

D_2 - D_1 phase transition of columnar liquid crystals

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The D_2 - D_1 phase transition in columnar liquid crystals of the HAT series [e.g., HAT11 (triphenylene hexa- n -dodecanoate)] is discussed within the framework of Landau theory. The order parameters which describe the transition are abstracted from a tensor density function, and are associated with two irreducible representations of the symmetry group of the high-temperature D_2 phase. A mechanism for a first-order transition is then suggested in accordance with both theoretical considerations and the experimental result for the D_2 - D_1 transition. Two possible arrangements of the herringbone structure of the D_1 phase are obtained, each of which gives six orientational states in the low-temperature D_1 phase.

I. INTRODUCTION

In the preceding paper¹ we investigated the possible low-temperature phases which may be obtained from the symmetry breaking of the D_2 phase by means of a second-order phase transition. The conclusion is that no herringbone structure can be obtained which is associated with only one irreducible representation (IR) of the high-temperature symmetry group G_0 (this is supposed to be necessary for the ordinary second-order phase transition according to Landau theory). Thus the results from the previous paper cannot be immediately used for discussing the D_2 - D_1 transition which is the subject of this paper. As we will see in the following, the order parameter which can be used to describe the symmetry breaking through the D_2 - D_1 transition is composed of two IR's of the symmetry group G_0 , and therefore is associated with a reducible representation of G_0 . As pointed out by Bak and Mukamel,² in this case, the phase transition is generally first order. We will give a possible way to realize such a first-order transition. The six orientational states we obtain here can be compared with those suggested by Safinya *et al.*³ Two possible arrangements of the herringbone structure are just as predicted by Levelut.⁴ For the sake of completeness, we briefly review some of the results from the preceding paper in the very beginning of Sec. II.

II. ORDER PARAMETERS AND SYMMETRY PATTERNS

By making use of the transition mechanism suggested by Safinya *et al.*,³ the present authors suggested¹ that the symmetry change through the D_2 - D_1 phase transition can be described by a symmetric second-order tensor density function $Q(\mathbf{r})$ which can be defined as

$$Q(\mathbf{r}) = \sum_{\alpha} \delta(\mathbf{r}_1 - \mathbf{r}_1^{\alpha}) q(\mathbf{r}_1^{\alpha}). \quad (2.1)$$

Here \mathbf{r}_1^{α} gives the location of α th column; and $q(\mathbf{r}_1^{\alpha})$ gives the average orientation of the molecules in the α th column and is a symmetric second-rank tensor.

In the D_2 phase, $Q(\mathbf{r}) \equiv Q_0(\mathbf{r})$. The symmetry group of D_2 phase can be identified as $G_0 = P6/mmm (D_{6h}^1)$. Then $Q_0(\mathbf{r})$ is an invariant under all the symmetry operations $g \in G_0$. In the low-temperature phase, the tensor density function takes the form

$$Q(\mathbf{r}) = Q_0(\mathbf{r}) + \delta Q(\mathbf{r}). \quad (2.2)$$

Each component of $\delta Q(\mathbf{r})$, which describes a phase transition from the D_2 phase to a rectangular columnar phase with the size of the unit cell doubled, can be expanded as¹

$$\delta Q_{ij}(\mathbf{r}) = \sum_{p=1}^3 T_{ij}^k \cos(\mathbf{k}_p \cdot \mathbf{r}), \quad (2.3)$$

where $\mathbf{k}_p \in \mathbf{k}_1^* \equiv \{ \frac{1}{2} \mathbf{b}_1, \frac{1}{2} \mathbf{b}_2, -\frac{1}{2} \mathbf{b}_1 + \frac{1}{2} \mathbf{b}_2 \}$ with $\mathbf{b}_1, \mathbf{b}_2$ being the basis of the reciprocal lattice of the D_2 phase. The free energy used in discussing the phase transition can be considered as a functional of T_{ij}^k .¹ For a second-order transition, or a transition which is nearly so, the free energy can be expanded as a power series of T_{ij}^k , which in each order consists of all the possible invariants of G_0 . With respect to the index \mathbf{k}_p , the T_{ij}^k transform as the IR $\Gamma^{[\mathbf{k}_1^*, \tau_1]}$ of G_0 where τ_1 is the unit IR of the group of \mathbf{k}_1 , and with respect to the indices i and j , they transform as a symmetric second-order tensor. So, the T_{ij}^k transform as the direct product of $\Gamma^{[\mathbf{k}_1^*, \tau_1]}$ and a second-order tensor. Generally, the T_{ij}^k give a reducible representation of G_0 , Γ . Suitable linear combinations of T_{ij}^k will reduce Γ into a direct summation of the IR's of G_0 . We found¹

