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Orientational ordering in discotic columnar phases
A theoretical approach

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We discuss the different transitions from hexagonal to lower symmetry phases which result from a molecular tilt inside the columns. Several orientational structures are explained and others are predicted. Our model also applies to other physical phenomena, in particular the elliptical deformation of columnar aggregates in lyotropic liquid crystals

1. Introduction

Columnar phases are obtained with flat molecules which we shall take to be disc-like. They actually have a discrete symmetry: in most cases it is defined by a binary or ternary, eventually quaternary, axis perpendicular to their plane. Numerous interesting features are associated with the corresponding degrees of freedom and will not be considered here. We note that in a columnar phase the molecules are stacked in columns which form a two dimensional lattice which is triangular or rectangular in practice. There is no positional long range order inside one column (that is a one dimensional liquid). Typically the sequence isotropic–nematic–triangular columnar is obtained so that the principal nematic axis yields the axis of the column. However, when the temperature is lowered, other columnar mesophases appear in which the orientational order is more complex. In other words the director departs from the column axis, for a review see [1, 2]. Figure 1 shows several typical orientational conformations encountered in various discotic compounds. In most cases the rectangular cell is obtained through a slight distortion of the triangular primitive cell. In this paper the driving mechanism of the transition is assumed to be orientational in nature. The crystallographic changes are simply induced, as they are in magnetic transitions for instance. For this reason they are hereafter neglected. However it should be recalled that such a coupling to the lattice distortion can change the order of the transition from two to one.

In view of the large number of parameters entering the problem, this paper is not intended to give an extensive study, which would involve complicated phase diagrams. Omitting computational details, we focus on the description of the theoretical ingredients required to explain the structures discovered so far. Emphasis is made on the similarities and differences with other physical systems exhibiting structures akin to those of discotics mesophases.

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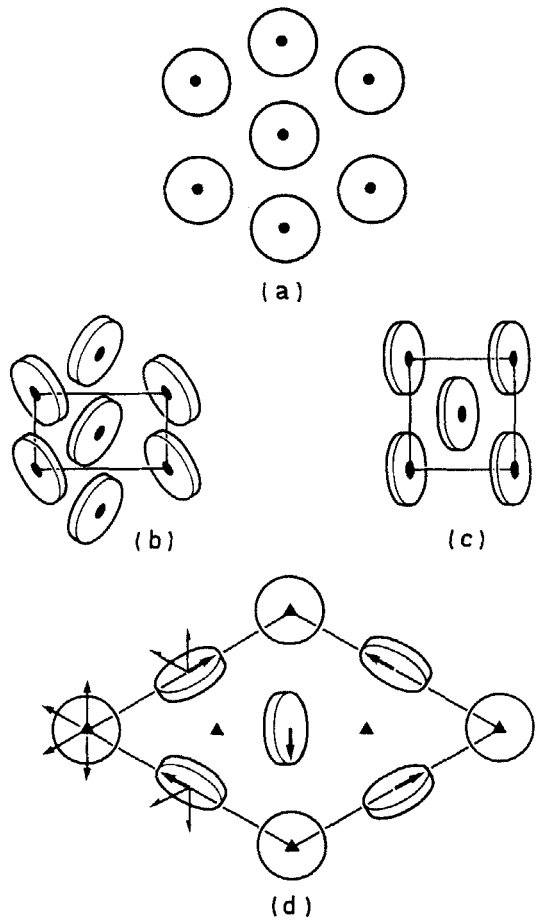

Figure 1. (*a*) The hexagonal phase D_{hd}. (*b*) The rectangular phase $D_{rd}(P2_1/a)$. (*c*) The rectangular phase $D_{rd}(C2/m)$. (*d*) The 4-sublattice phase of symmetry P(321). Experimental evidence for *a*, *b*, *c* can be found in [2], and for *d* in [9].

