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Molecular Crystals and Liquid Crystals

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Liquid Crystals as Inspirations for Fundamental Physics

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Liquid crystals fill the symmetry gap between the highest-symmetry homogenous and isotropic fluid phase and the lowest-symmetry crystalline lattices. As such they are ideal materials to probe fundamental concepts relating symmetry and conservation laws to generalized elastic distortions, broken-symmetry dynamics, and topological defects. This talk reviews how nematic and smectic phases illuminated these concepts.

Keywords Elasticity; hydrodynamics; symmetry of liquid crystal phases; topological defects

1. Introduction

Most of us learned in junior-high or high-school science classes that there are three states of matter: solids, liquids and gases distinguished by their flow and mechanical properties. Solids define their own boundaries and maintain their shape under the forces of gravity – they support shear; liquids need a container (or at least external forces) to define their boundaries under gravity, but they need not fill it; gases fill any container they occupy. This broad mechanical classification of the material world does in fact correspond to our everyday experience, but, as we now know, it is incomplete and naïve. A more sophisticated classification is based on symmetries. Liquids and gases are macroscopically homogeneous (all positions are equivalent) and isotropic (all directions are equivalent), and they are invariant under all of the operations – arbitrary translations and rotations – of the Euclidean group. This classification makes liquids and gases equivalent as they in fact are. The two phases can really only be distinguished when they coexist, and an interface or meniscus separates them. The liquid does not fill its container because it coexists with a gas above it. Solids come in many forms. Glasses, like liquids and gases, are macroscopically homogeneous and isotropic. They, however, are not equilibrium phases of matter. Equilibrium solids are crystals of some kind with a periodically repeated space-filling pattern of identical unit cells (ignoring the special case of quasicrystals in which more than one type of cell form quasi-periodic patterns), which in the

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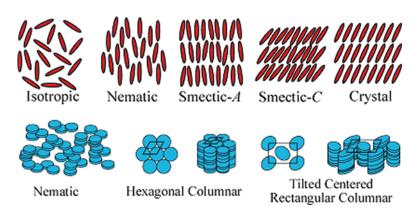


Figure 1. Top (bottom) schematic representation of liquid-crystalline phases of rod-like (disc-like) particles in order of decreasing symmetry. (Figure appears in color online.)

simplest cases contain one or only a few atoms but which can contain extremely large numbers. Crystals are neither homogeneous nor isotropic. A crystal translated along a symmetry direction by a distance equal to the appropriate unit cell length a is indistinguishable from the un-translated crystal. This translation, like an arbitrary translation in a liquid, is a symmetry operation of the crystal. Similarly, there is a discrete set of rotations and reflections that transform the crystal to itself and constitute the point group of the crystal. The combined set of all translations, rotations, and reflections that transform the crystal into itself constitute its space group.

The Euclidian group of homogeneous and isotropic fluids and the space groups of three-dimensional crystalline solids do not come close to exhausting the possible symmetry groups that equilibrium matter can exhibit. Liquid crystals (LCs), as their name suggests, are materials with properties that are both liquid-like and crystal-like and that have symmetries that fill the gap between that of the highest-symmetry fluid phases and the lowest symmetry crystalline phases [1]. Liquid crystals are composed of particles that exhibit some degree of anisotropy and that are generally complicated molecules or micron-scale colloidal particles (including viruses) that for many purposes can be modeled as rods or ellipsoids or as discs. Both rods and discs form uniaxial nematic phases in which mesogens align on average along a single direction, specified by a unit director **n**. Rods form smectic phases that consist (Fig. 1) of periodically repeated two-dimensional fluid-like layers; they are solid-like in one direction and fluid-like in the other two. Discs, on the other hand, generally form columnar phases consisting of a two-dimensional crystalline pattern of one-dimensional fluids (Fig. 1).