$$\Gamma = \Gamma^{[\mathbf{k}_1^*, \tau_3]} \oplus \Gamma^{[\mathbf{k}_1^*, \tau_5]} \oplus \Gamma^{[\mathbf{k}_1^*, \tau_7]} \oplus 3\Gamma^{[\mathbf{k}_1^*, \tau_1]}, \quad (2.4)$$

where the τ_i denote the IR of the group of \mathbf{k}_1 . For a second-order transition, only one of the IR is relevant to the expansion in the free energy. The possible second-order phase transitions and the corresponding symmetry patterns at the low-temperature phases induced by $\Gamma^{[\mathbf{k}_1^*, \tau_3]}$, $\Gamma^{[\mathbf{k}_1^*, \tau_5]}$, and $\Gamma^{[\mathbf{k}_1^*, \tau_7]}$ have been discussed by the present authors.¹ The results indicate that the herring-

bone structure cannot be induced by only one IR of G_0 . As noted in Ref. 1, the herringbone structure can be realized if the order parameter is composed of two IR's of G_0 , $\Gamma^{\{k_1^*, \tau_3\}}$ and $\Gamma^{\{k_1^*, \tau_3\}}$ or $\Gamma^{\{k_1^*, \tau_3\}}$ and $\Gamma^{\{k_1^*, \tau_7\}}$. This suggests that the D_2 - D_1 transition is induced by a reducible representation of G_0 , so the transition is usually first order² (a second-order transition associated with more than one IR of G_0 only corresponds to some isolated points in the phase diagram⁵).

The construction of the free energy is based on the following considerations.

(1) For the D_2 - D_1 transition, the appearance in the free energy of the variables being used as the order parameters is restricted to those which are associated with suitable IR's of G_0 , and which induce the correct symmetry of the D_1 phase as determined experimentally.

(2) A first-order transition mechanism requires suitable assumptions for the coefficients which appear in the power-series expansion of the free energy.

Let us first assume that the D_2 - D_1 transition is associated with IR's $\Gamma^{\{k_1^*, \tau_3\}}$ and $\Gamma^{\{k_1^*, \tau_3\}}$. Then, up to the fourth-order terms, the free energy F' can be expanded as

$$F' = \frac{1}{2}r \sum_{i=1}^3 \eta_i^2 + u \left[\sum_{i=1}^3 \eta_i^2 \right]^2 + v \sum_{i=1}^3 \eta_i^4 + \frac{1}{2}r' \sum_{i=1}^3 \xi_i^2 + u' \left[\sum_{i=1}^3 \xi_i^2 \right]^2 + v' \sum_{i=1}^3 \xi_i^4 + c_1 \sum_{i,j=1}^3 \eta_i^2 \xi_j^2 + c_2 \sum_{i=1}^3 \eta_i^2 \xi_i^2, \quad (2.5)$$

where all the coefficients are functions of temperature and pressure (T and P). The basic idea which yields a possible first-order transition is that, in some range of T and P , $u+v$ (or $u'+v'$) may turn to be negative, so that the IR of $\Gamma^{\{k_1^*, \tau_3\}}$ (or $\Gamma^{\{k_1^*, \tau_3\}}$) alone will lead to a finite jump of η (or ξ) through the transition, and the coupling [the c_1 and c_2 terms in Eq. (2.5)] gives rise to a nonzero jump of ξ (or η) simultaneously. Therefore, above the transition temperature both η and ξ are zero, and jump to finite nonzero values through the transition.

In the following, we suppose that, near the transition, $r, r' > 0$; $v, v' < 0$; $u, u' < 0$; $u'+v' > 0$; and $c_2, c_1+c_2 < 0$. Because $u+v < 0$, a stable state with nonzero η requires the addition of sixth-order terms in η_i to the free energy F' . We will simply take the sixth-order invariant to be isotropic, i.e., $w(\sum_{i=1}^3 \eta_i^2)^3$ with $w > 0$. And

$$F = F' + w \left[\sum_{i=1}^3 \eta_i^2 \right]^3. \quad (2.6)$$

The reason for neglecting the anisotropic sixth-order invariants is the assumption that the orientation of the parameter $\{\eta_i\}$ in its IR space is determined by the anisotropic fourth-order invariants only.