2. Model

We investigate the symmetry breaking that may occur if we start from a triangular columnar phase in which the molecular director is parallel to the column axis z. This has also been studied by Sun and Swift [12] on pure group theoretical grounds. To gain more insight into the mechanism of the transitions, we introduce a semi-microscopic model, to be studied à la Landau. No equilibrium state is assumed *a priori*, this stems from a rather realistic choice of interaction. We also predict new phases not obtained in [12, 13, 14].

In the absence of the others, each column has a full rotational invariance about z as well as through the reflection $z \rightarrow -z$. The irreducible representations of group D_∞ are complex one dimensional and labelled by an index n. Under rotation through an angle θ, ψ_i on column i transforms into

$$\psi_i \rightarrow \exp(in\theta)\psi_i.$$

Here we shall only discuss $n = 1$ and $n = 2$ although some of the results are valid for all n. In so doing, a tilt of the director with respect to z can be dealt with. Referring

to the general nematic order parameter, the tensor Q, we define

$$\psi^{(1)} = Q_{xz} + iQ_{yz}, \quad \text{(index 1)},$$
$$\psi^{(2)} = Q_{xx} - Q_{yy} + i(Q_{xy} + Q_{yx}), \quad \text{(index 2)};$$

note that if $z \to -z$, $\psi^{(1)} \to -\psi^{(1)}$, $\psi^{(2)} \to \psi^{(2)}$. The case $\psi^{(1)} = 0$, $\psi^{(2)} \neq 0$ represents a nematic order of the projection (n_x, n_y) of the director, e.g. a situation where the molecules are tilted by $\pm\alpha$ ($\pm$ at random) from the xy plane about a given axis in xy. In addition, the representation $n = 2$ describes two cases without tilt: (i) a secondary nematic order in the xy plane, e.g. if the molecules have a binary axis parallel to z (that is rectangular shaped molecules), and (ii) an elliptic deformation of the tubes as in lyotropic systems [3].

We now write a free energy form for the order parameter $\psi_i^{(n)}$

$$F^{(n)} = F_{\mathrm{L}}^{(n)} + F_{\mathrm{I}}^{(n)}.$$

$F_{\mathrm{L}}^{(n)}$ (L for local) takes into account the statistical mechanics of an isolated column, i.e. without interactions between $\psi_i^{(n)}$ and $\psi_j^{(n)}$ for different columns i and j. For instance, assuming rotational invariance for each isolated column, we can use the classical expression

$$F_{\mathrm{L}}^{(n)} = \sum_i \int dz \left[\left| \frac{d}{dz} \psi_i^{(n)} \right|^2 + \mu_n |\psi_i^{(n)}(z)|^2 + b_n |\psi_i^{(n)}(z)|^4 \right]. \tag{1}$$

The reader might object that this symmetry is not appropriate in a discrete lattice. This remark is relevant and is taken into account in §4. Formula (1) represents only a first order description (rather like the Bragg–Williams approximation). The interaction is contained in $F_{\mathrm{I}}^{(n)}$ for which we make the following choices:

$F_{\mathrm{I}}^{(n)}$ couples nearest neighbour columns (notation $\langle i, j \rangle$)

$$F_{\mathrm{I}}^{(n)} = \int dz \sum_{\langle i,j \rangle} U^{(n)}(\mathbf{R}_{ij}; \psi_i^{(n)}, \psi_j^{(n)});$$

$U^{(n)}$ is quadratic in $\psi^{(n)}$;

$U^{(n)}$ is invariant under any overall rotation $\mathscr{R}_\theta$ about z which is to be understood as

$$\mathbf{R}_{ij} \to \mathscr{R}_\theta \mathbf{R}_{ij}, \quad \psi_i^{(n)} \to \mathscr{R}_\theta \psi_i^{(n)} = \mathrm{e}^{\mathrm{i}n\theta} \psi_i^{(n)}, \quad \psi_j^{(n)} \to \mathscr{R}_\theta \psi_j^{(n)} = \mathrm{e}^{\mathrm{i}n\theta} \psi_j^{(n)}.$$