Many LC mesgens are chiral: their mirror images are not equivalent. Anisotropy axes of neighboring chiral mesogens tend to twist relative to each other, and the rate of change of the relative twist angle provide a new lengths scale in the problem that can be many times a molecular length. The combined effect of chirality and long-length scale leads to exotic phases that Bragg scatter light, including the cholesteric phase with a simple helical twist along one direction and the blue phase consisting of a regular array of double twist tubes that form a true three-dimensional crystal with a unit cell of 100,000 or more molecules. Combining chirality with the layered structure of smectics leads to a really astonishing variety of phases with different symmetries.

2. The Grand Synthesis

Over the 40 year or so, what I call a "grand synthesis" has emerged across essentially all fields of physics, particularly condensed-matter [2] and particle physics. This synthesis in the condensed matter context posits the following:

- 1. Equilibrium states of matter are characterized by their symmetry and their conservations laws. Each conservation law gives rise to one hydrodynamical mode with frequency ω that vanish with the wavenumber q.
- 2. Broken continuous symmetries give rise to excitations that tend to restore the system to its higher-symmetry states:
 - a. Elastic distortions with energies that vanish with wavenumber q of the distortions, usually but not always as q^2 ;
 - b. Goldstone hydrodynamics characterized by one dynamical modes whose frequency ω vanish as q tends to zero for each dimension of the continuous ground state manifold [3];
 - c. Topolgical defects;
- 3. Fluctuations, which become more important as spatial dimensionality d is decreased, can modify the above picture by destroying long-range order below a critical dimension d_L and by modifying the wavenumber dependence of excitations.

Liquid crystals with their wide range of symmetries have provided and continue to provide ideal materials to develop and test our understanding of the grand synthesis. The remainder to this article will look at some example the properties of liquid-crystalline phases can be extracted from the principles articulated in the grand synthesis.

3. Nematic Liquid Crystals

As shown in Figure 1, nematic liquid crystals are uniaxial, spatially homogeneous fluids. They are non-polar and thus invariant under inversion of the local director \mathbf{n} , specifying the anisotropy direction. In equilibrium, \mathbf{n} is spatially uniform. Rigid rotations of \mathbf{n} lead to different nematic states with the same free energy. The set of distinct nematic states with the equilibrium free energy is called the ground-state manifold or the order parameter space. In nematics this space, often denoted S^2/Z_2 or, equivalently as the projective plane RP^2 , is the unit sphere with opposite points identified as shown in Figure 2 [4].

Spatially uniform rotations of **n** in RP² do not change energy, but spatially non-uniform distortions of **n** do. Since distortions with wave vector **q** approach spatial uniformity as $q = |\mathbf{q}|$ tends to zero, the energy ε of these distortions will approach zero with q. The normal expectation is that ε will be analytic in q and, thus, that $\varepsilon \sim Kq^2$, where K is an elastic constant. Because the director and spatial position transform under the same rotation group, there are three distinct bulk elastic constants, the familiar splay (K_1) , twist (K_2) , and bend (K_3) constants of the Frank elastic energy [1].

The hydrodynamic limit is characterized by wavenumbers q and frequencies ω that are, respectively, much smaller than inverse mean free paths (λ^{-1}) and collision time (τ^{-1}) , i.e., by $q\lambda \ll 1$ and by $\omega \tau \ll 1$. The only variables that satisfy these hydrodynamic constraints in an isotropic fluid are the density of the conserved variables mass, energy, and momentm. These five conservations laws give rise to the Navier Stokes equations augmented by an energy diffusion equation that in turn have five hydrodynamical modes with frequency that vanish with wavenumber: an

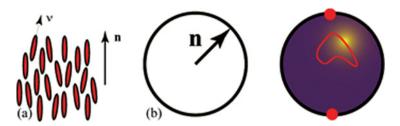


Figure 2. (a) Schematic representation of the nematic phase of rod-like molecules showing the unit vector ν specifying the direction of a particular molecule and the average direction n of all molecules. (b) The nematic order-parameter space showing first (right) the unit sphere S^2 and the unit sphere with opposite points identified (RP²). The closed path shows a possible path around a line not harboring a disclinations. (Figure appears in color online.)

energy (or temperature) diffusion mode with $\omega \sim -iq^2$, two transverse momentum diffusion modes with $\omega = -i (\eta/\rho)q^2$ (where η is the shear viscosity and ρ is the mass density), and two longitudinal sound modes with $\omega = \pm cq$.

