Orientational order and phase transitions in columnar liquid crystals

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An orientational order mechanism, which may lead to a symmetry change from a higher-temperature hexagonal columnar phase to lower-symmetry phases with two-dimensional rectangular symmetry, is considered. A tensor density function is invoked to describe the symmetry change through the phase transition. For a transition corresponding to a symmetry breaking in the plane perpendicular to the column axes, the molecular orientation in the plane at the low-temperature phase is determined. Transitions in which there is a symmetry breaking out of the plane are briefly discussed.

I. INTRODUCTION

The existence of D_1 and D_2 (or D_{rd} and D_{hd}) phases in hexa(n-alkanoyloxy)triphenylene (HAT) series has been reported by several authors.[1,2] If the molecular core and the attached chains are in a plane perpendicular to the column axes in both the D_1 and D_2 phases, the symmetry change through the transition from the D_2 to the D_1 phase can be considered to appear only in the plane perpendicular to the column axes; i.e., only the in-plane symmetry breaking needs to be considered. In this case, the seventeen two-dimensional space groups[3] are suitable to describe the symmetry change.[4,5] Based on these considerations, the authors have suggested a possible mechanism to realize the phase transition corresponding to the symmetry change from $P6mm$ to $P2gg$ due to the rearrangement of the molecular chains.[6] This procedure gives the symmetry patterns suggested by Destrade *et al.*[2] in their early classification of the mesophases of disclike mesogens.

However, qualitative optical observations[7] and a synchrotron x-ray study of freely suspended discotic strands[8] in triphenylene hexa (n-dodecanoate) (HAT 11) disclosed that the average orientation of the normal to the molecular planes in the D_1 phase is not parallel to the column axes, but tilted away from the column axes to give a herringbone structure.[5,8] Safinya *et al.*[8] found that the molecular core was tilted even in the D_2 phase at an angle of about 28° with respect to the column axis and remained almost the same across the transition to the D_1 phase. They suggested that, in the D_2 phase, the orientations of the molecular cores in each column were randomly distributed around the axis without long-range order. Here, we will start from a high-symmetry D_2 phase, and search for the possible low-symmetry phases with a two-dimensional (2D) rectangular structure due to an orientational order mechanism. Since the symmetry properties under reflection in the plane perpendicular to the column axes, as well as in-plane symmetries, may be different in the high- and low-temperature phases, the seventeen 2D space groups are not sufficient. For example, the symmetry of the D_1 phase was assigned[8] to be $P2_1/a$, which is one of the 80 space groups which have been suggested to describe meso-

phases in three-dimensional space with $\mathbb{Z}^2 \times \mathbb{R}$ translational structures.[9] Correspondingly, the symmetry of the D_2 phase can be assigned to be $P6/mmm$. To include orientations in a discussion of the transition, we introduce here a second-order tensor density function instead of a scalar density function.[6] The Landau free energy can then be expanded as a power series in the independent tensor elements, according to irreducible representations (denoted by IR's hereafter) of the $P6/mmm$ space group. We find that one of the IR's which corresponds to in-plane symmetry breaking will lead to the same results as our previous work.[6] However, the tensor density function makes possible the determination of the in-plane molecular orientation with respect to the 2D lattice structure in the low-temperature rectangular phase. This result may be compared with experiment. The lattice distortion in the low-temperature rectangular phase can be explained as a spontaneous strain effect.[10]

In Sec. II, we will introduce the tensor density function to describe the orientational symmetry breaking. All the irreducible representations are given by following considerations similar to those used in magnetic systems.[11] In Sec. III, we will consider the in-plane symmetry breaking which leads to the determination of the in-plane molecular orientation relative to the 2D lattice structure. Finally in Sec. IV, we will give a brief discussion of our results.