These prescriptions lead to

$$F_{\mathrm{I}}^{(n)} = \int dz \left[-C_0 \sum_{\langle ij \rangle} \psi_i^{(n)} \psi_j^{(n)*} - \sum_{\langle ij \rangle} C_{-2n}(\mathbf{R}_{ij}) \psi_i^{(n)} \psi_j^{(n)} + \text{c.c.} \right], \tag{2}$$

with the general form for interactions C_0 (isotropic) and C_{-2n} (tensorial)

$$C_0 = D_0 \exp(\mathrm{i}\delta_0),$$
$$C_{-2n}(\mathbf{R}_{ij}) = D_{-2n} \exp[-2ni(\theta_{ij} - \delta_{-2n})].$$

D_0 and D_{-2n} are positive numbers and θ_{ij} is the polar angle of vector $\mathbf{R}_{ij}$ in the xy plane with respect to an arbitrary direction. This model is not suitable to deal with the breaking of translational invariance along the z direction. This actually occurs in some compounds (e.g. helical order $//z$) and is left for a later study.

3. Landau theory for continuous transitions

In this section we investigate the different structures which may break the symmetry of the high temperature phase (nematic + triangular) assuming a second order

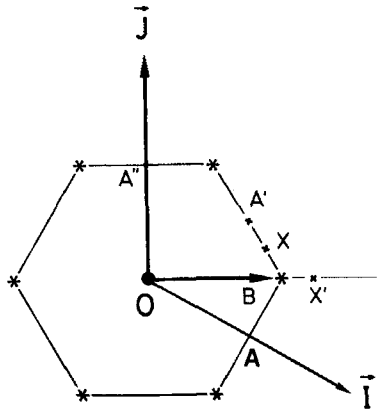

Figure 2. The first Brillouin zone of the triangular lattice. (**I**, **J**) is a basis of the dual lattice. $\mathbf{q}_A = \mathbf{OA}$ (or A', A''), $\mathbf{q}_B = \mathbf{OB}$, $\mathbf{q}(x) = \mathbf{OX}$, $\mathbf{q}'(x) = \mathbf{OX}'$.

transition. Then the order parameter corresponds to the lowest eigenvalue of the quadratic part of $F^{(n)}$ or equivalently $F_1^{(n)}$ (apart from the diagonal term $\mu|\psi_i|^2$). Since all solutions are independent of z, this variable is systematically dropped from now on. After Fourier transforming in the xy plane we find the eigenvalues of $F_1^{(n)}$

$$m^{\pm}(\mathbf{k}) = -\operatorname{Re} C_0(\mathbf{k}) \pm |C_{-2n}(\mathbf{k})|.$$

Due to the sixfold symmetry of the lattice it turns out that $C_{-2}(\mathbf{k})$ and $C_{-4}(\mathbf{k})$ are, apart from a phase factor, complex conjugate to each other and $m^{\pm}(\mathbf{k})$ are the same for $n = 1$ and $n = 2$. The critical modes are given by the wavevector $\mathbf{k}$ which minimizes $m^-(\mathbf{k})$. The stationary points of $m^-(\mathbf{k})$ in the first Brillouin zone (see figure (2)) are:

(i) $\mathbf{k} = 0$, $m^-(0) = -6D$;

(ii) $\mathbf{k} = \mathbf{q}_A$, $\mathbf{q}_{A'}$, $\mathbf{q}_{A''}$, $m^-(\mathbf{q}_A) = 2D(1 - 2x)$;

