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Unconventional states of liquid crystals: an analogy with unconventional superconductivity

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1. Introduction

The analogy with superconductivity has played an important role in the understanding of liquid crystals and in the prediction of new liquid-crystal states. The analogy was first pointed out by de Gennes [1] who noted that the nematic-smectic A scalar density-wave order-parameter is formally identical to the complex wave function in the Ginzburg-Landau theory of the normal-superconducting phase transition in metals [2]. Later, Renn and Lubensky [3] predicted on this basis the existence of the twist grain-boundary phase, as the liquid-crystal analog of the Abrikosov intermediate flux lattice phase of type II superconductors [4].

The preceding correspondences referred to conventional (BCS) superconductivity. Recently a further analogy was stressed [5] between the order-parameter symmetry of the two-dimensional d-pairing wave mechanism currently assumed for cuprate superconductors [6] and the vector density-wave order-parameter describing liquid crystal mesophases formed from achiral bent shaped molecules [7, 8]. This suggests the definition of a new class of unconventional liquid-crystal states characterized by the breaking of discrete symmetry operations of the parent state (inversion, reflections, two-fold rotations), in contrast to the conventional liquid-crystal states, which result from the breaking of a continuous (translational or rotational) symmetry.

2. Two-dimensional d-pairing wave superconductivity

The local symmetry of the normal state of a superconductor is given by $G_o = [U(1) \times T] \otimes G_R \otimes G_S$, where $U(1)$ and T are, respectively, the groups generated by the gauge (g_α) and time-reversal (T) operations. G_R and G_S are the groups formed by the symmetry operations of the system in real and spin spaces. Let us consider the situation, which is strongly suspected to be realized in oxide superconductors [9], of a two dimensional d-pairing-wave superconductor, i.e. a superconductor having a nonzero orbital momentum $L=2(S=0)$ in which the pair correlation length is much larger than

the thickness of the sample. Assuming the normal state to be structurally isotropic [$G_R \equiv G_S \equiv 0(2)$] the symmetry of the pairing-wave function of the condensate can be defined in the homogeneous case as:

$$\Psi_D(\vec{k}) = \eta_+ Y_2^+{}^2(\vec{k}) + \eta_- Y_2^-{}^2(\vec{k})$$

where \vec{k} is the normalized internal wave-vector of pairing particles. The functions $Y_2^\pm{}^2$ transform as the spherical harmonics of index 2. η_+ and η_- are the coefficients of the decomposition, which form with their complex conjugates η_+^* and η_-^* the four-component order-parameter associated with the normal-to-d-pairing superconducting transition. The symmetry of this order-parameter allows one to construct two independent invariants [10]: $\mathcal{F}_1 = \eta_+ \eta_+^* + \eta_- \eta_-^* = n$ and $\mathcal{F}_2 = \eta_+ \eta_+^* \eta_- \eta_-^* = (n^2 - L_z^2)/4$, where n has the meaning of the (scalar) condensate density of pairs, and L_z is a pseudoscalar representing the two-dimensional orbital momentum. Minimization of the corresponding Landau free-energy,

$$F(\mathcal{F}_1, \mathcal{F}_2) = a_1 \mathcal{F}_1 + a_2 \mathcal{F}_1^2 + \dots + b_1 \mathcal{F}_2 + b_2 \mathcal{F}_2^2 + \dots + c_{12} \mathcal{F}_1 \mathcal{F}_2 + \dots$$

yields three stable superconducting states, denoted I, II and III, whose equilibrium properties are summarized in table 1. The most remarkable features of these states are that they display either spontaneous orbital paramagnetism, or a spontaneous ferromagnetic like order of the orbital moments which results from a breaking of the time-reversal symmetry of the normal state. Thus, State I has the continuous polar symmetry ∞m and can be figured as formed by opposed magnetic domains with $L_z = |\eta_+|^2$ and $L_z = -|\eta_-|^2$. In State II the continuous symmetry is lowered to 4mm, which has the intrinsic tetragonality of d-pairs, and one has an orbital paramagnetic order for the infinite set of tetragonal domains transforming one another by the lost continuous rotations. The properties of State III intersect the properties of States I and II, i.e. it is structurally anisotropic and magnetically ordered. The location of the three states in the (a_1, b_1) plane is shown in figure 1.

