Bi-layer-dimerized chiral liquid crystals.

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Abstract

There is a large variety of bi-layered structures with smectic A type ordering. Results of this paper contribute to a theory of bi-layered phases. The difference between the $SmC^*_A$ and $SmC^*_\alpha$ phases is described in this latter paper and it is explained by existence and non-existence of the dipole moment pairs.

I. INTRODUCTION

In [1] authors discuss several types of bilayer smectic liquid crystals with ferroelectric and antiferroelectric properties in binary mixture of dimeric compounds. The mesomorphic behavior and phase structure were examined in [1] the mixture of two kinds of dimeric compounds, $\mu$-bis(4-alkoxyanilinebenzylidene-4-carbonyloxy)pentane (mOAM5AMOm), by optical microscopy, X-ray diffraction, polarization switching, and second-harmonic generation measurements. One compound is 4OAM5AMO4 with a short terminal alkyl chain that forms a single-layer smectic phase ($SmC_A$) with a random mixing of spacer and tail groups. Another compound is 16OAM5AMO16 with a long terminal alkyl chain that forms a chiral, anticlinic, and antiferroelectric bilayer phase ($SmC_{\alpha}$) with the bent molecules tilted to the bilayer. By mixing these two compounds, the $SmC_A$ phase of 4OAM5AMO4 is easily destabilized, leading to the wide content region of the bilayer phases. In the bilayer regime, three other smectic phases are newly induced. Two of them are antiferroelectric and ferroelectric phases in which the molecules lie perpendicularly with respect to the layer. The other shows no polar response to an external electric field and behaves like a smectic A. The new appearance of these bilayer phases is discussed as a mixing effect of long and short tail groups. Thus it is useful to discuss bi-layer-dimerized chiral liquid crystals theoretically.

Let us note that liquid crystal television display panel has application e.g. in tactical aircraft. Its key advantages are: (1) high contrast in small and large areas, (2) shade capability under all levels of illumination including direct-sunlight, (3) uniform high resolution over the entire display area, (4) interface similar to CRT TV display and (5) low power, weight, volume. Cockpit installations have been designed for the display which permit viewing under day and night conditions, see in [2].

Isotropic liquids represent the most symmetric phase: there is no discrete translational symmetry, the local point group symmetry is present only. Usually the point group in liquids is identified with the point symmetry of a group of molecules forming the unit, from which the liquid or liquid crystal is formed.

In the isotropic symmetry the unit has either full orthogonal symmetry group $K_h$, either a group $K$ of rotations around a point where inversion is absent.

A phase transition due to symmetry lowering of the isotropic liquid may occur, usually there are transitions to the nematic or the Sm A (centrosymmetric, nonchiral) phases ($K_h \rightarrow D_{\infty h}$), Sm A (without the inversion center, chiral) ($K \rightarrow D_{\infty}$), and to cholesteric phases. Symmetry groups $D_{\infty}$ and $D_{\infty h}$ correspond to all [3] known uniaxial liquid crystals with those phases. These liquid crystals are characterized by the fact that both directions along the axis of the full axial symmetry are equivalent. The director $\mathbf{n}$ orientation is equivalent to the opposite director orientation $-\mathbf{n}$.

The order parameter for these liquid crystals may be found using the well-known tensor $Q_{ij}$:

$$Q_{ij}(\mathbf{r}) = < l_i l_j > - \frac{1}{3} < l_k l_k > \delta_{ij},$$
which is formed as the local average of the quadratically mixed long molecular unit axis $l$ projections. It is possible to rewrite the above order parameter expression into the form

$$Q_{ij}(r) = Q(r)\langle n_i(r)n_j(r) - \frac{1}{3}\delta_{ij} \rangle.$$ \hfill (1)

Here the tensor amplitude $Q(r)$ tells us in which extent the molecules, forming the unit, are aligned in the direction given by the director $n$ (and equivalently $-n$).

Long molecules of lower molecular weight may have permanent electric dipole moments which are generally oriented to the long molecular axes. Ferroelectricity due to their alignment is not observed due to several main reasons [4]:

1. dipole-dipole interaction is weak and the thermal energy $k_BT$ destroys such an order,
2. when the molecular dipole moments are large, the fluidity leads to formation of dimers, in which an antiparallel orientation of the molecular dipoles exist, thus the effective dipole moment is canceled,
3. the flexoelectric effect may lead to deformation which as a consequence is relaxed by the formation of defects.