(iii) $\mathbf{k} = \mathbf{q}(x)$ or equivalently by symmetry $\mathbf{q}'(x) \| \mathbf{q}_B$ where

$$\cos\frac{q'(x)a}{2} = -\frac{1}{4}\frac{2-x}{1+x},$$

$$m^-(\mathbf{q}(x)) = 2D\,\frac{9x^2 + 12x + 12}{8(1+x)},$$

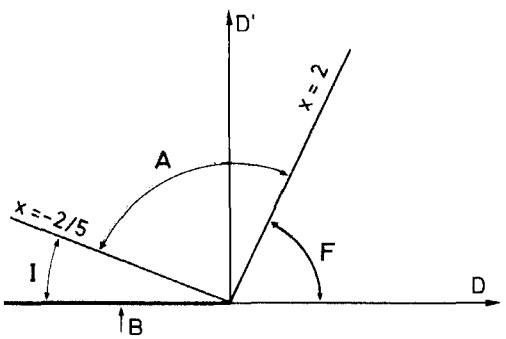

Figure 3. Diagram showing the critical wavevector as a function of the point ($D = D_0 \cos\delta_0$, $D' = D_{-2}$ or D_{-4}): $\mathbf{k} = 0$ (region F), $\mathbf{k} = \mathbf{q}_A$ (region A), $\mathbf{k} = \mathbf{q}(x)$ (region I), $\mathbf{k} = \mathbf{q}_B$ (on the half-line $D' = 0$, $D < 0$).

with the definition $D = D_0 \cos \delta_0$, $x = (D_{-2}/D)$ or (D_{-4}/D). Comparing the different values of m^- given here yields the critical wavevector as a function of D and D_{-2} (or D_{-4}). This is shown in the diagram (see figure 3)).

4. Discussion

4.1. *The ferro phase (region F)*

In the context of columnar phases, this would be the $D_{rd}(C2/m)$ (see figure 1(*c*)) structure, usually interpreted by a uniform tilt of the molecules. Note that with our model energies (1) and (2), this ferro solution at $q = 0$ has a full rotational degeneracy (this is a consequence of $C_{-2n}(k = 0) = 0$). Only anharmonic terms having the symmetry of the crystal can fix the orientation with respect to the lattice.

4.2. *Two-sublattice and four-sublattice phases (region A)*

(1) These phases are often encountered in various fields: adsorbed H_2 or N_2 molecules [4], hexatic smectics [5], lyotropic systems [3], and columnar phases. They are usually called chevron or herringbone structures. For elongated molecules adsorbed on a triangular lattice (such as grafoil), the molecular orientation on one sublattice is generally orthogonal to the other (i.e. the T structure) [4]. But in the other situations this property does not hold.

We now discuss how this feature can be taken into account in our models. In region (*A*) of figure (3) the order parameter $\psi^{(n)}$ ($n = 1, 2$) for which $F^{(n)}$ is minimum (with the simple quartic term $b_n|\psi|^4$) is

$$\psi_i^{(n)} = \psi_A^{(n)} \exp(i\mathbf{q}_A \cdot \mathbf{R}_i) \quad (\text{or } A', A''). \tag{3}$$

The ψs on the two sublattices are opposite to each other. For $n = 1$ we have an antiferromagnet like structure (shown in figure 4(*a*)). For $n = 2$ reversing the sign in $\psi^{(2)}$ amounts to a physical rotation of $\pi/2$: this is the *T* structure given in figure 4 (*b*)). The existence of this phase has been predicted theoretically by Harris and Berlinsky [4] for a pure electric quadrupolar interaction (their 2-*in* phase). In our model this would impose fixed relative values of $D_0^{(1)}$, $D_{-2}^{(1)}$, $D_0^{(2)}$ and $D_{-4}^{(2)}$. In equation (3) the