The nematic director has two independent degrees of freedom, each of which contributes a hydrodynamical mode whose frequency vanishes with q. For the parameters of real nematics, these are both diffusive modes with $\omega \sim -i (K/\eta)q^2$ [5,6].

Topological defects [4] are distortions of a system that cannot be removed without some kind of surgery that usually involves forcing the system out of its equilibrium state. In three dimensions, a system can in principle exhibit point and line defects, and a nematic exhibits both [7]. The line defect or disclination is characterized by a core around which the director describes a path between equivalent points in the order parameter space, i.e., between polar opposite points as shown in Figure 3(a). This figure shows what appear to be two distinct disclinations with charge $\pm 1/2$ because the director undergoes a rotation of $\pm 180^{\circ}$ in one circuit around the core. These two configurations are, however, topologically equivalent because one can be continuously deformed into the other by rotating the path between polar opposites from east to west.

Disclination lines can form closed loops. Figure 3(b) shows three such loops, the first of which keeps the director far from the loop parallel to the horizontal axis. The other two loops, however, create director configurations that in the far field visit

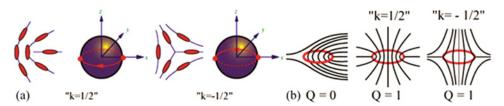


Figure 3. (a) Disclination configurations in the nematic and their associated paths in the order parameter space. In two-dimensions the configuration on the right and on the left would correspond to two distinct disclinations of respective charge +1/2 and -1/2. In three dimensions, the two are topologically equivalent because a continuous path converts one into the other. This path is represented in the order parameter space by rotating the path along a longitude connecting the poles from the front to the top to the back of the sphere. (Figure appears in color online.)

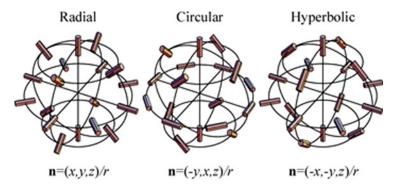


Figure 4. Three configurations for a+1 hedgehog. In the first, the headless director adopts the same configuration as the electric field around a point charge. The second and third are obtained from the first by rotating all directors about the z-axis through 90° and 180°, respectively. (Figure appears in color online.)

every point on the unit sphere S^2 . They are equivalent to point defects called hedge-hogs characterized by the number of times the director on a surface enclosing them wraps S^2 . Figure 4 shows three topologically equivalent hedgehog configurations.

A spherical particle whose surface is treated to favor alignment of a nematic director perpendicular to it surface (homeotropic boundary conditions) nucleates a radial-hedgehog configuration. If boundary conditions at infinity require the director to be parallel to a fixed axis (say the z-axis), then system must create configurations that cancel the hedgehog charge created by the spherical particle [8,9]. This can be done by drawing a hedgehog or a Saturn ring out of the sample as shown in Figure 5. The first two configurations break up-down symmetry and create a "topological dipole" whereas the third, "Saturn-ring" configuration does not and creates instead a topological quadrupole.

Figure 3(b) suggests that it might be possible to assign something like a hedgehog charge density to a disclinations loop by enclosing it in a torus and studying the

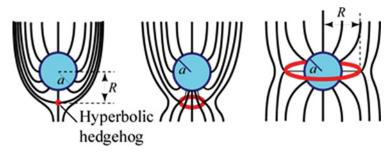


Figure 5. Schematic representation of the director field arising from a sphere with homeotropic boundary conditions embedded in a nematic in which the director is constrained to be along the vertical axis at infinity. On the left the hedgehog charge induced by the sphere is compensated by a hyperbolic point hedgehog drawn out of the nematic. In the middle and right, the sphere charge is balance by disclinations rings. The left and middle configurations break reflection symmetry and give rise to a topological dipole whereas the right-most configuration does not break this symmetry and gives rise to a topological quadrupole. (Figure appears in color online.)

