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SECOND ORDER TO FIRST ORDER PHASE TRANSITION

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MOLECULAR ORIENTATIONAL RE-ORDERING  
AND THE TRANSFORMATION OF A LANDAU SECOND ORDER PHASE TRANSITION  
TO FIRST ORDER IN A NEMATIC LIQUID CRYSTAL \*

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ABSTRACT

We consider the nature of the nematic to isotropic phase transition in terms of the molecular orientational re-ordering, expressed by the variation of the order parameter,  $s$ , in the light of Landau's theory of second order phase transition. Then, we show how the de Gennes modification to the Landau thermodynamic potential converts the transition to first order which is in better agreement with the experimental observations.

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## I. INTRODUCTION:

The subject of phase transitions in a liquid crystal commands a great deal of interest among materials scientists and condensed matter physicists partly because of the striking properties that arise and which have valuable theoretical, technological and industrial applications to date (Cf, Kelker, and Hatz, 1980).

In this paper, we want to understand better the nature of one such phase transition, in particular, the nematic to isotropic transition. We consider the ordering of the nematic molecules (which we assume here to be uniaxial although laboratory data show that certain nematics like the cyanobiphenyls are slightly biaxial (Bunning, et. al., 1986)), quantitatively defined in terms of the order parameter  $s$  (Cf Ponce, 1987), and relate it to the thermodynamic potential  $F$  which serves as basis for describing the phase transition in the style of Landau on second order phase transitions (SOPT), (Cf Landau, 1980). Then we show how de Gennes' retention of the cubic power term in the Landau free energy changes the nematic to isotropic transition to first order type, using the Ehrenfest classification scheme, (Cf Zanoni, 1979).

## II. MOLECULAR ORIENTATIONAL ORDERING IN A NEMATIC

The nematic liquid crystal has no positional ordering but possesses orientational ordering. Due to the elongated shape of the nematic molecules, their degree of orientational ordering or alignment along a preferred direction

plays an important role in the physical properties of the liquid crystal. In fact when the molecules become more or less aligned along a common direction given by the direction  $\hat{n}$ , also called the optic axis, what once was an isotropic liquid with direction-independent properties now becomes an anisotropic liquid or a liquid crystal with very remarkable properties (Cf Brown, 1983) which are of great theoretical and technological value. The degree of such alignment is measured quantitatively in terms of the order parameter,  $S$ .

## III. APPLICATION OF LANDAU'S SECOND ORDER PHASE TRANSITION THEORY TO THE NEMATIC-ISOTROPIC CASE

When the molecules are perfectly aligned with the director  $\hat{n}$ , then  $S = 1$ . On the other hand, when the molecules are completely randomly oriented, we have  $S = 0$ . Since these two arrangements can be regarded as two different states of symmetry (with the random arrangement corresponding to perfect symmetry, i.e., isotropic state and the perfectly aligned state corresponding to perfect order or lowest symmetry) then we can regard the order parameter  $s$  as a measure of deviation from the state of perfect symmetry (Cf Landau, 1980).

Following Landau, we postulate the existence of a thermodynamic potential function  $F$  which continually depends on temperature  $T$ , pressure  $p$  and order parameter  $s$ , i.e.,

$$F = F(p, T, s)$$

We assume that  $s$  is a continuous variable (at least near the transition point) likely to depend on pressure  $p$  and temperature  $T$  (although pressure studies show that the value of  $S$  at the transition point hardly depends on pressure (Cf. Chandrasekhar, 1977) and changes from a nonzero to a zero value at transition and therefore can be used to keep track of the transition. Since the free energy or thermodynamic potential  $F$  is a continuous function of  $s$ , we can Taylor series expand it about the  $s = 0$  or isotropic state such that:

$$(1) \quad F(p, T, s) = F_0(p, T) + \left. \frac{\partial F}{\partial s} \right|_{s=0} (s-0) + \frac{1}{2!} \left. \frac{\partial^2 F}{\partial s^2} \right|_{s=0} (s-0)^2 + \frac{1}{3!} \left. \frac{\partial^3 F}{\partial s^3} \right|_{s=0} (s-0)^3 + \frac{1}{4!} \left. \frac{\partial^4 F}{\partial s^4} \right|_{s=0} (s-0)^4 + \dots$$