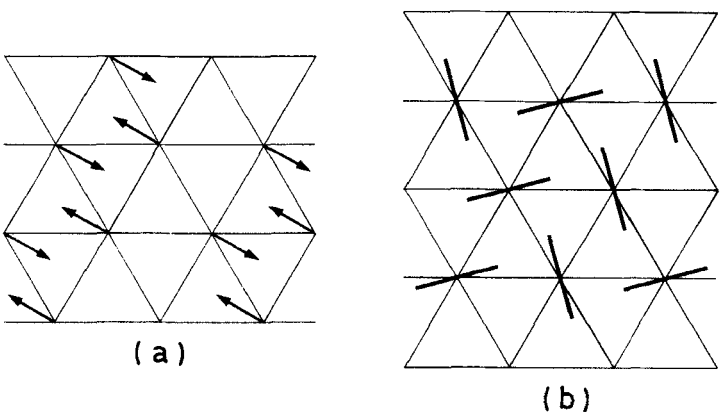

Figure 4. (*a*) Order parameter pattern for $n = 1$, $\mathbf{q} = \mathbf{q}_A$. The vector $(\mathrm{Re}\,\psi^{(1)}, \mathrm{Im}\,\psi^{(1)})$ is shown for $\delta_{-2} = 0$. (*b*) Order parameter pattern for $n = 2$, $\mathbf{q} = \mathbf{q}_A$, $\delta_{-4} = 0$. The rods represent $(\psi^{(2)})^{1/2}$.

orientation of $\psi^{(n)}$ with respect to the lattice is fixed by the anisotropic part D_{-2n}

$$\psi_A^{(n)} \propto \exp\left(-n\delta_{-2n} + \frac{n\pi}{3} - \frac{\pi}{2}\right).$$

(2) We further investigate the case $n = 2$. The existence of structures other than T has been reported both experimentally [3, 5] and theoretically [7, 8]. If the angle is different from $\pi/2$, summing ψ_i over the lattice gives a non-zero contribution and the structure has to be described with an extra Fourier component at $q = 0$, like a ferrimagnet for instance. Actually the T structure exists only if the coupling with the crystalline order is weak. For $n = 2$ the crystal field which has a sixfold symmetry, induces a cubic term

$$\sum_i \Gamma_{-6}(\psi_i^{(2)})^3 + \text{c.c.} = \gamma \sum_i \left(\sum_j e^{-6i\theta_{ij}}\right)(\psi_i^{(2)})^3 + \text{c.c.}$$

Fourier transformation of this yields the relevant couplings

$$\Gamma_{-6} \quad \psi^{(2)}(\mathbf{q} = 0)\psi^{(2)}(\mathbf{q}_A)\psi^{(2)}(\mathbf{q}_A), \tag{4}$$

$$\Gamma_{-6} \quad \psi^{(2)}(\mathbf{q}_A)\psi^{(2)}(\mathbf{q}_{A'})\psi^{(2)}(\mathbf{q}_{A''}). \tag{5}$$

This has several consequences: (i) because of (5) the transition is driven first order, and (ii) the ordered state is then such that

$$\psi^{(2)}(\mathbf{q}_A) = \exp(-2i\pi/3)\psi^{(2)}(\mathbf{q}_{A'}) = \exp(-4i\pi/3)\psi^{(2)}(\mathbf{q}_{A''}). \tag{6}$$

This superposition of three plane waves is a 4-sublattice structure (see figure 5). One of the sublattices is characterized by a zero order parameter. This phase has also been obtained in [6] for quadrupoles on a triangular lattice. It is strongly reminiscent of the configuration (P321) shown in figure 1(*d*)) seen by Levelut *et al.* [9]. (iii) If γ is small enough, the 2-sublattice phase is stabilized at a lower temperature because the quartic term then dominates. Moreover a uniform component at $q = 0$ is induced by equation (4). The orientations on the two sublattices are no longer orthogonal: this is the herringbone structure (see figure (6)). We note that the uniform component is

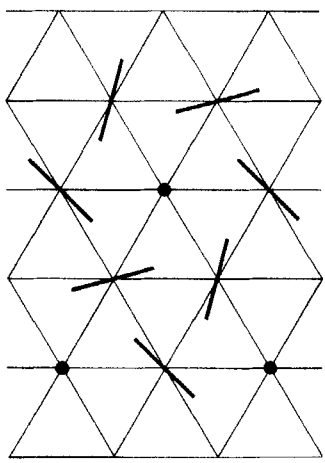

Figure 5. The 4-sublattice structure for $n = 2$ (rods).

