frac{1}{2}(k_2 - k_3)\frac{\mathrm{d}\theta}{\partial z} - k_2 q_0\right\}(k_-\mathrm{e}^{i\theta} + k_+\mathrm{e}^{-i\theta}) \\ &\quad + \frac{1}{2}(k_1 - k_2)(k_-\mathrm{e}^{i\theta} - k_+\mathrm{e}^{-i\theta})\frac{\mathrm{d}}{\mathrm{d}z}, \end{aligned} \tag{10}$$

$$\begin{aligned} A_{21} &= i\left\{\frac{1}{2}(k_1 + k_3 - 2k_2)\frac{\mathrm{d}\theta}{\partial z} + k_2 q_0\right\}(k_-\mathrm{e}^{i\theta} + k_+\mathrm{e}^{-i\theta}) \\ &\quad + \frac{1}{2}(k_1 - k_2)(k_-\mathrm{e}^{i\theta} - k_+\mathrm{e}^{-i\theta})\frac{\mathrm{d}}{\mathrm{d}z}. \end{aligned} \tag{11}$$

It is convenient to divide A_{11} into two parts,

$$A_{11} = {A_{11}}^{(0)} + {A_{11}}^{(1)},$$

where

$${A_{11}}^{(0)} = \frac{1}{2}(k_1 + k_3){k_\perp}^2 + \frac{k_2}{\kappa^2\xi^2}\left\{-\frac{\mathrm{d}^2}{\mathrm{d}u^2} + \kappa^2(\mathrm{sn}^2 u - \mathrm{cn}^2 u)\right\}, \tag{12}$$

$$A_{11}{}^{(1)} = \frac{1}{2}(k_3 - k_1)\mathrm{Re}(k_-^2\, \mathrm{e}^{2i\theta}), \tag{13}$$

in which cn x is Jacobi's cn-function, $u = z/\kappa\xi$ and $k_\perp{}^2 = k_x{}^2 + k_y{}^2$. At the eigenvalue equation of A_{11}, $A_{11}{}^{(1)}$ is treated as the perturbation for it is of the order k_-^2. By ignoring constant term proportional to $k_\perp^2$, the eigenvalue equation of $A_{11}{}^{(0)}$ is written explicitly with eigenvalue ε_0 as,

$$\left\{\frac{\mathrm{d}^2}{\mathrm{d}u^2} + \kappa^2(\mathrm{cn}^2 u - \mathrm{sn}^2 u)\right\}\varphi = \varepsilon_0\varphi, \tag{14}$$

which is known as the Lame's equation. This equation has been applied to the small oscillation in the Josephson junction [6,7] and to a special one-dimensional solid [8]. The general solution to Eq. (14) is expressed as [9]

$$\varphi(u) \propto H(u - u_0)/\Theta(u)\exp(-uZ(u_0)), \tag{15}$$

where $H(u)$, $\Theta(u)$, $Z(u)$ are the Jacobi's eta, theta and zeta functions, respectively. Because $\varphi(u)$ should be a hydrodynamic mode, u_0 is chosen to be $K(\kappa) + i(K'(\kappa) - \delta)$ with condition $\delta/K'(\kappa) << 1(K(\kappa)$; the complete elliptic integral of first kind and $K'(\kappa) = K(\sqrt{1-\kappa^2})$), and accordingly eigenfunction (15) is reduced to the form

$$\varphi(z) = a\exp(ik_z z)\mathrm{dn}\left(\frac{z}{\kappa\xi}\right)f(z, k_z), \tag{16}$$

in which dn x is Jacobi's dn-function, $f(z, 0) = 1$ and a is constant. Then, an eigenvalue for (14) associated with Eq. (16) is given by

$$\varepsilon_0 = \frac{(1-\kappa^2)K^2(\kappa)k_2}{E^2(\kappa)}k_z{}^2. \tag{17}$$

Then eigenvalue of A_{11}, ε, is obtained using eigenfunction (16) as

$$\begin{aligned}\varepsilon = {} & \frac{1}{2}(k_1 + k_3)k_\perp^2 + \frac{(1-\kappa^2)K^2(\kappa)k_2}{E^2(\kappa)}k_z^2 \\ & + \frac{1}{2}\alpha_1^{(1)}(k_3 - k_1)(k_x^2 - k_y^2),\end{aligned} \tag{18}$$

in which whole factors in A_{11} are taken into account and $\alpha_n{}^{(i)}$ is defined by

$$\alpha_n{}^{(i)} = \frac{(\kappa\xi)^{1-i}}{E(\kappa)}\int_0^{K(\kappa)} \cos 2n\theta(\kappa\xi u)(\mathrm{dn}\, u)^{i+1} du. \tag{19}$$