Table 1. Symmetries and equilibrium properties of the superconducting states I, II, III and their liquid crystal analogs. The columns have the following meaning: (a) list of the states in reference to the phase diagram of figure 1; column (b): equilibrium relationship between the order-parameter components; column (c): structural symmetries for the superconducting (upper line) and liquid crystal (lower line) states; column (d): symmetry operations which are lost at the transition from the normal state (upper line) or parent liquid crystal state (lower line).

(a)	(b)	(c)	(d)
I	$\left\{ \begin{array}{l} \eta_+ \neq 0, \quad \eta_- = 0 \\ \text{or} \\ \eta_- \neq 0, \quad \eta_+ = 0 \end{array} \right\}$	$\left\{ \begin{array}{l} \infty m \\ \infty 22 \end{array} \right\}$	$\left\{ \begin{array}{l} g_x, T \\ R_z, I, m \end{array} \right\}$
II	$ \eta_+ = \eta_- \neq 0$	$\left\{ \begin{array}{l} 4mm \\ mma \end{array} \right\}$	$\left\{ \begin{array}{l} g_x, c_\varphi \\ R_z, c_\varphi \end{array} \right\}$
III	$ \eta_+ \neq 0, \eta_- \neq 0$	$\left\{ \begin{array}{l} 4mm \\ 2_1 22 \end{array} \right\}$	$\left\{ \begin{array}{l} g_x, c_\varphi, T \\ R_z, c_\varphi, I, m \end{array} \right\}$

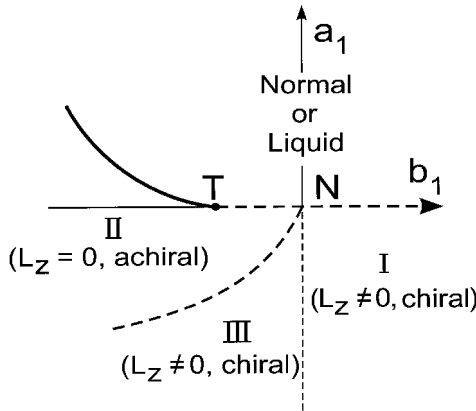


Figure 1. Theoretical phase diagram of a two-dimensional d-pairing wave superconductor or liquid crystal formed from bent-shaped molecules, for an isotropic parent state. The phase diagram is in the (a_1, b_1) plane of the coefficients in the Landau free-energy $F(\mathcal{F}_1, \mathcal{F}_2)$. Dashed and solid lines represent, respectively, second and first-order transition lines. T is a tricritical point, and N a four phase point.

3. Correspondence with liquid crystal mesophases formed from bent core molecules

There exists a one-to-one correspondence between the preceding d-pairing wave model of superconductors and the theoretical approach describing the liquid crystal mesophases which may arise below an isotropic liquid formed with achiral bent core molecules [7, 11]. The formation of the ordered mesophases can be depicted as driven by the condensation of a transverse polar vector

wave:

$$\vec{P}(z) = P_x \cos(kz + \varphi_x) \vec{e}_x + P_y \cos(kz + \varphi_y) \vec{e}_y,$$

corresponding to the local average of the permanent dipolar moments attached to the molecules. P_x and P_y are the components of the transverse wave amplitude. φ_x and φ_y are the initial phases. \hat{k} is the wave-vector chosen along the z axis, and (\vec{e}_x, \vec{e}_y) are the unit vectors in the (x, y) plane. $\vec{P}(z)$ plays the role of the wave function $\Psi_D(\vec{k})$, and the four components of the corresponding order-parameter are defined by the complex wave amplitudes: $\eta_+ = P_x e^{i\varphi_x} - iP_y e^{i\varphi_y}$, $\eta_- = P_x e^{-i\varphi_x} - iP_y e^{-i\varphi_y}$ and their complex conjugates η_+^* , η_-^* . They give rise to the same independent invariants \mathcal{F}_1 and \mathcal{F}_2 defined above, and to the same free-energy $F(\mathcal{F}_1, \mathcal{F}_2)$. Therefore one finds three stable mesophases which are the liquid crystal analogs of the d-pairing-wave states I, II, and III [7, 11]. Mesophase I corresponds to the chiral molecular configuration, shown in figure 2(a), represented by a circularly polarized wave in which the continuous rotations (c_φ), continuous translations along z (R_z), and inversion centre (I) are lost. Mesophase II is a biaxial achiral smectic phase corresponding to a linearly polarized wave forming a bilayer antiferroelectric structure (figure 2(b)). Mesophase III is associated with an elliptically polarized transverse vector wave, having a biaxial chiral symmetry and a ferroelectric order. Table 1 indicates the equilibrium properties of the three mesophases, which figure in the phase diagram of figure 1 together with their superconducting analogs.

