

# Simple Landau model of the smectic-A–isotropic phase transition

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**Abstract.** A simple Landau-type free energy function is presented to describe the smectic  $A$  - isotropic phase transition. Varying the coupling between orientational and positional order parameters a smectic  $A$  - isotropic or a nematic - isotropic phase transition occurs. Within this model the smectic  $A$  - isotropic phase transition is found to be always more strongly first order than the nematic - isotropic phase transition. The theoretical results are found to be in good agreement with all published experimental results.

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## 1 Introduction and motivation

Although the nematic-isotropic (NI) phase transition has been a topic of active theoretical and experimental studies over the past few decades [1], reports on the smectic  $A$  - isotropic (AI) transition are comparatively scarce. These include experimental [2–5] and theoretical [4, 6–9] studies.

The pretransitional behavior of dodecylcyanobiphenyl (12CB) exhibits a direct AI transition [2] which is different from those of NI transition. The existence of smectic  $A$  (smA) type cybotactic groups in the isotropic phase is detected with differential scanning calorimetry and low angle x-ray measurements. The high nonlinearity in a AI phase transition region is obtained due to the formation of the cybotactic groups of molecules. Ocko et al [3] measured the smectic ordering induced by the free surface in the isotropic phase of 12CB, which does not have a nematic phase but undergoes a first order transition from an isotropic to a smA phase at  $T_{AI}=57.7^\circ$  C, where  $T_{AI}$  is the AI transition temperature. Olbrich et al [4] measured the strain birefringence above a AI phase transition in liquid crystalline side-chain elastomers under an external static mechanical stress. They pointed out that both the nematic and smectic fluctuations become relevant above the AI phase transition. The same pretransitional behavior for the AI transition is also obtained in 10CB and 12CB [5, 10]. All the experimental observations described above show that the AI transition is more strongly first order than the NI transition, which is known to be very weakly first order. This indicates that the orientational order in the smA phase is much higher than that in the nematic phase. The vicinity of a smA phase has a noticeable influence on the pretransitional phenomena in the isotropic

phase. The value of the temperature difference  $T_{AI} - T^*$  is also higher than that of  $T_{NI} - T^*$ . Here  $T^*$  is the absolute stability limit of the isotropic phase. Below this temperature no stable isotropic phase is possible.

However there is no systematic theoretical study on the AI transition. Based on their lattice model Ronis and Rosenblatt [6] suggested that a direct AI transition is possible. In a separate paper Rosenblatt [7] predicted that in the presence of an intense magnetic field, materials, which in the absence of any field undergo a direct AI transition, can exhibit an intermediate nematic phase. Lelidis and Durand [9] described the AI transition based on a Landau model. We note, however, that there is a problem with their model which will be discussed below.

In none of the theoretical studies undertaken so far the key features of the AI transition were characterized. Thus it is interesting to see under what conditions the AI transition is more favorable than a NI transition. The purpose of the present note is to study the direct AI transition for the bulk state within the framework of a Landau phenomenological model.

## 2 Model

The starting point of our approach is to write down the Ginzburg-Landau free energy functional  $F$ . The nematic order parameter originally proposed by de Gennes [1] is a symmetric, traceless tensor described by  $Q_{ij} = \frac{1}{2}S(3n_i n_j - \delta_{ij})$ . The quantity  $S$  defines the strength of the nematic ordering (the modulus of the nematic order parameter) and is zero (one) for complete disorder (order). Thus in the isotropic phase  $S = 0$  and in the nematic phase  $S \neq 0$ . The smectic order parameter  $\psi(\mathbf{r}) = \psi_0 \exp(-i\phi)$  is a complex scalar quantity whose modulus  $\psi_0$ , is defined as the

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amplitude of a one dimensional density wave characterized by the phase  $\phi$ . The wave vector  $\nabla_i\phi$  is parallel to the director  $n_i$  in the smectic A phase. The layer spacing is given by  $d = 2\pi/q_0$  with  $q_0 = |\nabla\phi|$ .