For the  $s = 0$  or isotropic phase to be stable, it must have a minimum free energy  $F_0$ , hence we set  $\left. \frac{\partial F}{\partial s} \right|_{s=0} = 0$  and  $\left. \frac{\partial^2 F}{\partial s^2} \right|_{s=0} > 0$ . Thus, in the neighborhood of the transition point, for small nonzero values of  $s$ , we can neglect higher ordered terms and rewrite Eq (1) as:

$$(2) \quad F(p, T, s) = F_0(p, T) + A(p, T) s^2 + B(p, T) s^4 + C(p, T) s^3$$

where

$$(3-a) \quad A(p, T) \equiv \frac{1}{2!} \left. \frac{\partial^2 F}{\partial s^2} \right|_{s=0}$$

$$(3-b) \quad B(p, T) \equiv \frac{1}{4!} \left. \frac{\partial^4 F}{\partial s^4} \right|_{s=0}$$

$$(3-c) \quad C(p, T) \equiv \frac{1}{3!} \left. \frac{\partial^3 F}{\partial s^3} \right|_{s=0}$$

Symmetry arguments led Landau to assume that  $C(p, T) = 0$ . Temporarily, we do the same here in order to see through its consequences. (Later, we shall see how the restoration of this term by de Gennes, i.e., allowing  $C(p, T)$  to be nonzero leads to a correct result.) Thus, the Landau free energy has its final form given by:

$$(4) \quad F(p, T, s) = F_0(p, T) + A(p, T) s^2 + B(p, T) s^4$$

and the change in symmetry which occurs during the transition is mainly reflected in the change in sign of  $A(p, T)$ :  $A$  is positive when the system is in the symmetric state and  $A$  becomes negative when the system is in the more orderly or unsymmetric (i.e., less symmetric) state. Since, in general, higher temperatures are associated with more symmetry and lower temperatures accompany less symmetry, plus the fact that  $F$  and its higher derivatives are continuous functions of  $p$ ,  $T$  and  $S$ , we may power expand  $A(p, T)$  in the form

$$(5) \quad A(p, T) = \left. \frac{\partial A}{\partial T} \right|_{T_c} (T - T_c)$$

or  $A(p, T) = a(p)(T - T_c)$ ,  $a(p) > 0$ , about the transition temperature  $T_c$ , assuming that  $(T - T_c)$  is small, i.e., the system is very near the transition point.

Thus,  $A > 0$  when  $T > T_c$ ,  $A < 0$  where  $T < T_c$  and  $A = 0$  when  $T = T_c$ .

For the same reasons of stability and symmetry  $B(p, T) > 0$  always.

The following mathematical discussion lends support to the above considerations: the equilibrium values of  $s$ ,  $\tilde{s}$ , for a particular stable phase, is determined from the following conditions for : a given value of  $T$  and of  $p$ .

$$(6-a) \quad \left. \frac{\partial F}{\partial s} \right|_{p, T, \tilde{s}} = 0$$

$$(6-b) \quad \left. \frac{\partial^2 F}{\partial s^2} \right|_{p, T, \tilde{s}} > 0$$

Equation (4) into (6-a) gives:

$$s(2A + 4Bs^2) \Big|_{\tilde{s}} = 0$$

from which we get

$$(7-a) \quad \tilde{s} = 0$$

or

$$(7-b) \quad \tilde{s} = \sqrt{\frac{A}{-2B}} > 0$$

Now, observing that the only 2 possible phases are the isotropic liquid phase ( $\tilde{s} = 0$ ) and the nematic phase ( $\tilde{s} \neq 0$  and  $\tilde{s}$  is real), then with  $B > 0$ , one is led to infer that  $A$  must be negative in the nematic phase in order to obtain a nonzero and real, physically significant value of  $s$ , expected of the nematic phase. When  $A = 0$  in Eq (7-b), we get  $\tilde{s} = 0$ , the isotropic phase.

Do the values of  $\tilde{s}$  give a relative minimum(i.e., a stable state) for  $F$ ?

Substitution of Eq. (7-a) into (6-b) gives:

$$(8) \quad \left. \frac{\partial^2 F}{\partial s^2} \right|_{p, T, \tilde{s} = 0} = 2A + 12Bs^2 \Big|_{s=0} = 0$$

since  $A = 0$  for  $\tilde{s} = 0$ , or vice-versa.

