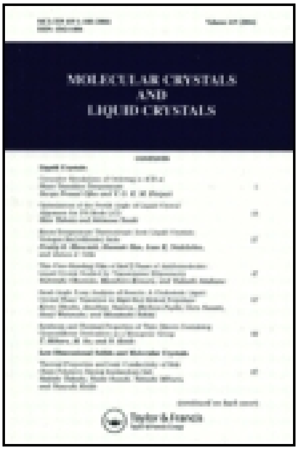

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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To cite this article: Harald Pleiner (1984): Broken Symmetries and Hydrodynamics of Hexatic B and Snectic F and I Liquid Crystal Phases, Molecular Crystals and Liquid Crystals, 114:1-3, 103-111

To link to this article: http://dx.doi.org/10.1080/00268948408071702

Mol. Cryst. Liq. Cryst.,, 1984, Vol. 114, pp. 103-111
0026-8941/84/1143-0103/$15.00/0

Printed in the United States of America

BROKEN SYMMETRIES AND HYDRODYNAMICS OF HEXATIC B AND SMECTIC F AND I LIQUID CRYSTAL PHASES

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Abstract The nature of the spontaneously broken symmetries in hexatic B and smectic F and I phases is investigated and the hydrodynamics of the associated degrees of freedom is discussed.

INTRODUCTION

In the past few years it has become clear[1-5] that there exists a class of smectic liquid crystals, which show no positional long-range order (liquid-like) within the layers, but which have a long-ranged orientational order of the bonds, i.e. the lines between neighboring molecules. This bond orientational order usually occurs in a hexagonal pattern from which the name hexatics is coined. There is one phase, hexatic B phase, which can be characterized as a smectic A phase with an additional hexagonal bond orientational order in the layers and two tilted phases (smectic F and I), which are smectic C like plus an additional hexagon-like bond orientational order in the layers. In the F phase and the I phase the $\hat{c}$-vector, which characterizes the tilt direction of the director, is parallel or perpendicular, respectively, to one of the directions of the bond orientational order.

It is the aim of this note to discuss the nature of the broken symmetries in these hexatic phases and to point out some of the major consequences for the hydrodynamics of these systems. For the full set of hydrodynamical equations, which have been derived in collaboration with H.R. Brand, I refer to Ref.6.

BROKEN SYMMETRIES AND HYDRODYNAMICAL VARIABLES

Hexatic B phase

Like in all smectic phases the existence of the layer structure breaks spontaneously the translational symmetry along the layer normal $\hat{p}$. However, it is well known[7-8] that such a one-dimensional order embedded in three-dimensional space cannot be truly long-ranged but only quasi-long-ranged. That means the correlation of the lamellar pattern is not constant in space but decreases with some power of the distance (algebraïc decay). If this decay is slow enough, this quasi-long-ranged order is pragmatically treated as a true long ranged one (having in view the finiteness of any experimental probe), i.e. the displacement u of the layers (along $\hat{p}$) is chosen as a hydrodynamic variable[9-11] (in some metastable situations, where the layers are not flat, a generalization of u, the phase ϕ , is used as variable[12-13]). This connection between spontaneously broken symmetries and hydrodynamic variables is well established[14-16,9] and can easily be understood: a homogeneous displacement of the layers (wave vector $\vec{k} = 0$) does not provoke any restoring force, i.e. an infinitely slow dynamics ($\omega = 0$), while an inhomogeneous displacement ($\vec{k} \neq 0$) results in a hydrodynamic motion with $\omega(\vec{k} \to 0) \to 0$; thus, u is a hydrodynamic variable.

The existence of the layered structure also breaks rotational invariance : rotations of the layer normal $\hat{p}$ lead to different states (with equal energy, however). Thus, rotational symmetry of $\hat{p}$ is spontaneously broken, while rotational symmetry about $\hat{p}$ is still unbroken. However, the appropriate hydrodynamical variable $\delta\hat{p}$ (with $\hat{p}.\delta\hat{p} = 0$) is not an independent hydrodynamic variable, but is given by gradients of u, i.e. in linear order by[10] $\delta\hat{p}_i = (\delta_{ij} - \hat{p}_i\hat{p}_j) \nabla_j u$. This relation is based on the very trivial fact, that the normal cannot be rotated without rotating the planes (otherwise the normal would not be any longer normal). Thus, the existence of layers breaks a combined translational-rotational symmetry. In hydrodynamics, where gradients serve as small expansion parameters, this combined broken symmetry shows up mainly as a broken translational symmetry, because rotations are always of higher order in gradients than translations.

Of course one can think of rotations of the director $\hat{n}$, the mean direction of the molecular long axes. However, in equilibrium $\hat{n}$ is parallel to $\hat{p}$ and any rotation of $\hat{n}$ (with fixed $\hat{p}$) would result in a restoring force, which does not vanish in the homogeneous limit. Therefore, the motion of $\hat{n}$ is not hydrodynamic but very rapid and does not show up in experiments below a certain frequency value[9].

