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RESEARCH ARTICLE

Landau theory for isotropic, nematic, smectic-A, and smectic-C phases

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We use the Landau theory of phase transitions to describe the phase diagram of a liquid crystal displaying the isotropic (i), nematic (N), smectic-A and smectic-C phases. The order parameter of the smectic-C phase is defined as the projection of the director on the plane of the smectic layers, vanishing in the smectic-A phase. We present a detailed phase diagram that shows transition between any two of these phases, containing a triple point INA, a Lifshitz point NAC, a tricritical at the NA line, and a critical end point IAC. As one approaches the NC line from the smectic-C phase, the tilt angle approaches a nonzero value, but if the AC line is approached, the tilt angle vanishes according to distance to the AC line to the power 1/2.

Keywords: phase transitions; Landau theory; nematic; smectic-A; smectic-C

1. Introduction

Molecular theories can successfully describe phase transitions occurring in liquid crystals [1–13]. The same can be said about the Landau theory of phase transitions [14], which is close related to the molecular theories [15, 16], and has also been applied to construct phase diagrams of liquid crystals [17–25]. The Landau theory consists in setting up a polynomial form for the free energy in terms of order parameters that describe the possible thermodynamic phases. Each term appearing in the polynomial is invariant under all transformations that leave the corresponding phases unaffected. Thus, the determining factor in the establishment a Landau free energy concerns the appropriate definition of the order parameter of each phase together with its symmetry operations.

Here we set up a Landau free energy to describe the phase diagram that includes the isotropic, the nematic, the smectic-A and the smectic-C phases. The phase diagram that includes the nematic, the smectic-A and the smectic-C, including the multicritical point were the the transition lines NA, NC and AC meet, were investigated experimentally, [26–30] and studied by molecular theories and other mean-field theories [28–41]. The nematic phase that concerns us here is the uniaxial nematic consisting of rod like molecules that are ordered in a given direction but without positional order. If the positional order is partially restored in such a way that the system is structured in layers with nematic order inside each layer, then the liquid crystal is a smectic. The two types of smectic states that will be treated here are the smectic-A and smectic-C. The former occurs when the orientation of the nematic inside the layers, described by the director $\vec{n}$, is normal to the smectic layers, and the latter when $\vec{n}$ is is tilted at an angle θ with respect to the normal.

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Phase transitions between all phases, isotropic (I), nematic (N), smectic-A, and smectic-C may occur. The discontinuous IN transition is described by the molecular theory introduced by Maier and Saupe [1–3]. The molecular theory of MacMillan [5] predicts for the NA transition a continuous phase transition with a tricritical point. Beyond the tricritical point, the NA phase transition is discontinuous. The AC transition was considered by de Gennes [42] who introduced the order parameter for the smectic-C phase, and also predict a directed NC transition. Ginzburg-Landau mean-field theories shown a continuous AC phase transition and a Lifshitz point at the point where the NA, NC, and AC lines meet [31, 32].

The phase transitions between the three phases, nematic, smectic-A and smectic-S was studied by the use a Landau-Ginzburg mean-field theory by Chen and Lubensky [31] and others [35, 36, 38] This theory gives a NA transition, a NC transition and a Lifshitz point at the boundary of these two transitions. Beyond the mean-field theory, the AC transition was studied by a theory that takes into account fluctuations by Grinstein and Pelcovitz who argued that the AC transitions belongs in the universality class of the three-dimensional XY model [32], a result first suggested by de Gennes [42]. Theories of the Landau type for the NAC phases containing two order parameter were developed [28, 33, 34, 37, 39] and they confirmed the existence of a multicritical point where the three phases meet, called a Lifshitz point. Our approach, including the phase diagram, treats not only these thre phases, nematic, smectic-A and smectic-C, but also the isotropic phase.