Keeping homogeneous terms up to quartic and gradients only to the lowest relevant order, the total free energy near the AI transition can be written as:

$$F = F_0 + \int \left[ \frac{1}{2} A Q_{ij} Q_{ij} - \frac{1}{3} B Q_{ij} Q_{jk} Q_{ki} + \frac{1}{4} C_1 (Q_{ij} Q_{ij})^2 + \frac{1}{4} C_2 Q_{ij} Q_{jk} Q_{kl} Q_{li} + \frac{1}{2} \alpha |\psi|^2 + \frac{1}{4} \beta |\psi|^4 + \frac{1}{2} \delta |\psi|^2 Q_{ij} Q_{ij} + \frac{1}{2} b_1 |\nabla_i \psi|^2 + \frac{1}{2} b_2 |\Delta \psi|^2 + \frac{1}{2} e_1 Q_{ij} (\nabla_i \psi) (\nabla_j \psi^*) \right] dV \quad (1)$$

where  $F_0$  is the free energy of the isotropic phase,  $A = a(T - T_{NI}^*)$  and  $\alpha = \alpha_0(T - T_{AI}^*)$ .  $T_{NI}^*$  and  $T_{AI}^*$  are the critical temperatures for a hypothetical second order transition to the nematic and the smectic state, respectively, in the absence of any cross coupling. All other coefficients, as well as  $a$  and  $\alpha_0$ , are assumed to be constants near the transition point. There is a direct, bi-quadratic coupling between the nematic and smectic order governed by the coupling coefficient  $\delta$ . As we shall see, a negative value of  $\delta$  favors the smA phase over the nematic phase, whereas a positive  $\delta$  favors the nematic phase over the smA phase. Naturally there exists also a term which is cubic in  $Q_{ij}$  and quadratic in  $\psi$ ; this term is not considered in this paper, since it would be of fifth order while all other spatially homogeneous terms in eq.(1) are at most of fourth order. We assume  $C_1$ ,  $C_2$ ,  $\beta$  and  $\beta C - \delta^2$  (with  $C = C_1 + C_2/2$ ) and  $b_2$  to be positive to guarantee the stability of the isotropic, homogeneous phase at high temperatures and  $B > 0$  to get  $S > 0$  in the nematic phase. Some higher gradient terms involving  $Q_{ij}$  as well as second order derivatives of  $\psi$ , which have been disregarded in eq.(1), do not qualitatively change the physical picture, but will be discussed in an Appendix for completeness. The isotropic gradient terms in (1) guarantee a finite wavelength  $q_0$  for the smectic density wave. Symmetry would allow another term,  $(b_3/2)|\nabla_i \nabla_j \psi|^2$ , which however does not lead to any new contribution (compared to that  $\sim b_2$ ) for the smectic A phase and has therefore been omitted here. The gradient term  $\sim e_1$  involving  $Q_{ij}$  governs the relative direction of the layering with respect to the director.

There is no direct linear coupling term  $\sim |\psi|^2 S$  [9] in the free energy (1), since such a term cannot exist in the isotropic phase: Written in the full order parameter  $Q_{ij}$  it would read  $\xi_{ij} |\psi|^2 Q_{ij}$ , which however is identically zero, since the material tensor  $\xi_{ij}$  takes in the isotropic phase the form  $\xi_{ij} = \xi \delta_{ij}$  and  $Q_{ij}$  is traceless. However, such a coupling term is possible near the nematic - smectic A transition, since one has uniaxial symmetry on both sides of the transition.

Here we consider phases in which the nematic and smectic order are spatially invariant,  $S = const.$  and  $\psi_0 = const.$ , and for the smectic A phase a spatially constant wave vector  $q_0$  with the layering along the director. In that

case eq.(1) reads

$$F - F_0 = \int \left[ \frac{3}{4} A S^2 - \frac{1}{4} B S^3 + \frac{9}{16} C S^4 + \frac{1}{2} \alpha \psi_0^2 + \frac{1}{4} \beta \psi_0^4 + \frac{3}{4} \delta \psi_0^2 S^2 + \frac{1}{2} b_1 \psi_0^2 q_0^2 + \frac{1}{2} e_1 \psi_0^2 S q_0^2 + \frac{1}{2} b_2 \psi_0^2 q_0^4 \right] dV \quad (2)$$