So far the situation is just like smectic A phases. In hexatic B phases the additional bond orientational order in the planes breaks also the rotational symmetry about $\hat{p}$. This is a spontaneous symmetry breaking since there is no energy which fixes in equilibrium the direction of the hexagonal orientational pattern. Therefore, only inhomogeneous rotations give rise to restoring forces and the appropriate dynamics has hydrodynamic character. Rotation of this hexagonal bond structure about $\hat{p}$ is described by an antisymmetric rotation matrix, which contains one additional hydrodynamic degree of freedom $\delta\theta$, the rotation angle. As far as linear hydrodynamics is concerned the rotation axis can be treated as constant (i.e. as the equilibrium direction of the layer normal $\hat{p}$). In a non-linear theory the direction of the rotation is also a hydrodynamic variable. However, since the group of finite rotation in three-dimensional space is non-Abelian, there is a difference between first rotationg the axis $\hat{p}$ ($\delta\hat{p}$ with $\hat{p}.\delta\hat{p} = 0$) and then rotating the bond structure ($\delta\theta \neq 0$) and the opposite sequence. This non-commutativity can be expressed by the relations ($p_x = -\nabla_x U$, $p_y = -\nabla_y U$)

$$\begin{aligned} \delta_1\delta_2 p_x - \delta_2\delta_1 p_x &= (\delta_1 p_y)(\delta_2\theta) - (\delta_1\theta)(\delta_2 p_y) \\ \delta_1\delta_2 p_y - \delta_2\delta_1 p_y &= (\delta_1\theta)(\delta_2 p_x) - (\delta_1 p_x)(\delta_2\theta) \qquad (1) \\ \delta_1\delta_2\theta - \delta_2\delta_1\theta &= (\delta_1 p_x)(\delta_2 p_y) - (\delta_1 p_y)(\delta_2 p_x) \end{aligned}$$

where δ_1, δ_2 stands for any first order differential operator. Eqs(1) have their well-known counterparts in the superfluid phases of ^{3}He[17-19] and in biaxial nematics[20].

Smectic F and I phases

The smectic F and I phases differ from hexatic B phases by the fact that the director $\hat{n}$ is tilted away from the layer normal $\hat{p}$. This is expressed by the $\hat{c}$-vector (like in smectic C phases[10]), which is a unit vector, lying in the planes and giving the tilt direction of $\hat{n}$ ($\hat{c} \equiv [\hat{n} - \hat{p}\,(\hat{p}.\hat{n})]\,[1 - (\hat{n}.\hat{p})^2]^{-1/2}$). The existence of the $\hat{c}$-vector breaks the rotational symmetry about $\hat{p}$. In smectic C phases this gives rise to an additional hydrodynamic degree of freedom[9,10]. In smectic F and I phases, however, the things are a little bit more complicated, since rotational symmetry about $\hat{p}$ is already broken by the ordered bond structure. Thus, one can either rotate (about $\hat{p}$) the bonds, i.e. the centers of mass of neighboring molecules or one can rotate the $\hat{c}$-vector, i.e. the long molecular axis of the molecules. The former operation is described by an antisymmetric rotation matrix with independent element $\delta\theta$, the latter by symmetric quadrupole tensors with one independent element $(\hat{p} \times \hat{c}).\delta\hat{c}$ describing rotations of $\hat{c}$ within the planes (leaving the tilt angle constant). Although these two operations are physically quite distinct, they are not independent dynamically. In equilibrium $\hat{c}$ and the directions of the bond structure are locked; it is of no importance for the dynamics how they are locked. Any relative rotation of one structure against the other $(\delta\theta \neq (\hat{p}\times\hat{c}).\delta\hat{c})$ would result in a restoring force even in the homogeneous limit. Thus, only $\delta\theta + (\hat{p} \times \hat{c}).\delta\hat{c} \equiv 2\delta\partial$ describing combined rotations of director and bond structure is a hydrodynamic variable, while the relative rotation $\delta\theta - (\hat{p} \times \hat{c}).\delta c \equiv 2\delta\phi$ is not. The latter may be a comparably slow (non-hydrodynamic) microscopic variable, if the lock-in energy between bond and director structure is small. In that case $\delta\phi$ can play a role in macroscopic dynamics quite similar to tilt angle variations near the phase transition (to the hexatic B or smectic A phases).

HYDRODYNAMIC CONSEQUENCES

When compared to smectic A phases, the hexatic phase and the smectic F and I phase (and the smectic C phase) have just one more hydrodynamical degree of freedom. In the following I will sketch briefly, how this degree of freedom manifests itself and discrimates these phases from one another and from the smectic A or C phases. Of course, there are further differences to the A and C phases not associated with broken symmetries, but based on microscopic properties (e.g. different values of the hydrodynamic parameters like viscosities, etc.. ; greater correlation length of the in-plane short-range positional order resulting in a reduction of the validity region of hydrodynamics; biaxiality in smectic F and I), which I will not discuss further.