2. Landau free energy

The Landau free energy describing a nematic phase is written in terms of the invariants of de Gennes traceless tensor order parameter Q [19]. For uniaxial nematics, the tensor Q can be written in terms of the director $\vec{n}$, which is a unit vector parallel to the axis that defines the nematic state. The tensor Q is given by

$$Q = \frac{q}{2}(3\vec{n}\vec{n} - I). \tag{1}$$

If we choose a Cartesian frame of reference such that the director is parallel to the z-direction then the tensor Q is diagonal. The diagonal entries are $Q_{11} = Q_{22} = -q/2$, $Q_{33} = q$. The tensor Q describes a uniaxial nematic. If $q > 0$, it describes a rod like object whose axis is parallel to $\vec{n}$; and if $q < 0$, it describes a disc like object whose axis is parallel to $\vec{n}$. When $q = 0$, the phase is isotropic.

The invariants of order two and three are, respectively, $I_2 = \operatorname{Tr} Q^2 = 3q^2/2$ and $I_3 = \operatorname{Tr} Q^3 = 3q^3/4$. The Landau free energy that describes the nematic and isotropic phases is written as a polynomial of the fourth degree,

$$f_n = a_2 q^2 + a_3 q^3 + a_4 q^4, \tag{2}$$

where a_2, a_3, and a_4 are parameters, and $a_4 > 0$ as required by stability of the nematic phase.

The smectic phases are characterized by modulation in density. Up to the first harmonic component, the density $\rho(\vec{r})$ at point $\vec{r}$ is written as

$$\rho = \rho_0 + \sigma \cos(\vec{k} \cdot \vec{r} + \varphi), \tag{3}$$

where $\sigma > 0$ is the density modulation amplitude, $\vec{k}$ is the wave vector, and φ is a phase shift. The wave vector $\vec{k}$, which determines the direction of modulation in space is normal to the smectic layers, as shown in figure 1. When the director $\vec{n}$ is parallel to $\vec{k}$, the density (3) describes a smectic-A liquid crystal. In this case, according to de Gennes [19], the smectic-A order parameter

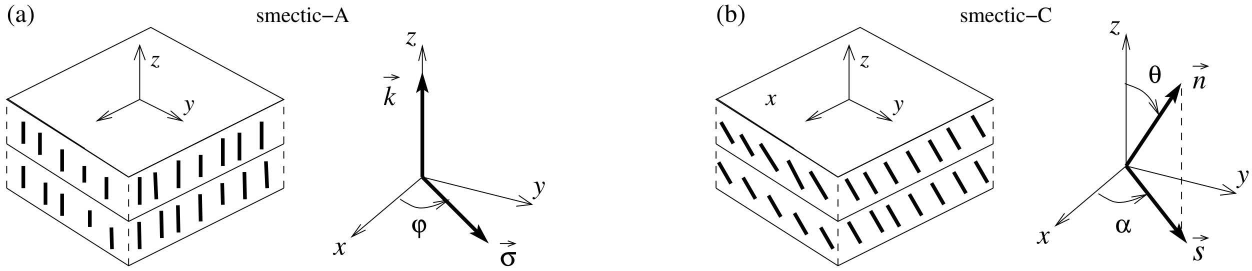

Figure 1. The Cartesian frame (x, y, z) is chosen in such a way that the z-direction is normal to the smectic layers and thus parallel to the wave vector $\vec{k}$ which determines the direction of space modulation. (a) The azimuthal angle φ is identified as the phase shift. The vector $\vec{\sigma}$ is the order parameter of the smectic-A phase and lies in the xy plane. (b) The vector $\vec{n}$ is the director which is tilted by an angle θ, and α is the azimuthal angle. The order parameter of the smectic-C phase is the vector $\vec{s}$ which is the projection of $\vec{n}$ onto the plane xy so that $s = \sin\theta$.

is described by the two-dimensional vector

$$\vec{\sigma} = \sigma \cos\varphi\, \hat{x} + \sigma \sin\varphi\, \hat{y}. \tag{4}$$

Any translation along the direction of modulation leaves the free energy invariant. Therefore, the free energy should not depend on the phase shift φ and the same is true for the invariants. The invariant of the lowest order is $\psi\psi^* = \sigma^2$. The other invariants are just powers of σ^2. A Landau free energy that describes a transition from a smectic state to a state with $\sigma = 0$, that might be the isotropic or the nematic, is given by the fourth degree polynomial

$$f_a = b_{20}\sigma^2 + b_{40}\sigma^4. \tag{5}$$

where b_{20} and b_{40} are parameters, and $b_{40} > 0$ as required by stability of the smectic-A phase.