A model which is widely accepted for the antiferroelectric smectic phases suppose that the molecules in the neighbouring layers tilt in the opposite directions, and so also the polarization point in the opposite directions. Two vectors $\theta_1$ and $\theta_2$, representing orientations of molecules in the alternating layers, may be defined

$$\theta_{1,2} = (-n_{1,2y}n_{1,2z}, n_{1,2x}n_{1,2z}).$$

where $n_{1,2}$ are the directors in the odd-numbered and even-numbered layers, respectively. The axis $z$ is perpendicular to the layer.

Such a model assumption corresponds with expectation that there are weaker interlayer correlations between molecules while inlayer interactions are stronger. These latter include also interactions leading to in-layer dimerization. Dipole-dipole forces may however strongly influence also interactions between molecules localized in different neighbouring layers. Then the interlayer dimerization between neighbouring layers may occur. Competition between the in-layer and inter-layer dimerization may be influenced by such factors as the in-layer space restrictions for molecular grouping and the value of the molecular dipole moment. In extreme the interlayer dimerization processes supported by the in-layer geometry restrictions may be leading forces driving formation of the layered liquid crystal structure when cooling the isotropic liquids.

It is this situation, in which pairing dimerization occurs predominantly between molecules on both sides of a layer interface, which we concern with in this paper. In traditional approach localization of these dimers is considered to be in the middle of every layer.

Due to interlayer pairing of molecules we expect that the invariance of the crystal with respect to the change of the direction $n$ to $-n$ in every layer is broken. Molecular groups (dimers), which are invariant with respect to the inversion are situated between two molecular layers.

One may expect that this new kind of liquid crystal ordering in which inter-layer dimerization prevails may, under appropriate conditions, occur when decreasing temperature of the isotropic liquid. In this case in the isotropic liquid tend to correlate more and more their movements. This process may be characterized by two correlation lengths: that of the above mentioned dimerization process and that of the interactions leading to a layered structure. Usually the characteristic dimerization length has been expected to be smaller or comparable with the interlayer forces the characteristic length of which is approximately equal to the interlayer spacing. If, however, the latter is smaller than the former then the inter-layer dimerization becomes realistic; i.e. dimerization groups centers are localized at the boundaries of neighbouring layers.

Our aim in the following part of the paper is to construct appropriate theory describing ordering and transitions just described, and to discuss experimental consequences and observations within the frame of this theory.

## II. ORDER PARAMETER

As a consequence of our expectation that molecular "dimer" groups are preferentially formed between two neighbouring smectic layers of molecules, two vectors $\theta'_1$ and $\theta'_2$, representing orientations of molecules in the alternating neighbouring layers, may be defined

$$\theta'_1 = (-n_{1,y}n_{2,z}, n_{1,x}n_{2,z}), \theta'_2 = (-n_{2,y}n_{1,z}, n_{2,x}n_{1,z}).$$

where $n_{1,2}$ are the directors in the odd-numbered and even-numbered layers, respectively. The axis $z$ is perpendicular to the layer.
TABLE I: Different behaviour of the smectic layered phases under inverse transformations.

| layer | 1 | 1 | 1 | 1 |
|-------|---|---|---|---|
|       | 2 | 1 | 1 | 1 |
| layered ordering type | [5] | 1 | 1 | 1 |
| this paper | 1 | -1 | -1 | 1 |

Symmetric (s) and antisymmetric (a) combinations of both vectors may be formed:

\[
s = \theta_1' + \theta_2', a = \theta_1' - \theta_2'.
\]  

While s does not change its sign under a basic translational symmetry: transformation shifting layers by one layer thickness, it is not the case of the antisymmetric combination a. The quantity s has symmetry properties of the in-bilayer ferroelectric polarization. The latter quantity represents the in-bilayer antiferroelectric polarization.

Both types of the mentioned axial vectors become the same quantity whenever the ordering of layers one and two is the same. However, they are different if the geometrical and dipolar forces tend to dimerize groups of molecules in adjacent layers.