Note that there is now an effective nematic-smectic coupling term linear in  $S$ . However it comes with a  $q_0^2$  factor and is zero in the non-smectic case. The presence of the cubic terms ( $\sim B$  and  $\sim e_1$ ) describes the first order character of the NI and AI transition. We note that a Landau energy of the same structure as the first line in eq.(2) has been used to describe the reentrant behavior in liquid crystals in [11] and for  $B \equiv 0$  in [12]. Minimization of Eq.(2) with respect to  $S$ ,  $\psi_0$  and  $q_0$  yields the following three phases:

$$\text{Isotropic : } S = 0, \quad \psi_0 = 0, \quad q_0 = 0 \quad (3)$$

$$\text{Nematic : } S_N = \frac{1}{6C} \left[ B + (B^2 - 24AC)^{1/2} \right] > 0, \\ \psi_0 = 0, \quad q_0 = 0 \quad (4)$$

$$\text{Smectic A : } S_A > 0, \quad \psi_0^2 = -\frac{1}{\beta} \left( \alpha^* - e^* S_A + \frac{3}{2} \delta^* S_A^2 \right), \\ q_0^2 = -\frac{1}{2b_2} (b_1 + e_1 S_A) \quad (5)$$

where  $S_A$  is defined by :

$$\frac{2\alpha^* e^*}{3\beta} + 2A^* S_A - B^* S_A^2 + 3C^* S_A^3 = 0$$

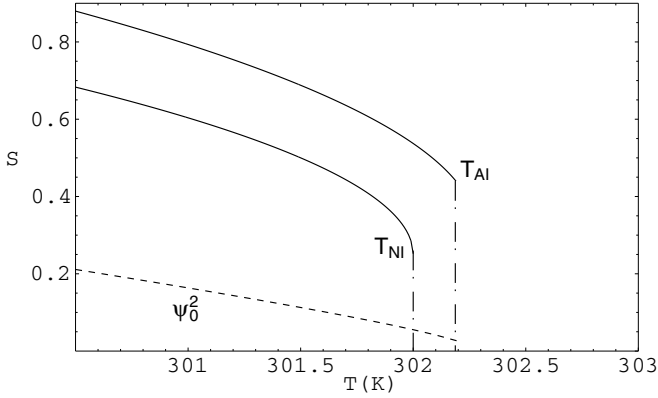
We use the abbreviations  $\alpha^* = \alpha - b_1^2/(4b_2)$ ,  $\delta^* = \delta - e_1^2/(6b_2)$ ,  $e^* = b_1 e_1/(2b_2)$  and  $C^* = C - \delta^{*2}/\beta$ ,  $B^* = B - 3e^* \delta^*/\beta$ , and  $A^* = A - \delta^* \alpha^*/\beta - e^{*2}/(3\beta)$ .

We show in Fig.1 the orientational order  $S_N$ ,  $S_A$  and the positional order  $\psi_0$  for the nematic and smA phases (4,5) plotted versus temperature. This is done for a set of parameters, where a direct isotropic to smA transition is possible. Fig.1 shows that both order parameters,  $S_A$  and  $\psi_0$  jump simultaneously at the AI transition point. We also see that orientational order in the smA phase is much higher than that of the nematic phase.

Necessary conditions for the different phases to be stable are ( $F = \int f dV$ )

$$\frac{\partial^2 f}{\partial S^2} > 0, \quad \frac{\partial^2 f}{\partial S^2} \cdot \frac{\partial^2 f}{\partial \psi_0^2} - \left( \frac{\partial^2 f}{\partial S \partial \psi_0} \right)^2 > 0 \\ \frac{\partial^2 f}{\partial \psi_0^2} > 0, \quad \frac{\partial^2 f}{\partial S^2} \cdot \frac{\partial^2 f}{\partial q_0^2} - \left( \frac{\partial^2 f}{\partial S \partial q_0} \right)^2 > 0, \\ \frac{\partial^2 f}{\partial q_0^2} > 0, \quad \frac{\partial^2 f}{\partial q_0^2} \cdot \frac{\partial^2 f}{\partial \psi_0^2} - \left( \frac{\partial^2 f}{\partial q_0 \partial \psi_0} \right)^2 > 0, \\ \text{and} \quad \det \left\| \frac{\partial^2 f}{\partial y_i \partial y_j} \right\| > 0 \quad (6)$$

were  $y_i \in \{S, \psi_0, q_0\}$  and  $i, j$  run from 1 to 3. The derivatives in (6) have to be taken at the values (3-5) for the



**Fig. 1.**  $S$  and  $\psi_0$  versus temperature. The upper (lower) solid line represents the orientational order parameter  $S_A$  ( $S_N$ ), while the dashed line represents the positional order parameter  $\psi_0^2$  in the smA phase. The values of the parameters in eq.(2) were taken to be  $\alpha_0 = .1J/K$ ,  $a = .1J/K$ ,  $B = .8J$ ,  $C = .53J$ ,  $\beta = 4J$ ,  $\delta = -.89J$  (for the AI transition) and  $\delta = .16J$  (for the NI transition).  $b_1 = .7J$ ,  $b_2 = .7J$  and  $e_1 = .1J$ .

appropriate phases. For the isotropic phase the stability conditions are simply  $\alpha > 0$  and  $A > 0$ . The nematic phase is stable, if  $2\alpha + \delta S_N^2 > 0$  and  $24AC < B^2$ .