This is a point of inflection for the  $F$  vs.  $s$  curve!

$$\text{For } s = \sqrt{\frac{A}{-2B}}, \quad A < 0, B > 0$$

$$= \sqrt{\frac{|A|}{2B}}$$

$$s^2 = \frac{|A|^2}{2B}$$

$$(9) \quad \left. \frac{\partial^2 F}{\partial s^2} \right|_{p, T, \tilde{s}} = \sqrt{\frac{|A|}{2B}} = 2A + 12B \frac{|A|}{2B} = -2|A| + 6|A| = 4|A| > 0$$

Thus,  $\tilde{s} = \sqrt{\frac{|A|}{2B}}$  is the equilibrium value of the order parameter in the stable nematic state, with  $A < 0$ , and  $B > 0$ . The isotropic state has  $s = 0$  with  $A = 0$ . The corresponding free energy values are (using Eq. (4)):

$$F(p, T, s) = F_0(p, T) \text{ for } s = 0, \text{ isotropic or symmetric phase}$$

and

$$F(p, T, s) = (F_0(p, T) + As^2 + Bs^4) \Big|_{\tilde{s}} = \sqrt{\frac{|A|}{2B}}, \quad A < 0$$

$$= F_0(p, T) + A \frac{|A|}{2B} + B \frac{|A|^2}{(2B)^2}$$

$$= F_0 - \frac{|A|^2}{2B} + \frac{|A|^2}{4B}$$

$$= F_0 - \frac{|A|}{4B}, \text{ for the nematic phase given by } \tilde{s} = \sqrt{\frac{|A|}{2B}} > 0.$$

Hence,  $F_{\text{nematic}} < F_{\text{isotropic}}$ , i.e., the nematic free energy is less than the isotropic free energy implying that more order means less free energy.

Note that when  $s$  approaches zero from the nematic side, for which  $A < 0$ , then  $\left. \frac{\partial^2 F}{\partial s^2} \right|_{s=0} = 2A = -2|A| < 0$ ,

meaning, the  $s = 0$  point is a local maximum, a state of unstable equilibrium. Figure (1) below shows the functional behavior of  $F$  vs.  $s$  for the three regimes:

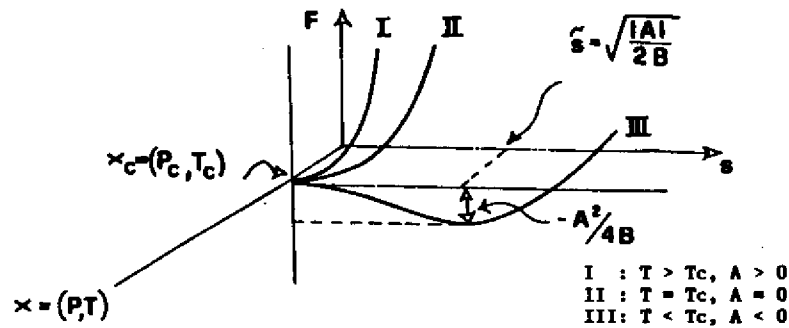


Fig. 1: The variation of the Landau free energy  $F$  with respect to order parameter  $s$  for the isotropic phase ( $T > T_c$ ), the transition point ( $T = T_c$ ) and the nematic phase ( $T < T_c$ ).

How does the  $F$  vs.  $s$  curve look for the transition region,  $T = T_c$  or  $A = 0$ ?

When  $A = 0$  at  $T = T_c$ , the Landau free energy, Eq. (4) becomes

$$(10) \quad F = F_0 + Bs^4$$

Equation (10) gives another parabola similar to that of the  $A > 0$ ,  $T > T_c$  case in Fig. (1).

The equation  $A(p, T) = 0$  gives  $T = T(p)$  or  $p = p(T)$  and determines the set of  $(p, T)$  points at which phase transition occurs. Every point on the  $(p, T)$  axis shown in Fig. (1) is such a point. This equation should also give the phase equilibrium curve at each point of which the nematic and isotropic phases are in equilibrium with each other. However, the above discussion shows that only the isotropic state exists