Now, we define that $\eta_i = \eta \gamma_i$ and $\xi_i = \xi \gamma'_i$ subject to the conditions $\sum_{i=1}^3 \gamma_i^2 = 1$ and $\sum_{i=1}^3 \gamma_i'^2 = 1$. Here $\{\gamma_i\}$ and $\{\gamma'_i\}$ represent the orientations of $\{\eta_i\}$ and $\{\xi_i\}$ in their own IR spaces. It is easy to see that for nonzero η and ξ ,

the possible orientations of $\{\eta_i\}$ and $\{\xi_i\}$ which may give the extreme minimum value of F are

$$[\{\gamma_i\}; \{\gamma'_i\}] = [\pm 1, 0, 0; \pm 1, 0, 0], \quad (2.7a)$$

$$[\{\gamma_i\}; \{\gamma'_i\}] = [0, \pm 1, 0; 0, \pm 1, 0], \quad (2.7b)$$

$$[\{\gamma_i\}; \{\gamma'_i\}] = [0, 0, \pm 1; 0, 0, \pm 1]. \quad (2.7c)$$

We should notice that the other possible combinations of $\{\gamma_i\}$ and $\{\gamma'_i\}$ are excluded by the existence of the coupling term $c_2 \sum_i \eta_i^2 \xi_i^2$.

In each case, we have

$$F = a\eta^2 + \frac{1}{2}b\eta^4 + \frac{1}{3}d\eta^6 + a'\xi^2 + \frac{1}{2}b'\xi^4 + c\eta^2\xi^2, \quad (2.8)$$

here

$$a = \frac{1}{2}r \quad (> 0), \quad a' = \frac{1}{2}r' \quad (> 0),$$

$$b = 2(u+v) \quad (< 0), \quad b' = 2(u'+v') \quad (> 0),$$

$$d = 3w \quad (> 0), \quad c = c_1 + c_2 \quad (< 0).$$

By introducing $\rho = \eta^2$ and $\rho' = \xi^2$, Eq. (2.7) can be further written as

$$F = a\rho + \frac{1}{2}b\rho^2 + \frac{1}{3}d\rho^3 + \frac{1}{2}b'\rho'^2 + c\rho\rho'. \quad (2.9)$$

The first-order phase transition appears when ρ and ρ' take the nonzero values $\bar{\rho}$ and $\bar{\rho}'$ which minimize F at $F=0$ (F is continuous through the transition). That means

$$\frac{\partial F}{\partial \rho} = a + b\rho + d\rho^2 + c\rho' = 0, \quad (2.10a)$$

$$\frac{\partial F}{\partial \rho'} = a' + b'\rho' + c\rho = 0, \quad (2.10b)$$

$$F = a\rho + \frac{1}{2}b\rho^2 + \frac{1}{3}d\rho^3 + a'\rho' + \frac{1}{2}b'\rho'^2 + c\rho\rho' = 0, \quad (2.10c)$$

with the condition $\rho, \rho' > 0$.

By eliminating ρ and ρ' from Eqs. (2.10a) through (2.10c), one can obtain a relation

$$\psi(\bar{a}, \bar{a}', \bar{b}, \bar{b}', \bar{c}, \bar{d}) = 0. \quad (2.11)$$

Equation (2.11) gives the first-order transition line in the T - P plane. The tilde variables indicate the values of these coefficients on the transition line in T - P plane.

With all the coefficients defined by Eq. (2.11), any two of the equations in Eqs. (2.10) will give a set of solutions for ρ and ρ' :

$$\bar{\rho}' = -\frac{\bar{a}'}{\bar{b}'} - \frac{\bar{c}}{\bar{b}'} \bar{\rho}, \quad (2.12a)$$

$$\bar{\rho} = \frac{-\bar{b} + \frac{\bar{c}^2}{\bar{b}'} + \left[\left(\bar{b} - \frac{\bar{c}^2}{\bar{b}'} \right)^2 - 4\bar{d} \left(\bar{a} - \frac{\bar{a}'\bar{c}}{\bar{b}'} \right) \right]^{1/2}}{2\bar{d}}. \quad (2.12b)$$

(Note: \bar{b} and \bar{c} are supposed to be negative.)