II. TENSOR DENSITY FUNCTION

We first consider a uniaxial nematic liquid crystal. The anisotropic properties of such a system can be described by a constant second-order tensor, Q. In the principal Cartesian coordinate system, it has the form:

$$Q = \begin{bmatrix} Q_\perp & 0 & 0 \\ 0 & Q_\perp & 0 \\ 0 & 0 & Q_\parallel \end{bmatrix} .$$

The D_2 phase (the disordered hexagonal columnar phase) can be imagined being condensed from the above uniaxial homogeneous system by locating the molecular

centers randomly along the discrete column axes to give a two-dimensional lattice order. The orientational anisotropy in the D_2 phase can then be described by a tensor function $Q_0(r)$, which has the form

$$Q_0(r) = \begin{bmatrix} Q_\perp(r) & 0 & 0 \\ 0 & Q_\perp(r) & 0 \\ 0 & 0 & Q_{||}(r) \end{bmatrix} .$$

Here, $Q_\perp(r)$ and $Q_{||}(r)$ are only nonzero when r is very close to the column axes.

The symmetry group of the D_2 phase is one of the 80 space groups which are suitable for describing $Z^2 \times R$ mesophases as suggested by Goshen et al.[9] We identify it as $G_0 = P6/mmm$. The symmetry operations $g \in G_0$ are denoted as $g = (h_\alpha \mid m\mathbf{a}_1 + l\mathbf{a}_2)$, where the h_α's $(\alpha = 1$ through 24) are the 24 rotational operations as defined by Kovalev;[13] also, $\mathbf{a}_1$ and $\mathbf{a}_2$ are the basis vectors of the 2D hexagonal lattice; and m and l are arbitrary integers. We choose the Cartesian coordinate system so that the x_3 axis is parallel to the column axes, with respect to which the tensor components are defined (Fig. 1). Now, $Q_0(\mathbf{r})$ should be invariant under all $g \in G_0$, i.e.,

$$D^{-1}(h_\alpha)Q_0(g^{-1}\mathbf{r})D(h_\alpha) = Q_0(\mathbf{r}) , \tag{1}$$

where $D(h_\alpha)$ is the matrix corresponding to the transformation of coordinates $x_i' = D_{ij}(h_\alpha)x_j$ (i and j run over the three Cartesian indices). Near the transition in the low-temperature phase, the tensor function is

$$Q(\mathbf{r}) = Q_0(\mathbf{r}) + \delta Q(\mathbf{r}) . \tag{2}$$

The components of $\delta Q(\mathbf{r})$, $\delta Q_{ij}(\mathbf{r})$, can be expanded as

$$\delta Q_{ij}(\mathbf{r}) = \sum_{n,\alpha} T_{ij}^{n,\alpha}\Phi_{n\alpha}(\mathbf{r}) , \tag{3}$$
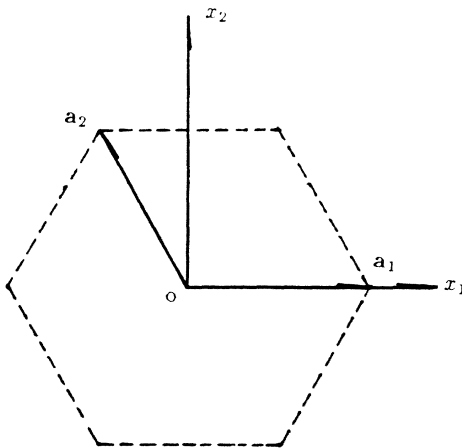

FIG. 1. $\mathbf{a}_1$ and $\mathbf{a}_2$ are the basis vectors of the two-dimensional hexagonal lattice structure (dotted lines), with respect to which the symmetry operations h_1 through h_{24} are defined; x_1, x_2, and x_3 define the Cartesian coordinate system with respect to which the tensor component Q_{ij} is defined; x_3 is along the direction perpendicular to the plane of the paper.

where $\Phi_{n\alpha}(\mathbf{r})$ is the αth basis function of the nth IR of G_0.

