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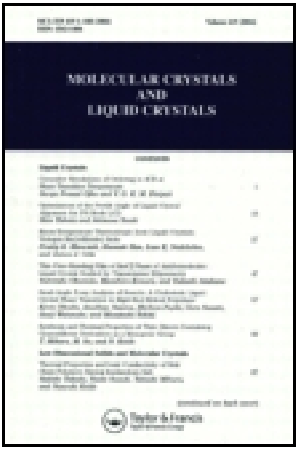

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Frustration between Ferroelectricity and Antiferroelectricity in Extremely Soft Chiral Smectic-C Like Phases of Liquid Crystals

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Frustration between Ferroelectricity and Antiferroelectricity in Extremely Soft Chiral Smectic-C Like Phases of Liquid Crystals

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In a group of compounds that exhibit antiferroelectric and/or ferrielectric liquid crystal phases, the bent molecular shape has been considered to cause unique interactions among smectic layers. The interactions are responsible for the successive phase transitions as described by the devil's staircase and the V-shaped switching, which are related with the interlayer ordering and the diminished tilting correlation, respectively. These two are apparently contradictory each other, but result from the same cause, *i.e.* frustration between ferroelectricity and antiferroelectricity in the Sm-C* like phase. The ANNNI model Yamashita and Miyazima introduced has been shown to describe the frustration appropriately provided that we undertake some refinements: the Ising spin corresponding to the tilting sense of the local in-plane director, the X-Y character of the spin, the tilt angle decrease toward Sm-A, and most importantly, the reassignment of the pseudo-spin to the biasing direction of the molecular rotation about their long axis.

Keywords: bent molecular shape; devil's staircase; disordered chiral smectic-C like phase; V-shaped switching; frustration between ferro- and antiferroelectricity; ANNNI model

INTRODUCTION

More than 1,000 compounds, which have similar molecular structures and exhibit antiferroelectric and/or ferrielectric liquid crystal phases, have been synthesized because of their potential application to displays[1]. Successive phase transitions are frequently observed, and at least five subphases are confirmed to exist; we speculated about the devil's staircase as a cause for the emergence of many subphases[2-4]. Frequently, the successive phase

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transitions induced by temperature or electric field are of first order; in homogeneous cells, an electric-field-induced change in optical light transmission due to the transition between Sm-C_A* and Sm-C* may show the tristable switching, which is characterized by DC threshold, hysteresis, and domain boundary movement[5]. In some of the compounds and mixtures that exhibit the antiferroelectric and/or ferrielectric phases in free-standing films, on the other hand, the thresholdless, hysteresis-free, V-shaped switching is observed in homogeneous cells[6-18]; it occurs quite uniformly with no boundary movement and gives very high contrast ratio of 300 or more. We speculated about a disordered Sm-C_R* phase and its Langevin-like alignment due to an applied electric field.

Before the discovery of antiferroelectric Sm-C_A*, it was considered that the packing entropy effect as well as the intermolecular interactions causes the tilting in the same direction and sense, aside from a slight precession from layer to layer due to chirality[19,20]; Sm-C* as well as Sm-C was the only fluid tilted smectic phase. The successive phase transitions indicate that several interlayer orderings can be stabilized in liquid crystal phases without long-ranged positional order[2-4]. On the contrary, the V-shaped switching seems to suggest that the tilting correlation between smectic layers may diminish to zero practically[6-18]. We believe that not only the interlayer ordering but also the diminished tilting correlation, though they are apparently contradictory each other, are caused by frustration between ferroelectricity and antiferroelectricity in the Sm-C* like phase [4,13,14]. Details are explained in the following.