The second term on the right-hand side of Eq. (2.12a) is positive, and the first term is negative. If $-\bar{c}\bar{\rho} > \bar{a}'$, $\bar{\rho}'$ will be positive. Then, it is not difficult to prove that Eqs. (2.12) give a stable state which minimizes the free energy

as shown in Fig. 1. On the other hand, if $-\tilde{c}\tilde{\rho} < \tilde{a}'$, $\tilde{\rho}'$ turns out to be negative which requires ξ to be imaginary ($\xi^2 = \rho'$). In this case, Eq. (2.10b) should be replaced by $\rho' \equiv 0$ (that is because $\xi = 0$ is also a solution of $\partial F / \partial \xi = 0$) and Eqs. (2.10a) and (2.10c) immediately reduce to

$$a + b\rho + d\rho^2 = 0, \quad (2.13a)$$

$$a\rho + \frac{1}{2}b\rho^2 + \frac{1}{3}d\rho^3 = 0. \quad (2.13b)$$

Equations (2.13) essentially mean that in this case, the two parameter sets $\{\eta_i\}$ and $\{\xi_i\}$ are decoupled from each other. The transition line is now simply determined by⁵

$$\psi'(\tilde{a}, \tilde{b}, \tilde{d}) = 16\tilde{a}\tilde{d} - 3\tilde{b}^2 = 0 \quad (2.14)$$

and

$$\tilde{\rho} = -\frac{3\tilde{b}}{4\tilde{d}}. \quad (2.15)$$

It will be interesting if there exists a point on the transition line determined by Eq. (2.11), in which $\tilde{\rho}' = 0$. Beyond this point, the transition line should be given by Eq. (2.14), and the symmetry pattern in the low-temperature phase will be that given in our previous work [i.e., Fig. 2(b) of Ref. 1]. A possible phase diagram is shown in Fig. 2. P_2 and P_3 are so-called tricritical points beyond which the first-order transition changes to the second order.⁵ Phase I is the high-temperature D_2 phase with the symmetry group $P6/mmm$. Now, we discuss the symmetry of phase III and phase II. To do this, we need the explicit expressions of the linear combinations of T_{ij}^k , which give $\{\eta_i\}$, $\{\xi_i\}$, and the other parameters transforming according to their IR's [see Eqs. (7) in Ref. 1]. Let us consider the orientational state corresponding to $[\{\gamma_i\}, \{\gamma'_i\}] = [1, 0, 0; 1, 0, 0]$ first. This requires

$$\eta_1 = \eta = (\tilde{\rho})^{1/2}, \quad \eta_2 = 0, \quad \eta_3 = 0;$$

$$\xi_1 = \xi = (\tilde{\rho}')^{1/2}, \quad \xi_2 = 0, \quad \xi_3 = 0.$$

and the rest of the parameters, which give IR's $3\Gamma^{\{k_1^*, \tau_1\}}$ and $\Gamma^{\{k_1^*, \tau_7\}}$, are equal to zero.

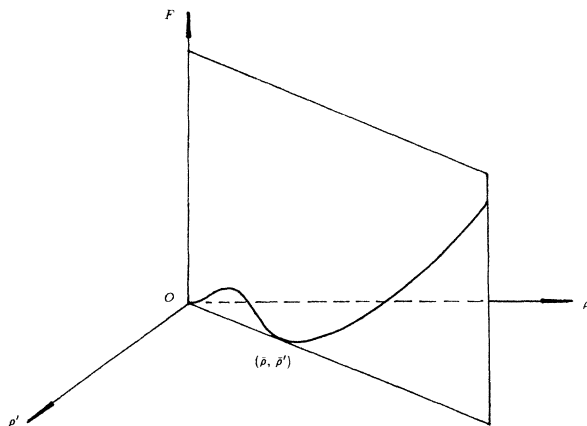


FIG. 1. The first-order D_2 - D_1 transition appears when ρ and ρ' take nonzero values $\tilde{\rho}$ and $\tilde{\rho}'$ which minimize the free energy F at $F = 0$.