4. Unconventional liquid crystal states

Phase transitions between *conventional* liquid-crystal mesophases are characterized by the breaking of a continuous translational or continuous rotational symmetry of the parent phase. This is the case, for example, for the isotropic-nematic, nematic-smectic A and Smectic A–Smectic C transition. In our considered examples, this traditional scheme applies only to the transition from the isotropic liquid to state II in which one has exclusively a breaking of the continuous translational (R_z) and rotational (c_φ) symmetries. In the analogous standard superconducting state II one finds a breaking of the continuous gauge (g_x) and continuous rotational (c_φ) symmetries of the normal state. More generally, one can verify from table 1 and figure 1 that there is an obvious correspondence between: (i) the group of continuous translations (R_z) which leave invariant the parent liquid crystal state and the continuous gauge symmetry of the normal state of superconductors, and (ii) the continuous rotational symmetry (c_φ) of the parent liquid crystal state, and the continuous ‘external’ rotations (c_φ) of the symmetry group G_R of the normal state.

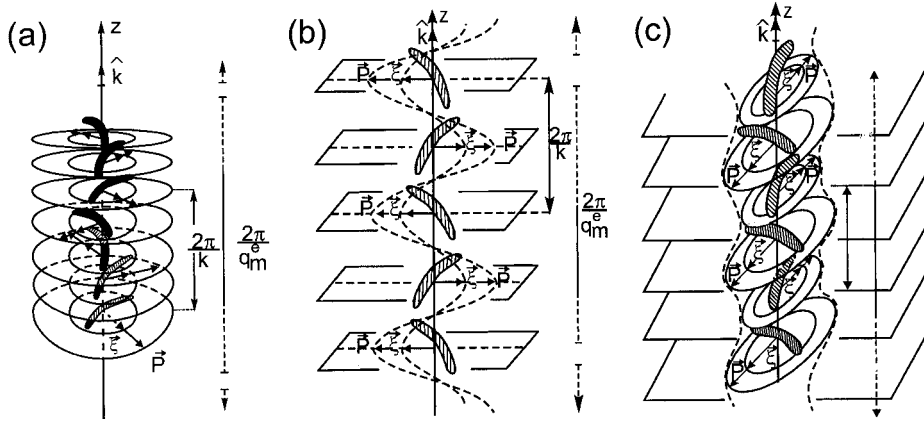


Figure 2. Structure of the molecular configurations I, II and III. (a) Circularly polarized configuration, which gives rise to a structure of chiral symmetry D_∞ . (b) Linearly polarized configuration II, which yields an antiferroelectric bilayer smectic structure for the mesophase. Each bilayer has the achiral symmetry mma . (c) Elliptically polarized ferrielectric bilayer configuration III with chiral symmetry $P2_122$.

In contrast, the transitions to the liquid crystal states I and III involve, in addition to the breaking of continuous symmetries (R_z or c_φ), the breaking of *discrete* symmetry operations (inversion, reflection, two-fold rotations) of the parent state. The *unconventional* states I and III are *chiral*, i.e. the achirality of the initial isotropic liquid is simultaneously broken. In the analogous unconventional superconducting states I and III the *discrete* time-reversal symmetry (T) is broken allowing the onset of a spontaneous orbital momentum L_z .

Summarizing, unconventional states of liquid crystals and superconductors are characterized in a unified way by a breaking of discrete symmetries of the respective parent states. The symmetry operation inverting the space coordinate z (inversion, reflection, two-fold rotations) in liquid crystals, coincide in superconductors with the time-reversal symmetry (T). Besides the onset of chirality in unconventional liquid crystal states corresponds, in unconventional superconducting states, to a spontaneous orbital magnetic order. Before analysing these correspondences at a different level, let us show that they are not specific to d-pairing superconductors, and liquid crystals formed from achiral polar molecules.