For each space group, different n's can be distinguished by two indices, a star of a wave vector, $\mathbf{k}_1^* = \{\mathbf{k}_1, \dots\}$, and an IR of the group of $\mathbf{k}_1$,[13-15] τ. Not all $\mathbf{k}_1^*$ are associated with the transition of interest. The only wave vector star which may give the rectangular lattice structure in the low-temperature phase is

$$\mathbf{k}_1^* = \{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3\} \equiv \{ \tfrac{1}{2}\mathbf{b}_1, \tfrac{1}{2}\mathbf{b}_2, -\tfrac{1}{2}\mathbf{b}_1 + \tfrac{1}{2}\mathbf{b}_2 \}$$

as shown in Fig. 1 of Ref. 6. Here $\mathbf{b}_1$ and $\mathbf{b}_2$ are the basis vectors of the 2D reciprocal lattice. So,

$$\delta Q_{ij}(\mathbf{r}) = \sum_{p=1}^{3} \sum_\tau T_{ij}^{\mathbf{k}_p,\tau}\Phi_{\mathbf{k}_p}^\tau(\mathbf{r}) , \tag{4}$$

where $\Phi_{\mathbf{k}_p}^\tau(\mathbf{r})$ are the basis functions of the IR $\Gamma^{\{\mathbf{k}_1^*,\tau\}}$, which is three dimensional. The real forms of the $\Phi_{\mathbf{k}_p}^\tau$ are

$$\Phi_{\mathbf{k}_p}^\tau(\mathbf{r}) = u_{\mathbf{k}_p}^\tau(\mathbf{r})\cos(\mathbf{k}_p \cdot \mathbf{r}) \tag{5}$$

where, $u_{\mathbf{k}_1}^\tau(\mathbf{r})$ is the basis function of the IR of the group of $\mathbf{k}_1$, and the $u_{\mathbf{k}_p}^\tau(\mathbf{r})$'s are invariant under the translation $\mathbf{t} = m\mathbf{a}_1 + l\mathbf{a}_2$.

Since the distribution of the molecular centers is the same in both the high- and low-temperature phase, only the unit representation τ_1 needs to be taken in the summation over τ.[11] So, we have

$$\delta Q_{ij}(\mathbf{r}) = \sum_{p=1}^{3} T_{ij}^{\mathbf{k}_p}\cos(\mathbf{k}_p \cdot \mathbf{r}) . \tag{6}$$

For a second-order phase transition (or a first-order transition with small latent heat), near the transition, the Landau free energy can be expanded as a power series in $T_{ij}^{\mathbf{k}_p}$, and each expansion term is invariant under G_0. All such invariants can be obtained from the transformation properties of $T_{ij}^{\mathbf{k}_p}$. In Eq. (6), the $\cos(\mathbf{k}_p \cdot \mathbf{r})$ are the basis of an IR $\Gamma^{\mathbf{k}_1^*,\tau_1}$ of G_0. In contrast to the case of a scalar density function, here the coefficients $T_{ij}^{\mathbf{k}_p}$ themselves transform like the components of a second-rank tensor. Thus, we need the direct product of the representation $\Gamma^{\mathbf{k}_1^*,\tau_1}$ and the representation formed by the components of a second-rank tensor. The direct product representation space is usually reducible. It can be reduced to a direct summation over several irreducible subspaces, labeled by an index τ, for fixed $\mathbf{k}_1^*$, by taking suitable linear combinations of the $T_{ij}^{\mathbf{k}_p}$. These linear combinations are

$$\eta_1^{\tau_1} = \frac{1}{\sqrt{2}}(T_{11}^{\mathbf{k}_1} + T_{22}^{\mathbf{k}_1}) , \tag{7a}$$

$$\eta_1^{\tau_1'} = \frac{1}{\sqrt{2}}(\tfrac{1}{2}T_{11}^{\mathbf{k}_1} - \tfrac{1}{2}T_{22}^{\mathbf{k}_1} + \sqrt{3}T_{12}^{\mathbf{k}_1}) , \tag{7b}$$

$$\eta_1^{\tau_1''} = T_{33}^{\mathbf{k}_1} , \tag{7c}$$

$$\eta_1^{\tau_3} = \frac{1}{\sqrt{2}}\left[\frac{\sqrt{3}}{2}T_{11}^{\mathbf{k}_1} - \frac{\sqrt{3}}{2}T_{22}^{\mathbf{k}_1} - T_{12}^{\mathbf{k}_1} \right] , \tag{7d}$$

$$\eta_1^{\tau_5} = \frac{1}{\sqrt{2}}(\sqrt{3}T_{13}^{k_1} + T_{23}^{k_1}) , \tag{7e}$$

$$\eta_1^{\tau_7} = \frac{1}{\sqrt{2}}(-T_{13}^{k_1} + \sqrt{3}T_{23}^{k_1}) . \tag{7f}$$