In the smectic-C phase the director $\vec{n}$ is tilted by an angle θ with respect to the normal to the smectic layers, as shown in figure 1. The order parameter $\vec{\zeta}$ of the smectic-C phase is defined by $\vec{\zeta} = \sigma\vec{s}$ where $\vec{s}$ is the projection of the director $\vec{n}$ onto the plane of the smectic layers, as illustrated in figure 1. Any rotation around the z-axis leaves the free energy invariant which means that the free energy does not depend on the azimuthal angle α. Thus any invariant of the smectic-C order parameter does not depend on α, which implies that the invariants are powers of $\zeta^2 = s^2\sigma^2$, where $s = \sin\theta$. A Landau free energy that describes the smectic-C is

$$f_c = b_{22}s^2\sigma^2 + b_{44}s^4\sigma^4, \tag{6}$$

where b_{22} and b_{44} are parameters and $b_{44} > 0$ as required by the stability of the smectic-C phase.

To describe the global phase diagram we need terms in the Landau free energy that couple the order parameters of the possible phases. A term that couple two types of order parameters should be invariant under the symmetry operations related to the two order parameters. The smectic-A and the smectic-C phases are coupled by the invariants $s^2\sigma^4$ and $s^4\sigma^2$. The nematic and the smectic-A and smectic-C phases are coupled by the invariants $q^2\sigma^2$ and $q^2\sigma^2 s^2$, respectively. The Landau free energy is then written as a sum of the free energies (2), (5), and (6), plus the coupling terms,

$$f = f_0 + a_2 q^2 + a_3 q^3 + a_4 q^4$$

$$+(b_{20} + b_{22}s^2 + b_{24}s^4)\sigma^2 + (b_{40} + b_{42}s^2 + b_{44}s^4)\sigma^4 - (c_4 + d_4 s^2)q^2\sigma^2. \tag{7}$$

The parameters a_i, b_{ij}, c_4 and d_4 are understood as thermodynamic fields or functions of thermodynamic fields. Since $s = \sin\theta$, the free energy f is understood as a function of q, σ and θ.