Secular Equation

The matrix element A_{22} is easily shown to be positive definite, and so $\psi(z)$ appears due to the off-diagonal terms of $\boldsymbol{A}$. The fluctuation vector $\Phi(\boldsymbol{r})$ is decomposed into two parts

$$\Phi(\boldsymbol{r}) = \Phi^{(0)}(\boldsymbol{r}) + \Phi^{(1)}(\boldsymbol{r}), \tag{20}$$

where $\Phi^{(0)}(\boldsymbol{r})$ is composed of $\varphi(z)$ of Eq. (16) multiplied by $\exp\{i(k_x + k_y y)\}$ and 0, and the transpose of $\Phi^{(1)}(\boldsymbol{r})$ is given by

$$\Phi^{(1)}(\boldsymbol{r})^{\mathrm{t}} = \mathrm{e}^{ik\cdot r}(\varphi'(z), \psi'(z)). \tag{21}$$

The contribution from φ' is proved easily of higher order than $k^2 (= k_\perp^2 + k_z^2)$ and neglected in Eq. (21). The term $\psi'(z)$ is expanded in the form,

$$\psi'(z) = \sum_{n=-\infty}^{\infty} b_n\, \mathrm{e}^{i(2n+1)\theta(z)} \mathrm{dn}\left(\frac{z}{\kappa\xi}\right) f(z, k_z), \tag{22}$$

Then, by carrying out some integral procedures, the first component of the eigenvalue equation, $\boldsymbol{A}\Phi = E\Phi$, is reduced to the following form,

$$\begin{aligned}(\varepsilon - E)a + \sum_{n=-\infty}^{\infty} \Bigg[& \left(\frac{1}{2}\{k_3 - k_2 - n(k_1 - k_2)\}\beta_{n+1}{}^{(2)} + k_2 \beta_{n+1}{}^{(1)}\right) k_- \\ & + \left(\frac{1}{2}\{k_3 - k_2 + (n+1)(k_1 - k_2)\}\beta_n{}^{(2)} + k_2 \beta_n{}^{(1)}\right) k_+ \Bigg] b'_n = 0,\end{aligned} \tag{23}$$

in which $b_n' = -iq_0 b_n$ and $\beta_n{}^{(i)} = \alpha_n{}^{(i)} q_0{}^{1-i}$. Similarly from the second component of the eigenvalue equation we obtain

$$\begin{aligned}&\Bigg[\left(\frac{1}{2}\{k_1 - 2k_2 + k_3 - m(k_1 - k_2)\}\beta_m{}^{(2)} + k_2 \beta_m{}^{(1)}\right) k_- \\ &\quad + \left(\frac{1}{2}\{k_1 - 2k_2 + k_3 + (m-1)(k_1 - k_2)\}\beta_{m-1}{}^{(2)} + k_2 \beta_{m-1}{}^{(1)}\right) k_+ \Bigg] a \\ &\quad + \sum_{n=-\infty}^{\infty} \Big[\{k_3 - 2k_2 - (2m-1)(2n+1)k_1\}\beta_{m+n}{}^{(3)} + 2k_2 \beta_{m+n}{}^{(2)} \\ &\quad + \frac{1}{4(q_0\xi)^2}\{2k_2 \beta_{m+n}{}^{(1)} + (2k_1 - k_2)(\beta_{m+n+1}{}^{(1)} + \beta_{m+n-1}{}^{(1)})\}\Big] b_n{}' = 0.\end{aligned} \tag{24}$$

The eigenvalue Eqs. (23) and (24) are rewritten in the form

$$\boldsymbol{B}\ \boldsymbol{a} = 0, \tag{25}$$

where the transpose of $\boldsymbol{a}$ is given by $(a, b_0', b_{-1}', b_1', b_{-2}', b_2', ...)$. Thus, we obtain a secular equation

$$\det \boldsymbol{B} = 0, \tag{26}$$

from which the eigenvalue E is derived.