The superscript τ labels the IR of the group of $\mathbf{k}_1$ and is the same as that of Kovalev.[13] In (7a), (7b), and (7c), τ_1, τ_1', and τ_1'' refer to the identity representation. For a given τ, η_2^{τ} and η_3^{τ}, which are associated with $\mathbf{k}_2$ and $\mathbf{k}_3$, are obtained from η_1^{τ} by the action of h_2 and h_3.[13,14]

Now we are ready to discuss the symmetry breaking through the phase transition.

III. THE IN-PLANE SYMMETRY BREAKING

According to Landau's theory,[15] an ordinary second-order phase transition is only associated with one of the IR's from which the free energy is constructed. The low-temperature phases induced by τ_1, τ_1', and τ_1'' will have higher symmetry than the phase in which we are interested. Thus we begin by considering the symmetry breaking due to τ_3. For compactness, $\eta_i^{\tau_3}$ is simply denoted as η_i. The Landau free energy will be

$$F = \frac{1}{2}r\sum_{i=1}^{3}\eta_i^2 + u\left[\sum_{i=1}^{3}\eta_i^2\right]^2 + v\sum_{i=1}^{3}\eta_i^4 \tag{8}$$

which has the same form as Eq. (12) of Ref. 6.

By minimizing F in the low-temperature phase, we find that the six solutions for $[\eta_1,\eta_2,\eta_3]$ are $[\pm\eta,0,0]$, $[0,\pm\eta,0]$, and $[0,0,\pm\eta]$ for $v<0$ and $u+v>0$.[6] Their orientations in the irreducible space are $[\pm1,0,0]$, $[0,\pm1,0]$, and $[0,0,\pm1]$, respectively. Let us consider the $[1,0,0]$ state. According to Eq. (7), this requires that

$$\eta_1(\equiv\eta_1^{\tau_3})=\eta, \quad \eta_2(\equiv\eta_2^{\tau_3})=0, \quad \eta_3(\equiv\eta_3^{\tau_3})=0 ,$$

and the rest of η_i^{τ}'s are equal to zero. So, $T_{11}^{k_1}$, $T_{22}^{k_1}$, and $T_{12}^{k_1}$ can be determined by putting $\eta_1^{\tau_3}(\equiv\eta_1)=\eta$, $\eta_1^{\tau_1'}=0$, and $\eta_1^{\tau_1'}=0$. Thus,

$$\frac{1}{\sqrt{2}}\left[\frac{\sqrt{3}}{2}T_{11}^{k_1} - \frac{\sqrt{3}}{2}T_{22}^{k_1}\right] = \eta , \tag{9a}$$

$$\frac{1}{\sqrt{2}}(T_{11}^{k_1} + T_{22}^{k_1}) = 0 , \tag{9b}$$

$$\frac{1}{\sqrt{2}}(\tfrac{1}{2}T_{11}^{k_1} - \tfrac{1}{2}T_{22}^{k_1} + \sqrt{3}T_{12}^{k_1}) = 0 . \tag{9c}$$

We obtain

$$T_{11}^{k_1} = \frac{\sqrt{6}}{4}\eta , \tag{10a}$$

$$T_{22}^{k_1} = -\frac{\sqrt{6}}{4}\eta , \tag{10b}$$

$$T_{12}^{k_1} = -\frac{\sqrt{2}}{4}\eta . \tag{10c}$$

Inserting this solution into Eq. (6), we have

$$\delta Q(\mathbf{r}) = \frac{\sqrt{2}}{4}\eta\begin{bmatrix} \sqrt{3} & -1 & 0 \\ -1 & -\sqrt{3} & 0 \\ 0 & 0 & 0 \end{bmatrix}\cos(\mathbf{k}_1\cdot\mathbf{r}) . \tag{11}$$