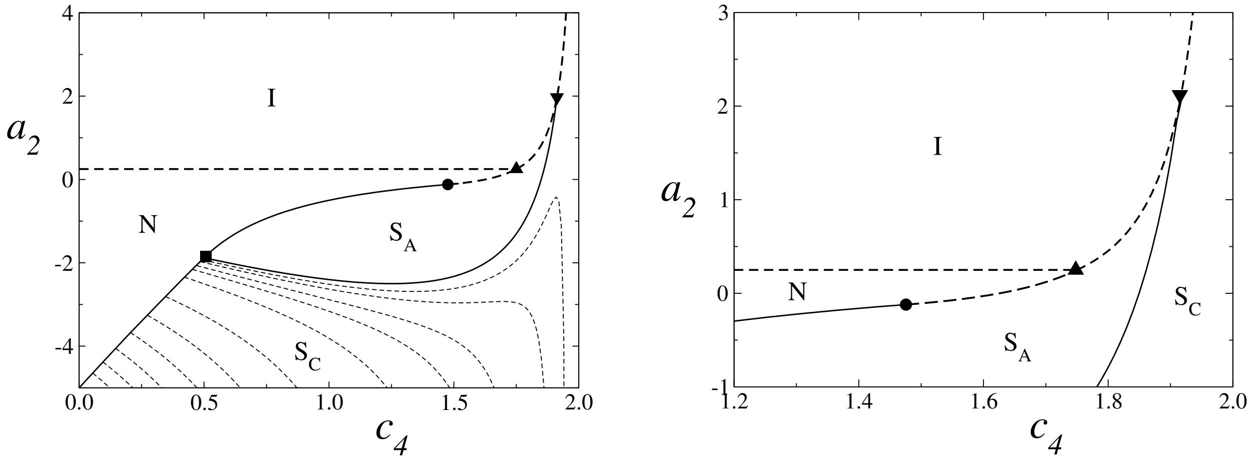

Figure 2. Phase diagram in the $c4$-$a2$ plane displaying the thermodynamic phases: isotropic (I), nematic (N), smectic-A, and smectic-C. The right panel is an enlargement of the left panel. The heavy dashed lines and the solid lines represent discontinuous and continuous phase transitions, respectively. The symbols represent a tricritical point (full circle), a triple point (up triangle), a critical end point (down triangle) and a Lifshitz point (square). The light dashed lines are lines of constant tilt angle.

The possible thermodynamic phases that can be described by the Landau free energy (7) are:
a) $q = 0$ and $\sigma = 0$, isotropic;
b) $q \neq 0$, $\sigma = 0$, nematic;
c) $q \neq 0$, $\sigma \neq 0$, $s = 0$, smectic-A;
d) $q \neq 0$, $\sigma \neq 0$, $s \neq 0$, smectic-C.
Notice that f_0 in equation (7) is identified as the free energy of the isotropic phase, because this phase is characterized by $q = 0$ and $\sigma = 0$

3. Phase diagram

The stability of the several phases, alone or in coexistence, requires that the actual free energy be the convex hull of the expression of the free energy (7). The construction of the convex hull is equivalent to minimize this expression. Thus, using a minimization procedure we find the possible thermodynamic phases and the transition lines between them, as well as the triple points and the multicritical points. In figure 2 we show the phase diagram in the plane of the parameters c_4 and a_2. The other parameters are set as follows: $a_3 = a_4 = b_{20} = b_{24} = b_{40} = b_{42} = b_{44} = d_4 = 1$ and $b_{22} = 2$. As seen in figure 2, the phase diagram displays the four phases: I, N, $\mathrm{S_A}$, and $\mathrm{S_C}$. The transition from the I phase to the other phases are always discontinuous. The N phase is separated from the $\mathrm{S_A}$ either by a discontinuous or by a continuous transition. The transition between the $\mathrm{S_A}$ and $\mathrm{S_C}$ is continuous. In addition, the phase diagram displays multicritical points and triple point. In the following we show how the several transition lines were determined.

3.1 *Transition I-N*

In this case it suffices to consider the free energy

$$f = f_0 + a_2 q^2 + a_3 q^3 + a_4 q^4. \tag{8}$$

Minimization of above equations gives

$$2a_2 + 3a_3 q + 4a_4 q^2 = 0. \tag{9}$$

At the I-N transition, which is discontinuous, the nematic free energy should be equal to the isotropic free energy, which equals f_0, leading to the equation

$$a_2 + a_3 q + a_4 q^2 = 0. \tag{10}$$

Replacing the nonzero solution of (9) in (10), we get

$$a_2 = \frac{a_3^2}{4a_4}, \tag{11}$$

which describes the I-N transition, as shown in figure 2. The jump in the order parameter along the transition line is $q = |a_3|/2a_4$.