EIGENVALUE CALCULUS IN WEAK FIELD EXPANSION

To obtain the eigenvalue E, the calculation of $\alpha_n{}^{(i)}$ (that is, $\beta_n{}^{(i)}$) is required. Though it is difficult in general to carry out the calculation of Eq. (19) practically, we can obtain $\beta_n{}^{(i)}$ in expansion forms of the field. From Eq. (5) ξ is related to κ by

$$\xi^{-1} = q_0\kappa + q_0\kappa^3/4 + \cdots, \tag{27}$$

and as an expansion parameter κ is utilized here. Then, $\theta(u)$ is rewritten as

$$\theta(u) = u + u_2\ \kappa^2 + u_4\ \kappa^4 + \cdots, \tag{28}$$

in which u_2 and u_4 are given, respectively, by

$$u_2 = (\sin 2u - 2u)/8, \tag{29}$$

$$u_4 = \sin 4u/256 + \sin 2u/16 - u(4\cos 2u + 5)/64. \tag{30}$$

Similarly, we obtain up to the order κ^4

$$\begin{aligned} \mathrm{dn}\, u = 1 &+ \kappa^2(\cos 2u - 1)/4 \\ &+ \kappa^4(\cos 4u + 4\cos 2u - 5 + 8u \sin 2u)/64. \end{aligned} \tag{31}$$

By using Eqs. (27)–(31) together with the expansion forms of $E(\kappa)$, calculations of $\beta_n^{(i)}$ are carried out.

By careful examination of element B_{mn} in Eq. (26) about the order of κ, the secular Eq. (26) is reduced up to the order κ^4 as

$$\det \boldsymbol{B}_5 = 0, \tag{32}$$

where $\boldsymbol{B}_5$ is 5×5 matrix composed of B_{mn} with indices $1 \le m, n \le 5$. The elements are given explicitly as

$$B_{11} = \varepsilon - E, \tag{33}$$

$$B_{22} = B_{33} = (k_1 - 2k_2 + k_3)\beta_0{}^{(3)} + 2k_2\beta_0{}^{(2)} + \frac{\kappa^2}{2}k_2\beta_0{}^{(1)}, \tag{34}$$

$$B_{44} = B_{55} = (9k_1 - 2k_2 + k_3)\,{\beta_0}^{(3)} + 2k_2\,{\beta_0}^{(2)} + \left(\kappa^2 + \frac{\kappa^4}{2}\right)\left\{\frac{1}{2}k_2\,{\beta_0}^{(1)} + \left(k_1 - \frac{1}{2}k_2\right){\beta_1}^{(1)}\right\}, \tag{35}$$

$$B_{12} = B_{31} = \left\{\frac{1}{2}(k_1 - 2k_2 + k_3)\,{\beta_0}^{(2)} + k_2\,{\beta_0}^{(1)}\right\}k_+ + \left\{\frac{1}{2}(-k_2 + k_3)\,{\beta_1}^{(2)} + k_2\,{\beta_1}^{(1)}\right\}k_-, \tag{36}$$

$$B_{13} = B_{21} = \left\{\frac{1}{2}(-k_2 + k_3)\,{\beta_1}^{(2)} + k_2\,{\beta_1}^{(1)}\right\}k_- + \left\{\frac{1}{2}(-k_2 + k_3)\,{\beta_1}^{(2)} + k_2\,{\beta_1}^{(1)}\right\}k_+, \tag{37}$$

$$B_{14} = B_{51} = \left\{\frac{1}{2}(2k_1 - 3k_2 + k_3)\,{\beta_1}^{(2)} + k_2\,{\beta_1}^{(1)}\right\}k_+ + \left\{\frac{1}{2}(-k_2 + k_3)\,{\beta_2}^{(2)} + k_2\,{\beta_2}^{(1)}\right\}k_-, \tag{38}$$

$$B_{15} = B_{41} = \left\{\frac{1}{2}(2k_1 - 3k_2 + k_3)\,{\beta_1}^{(2)} + k_2\,{\beta_1}^{(1)}\right\}k_- + \left\{\frac{1}{2}(-k_2 + k_3)\,{\beta_2}^{(2)} + k_2\,{\beta_2}^{(1)}\right\}k_+, \tag{39}$$

$$B_{23} = B_{32} = (-k_1 - 2k_2 + k_3)\,{\beta_1}^{(3)} + 2k_2\,{\beta_1}^{(2)} + \left(\kappa^2 + \frac{\kappa^4}{2}\right)\left\{\frac{1}{2}k_2\,{\beta_1}^{(1)} + \left(\frac{1}{2}k_1 - \frac{1}{4}k_2\right){\beta_0}^{(1)}\right\}, \tag{40}$$