It is easy to check that the remaining symmetry operations of $P6/mmm$ which keep $\delta Q(\mathbf{r})$ invariant are $\tilde{g}\in G\subset G_0$ with

$$\tilde{g} = \begin{cases} (h_\alpha \mid 2m\mathbf{a}_1 + l\mathbf{a}_2), & \alpha=1,4,13,16 \\ (h_\alpha \mid (2m+1)\mathbf{a}_1 + l\mathbf{a}_2), & \alpha=7,10,19,22 . \end{cases} \tag{12}$$

Now we choose $\mathbf{a}=2\mathbf{a}_1+\mathbf{a}_2$, $\mathbf{b}=-\mathbf{a}_2$ as the basis vectors in the low-temperature phase, and define h_α's in this rectangular reference system.[13] Equation (12) can then be rewritten as

$$\tilde{g}=(h_\alpha \mid \sigma_\alpha+k\mathbf{a}+l\mathbf{b}), \quad \alpha=1,2,3,4,25,26,27,28 , \tag{13a}$$

where

$$\sigma_\alpha = \begin{cases} (a+b)/2, & \text{if } \alpha=2,3,26,27 \\ 0, & \text{if } \alpha=1,4,25,28 . \end{cases} \tag{13b}$$

We can verify that all the symmetry operations $\tilde{g}\in G$ belong to space group $Pbam$. The existence of h_{28} in Eq. (13) [or h_{16} in Eq. (12)] means that the out-of-plane symmetry is not broken in the low-temperature phase. This result is the same as our previous work,[6] except that we use the space group notation $Pbam$ instead of the 2D space group notation $P2gg$. However, the equilibrium form of the tensor density function Eq. (11) has the in-plane molecular orientation with respect to the 2D lattice structure included. To see this, we only need to check the local orientation near a given column, say at $\mathbf{r}=0$, which gives

$$\delta Q(0) = \frac{\sqrt{2}}{4}\eta\begin{bmatrix} \sqrt{3} & -1 & 0 \\ -1 & -\sqrt{3} & 0 \\ 0 & 0 & 0 \end{bmatrix} . \tag{14}$$

By rotating the coordinate system about the x_3 axis by an angle ϕ, we find a new reference system in which $\delta Q(0)'$ is diagonal. It is not difficult to find that

$$\phi=15° \tag{15a}$$

and

$$\delta Q'(0) = \frac{\sqrt{2}}{4}\eta\begin{bmatrix} 2 & 0 & 0 \\ 0 & -2 & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{15b}$$

as shown in Fig. 2(a).

The new coordinate system $x_1' - x_2'$ is now the principal system, so that x_1' and x_2' are along the longer and shorter axes of the ellipselike molecular pattern. Taking the lattice distortions in the low-temperature phase into account, we find that the angle ψ [Fig. 2(a)] is not equal to 60°, so that ϕ will deviate from 15° and can be determined by simple geometric considerations to be