3.2 *Transition I-S*$_\mathrm{A}$

In both phases, I and $\mathrm{S_A}$, s vanishes and the Landau free energy is reduced to

$$f = f_0 + a_2 q^2 + a_3 q^3 + a_4 q^4 + b_{20}\sigma^2 + b_{40}\sigma^4 - c_4 q^2 \sigma^2. \tag{12}$$

The minimization leads to the equations

$$2a_2 + 3a_3 q + 4a_4 q^2 - 2c_4\sigma^2 = 0, \tag{13}$$

$$b_{20} - c_4 q^2 + 2b_{40}\sigma^2 = 0, \tag{14}$$

whose solution gives q and σ of the $\mathrm{S_A}$ phase. At the transition, the free energies of both phases are equal, which gives

$$(4a_4 b_{40} - c_4^2)q^4 + 4a_3 b_{40} q^3 + 2(2a_2 b_{40} + b_{20}c_4)q^2 - b_{20}^2 = 0, \tag{15}$$

where we have taken into account that the free energy of the I phase is f_0. These three equations give parametrically the line of transition I-$\mathrm{S_A}$, which is a discontinuous transition as shown in figure 2. The order parameters q and σ jump from a finite values at $\mathrm{S_A}$ to zero values at the isotropic phase.

3.3 *Transition I-S*$_\mathrm{C}$

The minimization of the full Landau free energy (7) gives

$$2a_2 + 3a_3 q + 4a_4 q^2 - 2(c_4 + d_4 s^2)\sigma^2 = 0, \tag{16}$$

$$(b_{20} + b_{22}s^2 + b_{24}s^4) + 2(b_{40} + b_{42}s^2 + b_{44}s^4)\sigma^2 - (c_4 + d_4 s^2)q^2 = 0, \tag{17}$$

$$b_{22} + 2b_{24}s^2 + (b_{42} + 2b_{44}s^2)\sigma^2 - d_4 q^2 = 0. \tag{18}$$

At the transition, the free energy (7) equals the isotropic free energy f_0 and we get

$$a_2 q^2 + a_3 q^3 + a_4 q^4 + (b_{20} + b_{22}s^2 + b_{24}s^4)\sigma^2 + (b_{40} + b_{42}s^2 + b_{44}s^4)\sigma^4 - (c_4 + d_4 s^2)q^2\sigma^2 = 0. \tag{19}$$

The discontinuous transition line, shown in figure 2, is found by replacing into (19) the values of q, σ and s obtained by solving the equations (16), (17), (18).

3.4 *Transition N-S*$_{\mathrm{A}}$

The minimization of the nematic N free energy,

$$f_n = f_0 + a_2 q_n^2 + a_3 q_n^3 + a_4 q_n^4, \tag{20}$$

gives

$$2a_2 + 3a_3 q_n + 4a_4 q_n^2 = 0. \tag{21}$$

The free energy of the smectic-A phase is

$$f_a = f_0 + a_2 q_a^2 + a_3 q_a^3 + a_4 q_a^4 + b_{20}\sigma^2 + b_{40}\sigma^4 - c_4 q^2 \sigma^2, \tag{22}$$

and its minimization gives

$$2a_2 + 3a_3 q_a + 4a_4 q_a^2 - 2c_4\sigma^2 = 0, \tag{23}$$

$$b_{20} - c_4 q_a^2 + 2b_{40}\sigma^2 = 0. \tag{24}$$

The transition N-S$_{\mathrm{A}}$ can be continuous or discontinuous. The continuous transition is obtained by first taking the limit $\sigma \to 0$ in equation (24) to get

$$b_{20} - c_4 q_a^2 = 0. \tag{25}$$

At the continuous transition, $q_n = q_a$, which together with equations (21) and (23) give the critical line. To find the discontinuous transition line, we solve the equation (21) for q_n, and the equations (23) and (24) for q_s and σ. After, we replace them into the equation $f_n = f_s$ to get the discontinuous transition line. The continuous and the discontinuous lines are separated by a tricritical point as shown in figure 2.