$$B_{24} = B_{42} = B_{35} = B_{53} = (3k_1 - 2k_2 + k_3)\,{\beta_1}^{(3)} + 2k_2\,{\beta_1}^{(2)} + \left(\kappa^2 + \frac{\kappa^4}{2}\right) \times \left\{\frac{1}{2}k_2\,{\beta_1}^{(1)} + \left(\frac{1}{2}k_1 - \frac{1}{4}k_2\right){\beta_0}^{(1)}\right\}, \tag{41}$$

$$B_{25} = B_{52} = B_{34} = B_{43} = (-3k_1 - 2k_2 + k_3)\,{\beta_2}^{(3)} + 2k_2\,{\beta_2}^{(2)} + \kappa^2\left(\frac{1}{2}k_1 - \frac{1}{4}k_2\right){\beta_1}^{(1)}, \tag{42}$$

$$B_{45} = B_{54} = O(\kappa^6). \tag{43}$$

Here, ${\beta_n}^{(i)}$'s are written explicitly in the expansion forms of κ as

$$ {\beta_n}^{(1)} = \delta_{n.0} + 1/8\,\delta_{n.\pm1}\kappa^2 + (1/16\,\delta_{n.\pm1} - 1/128\,\delta_{n.\pm2})\kappa^4 + \cdots, \quad (44) $$

$$ {\beta_n}^{(2)} = \delta_{n.0} + 1/4\,\delta_{n.\pm1}\kappa^2 + (1/32\,\delta_{n.0} + 1/8\,\delta_{n.\pm1})\kappa^4 + \cdots, \quad (45) $$

$$ \begin{aligned} {\beta_n}^{(3)} = {} & \delta_{n.0} + 3/8\,\delta_{n.\pm1}\kappa^2 \\ & + (3/32\,\delta_{n.0} + 3/16\,\delta_{n.\pm1} + 3/128\,\delta_{n.\pm2})\kappa^4 + \cdots. \end{aligned} \quad (46) $$

Eventually, the eigenvalue E, which is nothing but the excess free energy, is derived from Eq. (32) as

$$ E = \left\{\frac{k_2}{2}\kappa^2 + \left(\frac{5}{16}k_2 - \frac{{k_2}^2}{2(k_1+k_3)}\right)\kappa^4\right\}k_y^2 + \frac{k_2(1-\kappa^2)K^2(\kappa)}{E^2(\kappa)}k_z^2, \quad (47) $$

or, in terms of nondimensional field $h(=(\chi_a/k_2)^{1/2}H/q_0)$,

$$ E = \left\{\frac{k_2}{2}h^2 + \left(\frac{1}{16}k_2 - \frac{{k_2}^2}{2(k_1+k_3)}\right)h^4\right\}k_y^2 + \left(1 - \frac{1}{8}h^4\right)k_2k_z^2, \quad (48) $$

where we use

$$ \kappa = h - \frac{1}{4}h^3 + \cdots. \quad (49) $$

STABILITY OF CHOLESTERIC PHASE

While the term proportional to k_x^2 vanishes, the one proportional to ${k_y}^2$ is shown to persist in E so long as the field does not vanish, that stabilizes the cholesteric phase [3]. Here, we mention this briefly. In the absence of the magnetic field terms proportional to $k_\perp^4$ and ${k_\perp}^2k_z^2$ appear together with k_z^2 [1]. So, the following form is derived,

$$ E = d_1{k_y}^2 + d_2{k_z}^2 + d_3{k_\perp}^4 + d_4{k_\perp}^2{k_z}^2, \quad (50) $$

where the coefficients d_1 and d_2 are given in Eq. (47), and d_3 and d_4 are

$$ d_3 = {q_0}^{-2} + O(\kappa^2), \quad (51) $$

$$ d_4 = {q_0}^{-2} + O(\kappa^2). \quad (52) $$

The form of Eq. (50) is common for any values of the elastic constants, k_1, k_2 and k_3, and accordingly an estimate of auto-correlation function of $\Phi(r)$ is obtained by the same method of the previous article [3],

$$<|\Phi|^2> \simeq k_{\rm B}T \int \frac{d^3k}{E}, \tag{53}$$

which is shown to be bounded in the zero limit of wave number k_0 that is the inverse of the system size.

CONCLUSION

The fluctuation spectrum of the cholesteric phase exposed to the magnetic field with positive anisotropy is studied on the basis of the Frank's elastic theory with the general values of three Frank's elastic constants. Euler-Lagrange equation for the fluctuation is reduced to an eigenvalue equation, from which the eigenvalue E is calculated here up to the order H^4 of the external field H. The stability of the cholesteric phase with respect to the long wave fluctuation is certified irrespective of values of elastic constants.

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