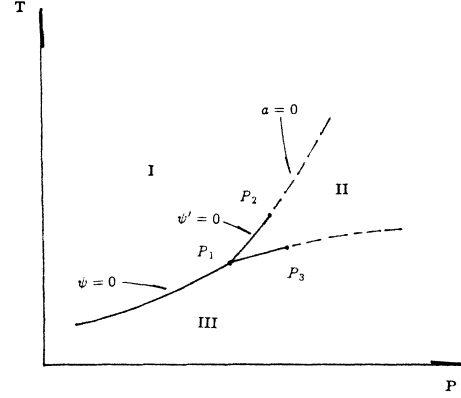


FIG. 2. Phase diagram in the T - P plane. The solid lines represent first-order transition lines, and the dashed lines, second-order transition lines; phase I is the high-temperature D_2 phase with $P6/mmm$ (D_{6h}^1) symmetry, and phases II and III are the low-temperature phases with $Pbam$ (D_{2h}^9) and $P2_1/a$ (C_{2h}^5) symmetries, respectively.

The nonzero tensor components we obtained are

$$T_{11}^{k_1} = \frac{\sqrt{6}}{4}\eta, \quad T_{22}^{k_1} = -\frac{\sqrt{6}}{4}\eta, \quad T_{12}^{k_1} = -\frac{\sqrt{2}}{4}\eta \quad (2.16)$$

and

$$T_{13}^{k_1} = \frac{\sqrt{6}}{4}\xi, \quad T_{23}^{k_1} = \frac{\sqrt{2}}{4}\xi. \quad (2.17)$$

Substituting Eqs. (2.16) and (2.17) to Eq. (2.3) we have

$$\delta Q(\mathbf{r}) = \frac{\sqrt{2}}{4} \begin{bmatrix} \sqrt{3}\eta & -\eta & \sqrt{3}\xi \\ -\eta & -\sqrt{3}\eta & \xi \\ \sqrt{3}\xi & \xi & 0 \end{bmatrix} \cos(\mathbf{k}_1 \cdot \mathbf{r}). \quad (2.18)$$

It is not difficult to check that the remaining symmetry operations of G_0 ($P6/mmm$) which keep $\delta Q(\mathbf{r})$ invariant are

$$\tilde{g} = (h_\alpha | \sigma_\alpha + \mathbf{k}\mathbf{a} + l\mathbf{b}), \quad (2.19)$$

where

$$\sigma_\alpha = \begin{cases} 0 & \text{if } \alpha = 1, 2, 5 \\ \frac{\mathbf{a} + \mathbf{b}}{2} & \text{if } \alpha = 2, 2, 6 \end{cases}$$

and \mathbf{a} and \mathbf{b} are the unit basis vectors of the low-temperature $[1, 0, 0; 1, 0, 0]$ state as defined in Ref. 6. The h_α 's are the rotational operations (including reflection and inversion) defined with respect to a rectangular coordinate system (the x and y axes coincide with \mathbf{a} and \mathbf{b} , respectively) as given by Kovolev.⁷ One can identify that all \tilde{g} 's form the $P2_1/a$ space group. The symmetry pattern is shown in Fig. 3(a) and fits the herringbone structure. For the $[\{\gamma_i\}; \{\gamma'_i\}] = [-1, 0, 0; 1, 0, 0]$ state, we will essentially have the same results as for $[1, 0, 0; 1, 0, 0]$. This only corresponds to a different way of choosing the unit cell.

For the solution $[\{\gamma_i\}; \{\gamma'_i\}] = [1, 0, 0; -1, 0, 0]$, the symmetry group in the low-temperature phase is again

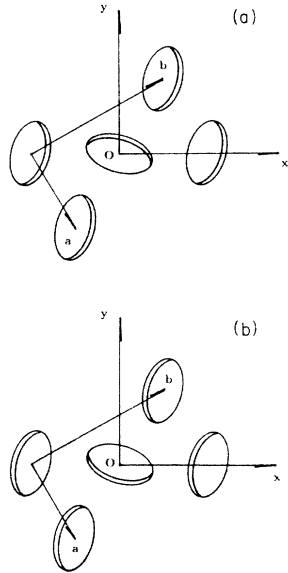


FIG. 3. Symmetry patterns of the low-temperature D_1 phase ($P2_1/a$) in the orientational states (a) $[1,0,0;1,0,0]$ and (b) $[1,0,0;-1,0,0]$; **a** and **b** are the basis vectors of the two-dimensional rectangular lattice structure. (a) and (b) can be mapped from one to another by a mirror reflection in the x - y plane.