Another comparison, results of which are displayed in Table 1, shows different behaviour of the bilayer vectors s and a when the inverse transformations i (\( n_{1,2} \rightarrow -n_{1,2} \)) are performed in all four combinations for two adjacent layers: layers 1 and 2 remained unchanged, the layer 1 inverted only (the remaining layer 2 is unchanged), the layer 2 inverted only (the remaining layer 1 is unchanged), and the layers 1 and 2 simultaneously inverted.

In the Table 1 we denote by 1 and by i the identity and the in-layer inverse transformations. The third and fourth rows contain results of this transformations on vectors s and a obtained when the ferroelectric and antiferroelectric phases are defined by the way used in [5] and in this paper. Symbol 1 denotes resulting identity transformation in the axial vectors s, a space: (s ↔ s) and (a ↔ a) ; and symbol -1 represents result of inverse transformation in the axial vectors s,a space: (s ↔ -s) and (a ↔ -a).

The most pronounced difference between our new order parameters and the traditional ones becomes apparent whenever phases with modulation of the ground state in the direction perpendicular to the smectic layers occur. If this modulation is weak and with long wavelength with respect to the interlayer distance a, then one can relate director vectors in neighbouring layers:

\[
n_{2,\alpha} = n_{1,\alpha} + a \frac{\delta n_{1,\alpha}}{\delta z}, n_{1,\alpha} = n_{2,\alpha} - a \frac{\delta n_{2,\alpha}}{\delta z}.
\]  

Our new order parameters \( \theta_1' \) and \( \theta_2' \) are related to the well-known order parameters \( \theta_1 \) and \( \theta_2 \):

\[
\theta_1' = \theta_1 + a(-n_{1,y} \frac{\delta n_{1,\alpha}}{\delta z}, n_{1,x} \frac{\delta n_{1,\alpha}}{\delta z}), \theta_2' = \theta_2 - a(-n_{2,y} \frac{\delta n_{2,\alpha}}{\delta z}, n_{2,x} \frac{\delta n_{2,\alpha}}{\delta z}).
\]  

Both sets of order parameters \( \theta \) and \( \theta' \) become identical, whenever there is no gradient change of the director orientation from layer to layer. If this modulation is not weak then one can relate director vectors in neighbouring layers using finite differences of directors:

\[
n_{2,\alpha} = n_{1,\alpha} + \delta,
\]  

and obtains, that our new order parameters \( \theta_1' \) and \( \theta_2' \) are related to the order parameters \( \theta_1 \) and \( \theta_2 \):

\[
\theta_1' = \theta_1 + (-n_{1,y}\delta, n_{1,x}\delta), \theta_2' = \theta_2 - (-n_{2,y}\delta, n_{2,x}\delta).
\]  

Both sets of order parameters \( \theta \) and \( \theta' \) become identical, whenever the finite difference \( \delta \) vanishes.

Thus we conclude, that our order parameters are more general than those traditionally used, the latter mentioned are special realizations of the former.
III. FREE ENERGY

Up to the fourth order the free energy $F$ expansion in small amplitudes of the order parameters $s$ and $a$ has the form

$$F = \frac{\alpha}{2} I_1 + \frac{a}{2} I_2 + cI_1 I_2 + dI_3,$$  \hspace{1cm} (8)

due to the fact that there are only the following second order invariants

$$I_1 = s^2, I_2 = a^2,$$

and the fourth order invariants

$$I_3 = (s,a)^2, I_1 I_2, I_1^2, I_2^2.$$  \hspace{1cm} (9)

There are five ordered low-symmetry phases: the ferroelectric phase

$$s \neq 0, a = 0;$$  \hspace{1cm} (9)

the antiferroelectric phase

$$a \neq 0, s = 0;$$  \hspace{1cm} (10)

the ferrielectric phase A ($a \parallel s$)

$$a \neq 0, s \neq 0;$$  \hspace{1cm} (11)

the ferrielectric phase B ($a \perp s$)

$$a \neq 0, s \neq 0;$$  \hspace{1cm} (12)

the ferrielectric phase C ($0 < a.s < |a|.|s|$)

$$a \neq 0; s \neq 0.$$  \hspace{1cm} (13)

Until now we did not consider any difference between ordering in which both neighbouring layers are ordered in the same way (uni-layer-ordering) and ordering in which neighbouring layers are ordered in a different way (bi-layer-ordering). Our free energy (8) does not contain a term which breaks the symmetry between bi-layer-ordering and uni-layer-ordering. This term will be discussed latter on.