(i.e., is stable) at transition. To determine the order of the nematic to isotropic transition, let us recall the Ehrenfest scheme of classifying phase transitions (Cf. Zannoni, 1979): a transition is  $n$ th order if its  $n$ th derivative across the phase transition curve is discontinuous while its first  $(n-1)$  derivatives are continuous. Hence, by differentiating the free energy with respect to temperature  $T$  to obtain entropy  $S$ , we get

$$(11) \quad S = - \frac{dF}{dT} = - \left[ \frac{\partial F}{\partial T} + \frac{\partial F}{\partial s} \right]_{s=0} \frac{\partial s}{\partial T}$$

which with Eq (4) together with  $\frac{\partial F}{\partial s} \Big|_{s=0} = 0$  becomes

$$(12) \quad S = - \frac{\partial F}{\partial T} = - \frac{\partial F_0}{\partial T} - \frac{\partial A}{\partial T} s^2$$

since  $A(p, T) = a(p)(T - T_c)$ ,  $a(p)$  and  $A < 0$ , i.e., near the transition point but in the

$$\frac{\partial A}{\partial T} = - a(p)$$

nematic phase

and  $s^2 = \frac{|A|}{2B} = - \frac{A}{2B}$ ,  $A < 0$  (see Eq. 7-b) then (12) becomes

$$S = - \frac{\partial F_0}{\partial T} + \frac{|A|a}{2B}$$

$$= S_0 + \frac{A^2}{2B(T - T_c)}$$

$$(13) \quad = S_0 + \frac{a^2(T - T_c)}{2B}$$

entropy near the transition point, at temperature  $T$ , from the nematic side

Approaching the phase transition from the isotropic liquid ( $T > T_c$ ) side,  $s = 0$  and

$$(14) \quad S = - \frac{\partial F_0}{\partial T}$$

$$= S_0 \quad \text{isotropic fluid phase}$$

At transition,  $T = T_c$  and Eqs. (13) and (14) become identical. This makes the first derivative of free energy  $F$  continuous across the phase equilibrium curve. From the second derivative of  $F$ , we obtain the specific heat for the isotropic phase:

$$(15) \quad C_{p_0} = -T \frac{\partial^2 F_0}{\partial T^2} \\ = T \frac{\partial S_0}{\partial T}, \quad T > T_c$$

and that for the nematic phase:

$$(16) \quad C_p = +T \left[ - \frac{\partial^2 F}{\partial T^2} + \frac{a^2}{2B} \right] \\ = T \frac{\partial S}{\partial T} + T \frac{a^2}{2B}, \quad T < T_c$$

At transition,  $T = T_c$  and Eq. (15) becomes

$$C_{p_0} = T_c \frac{\partial S_0}{\partial T}$$

while Eq. (16) becomes

$$(17) \quad C_p = C_{p_0} + \Delta C_p, \quad \text{where}$$

$$(18) \quad \Delta C_p = \frac{a^2 T_c}{2B} > 0, \quad \text{difference between the nematic} \\ \text{and isotropic phases at } T = T_c.$$

So we see that the specific heat at constant pressure,  $C_p$  of the system jumps up as it changes from isotropic to the nematic phase.

This, therefore, classifies the isotropic to nematic transition as second order based on a Landau type free energy.

Experimental data, however, show a nonzero latent heat for the isotropic nematic transition as affirmed for example by the following data for p-azoxyanisole (PAA), a well known nematic (Cf. Chandrasekhar, 1977; Martire, 1979).

Table 1

Nematic Compound	Solid-Nematic Transition Latent Heat (J/mole)	Nematic-Isotropic Transition Latent Heat (J/mole)
PAA	29,570 $T_c = 391 \text{ }^\circ\text{K}$	574 $T_c = 408 \text{ }^\circ\text{K}$

The above statistics show that in comparison to the solid-nematic transition, the nematic-isotropic transition is weakly first order since its latent heat,  $H$ , where

$$(19) \quad H = T_c \Delta S = 574 \text{ joules/mole}$$

gives a small discontinuity in entropy

$$\Delta S = \frac{574 \text{ J/mole}}{408 \text{ }^\circ\text{K}} = 1.41 \text{ J/mole }^\circ\text{K}$$

for PAA. The corresponding value for the solid-nematic transition is  $\Delta S = \frac{29,570 \text{ J/mole}}{391 \text{ }^\circ\text{K}} = 75.62 \text{ J/mole }^\circ\text{K}$ .

Therefore, we ask: how can the Landau model for the  $N \leftrightarrow I$  transition be modified in order to arrive at a prediction consistent with experiment?