5. Unconventional $S+S'$ superconducting state and the SmC* state with longitudinal polarization

There exists also a one-to-one analogy between the $S+S'$ unconventional pairing wave model of superconductors [12] and the description proposed in [13] for the transition from a chiral smectic A^* phase, formed with stick-like molecules, to polar smectic phases. The $S+S'$ model assumes the coupling of two *dephased* s-pairing wave order-parameters ($L=0$, $S=0$) associated with the complex wave functions $\Psi_1 = |\Psi_1|e^{i\theta_1}$ and $\Psi_2 = |\Psi_2|e^{i\theta_2}$. From the transformation properties of Ψ_1 and Ψ_2 one finds for any continuous or discrete symmetries

of $G_R \otimes G_S$, the following invariant monomials: $\mathcal{F}_1 = \Psi_1^2$, $\mathcal{F}_2 = \Psi_2^2$ and $\mathcal{F}_3 = |\Psi_1||\Psi_2| \cos(\theta_1 - \theta_2)$. Accordingly, the corresponding Landau expansion has the form:

$$F(|\Psi_i|, \theta_i) = a_1 \mathcal{F}_1 + a_2 \mathcal{F}_1^2 + \dots + b_1 \mathcal{F}_2 + b_2 \mathcal{F}_2^2 + \dots \\ + c_1 \mathcal{F}_3 + c_3 \mathcal{F}_3^2 + \dots + d_{12} \mathcal{F}_1 \mathcal{F}_2 + \dots$$

Minimization of F with respect to the $|\Psi_i|$ and θ_i shows that, in addition to the normal state ($|\Psi_1| = |\Psi_2| = 0$) two distinct superconducting states can be stabilized [12]:

- (1) The state denoted I in figure 3(a) for $|\Psi_1| \neq 0$, $|\Psi_2| \neq 0$ and $\theta_1 - \theta_2 = 0, \pi$. This is the conventional (Ginzburg-Landau) superconducting s-pairing wave state, in which only the gauge symmetry of the normal state is lost. figure 3(a) shows that state I is separated from the normal state by a line of second-order transitions.
- (2) A superconducting state II which is stabilized for $|\Psi_1| \neq 0$, $|\Psi_2| \neq 0$ and $(\theta_1 - \theta_2) \neq 0, \pi$. In this unconventional state the discrete time-reversal symmetry is broken giving rise to a spontaneous polarization induced by a second-order magnetoelectric effect:

$$\vec{P} = \eta(\vec{E} \times \vec{H})$$

where \vec{E} and \vec{H} are non-colinear electric and magnetic field, and $\eta = |\Psi_1||\Psi_2| \sin(\theta_2 - \theta_1)$ is a third-rank magnetoelectric tensor.

State II can be reached from the normal state across the region of stability of state I (figure 3(a)). Different microscopic mechanisms have been suggested [12] for the realization of dephased s-pairing waves, as for example the coupling between pairs of electron and pairs of holes in a system with disconnected Fermi surfaces in the normal state.

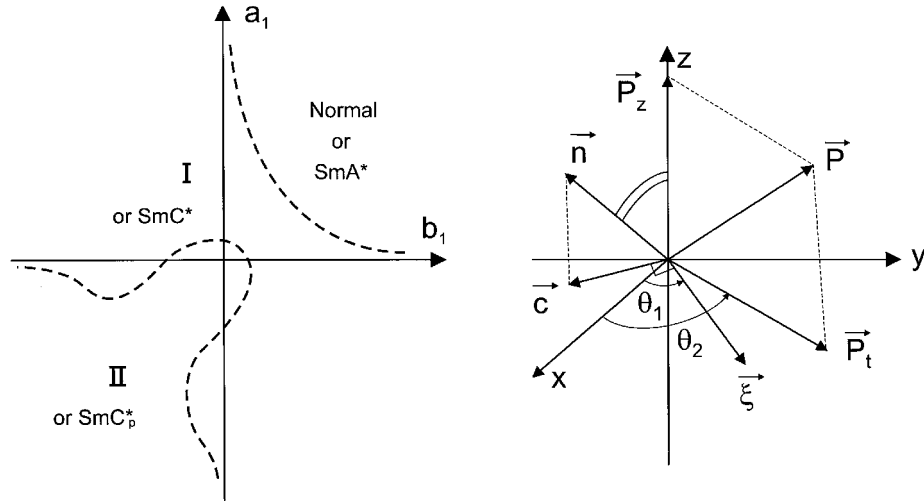


Figure 3. (a) Phase diagram in the (a_1, b_1) plane corresponding to the Landau expansion $F(\mathcal{F}_1, \mathcal{F}_2, \mathcal{F}_3)$, truncated at the fourth degree. The dashed lines represent second-order transition lines. (b) Local configuration of the SmC_P^* phase.