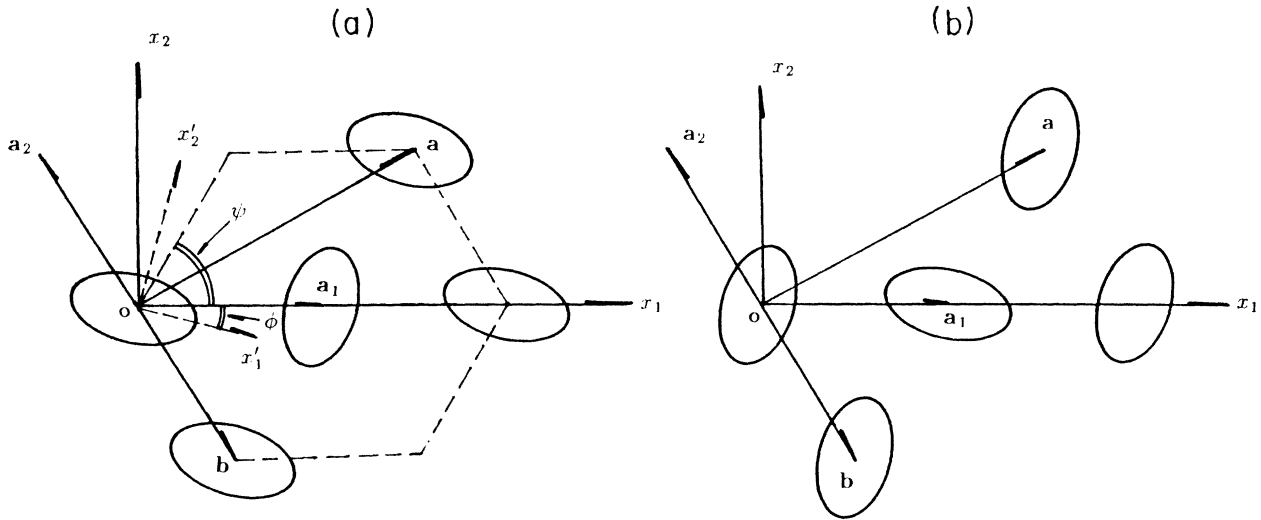$$\phi=45° - \arctan\frac{b}{a} . \tag{16}$$

FIG. 2. The symmetry patterns of the low-temperature phase induced by τ_3; (a) and (b) correspond to [1,0,0] and [−1,0,0] orientational states, respectively; $x_1' - x_2'$ represents the principal coordinate system in which the local orientational tensor $\delta Q_{ij}(0)$ is diagonal; $a=2a_1+a_2$ and $b=-a_2$.

Up to now, no measurement of this ϕ has been reported, so this is a prediction to be compared with experiment.

For the [−1,0,0] state, similar procedures will give the low-temperature pattern shown in Fig. 2(b). Figure 2(b) is essentially the same as Fig. 2(a). The two figures correspond to different ways of choosing the unit cell. So, [−1,0,0] gives the same state as [1,0,0]. Two other states can be obtained by rotating Fig. 2(a) [or 2(b)] 60° and 120° about x_3 axis. These correspond to the solutions $[0, \pm 1, 0]$ and $[0,0,\pm 1]$, respectively, and are the so-called three orientational states.[10,12] They may coexist in the low-temperature system. Each state forms a single domain

and is separated from the others by domain walls.[7,8]

We can take the spontaneous strain into account as in Ref. 10. So, we write down the total free energy as:

$$F = \tfrac{1}{2} r \sum_{i=1}^{3} \eta_i^2 + u \left[\sum_{i=1}^{3} \eta_i^2 \right]^2 + v \sum_{i=1}^{3} \eta_i^4$$