The ordering of molecules which corresponds to these five phases can be conveniently described introducing usual spherical coordinates $\phi$ and $\Theta$ in each individual layer

$$n_i = (\cos(\phi_i)\sin(\Theta_i), \sin(\phi_i)\sin(\Theta_i), \cos(\Theta_i)).$$  \hspace{1cm} (14)

Note, that while the azimuthal angle $\phi$ takes values from 0 to $2\pi$ (excluding the last value), the tilt angle takes values from 0 to $\pi$ (excluding the last value) due to absence of the inversion in-layer symmetry.

Let us discuss all possible molecular ordering corresponding to individual phases. For the ferroelectric phase (9) we obtain from the condition $a=0$ that the rotation and tilt angles for the layer 1 and the layer 2 are related in this phase via the following equations:

$$\sin(\phi_1)\sin(\Theta_1)\cos(\Theta_2) = \sin(\phi_2)\sin(\Theta_2)\cos(\Theta_1)$$

and

$$\cos(\phi_1)\sin(\Theta_1)\cos(\Theta_2) = \cos(\phi_2)\sin(\Theta_2)\cos(\Theta_1).$$

In correspondence with the mentioned double degeneracy there are two possibilities how to satisfy these equation conditions. The first one is that in which there is equal rotation angle in both layers $\phi_1 = \phi_2$, and equal tilt angle in both layers $\Theta_1 = \Theta_2$. This state corresponds to the usually considered description of the smectic ferroelectric phase. In the other one the rotation angles in neighbouring adjacent layers are differing by 180 degrees $\phi_1 = \phi_2 \pm \pi$, and tilt angles in both layers are complementary to 180 degrees differing $\Theta_1 = \pi - \Theta_2$. This state does not corresponds
to any traditionally accepted ordering of the smectic ferroelectric phase. It is a new state, let us name this state as bi-layered-ferroelectric state. Both types of the ferroelectric states are schematically visualized in Fig.1.

In the antiferroelectric phase the condition \( s=0 \), ref9, holds and we obtain that the rotation and tilt angles for the layer 1 and the layer 2 are related in this phase via the following equations:

\[
\sin(\phi_1)\sin(\Theta_1)\cos(\Theta_2) = -\sin(\phi_2)\sin(\Theta_2)\cos(\Theta_1)
\]

and

\[
\cos(\phi_1)\sin(\Theta_1)\cos(\Theta_2) = -\cos(\phi_2)\sin(\Theta_2)\cos(\Theta_1).
\]

There are again two possibilities how to satisfy these equation conditions. The first one is that in which rotation angles in neighbouring adjacent layers are differing by 180 degrees \( \phi_1 = \phi_2 \pm \pi \), and tilt angles in both layers are complementary to 180 degrees also differing \( \Theta_1 = \Theta_2 \). This state corresponds to the usually accepted description of the smectic antiferroelectric phase.

The other one is characterized by equal rotation angle in both layers \( \phi_1 = \phi_2 \), and equal tilt angle in both layers \( \Theta_1 = \pi - \Theta_2 \). This state does not corresponds to any traditionally considered smectic antiferroelectric phase. It is a new state, let us name this state as bi-layered-antiferroelectric state. Both possibilities in which antiferroelectric ordering realizes are schematically visualized in Fig.2.

The ferrielectric phases, (11) - (13), A where \((\sigma || s)\), B where \((\sigma \perp s)\), C where \((0 < a.s < |a||s)\) may be similarly discussed as ferroelectric and antiferroelectric phases above.

It is possible to discuss phenomenological theory of ordering transitions between just described phases, where also inhomogeneous terms contributing to the free energy play important role. Such a description will be made in another paper together with discussion of behaviour of relevant quantities.

Here we concentrate our attention on the bi-layered antiferroelectric phases. Our aim is to discuss mechanisms which, on the semiphenomenological level, lead to bilayered structures. In the next section we develop a semiphenomenological theory of the McMillan type which enables to obtain better insight how new kind of antiferroelectric ordering discussed above may evolve from the isotropic phase. The case of ferroelectric and ferrielectric phases is not considered in this paper in order to simplify illustration of our model assumptions and consequences.