For the smectic A phase the first three stability conditions

$$A - BS_A + \frac{9}{2}CS_A^2 + \delta\psi_0^2 > 0 \quad (7)$$

$$\alpha^* + \frac{3}{2}\delta^*S_A^2 < e^*S_A \quad (8)$$

$$b_1 + e_1S_A < 0 \quad (9)$$

ensure  $S_A$ ,  $\psi_0^2$  and  $q_0^2$  to be indeed positive quantities. The Cauchy conditions in (6) lead to only two additional stability criteria (since  $\partial^2 f / (\partial q_0 \partial \psi_0) = 0$ )

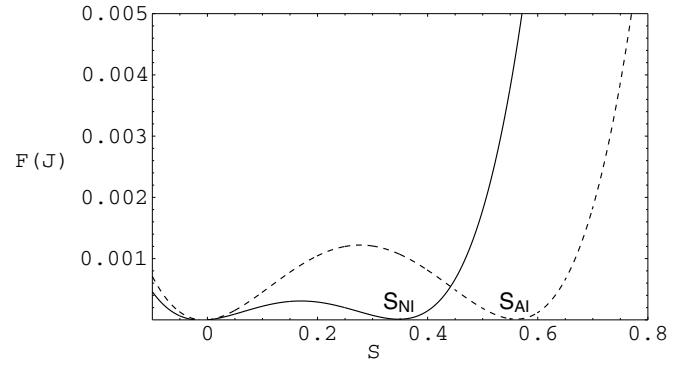
$$A^* - B^*S_A + \frac{9}{2}C^*S_A^2 + \frac{e_1^2}{6b_2}\psi_0^2 > 0 \quad (10)$$

$$A - BS_A + \frac{9}{2}CS_A^2 + \delta^*\psi_0^2 > 0 \quad (11)$$

while the determinant condition is

$$A^* - B^*S_A + \frac{3}{2}C^*S_A^2 > 0 \quad (12)$$

Since  $\delta^* < \delta$  and  $b_2 > 0$ , conditions (12) and (11) are stronger than (10) and (7), respectively. These stability conditions determine the existence ranges (in terms of temperature) of the different phases rather implicitly. Although they are involved, one important qualitative feature can be extracted immediately. It is obvious that a continuous AI transition ( $S_A = 0 = \psi_0$  at the transition temperature) is not possible, except for the very special case  $B = 0 = e_1$  and  $T_{NI}^* = T_{AI}^* + b_1^2 / (4b_2\alpha_0)$ . (As one can see from these conditions, a total of seven coefficients must either vanish or obey a very special relationship. That all of these conditions are met is very unlikely.) In the general case there is a jump in  $S_A$  and  $\psi_0$  at some temperature



**Fig. 2.** The free energy  $F$  as a function of the order parameter  $S$  for different values of the coupling constant  $\delta$ . The solid line ( $\delta = 0.16J$ ) represents the nematic- isotropic transition while the dashed line ( $\delta = -0.89J$ ) represents the isotropic - smectic A transition. The other parameter values are the same as in Fig.1

$T_{AI} > T_{AI}^*$ . Since the transitions involved are first order, the edges of the stability ranges (where ' $> 0$ ' or ' $< 0$ ' is replaced by ' $= 0$ ' in (7-12)) are not the phase transition lines. However, any phase discussed has to be within its stability range, in order to be a real physical phase.

Fig.2 shows the influence of the coupling term  $\delta$  on the NI and AI transition in the free energy versus orientational order parameter. The dashed curve is calculated for a negative  $\delta$ . The transition is always isotropic - smectic A. At  $T = T_{AI}$ , the free energy has two minima at  $S = 0$  and  $S \neq 0$  (also  $\psi_0 \neq 0$ ) corresponding to the isotropic and the smA phase. There is no third minima for the nematic phase for this particular value of  $\delta$ .