BENT MOLECULAR SHAPE, SM-C*, and SM-C_A*

The characteristic feature of constituent molecules of the compounds under consideration is that the chiral chain is projecting obliquely from the main core part. This bent molecular shape has been confirmed in Sm-A, Sm-C*, Sm-C_A*, Sm-I*, and Sm-I_A* by X-ray diffraction[21], polarized IR spectroscopy[22,23], ^{13}C-NMR[24], and ^{2}H-NMR[25]. Okuyama *et al.*[26] reported the herringbone molecular arrangement in a crystal structure of MHPBC, which allows us to imagine the arrangement in Sm-C_A*. It is useful to visualize the components of dipole moment density averaged over the X and Y directions, $p_X(Z)$ and $p_Y(Z)$. Here X, Y, and Z constitute the right-handed Cartesian coordinate system and the Y and Z axes are perpendicular to the tilt plane and the smectic layer,

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respectively. Although detailed calculations have not yet been performed, Fig. 6 of Ref. [26] clearly indicates that both $|p_X(Z)|$ and $|p_Y(Z)|$ have large values near the boundary and becomes almost zero near the center of smectic layers.

This tendency does not change much even in Sm-C$_A$*, although thermally activated molecular motions make $p_X(Z)$ and $p_Y(Z)$ more diffuse. There are various motions, such as rotations about the molecular long and short axes, azimuthal angle rotation about the smectic layer normal, and translations within a smectic layer and between smectic layers. Since large transverse dipole moments exist near the chiral center, rotation about the molecular long axis (the eigen axis of the smallest moment of inertia) affect $p_X(Z)$ and $p_Y(Z)$ most seriously. This rotation couples strongly with the azimuthal angle rotational freedom about the smectic layer normal in Sm-C* like phases, because the chiral chain is bent. The coupling biases the rotation about the molecular long axis and produces in-plane spontaneous polarizations at smectic layer boundaries, P_Y in Sm-C* and P_X in Sm-C$_A$*. Irrespective of the coupling, the longitudinal dipole moments also produce P_X in Sm-C$_A$*. Symmetry requires that the in-plane spontaneous polarizations at boundaries should be parallel and perpendicular to the tilt plane; but the biased directions may not be exactly parallel or perpendicular because of the two-fold symmetry around the X and Y axes[1].

SUBPHASES AND ISING / *X*-*Y* SPINS

When Isozaki *et al.*[2] noticed three ferrielectric and one antiferroelectric subphases between Sm-C$_A$* and Sm-C* by drawing electric field - temperature (E - T) phase diagrams, they considered that these subphases constitute a part of the devil's staircase formed by frustration (competition) between ferro- and antiferro-electricity in liquid crystals[2-4]. Since then many experimental investigations on the subphases have been performed, and a possible, most general subphase sequence has been given as

– Sm-C$_A$* – spr3 – Sm-C$_\gamma$* – spr2 – AF – spr1 – Sm-C* – Sm-C$_\alpha$* – Sm-A –

The fundamental phases are Sm-C$_A$* and Sm-C*; the others between them together with Sm-C$_\alpha$* are the subphases; Sm-C$_\alpha$* may emerge on the high temperature side of Sm-C* and hence must be slightly different from the other subphases. Ferrielectric Sm-C$_\gamma$* and antiferroelectric AF phases seem to be

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secondarily fundamental. At least one ferrielectric subphase was observed in each subphase region, spr1, spr2, and spr3. Identification as a ferro-, ferri-, or antiferro-electric phase has been made by an electric-field-induced change in the conoscopic figure; in particular, whenever the melatopoes appear parallel to an applied field, the phase is considered as ferrielectric. It is worthwhile insisting that at least four different types of ferrielectric phases have been confirmed to exist[2-4].

Two statistical mechanics models illustrating the emergence of the devil's staircase have been developed; the one by weak long-range forces is the one-dimensional Ising model and the other by thermal fluctuations is the so-called ANNNI (axial next-nearest neighbor Ising) model with competing nearest and next-nearest neighbor coupling[27,28]. Isozaki *et al.*[2] explained the observed sequence of the subphases in terms of the Ising model with the long-range repulsive interactions, but Fukuda *et al.*[3] and Itoh *et al.*[4] pointed out several difficulties in the explanations. Yamashita and Miyazima[29] tried to apply the ANNNI model to this problem. Their trial together with the subsequent developments has been successful in explaining various experimental facts [29-34]. We now believe that the ANNNI model can describe the complicated phenomena under consideration appropriately if it is refined by taking into account of the *X*-*Y* character of the spins and the tilt angle decrease toward Sm-A. Several investigators have, in fact, also made some explanations in terms of the *X*-*Y* model; unification of the Ising model and the *X*-*Y* model must be necessary in some way [35-39]. Since Yamashita and his collaborators developed the ANNNI model to a considerable extent, we will use their theoretical treatments in this paper as a guideline to understand the complicated phenomena under consideration.