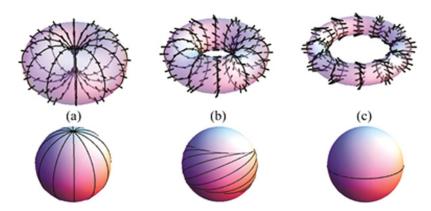


Figure 6. (a) Top – director on a torus and bottom associated paths in the order parameter space. The director on the torus covers S^2 and carries a recognizable hedgehog charge. (b) and (c) – continuous distortions on smaller tori of the configuration in (a) that cover a smaller and smaller fraction of S^2 with (c) covering only the equator – a set of measure zero. (Figure appears in color online.)

mapping of the director on that torus to the order parameter space. In the configuration shown in Figure 3(b), the director describes a path from the north pole to the south pole along a different longitude on each cross section of the helix and thus covers the entire sphere S²as shown in Figure 6(a). Figures 6 (b) and 6 (c) show, however, that a continuous distortion of the director can produce a configuration in which the director is confined to the equatorial plane. This raises the question of what general properties of mappings from a torus enclosing a disclinations loop to RP². This question was answered in mathematical terms by Janich [10], who found that there are four and only four topologically distinct configurations on such a torus, one of which is the chargeless configuration shown in Figure 3(b), one of which carries a hedgehog charge, and two of which correspond to loops penetrated by a disclination line. Reference [11] provides more details along with actual depictions of these configurations.

4. Liquid Crystal Elastomers

Liquid crystal elastomers are crosslinked liquid crystal polymers that combine the mechanical properties of rubber with the anisotropic properties of liquid crystals [12]. They exhibit essentially all of the thermodynamic phases as their uncrosslinked cousins. In an ideal limit, they undergo a symmetry-lowering transition from an isotropic rubber phase to a nematic phase in which the director points in an arbitrary direction. As we have argued, one expects the emergence of new low-energy elastic distortions and dynamic modes in the nematic phase, but what are they? Unlike the isotropic fluid, the isotropic rubber supports shear, so it already has the phonon modes of a solid with energies vanishing as q^2 . How can this energy be lowered by symmetry breaking? The answer lies in the vanishing of one of the five elastic constants of a uniaxial solid [13]. The nematic elastomer is uniaxial, and naively, one would expect it to be characterized by five independent elastic constant, but Ward identities [14,15] following from the broken symmetry show that the elastic modulus C_5 measuring the energy associated with strains u_{xz} in planes containing the

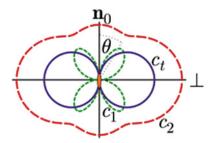


Figure 7. Sound velocities as a function of angle θ of propagation relative to the anisotropy axis of the three acoustic modes of a nematic elastomer. (Figure appears in color online.)

anisotropy axis along z vanishes identically. This leads to a modification of the dispersion of phonon modes. The two longitudinal and four transverse sound modes of an isotropic rubber with isotropic velocities develop anisotropic sound velocities, one of which vanishes for \mathbf{q} either parallel or perpendicular to the anisotropy axis and one of which vanishes only for \mathbf{q} parallel to that axis as shown in Figure 7. Interestingly, as is the case for smectic liquid crystals [16], nonlinearities along with thermal fluctuations render the naïve elastic moduli singular [17] in the limit of q approaching zero with $C_5 \sim (\ln \mathbf{q}_p)^{-4/59}$ and the Frank elastic constant $K \sim (\ln \mathbf{q}_p)^{38/59}$, where q_p is the magnitude of the component of \mathbf{q} perpendicular to the anisotropy axis.

The ideal nematic elastomer phase discussed above is never realized in real physical system because microscopic, random inhomogenities, prevent the formation of a monodomain nematic. Samples crosslinked once in the isotroic phase and then crosslinked a second time under unixial stress do form essentially monodomain samples [18], but there is no longer a spontaneously broken symmetry, and these samples are simply uniaxial solids with five elastict constants and small strains.