The additional degree of freedom in the hexatic phases is of rotational character, a rotation of the bond structure in hexatic B, $\delta\theta$, and a combined rotation of bond structure and director in smectic F and I, $\delta\partial$ (while a pure director rotation in smectic C, $(\hat{p} \times \hat{c}).\delta\vec{c}$). Since the bond structure probably couples only very weakly to external electric or magnetic fields, this additional degree of freedom in hexatic B is much harder to detect than the director fluctuations in nematics or smectics C. The influence of the former can only be studied indirectly, through its coupling to other hydrodynamic variables. Thus, the simplest way is to look for the normal mode structure. One finds[6] that $\delta\theta$ couples only to $\hat{p}.\vec{\omega}$, the vorticity perpendicular to the layers (provided that the wave vector $\vec{k}$ is not parallel to $\hat{p}$, in that case $\delta\theta$ is completely decoupled from the rest). This coupled motion (bond-vorticity mode) obeys the equations

$$\begin{aligned} &\delta\dot{\theta} - \hat{p}.\vec{\omega} = \mu h \\ &\rho_o\, \hat{p}.\dot{\vec{\omega}} - \frac{1}{4}\,\Delta_{tr}\, h = -\,\nu_{ij}\nabla_i\nabla_j\, \hat{p}.\vec{\omega} \end{aligned} \qquad (2)$$

where $h \equiv \nabla_i(\partial f/\partial\nabla_i\theta)$, $\Delta_{tr} \equiv (\delta_{ij} - \hat{p}_i\hat{p}_j)\nabla_i\nabla_j$

and $f = \frac{1}{2} M_\perp (\nabla_\perp\theta)^2 + \frac{1}{2} M_{\parallel} (\nabla_{\parallel}\theta)^2$

is the distortion free energy density of the bond structure. If $M_{\parallel}$ is zero (i.e. no correlation of the bond structure of different layers, one has to add $\tilde{K}(\nabla_{\parallel}^2\,\theta)^2$ to f and to replace $M_{\parallel}\,k_{\parallel}^2$ by $\tilde{K}\,k_{\parallel}^4$ in the following. The dispersion relation

$$\omega = \frac{i}{2}\,\alpha_+ \pm \frac{1}{2}\,(\beta^2 - \alpha_-^2)^{1/2} \qquad (3)$$

with

$$\alpha_\pm = \mu\,(M_\perp k_\perp^2 + M_\parallel k_\parallel^2) \pm \rho_o^{-1}\,(3\nu_2\,k_\perp^2 + \nu_3\,k_\parallel^2)$$

$$\beta^2 = \rho_o^{-1}\,k_\perp^2\,(M_\perp k_\perp^2 + M_\parallel k_\parallel^2)$$

has the same structure as for the director-shear mode in nematics[16] or the "orbit-wave" mode in ^{3}He-A[21]. It can be a propagating, though dispersive and strongly damped mode (for $\beta^2 > \alpha^2$) or a coupled diffusion ($\beta^2 < \alpha^2$). In the limit $k_\parallel = 0$, $\mu\nu_2 << 1$ propagation is obtained provided the transverse stiffness constant $M_\perp$ obeys[6]

$$\mu^{-2} < \rho_o\,M_\perp < \frac{3}{4}\,\nu_2^2 \qquad (4)$$

This mode can be excited by an inhomogeneous rotation ($(\mathrm{curl}\,\vec{v})_z \neq$ const.) about the layer normal (e.g. a fixed inner cylinder and an oscillating outer one). As already mentioned the bond structure does probably not orient in an external electric or magnetic field. Nor does it show a flow alignment effect, since it is not due to the orientation of unsymmetrical molecules but of the centers of mass of neighboring molecules. Thus, the usual techniques to investigate a nematic-like degree of freedom do not apply to $\delta\theta$. Furthermore, the bond structure is sixfold, so the isotropy within the layers (with respect to second rank tensors) is still preserved and there is no dramatic enhancement of light scattering intensity due to (orientational) fluctuations of $\delta\theta$. (The dependence of the magnitude of the dielectric tensor on $\delta\theta$ is probably negligible).

If, on the other hand, there could be found a smectic phase with i.e. rectangular bond orientational order, this phase would be biaxial. The additional preferred direction $\hat{c}(\perp \hat{p})$ would be orientable by external fields and there should be a flow alignment effect according to the dynamical equation

$$\dot{\theta} - \hat{p}.\vec{\omega} + \frac{1}{2}\lambda(\hat{p} \times \hat{c})_i\, \hat{c}_j(\nabla_j v_i + \nabla_i v_j) = \mu h \qquad (5)$$

Of course, there is an appropriate back flow effect (in the equation for $\dot{g}_i$) due to the non-vanishing λ.

The situation is slightly different in smectic F and I phases. There the degree of freedom $\delta\partial$ contains rotations of the director and this part of $\delta\partial$ is accessible to experiments like any nematic-like degree of freedom (e.g. flow alignment like in smectic C phases[22]). However, in $\delta\partial$, $\delta\vec{c}$ is combined with $\delta\theta$, and the rotations $\delta\vec{c}$ can only be seen, if $\delta\theta$ is able to follow them. Such experiments would give, therefore, new information on the strength of the coupling between bond structure and director structure or on the forces (due to walls, defects, etc..) by which the bond structure is hindered to rotate.

Acknowledgement

A grant from the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

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