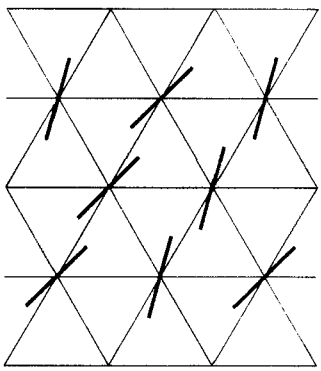

Figure 6. The herringbone (or chevron) structure for $n = 2$.

absent in the 4-sublattice phase because of (6):

$$(\psi^{(2)}(\mathbf{q}_A))^2 + (\psi^{(2)}(\mathbf{q}_{A'}))^2 + (\psi^{(2)}(\mathbf{q}_{A''}))^2 = 0.$$

It seems to us that the intermediate 4-sublattice phase has been missed in [7]. Let us stress that the preceding considerations could apply in a straightforward manner to the elliptic deformation of column-shaped aggregates of lyotropic molecules [3].

(3) We now discuss more precisely the case of tilted chevron phases in discotic columns; this is slightly more complex. The tilt is described by $\psi^{(1)}$ and reflection invariance ($z \to -z$) rules out odd terms in $\psi^{(1)}$. Thus the previous argument for the existence of the $\psi^{(2)}$ chevron structure cannot hold. However there certainly exists a coupling between $\psi^{(1)}$ and $\psi^{(2)}$ which at lowest order is

$$\beta \sum_i (\psi_i^{(1)})^2 (\psi_i^{(2)})^* + \text{c.c.} \tag{7}$$

whatever the lattice structure [15]. The different types of coupling between modes are

$$\beta\psi^{(1)}(\mathbf{q}_A)\psi^{(1)}(\mathbf{q}_A)\,(\psi^{(2)}(\mathbf{q} = 0))^*,$$
$$\beta\psi^{(1)}(\mathbf{q} = 0)\psi^{(1)}(\mathbf{q} = 0)\,(\psi^{(2)}(\mathbf{q} = 0))^*,$$
$$\beta\psi^{(1)}(\mathbf{q} = 0)\psi^{(1)}(\mathbf{q}_A)\,(\psi^{(2)}(\mathbf{q}_A))^*,$$
$$\beta\psi^{(1)}(\mathbf{q}_A)\psi^{(1)}(\mathbf{q}_{A'})\,(\psi^{(2)}(\mathbf{q}_{A''}))^*,$$

Classically this would lead to first order transitions if β is large enough (contrary to γ there is no reason to regard β as a perturbation because the coupling (7) does not rely on the existence of a lattice and is always present). Two types of structures are then expected:

(i) tilted herringbone (see figure (7)) with

$$\psi^{(1)}(\mathbf{q}_A) \neq 0,\ \psi^{(1)}(0) \neq 0,\ \psi^{(2)}(\mathbf{q}_A) \neq 0,\ \psi^{(2)}(0) \neq 0,$$
$$\psi^{(1)}(\mathbf{q}_{A'}) = \psi^{(1)}(\mathbf{q}_{A''}) = \psi^{(2)}(\mathbf{q}_{A'}) = \psi^{(2)}(\mathbf{q}_{A''}) = 0;$$

(ii) 4-sublattice tilt (see figure (8))

$$\psi^{(1)}(\mathbf{q}_A) = -\exp(-i\pi/3)\psi^{(1)}(\mathbf{q}_{A'}) = \exp(-2i\pi/3)\psi^{(1)}(\mathbf{q}_{A''}) \neq 0,$$
$$\psi^{(2)}(\mathbf{q}_A) = \exp(-2i\pi/3)\psi^{(2)}(\mathbf{q}_{A'}) = \exp(-4i\pi/3)\psi^{(2)}(\mathbf{q}_{A''}) \neq 0,$$
$$\psi^{(1)}(0) = \psi^{(2)}(0) = 0.$$

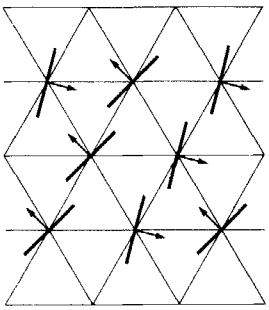

Figure 7. The tilted herringbone phase (e.g. discotics). The arrows picture $\psi^{(1)}$ and the rods $(\psi^{(2)})^{1/2}$. For the sake of clarity we have drawn the rods orthogonal to the arrows. That would correspond to a choice $\beta > 0$.