THE ANNNI MODEL

The Hamiltonian Yamashita and Miyazima [29] assumed is

$$\mathcal{H} = -J \sum_{(i,j)} s_i s_j - J_1 \sum_{i}^{A} s_i s_{i+1} - J_2 \sum_{i}^{A} s_i s_{i+2} - J_3 \sum_{i}^{A} s_i s_{i+3} \qquad (1)$$

Here the Ising spin s_i takes a value of ±1 corresponding to the tilting senses of the local in-plane director in the *i*th smectic layer; the second (next)-nearest neighbor interaction J_2 should be negative to ensure frustration, and the third-

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nearest neighbor interaction J_3 (>0 or <0) is included for the possible wide stability of Sm-C_γ*. Two fundamental questions about their treatment are: (1) Why can we disregard the *X-Y* character of the spins and consider the Ising spins? and (2) Is there any realistic physical grounds for the rather long-range interactions, J_2 and J_3? The first question will be answered by disclosing the stabilization mechanism of Sm-C_A* and by showing how easily that mechanism suppresses the *X-Y* character. Some theoretical trials have already been done, clarifying that the phase of Ising symmetry with a large azimuthal angle fluctuation always appears in the high temperature region [34]. As for the second question, Yamashita noticed a freedom regarding the senses of the molecular long axes which was already introduced by Koda and Kimura [40,41], decimated in the partition function the pseudo-spins describing the senses, and eventually obtained the effective long-range interactions, J_2 and J_3.

His treatment inevitably introduces the ordering of the sense of molecular long axes in the subphases [30]. The ordering must be produced by the molecular rotation about their short axes and/or the translational diffusion

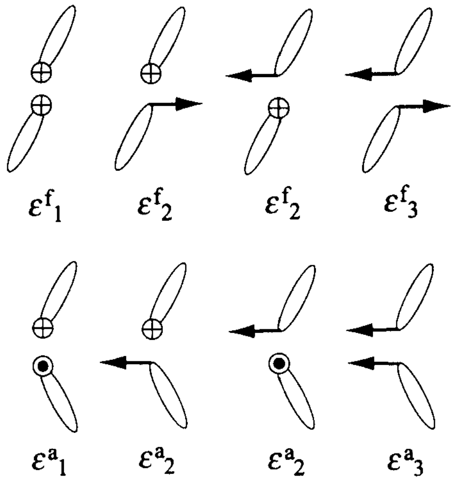

Figure 1 By reassigning the pseudo-spin $t_i = \pm 1$ to the biasing directions of the molecular rotation about their long axes, perpendicular and parallel to the tilt plane, respectively, we can consider the energy parameters as Yamashita did in Fig. 1 of Ref. [30].

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between smectic layers. However, these are not the relevant molecular motions. Moreover, the head-head (chiral chain - chiral chain) interactions must be remarkably strong, as is seen in Fig. 6 of Ref. [26]. It is realistic to consider the head-head interactions only and to relate the ordering with the much more relevant rotational motions about the molecular long axis and about the smectic layer normal. Consequently, we try to reassign the pseudo-spin t_i to the biasing direction of the molecular rotation about the long axis; for simplicity, the rotation is assumed to be completely frozen in either of the directions parallel or perpendicular to the tilt plane. We can assign the energy parameters given in Figure 1, as Yamashita did in Fig. 1 of Ref. [30]. Then we obtain the effective long-range interactions, J_2 and J_3, because all the formulations given by Yamashita also hold for the current energy parameters. In the ferrielectric Sm-C_γ* and antiferroelectric AF phases, the in-plane spontaneous polarizations, parallel and perpendicular to the tilt plane, must be different from those in Sm-C* and Sm-C_A*. This fact is described as a change in the molecular rotational states about their long axes, *i.e.* a change in the number of molecules biased parallel and perpendicularly to the tilt plane. Note that, in the achiral or racemic system, the simple reasingnment is not possible because both senses of the perpendicular direction become equivalent. It is an open problem to describe the molecular rotation more realistically.