The liquid crystal analog of the superconducting state II correspond to a polar smectic phase (denoted SmC_P^* in [13]) which exhibits a longitudinal component of the polarization (P_z) resulting from the coupling of non-orthogonal transverse polarization \vec{P}_t and tilt vector $\vec{\xi}$, which play the role of the complex wave functions Ψ_1 and Ψ_2 . The dephasing $\theta_1 - \theta_2 \neq 0, \pi$ between \vec{P}_t and $\vec{\xi}$, shown in figure 3(b), induces a longitudinal polarization component:

$$P_z \vec{e}_z = \lambda (\vec{P}_t \times \vec{\xi})$$

where λ is a third-rank piezoelectric tensor, i.e. the longitudinal component P_z results from a second-order piezoelectric effect which is the analog of the second-order magnetoelectric effect depicted here above. Figure 3(a) shows the location of the standard SmC^* ferroelectric phase (with colinear \vec{P}_t and $\vec{\xi}$ vectors) and of the SmC_P^* phase, which displays a triclinic C_1 symmetry, involving a breaking of the two-fold rotations of the parent (D_∞) SmA^* phase. Note that three distinct orientations of the total polarization $\vec{P} = \vec{P}_t + P_z \vec{e}_z$ may occur, which are, respectively, orthogonal to the director \hat{n} (preserving the $\hat{n} \rightarrow -\hat{n}$ symmetry), orthogonal to the layer normal \hat{k} (breaking the $\hat{n} \rightarrow -\hat{n}$ symmetry), or general (breaking both the $\hat{n} \rightarrow -\hat{n}$ and $\hat{k} \rightarrow -\hat{k}$ symmetries) as represented in figure 3(b). Note also that the existence of a longitudinal polarization component has been also predicted in antiferroelectric and ferroelectric mesophases [14] and was recently suggested in liquid crystals formed from bent shaped molecules [15].

6. Unconventional effects in liquid crystals

The preceding example confirms the correspondence between the time-reversal symmetry in superconductors and the discrete symmetry operation (two-fold rotation)

which reverses the space variable corresponding to the layer normal (or to the director's orientation) in liquid crystals. The time-reversal operation transform the superconducting wave function $\Psi(t) = |\Psi| e^{-i(t/\hbar) E_F t}$ where E_F is conventionally chosen as the Fermi energy, into its complex conjugate, in the same way that the helical polarization $\vec{P} = \vec{P}_0 e^{ikz}$ of a SmC^* phase is transformed into its complex conjugate by the two-fold rotations normal to the z -axis. On the other hand shifts, Δt and Δz of the time and space variables multiply $\Psi(t)$ and $\vec{P}(z)$ by phase factors ($e^{-i(t/\hbar) E_F \Delta t}$ and $e^{ik \Delta z}$, respectively) which can be figured as rotations in the complex plane of the wave function, and in the (x, y) plane. Accordingly, $\Psi(t)$ and $\vec{P}(z)$ possess the same symmetry groups, which is D_∞^t for superconductors (combinations of temporal translations with gauge transformations) and D_∞^z for liquid crystals (combination of translations along z with continuous rotations in the plane perpendicular to z).

The equivalence between the time variable in superconductors and a space variable in liquid crystals allows prediction of remarkable effects in liquid crystals. For example, a Josephson-like effect was recently described theoretically and verified experimentally at the 'junction' between two chiral mesophases (B2 and B7) stabilized in two distinct liquid crystals formed from bent shaped molecules [16]. It consists in the spontaneous formation of an incommensurate modulated structure in the contact region between two-helical mesophases displaying distinct helical structures. This is the analog of the Josephson alternative current which flows across the junction between two different superconductors and results from a difference of electric potential ΔV between the junction walls. In each superconductor there is a precession in time of the phase of $\Psi_S(t)$, which can

be viewed as a helical structure arising in the three-dimensional space formed by the complex plane of the superconducting wave-function Ψ_S and the time axis. ΔV induces a difference in the precession times on the sides of the junction which ultimately give rise to the alternative current. This effect related to the temporal behaviour of superconductors while the effects previously predicted in liquid crystals (Meissner-type effect [1] and TGB-Vortex state [3]) used the analogy with the spatial behaviour of superconductors.

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