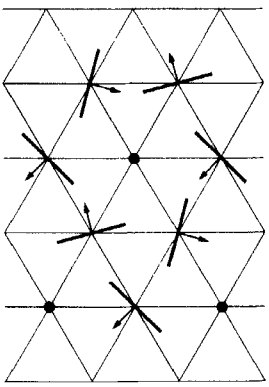

Figure 8. The tilted 4-sublattice phase.

The former is identified with $D_{\mathrm{rd}}(\mathrm{P2_1/a})$ and the latter with the P(321) phase of [9].

The transition

$$D_{\mathrm{rd}}(\mathrm{P2_1/a}) \rightarrow D_{\mathrm{hd}}$$
$$\rightarrow T \nearrow \rightarrow$$

is so accounted for. However in some compounds it is split as [10]

$$D_{\mathrm{rd}}(\mathrm{P2_1/a}) \rightarrow D_{\mathrm{rd}}(\mathrm{C2/m}) \rightarrow D_{\mathrm{hd}}$$
$$\rightarrow T \nearrow \rightarrow .$$

In our model this is interpreted as follows; when the temperature decreases a ferro order first sets in (see §4.1)):

$$\psi^{(1)}(q = 0) \neq 0, \quad \psi^{(2)}(q = 0) \neq 0.$$

This is possible if $\mu_1 + m_1^-(0)$ reaches zero well before $\mu_2 + m_2^-(q_A)$. In spite of the coupling (7) it has been checked that the possibility of a linear instability remains, in particular if $(D_0^{(2)}, D_4^{(2)})$ lies in region A. It leads to

$$\psi^{(2)}(\mathbf{q}_A) \neq 0, \quad \psi^{(1)}(\mathbf{q}_A) \neq 0$$

which describes a tilted herringbone structure. It is worth stressing that this ferro-herringbone transition can be second order in spite of the existence of the cubic term

(7): in the ferro phase the $\pi/3$ rotation invariance is lost and the 3 wavevectors $\mathbf{q}_A$, $\mathbf{q}_{A'}$, $\mathbf{q}_{A''}$, are no longer degenerate. Thus there are no star contributions such as

$$\psi^{(1)}(\mathbf{q}_A)\psi^{(1)}(\mathbf{q}_{A'})(\psi^{(2)}(\mathbf{q}_{A''}))^*$$

to force the transition to be first order. Experimentally it turns out that the corresponding transition is second order or almost second order contrary to the hexagonal → chevron transition which is clearly first order [10].

4.3. *The incommensurate phase (region I)*

So far there is no experimental evidence for such a phase. We hope this paper will stimulate experimental research in this direction.

4.4. *Antiferro XY model (region B)*

The $\mathbf{q}_B$ structure is obtained only for a negative ($C_0 < 0$) isotropic ($C_{-2n} = 0$) interaction. This is the triangular antiferro XY model, the solution of which is classical in the context of frustrated systems [11].

5. Conclusion

We have developed a model of interacting columns allowing us to go beyond a simple phenomenological approach. The underlying lattice structure enters the problem through the intercolumnar couplings. The ordered phases and the corresponding phase diagram can, in principle, be obtained by varying the parameters. Conversely the experimental results might shed light on the hierarchy of the coupling strengths. After slight modifications the same model can take into account the case of ternary molecules (i.e. $n = 3$ for the order parameter) and the helical order often associated with them. Finally let us stress that an original mechanism for the transition to the herringbone has been exhibited. Also the 4-sublattice structure is accounted for and an incommensurate phase predicted.

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