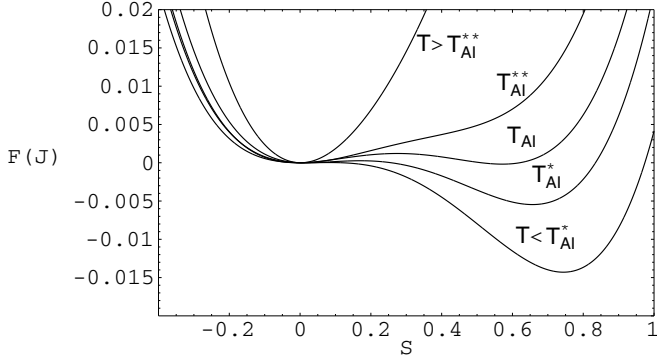
### 3 Direct isotropic-to-smectic-A transition

The smectic A phase is in competition with possible isotropic and nematic phases. The existence ranges of all three phases generally overlap. That phase with the lowest free energy is the stable one. A (first order) transition takes place, when 2 free energies are identical. In order to study the direct AI transition in more detail, we substitute the solution (5) for the smectic order parameter  $\psi_0 \neq 0$  and for  $q_0^2 \neq 0$  into the free energy (2). We get the free energy density for the smectic A phase as a function of  $S$  alone, which can be written as

$$f = f_0 - \frac{\alpha^{*2}}{4\beta} + \frac{\alpha^*e^*}{2\beta}S + \frac{3}{4}A^*S^2 - \frac{1}{4}B^*S^3 + \frac{9}{16}C^*S^4 \quad (13)$$

where the starred coefficients are defined after (5). Since  $\delta^* < \delta < 0$ , we can infer  $C^* > 0$  from  $\beta C > \delta^2$ ; similarly  $B > 0$  leads to  $B^* > 0$ . Of the 3 possible extrema ( $\frac{\partial f}{\partial S} = 0$  leading to a real solution for  $S$ ) only those are relevant that exist within the stability range of the smectic A phase (since we assumed  $\psi_0 \neq 0$  and  $q_0^2 \neq 0$  when deriving (13)).

In order to make the discussion of the temperature dependence of the transition more transparent, we plotted



**Fig. 3.** The free energy  $F$  as a function of the order parameter  $S$  for the superheated smectic temperature  $T_{AI}^{**}$ , the transition temperature  $T_{AI}$ , and the temperature  $T_{AI}^*$ , where the isotropic state becomes unstable, for  $\delta = -0.89J$ . The other parameter values are the same as in Fig.1

in Fig.3 the free energy (eq.(13)) versus the orientational order for different temperatures taking a negative value for  $\delta$ .

If there are three extrema two are minima (stable or metastable phases), one with large and one with small  $S$ , and there is a maximum (unstable solution) in between. The minimum at the larger  $S = S_A$  is the deeper one, since  $B^* > 0$ , and represents thus the relevant possible smectic A phase.

At the AI transition the isotropic minimum at  $S = 0$  and the smectic A minimum at  $S_A$  have equal free energies, but the two phases are separated by a barrier height. These conditions,  $F(S_A) = F_0$  and  $\delta F/\delta S|_{S_A} = 0$ , fix the transition temperature  $T_{AI}$  and give the jump in  $S$  as  $\Delta S = S_A(T_{AI})$ . Some implicit formulas for these quantities are

$$f_1 g_2 + f_2 (g_1^2 - f_1 h_1)^{1/2} = f_2 g_1 + f_1 (g_2^2 - f_2 h_2)^{1/2} \quad (14)$$

with

$f_1 = 3(-18A^*C^* + B^{*2})$ ,  $g_1 = -3(A^*B^* + 9\alpha^*e^*\beta^{-1}C^*)$ ,  $h_1 = (2\alpha^*/\beta)(18\alpha^*C^* - e^*B^*)$ ,  $f_2 = 18\alpha^*C^* - e^*B^*$ ,  $g_2 = 3(A^*e^* - \alpha^*B^*)$ ,  $h_2 = 6\alpha^*(2A^* + e^{*2}/\beta)$ , where  $\alpha^*$  and  $A^*$  (or rather  $\alpha$  and  $A$ ) have to be taken at  $T = T_{AI}$ . Thus, eq.(14) is an implicit equation for  $T_{AI}$ . In terms of  $T_{AI}$  the jump  $\Delta S$  is given by

$$\Delta S = f_2^{-1}(-g_2 + [g_2^2 - f_2 h_2]^{1/2}) \quad (15)$$

Having calculated  $T_{AI}$  and  $\Delta S$  we can go back to eq.(5) and calculate the finite smectic order  $\psi_0(T_{AI})$  and the layer wavelength  $q_0(T_{AI})$  at the transition temperature. Of course, one has to check that  $T_{AI}$  is within the existence range (6) of the smectic phase, e.g. that the resulting quantities  $\psi_0^2$  and  $q_0^2$  are positive. In that case there is always a direct AI transition possible within the framework and assumptions of our mean-field model.