PHASE DIAGRAMS

Figure 2 is an example of the phase diagrams obtained from the ANNNI model. The temperature variation of a particular compound is given by a straight line

$$J_1 = ST - J_0 \tag{2}$$

Several phase sequences actually observed are illustrated by lines (a), (b), (c), and (d). If we choose the parameter values appropriately, we will obtain phase diagrams in which Sm-C_γ* and/or AF will practically disappear[32]. The phase sequences Sm-C_A* – Sm-C* in partially racemized MHPOBC and TFMHPOBC and Sm-C_A* – Sm-C_γ* – Sm-C* in enantiomeric MHPOBC can be illustrated; Sm-C* may be replaced by a phase in sprl. In a few compounds FI in sprl has unambiguously been confirmed to exist[4, 42-44]. When the lines are located far distant from the critical curve, the corresponding phase sequences are to be observed stably irrespective of interface effects in homogeneous as

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well as homeotropic cells. When the lines approaches to the critical curve and phases in spr's emerge in wide temperature ranges, the interface effects plays an important role; free-standing films are most suitable for making observations almost free from the effects[4]. This fact is also in accordance with the temperature variation of the order parameters obtained theoretically by Yamashita[31]. Figure 2 indicates the emergence of many subphases in spr's, but experimentally a single phase is observed. Yamashita and Tanaka theoretically showed that an applied electric field stabilizes one of these subphases with q = 2/5 and 2/7 predominantly [33]; note that we always applied an electric field in observing conoscopic figures to identify the subphases. Itoh *et al.* experimentally showed that the apparent tilt angle in the q=2/5 subphase is nearly equal to 1/5 of the tilt angle in Sm-C* induced by an electric field at the same temperature[4]. In this way, the ANNNI model seems to explain most of the phase sequences actually observed.

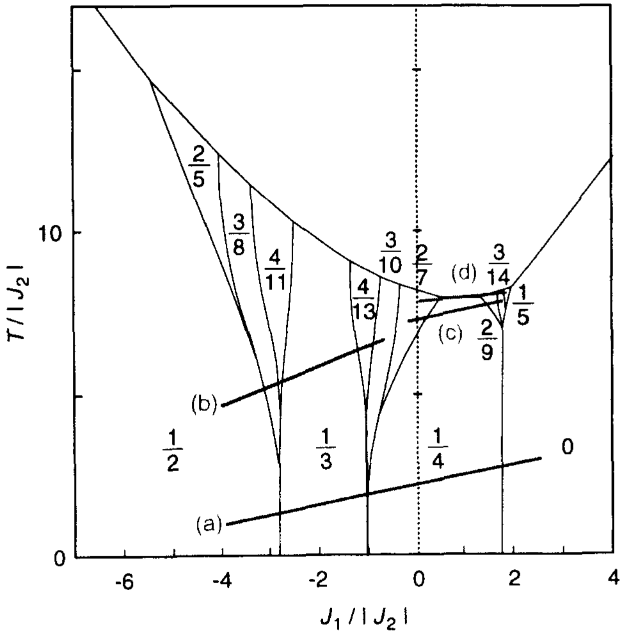

Figure 2 An example of the phase diagrams obtained by Yamashita from the ANNNI model[29-34].