IV. GENERALIZED McMILLAN THEORY OF STRONGLY DIPOLAR LIQUIDS OF THE SM A TYPE

In this section we discuss a generalized form of the McMillan theory of strongly dipolar liquids. In its original form it is describing liquids in which there are weak inter-layer correlations between molecules while strong in-layer interactions are responsible for the mechanism of the molecular ordering with prevailing in-layer dimerization. The theory presented below is our generalization of the McMillan version [6] of the mentioned theory for traditional type of Sm A ordering described in to those materials in which dimerisation effects between neighbouring layers are strong. McMillan generalized the Maier-Saupe theory introducing a new order parameter, which characterizes one dimensional translational periodicity of the layered smectic structure. In the next section we generalize this McMillan theory by introducing another new order parameter, which characterizes bi-layered ordering due to strong dipolar interactions resulting in interlayer dimerisation of molecules.

A. The effective potential

Anisotropic part of the pair interaction potential is assumed to have the form

\[
V_{12} = -\frac{V_0}{2r_0\sqrt{\pi}} \exp\left(-\frac{r_{12}^2}{r_0^2}\right)(3\cos^2(\theta_{12}) - 1)
\]

where short range character of the interactions is described by the exponential part of the potential and where \( r_{12} = r_1 - r_2 \) is the distance between centers of two interacting molecules 1 and 2, \( \theta_{12} = \theta_1 - \theta_2 \) is the angle between long axis of these molecules, and where \( V_0 \) is the interaction potential energy constant, \( r_0 \) is a characteristic molecule length, \( \theta_{1,2} \) and \( r_{1,2} \) are tilt angles and positional vectors of the molecules 1 and 2. The potential (15) leads, adopting steps similar as in [6], to a semiphenomenological form of the effective potential describing interaction of a given molecule with the effective medium in layers of the thickness \( a \):

\[
V = -V_0(\Sigma + \sigma a \cos(\frac{2\pi z}{a}))\frac{1}{2}(3\cos^2(\Theta) - 1),
\]

(16)
where $V_0$ is the phenomenological energy constant, $z$ is a coordinate in the ordering direction, $\alpha$ is a phenomenological constant describing relative strength of energy scales for nematic and smectic ordering, $\alpha = 2 \exp(-\frac{(\pi z)^2}{a})$, $\theta$ is a tilt angle of a given molecule with respect to the effective medium, $\Sigma$ is the nematic order parameter, $\sigma$ is the smectic order parameter.

For $\Sigma$ nonzero this potential prefers nonlayered nematic-type dipole ordering. For $\sigma$ nonzero layered ordering of the is smectic type is preferred. In the Fig.3 the most advantageous orientations of long molecules (dimerization) in the traditional, case (I.) and in the case considered here (II.) are shown.

To model bi-layered ordering we assume that there exists an additional anisotropy potential due to dipolar forces in neighbouring layers and due to geometrical restrictions in layered ordering. When constructing that potential we have in mind that strongly polar molecules are characterized by the short range interactions of the Van der Waals type and by dipolar interactions. While the first type of interactions is already included in the form of the effective potential, the latter is not. The effective medium, which interacts with the given molecule is formed by the nonpolar background and a polar group of molecules in the neighbouring layer. This latter interaction may be described by the same way as in nematics when studying the antiferroelectric short range order. Thus it is expected to be proportional to $\cos(\Theta)$. In other words we expect that there is a tendency to orient the molecule head with respect to the neighbouring layer. To describe the layer forming tendency we expect, that a term $\cos(2\pi z a)$ will be the most important term. Note that in this term there is the length of two layer separation 2a due to the fact that we expect prevailing orientation in a smectic layer with the molecular heads up (down) and in the next layer down (up). Namely this bi-layered structure has periodicity 2a. Thus summarizing we expect that a new effective potential term has the form

$$V' = -V_1 \cos(\theta)\cos(\frac{\pi z}{a}),$$  \hspace{1cm} (17)$$

where $V_1$ is assumed to be positive. This potential prefers orientation of molecules $\Theta = 0$ for even layers $z=2$ (etc.) and $\Theta = \pi$ for odd layers $z=1$ (etc.) The total effective potential is composed from the basic part (16) and from the additional anisotropy part (17)

$$V_T = -V_0((\Sigma + \alpha \cos(\frac{2\pi z}{a})) \frac{1}{2}(3 \cos^2(\Theta) - 1) + S \gamma \cos(\theta)\cos(\frac{\pi z}{a}))$$  \hspace{1cm} (18)$$

where

$\gamma = \frac{V_1}{V_0}$.  