$P2_1/a$. But the matrix structure of $\delta Q(\mathbf{r})$ is different. By rotating the coordinate system to diagonalize local tensor $\delta Q(\mathbf{0})$ at $\mathbf{r}=\mathbf{0}$, we find that molecular orientations in this state [Fig. 3(b)] are different from those in Fig. 3(a). Figure 3(b) can be obtained from Fig. 3(a) by a mirror reflection in the plane perpendicular to the column axis. So, in the sense of Aizu,^{8,9} Figs. 3(a) and 3(b) refer to two different orientational states. The other four orientational states correspond to $[\{\gamma_i\}; \{\gamma'_i\}] = [0,1,0;0,\pm 1,0]$ and $[0,0,1;0,0,\pm 1]$. Their symmetry patterns can be obtained by rotating Figs. 3(a) and 3(b) by 60° and 120° about the x_3 axis. Therefore, we have six orientational states in the low-temperature phase.

If we assume that the symmetry breaking is associated with $\Gamma^{\{\mathbf{k}_1^*, \tau_3\}}$ and $\Gamma^{\{\mathbf{k}_1^*, \tau_7\}}$, then by following similar procedures, we can obtain a low-temperature phase with the symmetry $P2_1/b$ which just exchanges the positions of the two glide planes with the two screw axes as shown in Fig. 7 of Ref. 4.

Phase II is obtained by the symmetry breaking associated with $\Gamma^{\{\mathbf{k}_1^*, \tau_3\}}$ alone, so that the low-temperature symmetry pattern will be the same as our previous work^{1,6} which gives $Pbam$ symmetry. According to Fig. 2, the transition may be first or second order depending on the different temperature and pressure ranges. For the transition from phase II to phase III, one should take $Pbam$ as the high-temperature symmetry group and follow the discussion in Ref. 10.

Finally, we take spontaneous strain effects into account, i.e., the homogeneous elastic distortions which have been measured experimentally.³ Taking only $\{\eta_i\}$ as the order

parameter, we already gave the spontaneous strain variables and the coupling of the order parameter to the strain variables in Ref. 11. Now, we have both $\{\eta_i\}$ and $\{\xi_i\}$ as the order parameters and the low-temperature D_1 phase has the point symmetry C_{2h} which is lower than that in the $Drd(pbam)$ phase (D_{2h}). Fortunately, the allowable spontaneous strain variables are still χ_1 and χ_2 (see Ref. 11) which give the distortion of the two-dimensional lattice structure. Another possible spontaneous strain variable is the shear deformation in the plane perpendicular to C_2 axis. However, this corresponds to sliding the column axis which in a columnar system costs no energy. In other words, such a shear deformation cannot be supported by sliding the column axes. The coupling between $\{\xi_i\}$ and $\{\chi_i\}$ has the same form as that between $\{\eta_i\}$ and $\{\chi_i\}$. Therefore, the total free energy F_{tot} is

$$F_{\text{tot}} = F + F_e + F_c, \quad (2.20)$$

$$F_e = \frac{1}{2} D^0 (\chi_1^2 + \chi_2^2), \quad (2.21)$$

$$F_c = \delta_1 [\chi_1 (2\eta_1^2 - \eta_2^2 - \eta_3^2) + \sqrt{3}\chi_2 (\eta_2^2 - \eta_3^2)] \\ + \delta_2 [\chi_1 (2\xi_1^2 - \xi_2^2 - \xi_3^2) + \sqrt{3}\chi_2 (\xi_2^2 - \xi_3^2)]. \quad (2.22)$$

The rest of the discussion is the same as in Ref. 11. For example, the spontaneous strains in $[1,0,0;\pm 1,0,0]$ states are

$$\chi_1^0 = -\frac{2}{D^0} (\delta_1 \eta + \delta_2 \xi), \quad (2.23a)$$

$$\chi_2^0 = 0. \quad (2.23b)$$

III. CONCLUSION

We have taken two IR's of the symmetry group of the high-temperature D_2 phase as the order parameters for the D_2 - D_1 phase transition in the HAT columnar liquid crystals. By minimizing the free energy we constructed, we have shown that the herringbone structure in the low-temperature D_1 phase can be obtained by crossing a first-order transition line, $\psi=0$ (transition between phases II and III in Fig. 2). Two possible arrangements of the herringbone structure and six orientational states for each are just as previously pointed out by other authors.^{3,4} A possible phase diagram was also given. This suggests that, in some range of temperature and pressure, the two IR's may be decoupled from each other so that the transition turns out to be associated with only one IR and the induced symmetry pattern in the low-temperature phase will be the same as our previous work.^{1,6}

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