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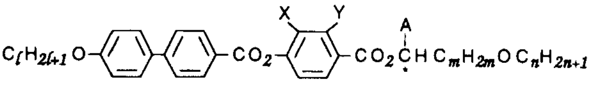
Near the second order critical curve above which the disordered phase emerges, however, some discrepancies may appear. In this connection, the temperature-concentration (*T*-*x*) phase diagram of two component mixtures obtained by Seomun *et al.*[17] is interesting. In the concentration range $0 \leq x \leq 0.4$, the phase sequence is –FI_H–AF–FI–Sm-C*–, which is illustrated by line (c) in Figure 2. Then the AF phase rather abruptly disappears, and ferrielectric phases predominantly emerge. Near $x = 0.5$, Sm-C* seems to exist in the high temperature region. Since not only the helicoidal pitch but also the melatopes of the conoscopic figures continuously change with temperature, however, it is not clear whether Sm-C* really exists and where the transition occurs, if any. Moreover, an applied electric field induces a continuous switching from a ferrielectric phase to ferroelectric Sm-C*. Sometimes, at $x = 0.7$, for example, the helicoidal pitch shows a small discontinuous change as illustrated in Fig. 8 of Ref. [17]; this fact indicates that there exist at least three ferrielectric phases with quite similar structures. In this way, the AF phase does not seem to exist near the minimum of the critical curve; instead, a variety of ferrielectric phases may emerge there; some of them may have structures, which show conoscopic figures with melatopes perpendicular to an applied electric field as Sm-C* does. We speculate that azimuthal angle fluctuations may cause the appearance of these ferrielectric phases. Unification of the Ising model with the *X*-*Y* model is one of the interesting future theoretical problems.

V-SHAPED SWITCHING

The emergence of a single ferrielectric phase in a wide temperature range after the first order phase transition from Sm-A [8] has also been confirmed in free-standing films of a series of compounds with the general chemical structure,



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In homogeneous cells, however, substrate interfaces produce a single novel phase that is different from the ferrielectric one [8,18]. The similar drastic interface effect has been noticed in the two component mixtures, as become clear by comparing Figs. 2 and 6 in Ref. [17]. When an electric field is applied, the thresholdless, hysteresis-free, "V-shaped switching" is observed. Its characteristic features attractive from an application point of view are that

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switching occurs so uniformly without accompanying any boundary motions and that the contrast ratio is as high as 300 or more. The V-shaped switching was first reported in connection with liquid crystal displays (LCDs) [6-8]; Toshiba and Casio have already prototyped the LCDs using the V-shaped switching[45-47].

By analyzing the pretransitional phenomenon in the tristable switching, we speculated about a disordered Sm-C_R* phase where molecular tilting directions (azimuthal angles) are random from layer to layer with a constant polar angle. Because of the extremely anisotropic tilting correlation lengths, $\xi_\perp >> \xi_{||}$, local in-plane spontaneous polarizations may exist at smectic layer boundaries with random orientations and variable magnitudes, and may show the Langevin-like alignment (V-shaped switching) when applying an electric field [1,6,7,14]. The in-plane spontaneous polarization on a boundary is parallel to the bisector of C-directors in the upper and lower layers and its magnitude depends on the C-directors [13]. Such a disordered phase as Sm-C_R* may emerge at least in restricted conditions created by interface effects sometimes in cooperation with an applied electric field [11]; in the bulk, materials so far reported exhibit the ordinary ferro-, ferri, and antiferro-electric phases. It is understandable that Sm-C_R* scarcely emerges, because diminished tilting correlation increases packing entropy and endangers the existence of the tilted phase itself. Extremely anisotropic correlation lengths may compromise the diminished tilting correlation and an increase in packing entropy and allows the existence of Sm-C_R*. A mechanism by which interfaces induce randomization in azimuthal angle distribution has already been discussed [11,14].