For  $T < T_{AI}$  the smectic A minimum represents the stable state. The isotropic state is then a metastable one, and at even lower temperatures, it becomes unstable (a local energy maximum).

Of course, the NI transition could come first before  $T_{AI}$  is reached. To prevent this,  $T_{AI}$  has to be larger than  $T_{NI} = T_{NI}^* + B^2/(27Ca)$ , where  $T_{NI}$  is that temperature for which  $F_N = F_0$  with  $F_N$  the free energy (2) taken at the nematic solution (4).

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## Appendix A

In the Ginzburg-Landau expansion (1) we have kept homogeneous terms that give rise in (2) to contributions  $\sim S^n \psi_0^m$  with  $n+m \leq 4$  only. If we then allow for gradient terms up to the same  $S$ - or  $\psi_0$ -order with two or four gradients (of  $\phi$ ), the following additions to (1) should be made:  $(f_1/2)Q_{il}Q_{jl}(\nabla_i\psi)(\nabla_j\psi^*) + (e_2/2)Q_{ij}(\nabla_i\nabla_l\psi)(\nabla_j\nabla_l\psi^*) + (d_3/4)|\nabla_i\psi|^4 + (f_3/2)Q_{ij}Q_{kl}(\nabla_i\nabla_j\psi)(\nabla_k\nabla_l\psi^*)$ . There are variants of these terms, e.g.  $\sim Q_{il}Q_{il}(\nabla_j\psi)(\nabla_j\psi^*)$ ,  $\sim Q_{ij}(\nabla_k\nabla_k\psi)(\nabla_i\nabla_j\psi^*) + c.c.$ , or  $Q_{ij}Q_{kl}(\nabla_i\nabla_k\psi)(\nabla_j\nabla_l\psi^*)$ , which do not lead to any new contribution (compared to the above ones) for the smectic A phase. In (2) the new terms give rise to the additional free energy contributions

$$\Delta F = \frac{1}{2}f_1 S^2 \psi_0^2 q_0^2 + \frac{1}{2}e_2 S \psi_0^2 q_0^4 + \frac{1}{4}d_3 \psi_0^4 q_0^4 + \frac{1}{2}f_3 S^2 \psi_0^2 q_0^4 \quad (16)$$

which however, does not change qualitatively the results (especially in sec.3) of this paper. This is most easily demonstrated for the  $f_1$ -contribution. Eq.(5) is slightly changed into

$$\begin{aligned} \psi_0^2 &= -\frac{1}{\beta} \left( \alpha^* - e^* S_A + \frac{3}{2} \delta^{**} S_A^2 + O(S_A^3) \right), \\ q_0^2 &= -\frac{1}{2b_2} (b_1 + e_1 S_A + f_1 S_A^2 + O(S_A^3)) \end{aligned} \quad (17)$$

with  $\delta^{**} = \delta^* - f_1 b_1 / (3b_2)$ . This leaves unchanged the form of the free energy (13) relevant for the direct isotropic to smectic A transition. Only the coefficients  $A^*$ ,  $B^*$  and  $C^*$  are replaced by  $A^{**}$ ,  $B^{**}$  and  $C^{**}$ , respectively, for which we find in detail

$$A^{**} = A^* + \frac{1}{3} \frac{\alpha^* b_1 f_1}{\beta b_2} \quad (18)$$

$$B^{**} = B^* - \frac{e_1 f_1}{\beta b_2} \left( \alpha^* - \frac{1}{2} \frac{b_1^2}{b_2} \right) \quad (19)$$

$$C^{**} = C^* + \frac{2}{9} \frac{f_1}{\beta b_2} \left( f_1 \left( \alpha^* - \frac{1}{2} \frac{b_1^2}{b_2} \right) + 3b_1 \left( \delta^* - \frac{2}{3} \frac{e_1^2}{b_2} \right) \right) \quad (20)$$

Thus, the nature of the direct transition as discussed in section 3 is unchanged, only the dependence on the Ginzburg-Landau parameters becomes more complicated, when higher order terms are included.

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