$$+ \tfrac{1}{2} D^0 (\chi_1^2 + \chi_2^2) + \delta [\chi_1(2\eta_1^2 - \eta_2^2 - \eta_3^2)$$
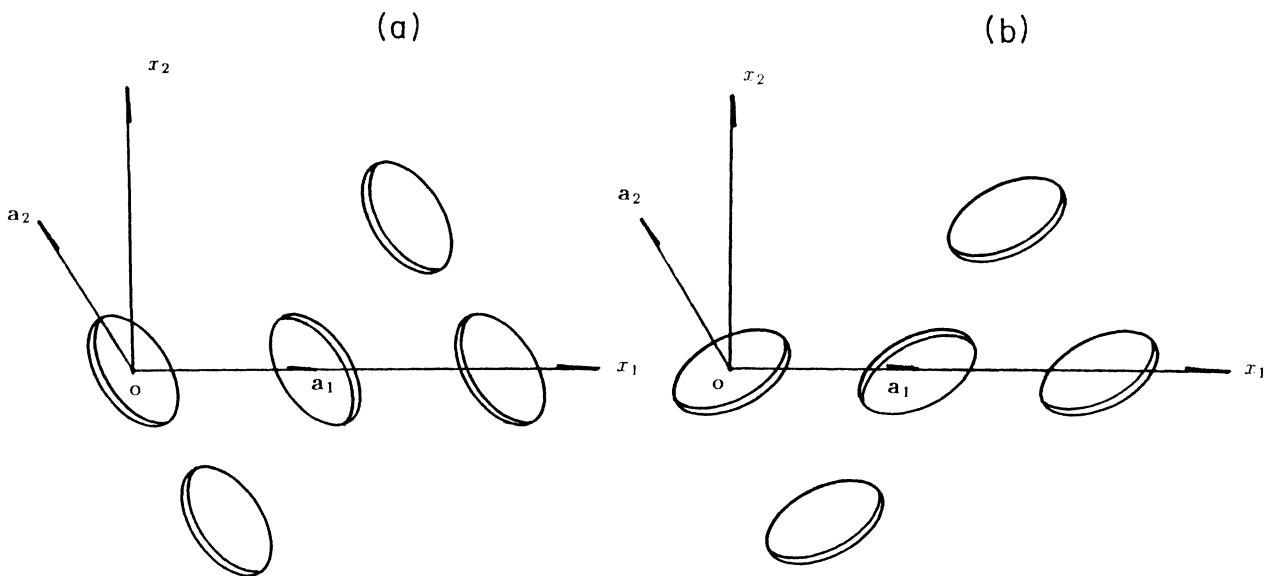
$$+ \sqrt{3}\chi_2(\eta_2^2 - \eta_3^2)] , \qquad (17)$$

FIG. 3. (a) and (b) correspond to the symmetry patterns of the low-temperature phases induced by τ_5 and τ_7, respectively.

where $\mathcal{X}_1$ and $\mathcal{X}_2$ are the strain variables.[10]

Note that, in obtaining Eq. (24), we have already rotated the reference frame $x_1 - x_2$ in Fig. 1 by 60° about the x_3 axis, so that the spontaneous strain variables $\mathcal{X}_1$ and $\mathcal{X}_2$ have the same definition as in Ref. 10. The equilibrium values of $\mathcal{X}_1$ and $\mathcal{X}_2$ in the given orientational state, e.g., [1,0,0], are

$$\mathcal{X}_1^0 = -\frac{2\delta\eta}{D^0}, \quad \mathcal{X}_2^0 = 0 . \tag{18}$$

(See Fig. 1 of Ref. 10.)

IV. DISCUSSION

A. Out-of-plane symmetry breaking

In Sec. III, we discussed only the in-plane symmetry breaking. This led to a state with *Pbam* symmetry. Among the six IR's (see Sec. II), only τ_5 and τ_7 can be invoked to describe the out-of-plane symmetry breaking. Using procedures similar to those in Sec. III, we can obtain low-temperature phases with *Pmna* symmetry as shown in Figs. 3(a) and 3(b) (in the [1,0,0] state), both of which are different from the D_1 phase ($P2_1/a$). In fact, the herringbone structure with $P2_1/a$ symmetry can be only obtained in our work if the symmetry breaking is associated with two irreducible representations, τ_3 and τ_5 or τ_3 and τ_7. For these two cases, the symmetry patterns are the same as Fig. 7 of Ref. 16. However, according to

Landau's theory,[15] if a phase transition is associated with two IR's, it either corresponds to an isolated point in the phase diagram if the transition is second order,[17] or corresponds to a first-order transition.[18] The former is apparently not the case experimentally, and the latter will be discussed in the following paper.[19]

B. Order of transition

The results obtained in the previous sections are based on the framework of Landau's theory. According to Eq. (8), the phase transition is continuous (there is no cubic term in η_i). Theoretically, the correct answer as to whether the transition is first order or second order can not be given until the problem is worked out in the framework of the renormalization group, in which important fluctuation effects near the transition are included. We will not present the renormalization group calculation in this paper, but simply note that a strong coupling between the order parameter and the spontaneous strain variables may give a possible mechanism for driving the transition to be first order.[10,20]

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