#### IV. TRANSFORMATION OF THE LANDAU SECOND ORDER NEMATIC-ISOTROPIC TRANSITION TO FIRST ORDER VIA THE DE GENNES MODIFICATION

We try the de Gennes modification (Cf. de Gennes, 1974)

to the Landau free energy  $F$  by removing the restriction to the value of  $C(p,T)$  in Eq. (2), i.e., make it nonzero! Then the free energy  $F$  regains its original form before symmetry considerations are ever made:

$$(20) \quad F(p,T,s) = F_0(p,T) + A(p,T)s^2 + B(p,T)s^4 + C(p,T)s^3$$

De Gennes' reason for such a modification is the observation that the molecular ordering corresponding to  $-s$  is utterly different from that of  $s$  such that no symmetry operation or coordinate transformation would directly change  $s$  to  $-s$ ; the result of this is that  $s$  and  $-s$  correspond to different energies i.e.,  $F(s) \neq F(-s)$  since the molecules of the  $(-s)$  configuration are more nearly perpendicular to the optic axis  $\hat{n}$  while those of  $s$  are more nearly aligned with  $\hat{n}$  (Cf, de Gennes, 1974; Chandrasekhar, 1977).

Thus, in a stable phase, characterized by  $\tilde{s}$ , the following are true:

$$(21) \quad \left. \frac{\partial F}{\partial s} \right|_{\tilde{s}} = 2As + 4Bs^3 + 3Cs^2 \Big|_{\tilde{s}} = 0$$

and

$$(22) \quad \left. \frac{\partial^2 F}{\partial s^2} \right|_{\tilde{s}} = 2As + 12Bs^2 + 6Cs \Big|_{\tilde{s}} > 0$$

From Eq (21) we obtain:

$$\tilde{s} = 0 \text{ or } \tilde{s} = \frac{-3C \pm [9C^2 + 32|A|B]^{1/2}}{8B}, \quad A < 0$$

For  $s$  to have a nonzero and positive value (nematic phase), since  $[9C^2 + 32|A|B]^{1/2} \geq 3|C|$ , then it's necessary for  $C$  to be negative. Consequently,

$$\tilde{s} = \frac{3|C| \pm [9C^2 + 32|A|B]^{1/2}}{8B}$$

$$\text{let } \tilde{s}_+ = \frac{3|C| + [9C^2 + 32|A|B]^{1/2}}{8B} > 0$$

and

$$\tilde{s}_- = \frac{3|C| - [9C^2 + 32|A|B]^{1/2}}{8B} < 0$$

both nematic phases since a nematic phase is defined by a nonzero  $s$ .

During transition,  $T = T_c$  and  $A = 0$  and the above expressions for  $\tilde{s}_+$  and  $\tilde{s}_-$  reduce to

$$(23) \quad \tilde{s}_+ = \frac{3|C|}{4B}, \text{ nematic phase}$$

$$(24) \quad \tilde{s}_- = 0, \text{ isotropic phase}$$

Eqs. (23) and (24) tell us that 2 stable phases, the isotropic and nematic phases are present and are in equilibrium with each other during transition.

Do  $\tilde{s}_+$  and  $\tilde{s}_-$  satisfy Eq. (22)?

$$\left. \frac{\partial^2 F}{\partial s^2} \right|_{\tilde{s}_+} = 2A + 12Bs^2 + 6Cs \Big|_{\tilde{s}_+} = \frac{3|C|}{4B}, \quad A = 0$$

$$= 0 + \frac{27}{4} \frac{|C|^2}{B} - \frac{18}{4} \frac{|C|^2}{B} = \frac{9}{4} \frac{|C|^2}{B} > 0$$

$\therefore \tilde{s}_+$  gives a relative minimum for  $F$  and indicates the existence of a stable nematic phase at transition.

On the other hand,

$$\left. \frac{\partial^2 F}{\partial s^2} \right|_{\tilde{s}_-} = 2A + 12Bs^2 + 6Cs \Big|_{\tilde{s}_-} = 0$$

$$A = 0$$

$$= 0 + 0 + 0 = 0$$

$s_- = 0$  gives a point of inflection for the the  $F$  vs.  $s$  curve.