The total effective potential depends on the orientation of molecules $\theta$ within the layers.

**B. Thermodynamics**

The single particle effective distribution function has the form

$$f(\theta, z) = \exp(-\frac{V_T(\theta, z)}{k_B T})$$  \hspace{1cm} (19)$$

Selfconsistency conditions are

$$\Sigma = <\frac{3 \cos^2(\theta) - 1}{2} >$$

$$\sigma = <\cos\left(\frac{2\pi z}{a}\right) \frac{3 \cos^2(\Theta) - 1}{2} >$$

$$S = <\cos\left(\frac{\pi z}{a}\right) \cos \theta >$$  \hspace{1cm} (20)$$

The free energy $F=U-TS$, where $U$ is the internal energy, $S$ is the entropy and $T$ is temperature, may be easily calculated. Here

$$U = -\frac{1}{2} NV_0(\Sigma^2 + \alpha \sigma^2 + S^2 \gamma),$$  \hspace{1cm} (22)$$

and

$$-TS = NV_0(\Sigma^2 + \alpha \sigma^2 + S^2 \gamma) - Nk_B T \ln \frac{1}{2a} \int_0^{2a} dz \int_0^1 d(cos(\theta)) f(z, \theta).$$  \hspace{1cm} (23)$$
C. High temperatures

High temperature free energy expansion may be easily calculated from (22) and (23):

\[
F = \frac{N V_0}{2} \left[ (1 - \frac{T_0 \Sigma}{T}) \Sigma^2 + \alpha (1 - \frac{T_0 \sigma}{T}) \sigma^2 + \gamma (1 - \frac{T_0 S}{T}) S^2 \right] + \text{higher order terms}
\]

where

\[ T_0 \equiv \frac{V_0}{k_B}, \]

\[ T_0 \Sigma \equiv \frac{T_0}{5}, \]

\[ T_0 \sigma \equiv \frac{T_0 \alpha}{10} < T_0 \Sigma \]

with

\[ 0 \leq \gamma < 1, \]

and where

\[ T_0 S \equiv \frac{T_0 \gamma}{6} \]

and where

\[ \frac{5 \gamma}{6} < 1 \]

D. Phase transitions

Within the McMillan theory presented above there are two possible sequences of phase transitions. If

\[ \frac{5 \gamma}{6} > \frac{\alpha}{10} \]

then

\[ I \rightarrow N \rightarrow Sm_A \rightarrow Sm(AF) \]

Note that the condition above is equivalent to

\[ \gamma > \frac{6}{25} \exp \left( \frac{-\pi r_0}{a} \right)^2, \]

which holds for molecules with characteristic length \( r_0 \) much larger than the interlayer spacing \( a \), and systems in which additional anisotropy forces are not negligible. The phase \( Sm_A \) is a new bi-layered phase, the phase \( Sm(AF) \) is of the antiferroelectric type described above. The McMillan theory presented here is not constructed to describe the ferrielectric phases mentioned in the first part of the paper.

Another possibility is:

If

\[ \frac{5 \gamma}{6} < \frac{\alpha}{10} \]

then

\[ I \rightarrow N \rightarrow Sm(AF) \rightarrow Sm_A \]
Structure and phase transitions in smectic A liquid crystals with polar and sterical asymmetry are discussed in [7] and [8]. There is a large variety of bi-layered structures with smectic A type ordering. As far as we know a theory which describes as well traditional ferro-, antiferro- and ferri-electric phases as corresponding new bi-layered phases is absent. Results of this paper may, at least partially, cover this gap. Antiferroelectric chiral smectic liquid crystals are discussed in [4]. The difference between the SmC*\textsubscript{A} and SmC*\textsubscript{a} phases is described in this latter paper and it is explained by existence and non-existence of the dipole moment pairs.

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