3.5 *Transition N-S*$_{\mathrm{C}}$

This is a continuous transition in the sense that q is continuous and σ vanishes continuously. The tilt angle θ on the other hand has a finite value at the transition and so does $s = \sin\theta$. We may in an equivalent way say that s has a jump from a nonzero at the smectic-C phase to a zero value at the nematic phase. The equations that minimizes the full Landau free energy were already obtained and are given by equations (16), (17), and (18). If we let $\sigma = 0$, in these three equations we find

$$2a_2 + 3a_3 q + 4a_4 q^2 = 0, \tag{26}$$

$$b_{20} + b_{22}s^2 + b_{24}s^4 - (c_4 + d_4 s^2)q^2 = 0, \tag{27}$$

$$b_{22} + 2b_{24}s^2 - d_4 q^2 = 0. \tag{28}$$

The elimination of q and s in these equations gives the transition line shown in figure 2. Along the N-S_C transition line, s varies and vanishes at the Lifshitz point. Denoting by c_4^* the value of c_4 at the Lifshitz point, the behavior of s along the transition line near the Lifshitz point is $s \sim (c_4^* - c_4)^{1/2}$. Taking into account that $s = \sin\theta$, the same behavior is valid for the tilt angle, $\theta \sim (c_4^* - c_4)^{1/2}$.

3.6 *Transition S_A-S_C*

Let us write the full Landau free energy (7) in the form

$$f = f_a + (b_{22} + b_{42}\sigma^2 - d_4 q^2)\sigma^2 s^2 + (b_{24} + b_{44}\sigma^2)\sigma^2 s^4, \tag{29}$$

where f_a, understood as the free energy of the smectic-A phase, is given by

$$f_a = f_0 + a_2 q^2 + a_3 q^3 + a_4 q^4 + b20\sigma^2 + b_{40}\sigma^4 - c_4 q^2\sigma^2. \tag{30}$$

In phase S_C, the order parameters q, σ, and s are nonzero. In phase S_A, q and σ are also nonzero but s is zero. As one approaches the S_A-S_C transition line from the S_C phase, the order parameter s vanishes continuously. The transition point occurs when the coefficient of s^2 of the free energy (29) vanishes, that is, when

$$b_{22} + b_{42}\sigma^2 - d_4 q^2 = 0. \tag{31}$$

At the transition line, q and σ are determined by the minimization of f_a, that is,

$$2a_2 + 3a_3 q + 4a_4 q^2 - 2c_4\sigma^2 = 0, \tag{32}$$

$$b_{20} - c_4 q^2 + 2b_{40}\sigma^2 = 0. \tag{33}$$

To find the transition line S_A-S_C it suffices to eliminate q and σ from equations (31), (32), and (33). The result is the continuous transition line shown if figure 2.

In figure 2, we have also drawn lines of constant tilt angle θ. Along each one of these lines, the amplitude σ varies and vanishes as one approaches the N-S_C line. On the other hand, if one approaches the S_A-S_C line from the smectic-C region, the tilt angle θ vanishes, but the amplitude σ remains finite. As one approaches the AC line from the smectic-C the tilt angle vanishes as

$$\theta \sim \varepsilon^\beta \tag{34}$$

where $\varepsilon = c_4^* - c_4$ is the distance from the AC line and $\beta = 1/2$. Indeed, there are experimental investigations on the AC transitions showing that the order parameter exponents $\gamma = 0.5$ [43–45]. However, there are experiments showing exponents smaller that this mean-field exponent and closer to the prediction of the de Gennes model for the AC transition [42, 45].

4. Conclusion

In conclusion, we have set up a Landau free energy that describes the isotropic, nematic, smectic-A and smetic-C phases. This was accomplished by using invariants of the order parameters of the several phases. The order parameter of the smectic-C phase was defined in a way similar to that of the smectic-A phase. From the minimization of the free energy we obtained the phase transition lines and constructed the phase diagram. The topology of this diagram is as follows. The IN, IA, IC are lines of discontinuous phase transition. The NA transition line is continuous up to the tricritical point when it becomes discontinuous. The NC is continuous with a vanishing tilt angle along the line, but it vanishes as one approaches the Lifshitz point where the line NC meets the AC line. The AC transition line is continuous with a vanishing tilt angle along the line.

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