The corresponding values of  $F$  at transition  $A = 0$  are:

$$F(\tilde{s}_+) = F_0 + Bs^4 + Cs^3 \Big|_{\tilde{s}_+} = \frac{3}{4} \frac{|C|}{B}$$

$$= F_0 - \frac{27}{256} \frac{C^4}{B^3} = F_0 + \Delta F(\tilde{s}_+)$$

$$F(\tilde{s}_-) = F_0 + Bs^4 + Cs^3 \Big|_{\tilde{s}_-} = 0$$

$$= F_0$$

Since, it is the differences in  $F$  and not the absolute values of  $F$  which are of interest to us, we reset  $F_0$  to zero and find the values of  $s$  for which  $F$  is zero (i.e., we look for the zeroes of  $F$ ) when  $A = 0$

$$F = Bs^4 + Cs^3 = 0$$

gives  $s = 0, -\frac{C}{B}, C < 0$

With the above result, the variation of  $F$  with respect to  $s$  for the transition region typified by  $A = 0, T = T_c$  can be seen in Fig. 2 below:

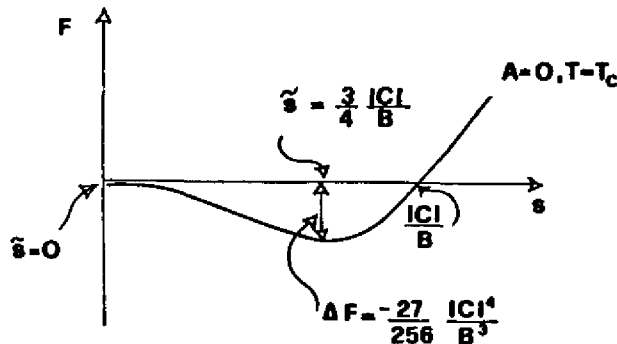


Fig. 2. Behavior of  $F$  during transition  $A = 0, T = T_c$

Recalling Eq. (20), where  $B = B(p)$  at transition, the entropy  $S$  becomes

$$S = - \frac{dF}{dT} = - \left[ \frac{\partial F}{\partial T} + \frac{\partial F}{\partial s} \right]_{T=T_c} \frac{\partial s}{\partial T} \quad \text{where } \frac{\partial F}{\partial s} \Big|_{T=T_c} = 0$$

$$(25) S = - \frac{\partial F}{\partial T} = - \frac{\partial F}{\partial T} + \frac{\partial A}{\partial T} s^2 + \frac{\partial C}{\partial T} s^3$$

recall,  $A = a(p)(T - T_c)$  with  $a(p) > 0$  and  $A < 0$  for  $T < T_c$  then  $\frac{\partial A}{\partial T} = -a(p)$ ; therefore,

$$(26) S = - \frac{\partial F_0}{\partial T} - a(p) s^2 + \frac{\partial C}{\partial T} s^3 \quad \text{near transition in the nematic phase}$$

since  $s = 0, \frac{3}{4} \frac{C}{B}$  in the transition point,  $T = T_c$ ; then  $S_{\text{isotropic}} = - \frac{\partial F_0}{\partial T} = S_0$

and

$$S_{\text{nematic}, T = T_c} = - \frac{\partial F_0}{\partial T} - \frac{9}{16} a(p) \frac{|C|^2}{B^2} + \frac{27}{64} \frac{|C|^3}{B^3} \frac{\partial C}{\partial T}$$

$$s = \frac{3}{4} \frac{|C|}{B}$$

where the change  $\Delta S = S_0 + \Delta S$  in entropy is

$$(27) \Delta S = - \frac{9}{16} a(p) \frac{|C|^2}{B^2} + \frac{27}{64} \frac{|C|^3}{B^3} \frac{\partial C}{\partial T}$$

Eq. (27) shows a discontinuity in entropy implying that the isotropic-nematic transition is of first order. Since Eq. (19) gives us a small value for  $\Delta S = 1.41 \text{ J/mole } ^\circ\text{K}$  then the coefficient  $C$  must be small.

## V. CONCLUSION

We have just seen in this paper how molecular orientational reordering, indicated by the variation of order parameter  $s$



from a nonzero to zero value gives way to the nematic - isotropic transition. We have also shown how de Gennes' restoration of the cubic power term in the free energy  $F$  leads to the transformation of a Landau second order nematic-isotropic phase transition to first order to be able to arrive at results more compatible with experimental data.

The role of  $s$  as a measure of deviation from symmetry needs to be examined further though, in so far as its effect on the symmetry of the free energy  $F$  is concerned. Does the symmetry of  $F$  reflect the state of symmetry of the system as  $s$  approaches the perfect symmetry state? Is it absolutely necessary to talk of the phase transition mechanism in terms of symmetry considerations or can we attain the same results ignoring symmetry altogether? The last question is raised because it is through the considerations of symmetry that leads one to conclude that the nematic-isotropic transition is second order instead of being first order.

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