A DISORDERED SM-C* LIKE PHASE IN THE BULK

The point at issue is whether the disordered chiral smectic-C like phase, Sm-C_R*, is observed in the bulk sample, e.g. in a free-standing film. We may be able to conclude that Sm-C_R* is almost realized as their bulk property at least in a compound **1** with ℓ=11 X=H, Y=F, A=CF_3, m=4, and n=2. Apparently, a single phase emerges and it is ferrielectric. The helicoidal pitch is rather short at low and high temperatures and in-between it diverges as given in Fig. 2 of Ref. [18]. It is tempting to speculate that J_1 varies with temperature near the minimum of the critical curve as illustrated by line (d) in Figure 2. In the temperature range where the helicoidal structure disappears, the phase may

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become Sm-C_R* and the tilting correlation in adjacent smectic layers must be diminished; the helicoidal structure cannot help existing. Suppose the twisting power stays constant, the less correlation may make the helicoidal pitch much shorter. Accordingly, we can conclude without contradiction that both of the short pitch and its divergence result from the reduced or diminished tilting correlation in adjacent smectic layers.

In this way, Sm-C_R* in the bulk would have no helicoidal structure, exhibit no threshold to an applied electric field, and continuously switch into ferroelectric Sm-C*; a uniform uniaxial texture is expected to be observed as in Sm-A. Actually, however, the in-plane coherence length $\xi_\perp$ larger than the visible wavelength may easily disturb the uniform uniaxial texture. Moreover, the spontaneous C-director bent caused by a linear term in the distortion free energy frequently produces hexagonal and/or rectangular patterns in the texture [48]. Since helicoidal pitch divergence results from several causes, systematic investigations need to be performed in a series of compounds where a single ferrielectric phase emerges in a wide temperature range. In three such compounds preliminarily studied, one shows divergence and the other two have short helicoidal pitches. In Fig. 8 of Ref. [17], the helicoidal pitch also diverges in the ferri 2 phase. Following our speculation based on Figure 2, any of antiferro-, ferri-, and ferro-electric phases may slightly enter into the disordered phase, Sm-C_R*, and the helicoidal pitch may become infinite. Whether it abruptly jumps or gradually diverges to infinity may depend on the structure of an ordered phase under consideration. In a compound **1** with $\ell=11$, X=H, Y=H, A=CF_3, $m=5$, and $n=2$, we observe a jump in Sm-C* but a gradual divergence in an antiferroelectric phase. The relation between the helicoidal pitch behavior and the reduced or diminished tilting correlation in ferro-, ferri-, and antiferro-electric phases seems to be one of the interesting future problems and should be investigated by taking account of azimuthal angle fluctuations.

CONCLUDING REMARKS

As has been explained above, it is natural to consider that frustration between ferro- and antiferro-electricity causes the V-shaped switching [49]. We speculated an ideal cylindrical distribution of in-plane directors around the smectic layer normal on the assumption of a disordered chiral smectic-C like phase, Sm-C_R* [6,7]. What is really essential is that the phase becomes

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extremely soft. Consequently, interfaces may drastically change the in-plane director alignment. In fact, Clark *et al.*[50] insisted that the molecular arrangement at the tip of V is the so-called twisted state stabilized by large spontaneous polarization. We share the experience that the twisted state is easily formed when polar interfaces, such as ITO, are used and that an apparently V-shaped switching is observed. Our accumulated experience clearly indicates, however, that non-polar interfaces are favorable for realizing the V-shaped switching attractive in display applications[9]. Nevertheless, strong anchoring may cause the alignment along the rubbing direction as considered by Pauwells *et al.*[51]. The dielectric property of Sm-C_R* had better be called "frustrated electricity" or "frustoelectricity" although it was initially designated as thresholdless antiferroelectricity. To commercialize liquid crystal displays using frustorelectricity with active matrix electrodes (AM-FR-LCDs), it is important to assure appropriate randomization by balancing ferro- and antiferro-electricity in liquid crystal materials and by developing attractive polyimide aligning films so that the frustration manifest itself to its fullest extent. For checking the randomization, it is useful to analyze in detail, not only the ordinary switching process but also the second harmonic generation (SHG) together with its coherence characteristics[15]. The speculated cylindrical distribution of in-plane directors around the smectic layer normal may be confirmed by polarized IR and /or Raman spectroscopy.

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