5. Smectic Liquid Crystals

Smectic liquid crystals are one-dimensional solids [1]. Translations of the structure a distance u normal to the layers leaves the energy invariant and translations by a distance equal to an integral multiple of the layer spacing d leave the configuration unchanged. Thus the order parameter space is an infinite line with all points separated by distances nd, where n is an integer, identified. There is, therefore, an elastic energy that depends on the single displacement variable u. The arguments used to derive the nematic Frank elastic energy would lead one to expect an energy proportional to q^2 , but because the smectic phase exhibits both broken translational and rotationally symmetry, this is not the case. A uniform growth of u in a direction parallel to the layers, say $u = \theta x$, where the layer normal is along the z direction, merely rotates the smectic and does not change its energy. The result is that the elastic energy takes the Landau-Peierls form [13]

$$F_{\rm sm} = \frac{1}{2} \int d^3x \left[B u_{zz}^2 + K_1 \left(\nabla_{\perp}^2 u \right)^2 \right],$$

where $u_{zz} = \partial_z u - \frac{1}{2} (\nabla u)^2$ and $\nabla_{\perp}^2 = \partial_x^2 + \partial_y^2$.

The single additional hydrodynamic variable u leads to one additional hydrodynamical variable beyond those of an isotropic fluid [3]. The associated mode is purely

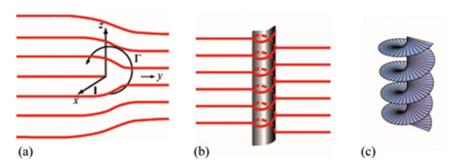


Figure 8. Schematic representation (a) of and edge dislocation and (b) a screw dislocation in a smectic. (c) A screw dislocation represented as a helicoidal surface. (Figure appears in color online.)

diffusive with $\omega \sim -iq^2$ for $\mathbf{q} = (q, 0, 0)$ or $\mathbf{q} = (0, q_x, q_y)$ and is dominated by permeation, which is a kind of vacancy diffusion in which mass is transported while leaving the smectic layer structure intact. For other values of \mathbf{q} , there is a transverse sound mode (resulting from the mixing of u with the transverse momentum) with $\omega^2 = (B/\rho)(q_z^2q_p^2/q^2)$, where $q_p^2 = q_x^2 + q_y^2$ [3,19].

The topological line defects in a smectic are dislocations in which the displace-

The topological line defects in a smectic are dislocations in which the displacement u undergoes a change of nd in one circuit around the core [1,2,4,7]. Figure 8 depicts strength-one edge and screws dislocations

A planar array of dislocations creates a grain boundary across which there is a discontinuous jump of the direction of the layer normal. A periodic array of grain

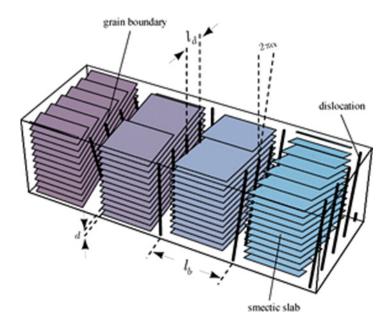


Figure 9. Schematic representation of a twist-grain-boundary phase showing grain boundaries, separated by l_b , consisting of a planar array of dislocations separated by a distance l_d . (Figure appears in color online.)

boundaries can rotate the layer normal and thus the nematic director in a helical pattern similar to that encountered in the cholesteric liquid crystal leading to the twist-grain-boundary phase [20,21] shown in Figure 9.

Liquid crystals continue to produce examples of phases, from biaxial nematics to ferro-electric banana to Blue-phase-TGB phases, and more, whose properties can largely understood in through the construction of phenomenological elastic and dynamic theories that reflect their conservations laws and symmetries. This is a fortunate situation. Even with today's most powerful computers, it is difficult if not impossible to calculate macroscopic properties from first-principle treatments of interaction among the very complicated liquid crystalline mesogen.

Acknowledgment

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