Liquids with Chiral Bond Order

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I describe new phases of a chiral liquid crystal with nematic and hexatic order. I find a conical phase, similar to that of a cholesteric in an applied magnetic field for Frank elastic constants \( K_2 > K_3 \). I discuss the role of fluctuations in the context of this phase and the possibility of satisfying the inequality for sufficiently long polymers. In addition I discuss the topological constraint relating defects in the bond order field to textures of the nematic and elucidate its physical meaning. Finally I discuss the analogy between smectic liquid crystals and chiral hexatics and propose a defect-riddled ground state, akin to the Renn-Lubensky twist grain boundary phase of chiral smectics.

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1. Introduction and Summary

Since time immemorial [1] people have looked for liquid crystal phases with bond-orientational order [2]. Aside from the possibility of a new type of liquid crystal, hexatic-type order can be an intermediate stage in the continuous freezing of a liquid in three dimensions. Much more recently there has been an explosive progress in new chiral phases in both two [3,4,5], and three dimensions [6,7]. In this paper I propose both a uniform and defect laden ground state of a liquid crystal with both hexatic order and chirality.

Toner [1] has proposed that nematic liquid crystals, upon cooling, could form a liquid crystalline phase with nematic order and hexatic order in the plane perpendicular to it. In section 2 I will consider the melting of a chiral columnar phase [7] into a chiral liquid with hexatic order. I discuss both the Landau theory as well as the melting of the crystal through dislocation loop unbinding. When a proliferation of edge and screw dislocations develops, the crystal melts, leaving a normal liquid with hexatic order and no residual constraints associated with the crystal rigidity. Typically, long thin molecules do not have columnar phases, but freeze via smectic phases. I expect then that chiral, disc shaped molecules could participate in the structures discussed here.

It is well known [8] that cholesteric phases, under applied magnetic fields can unwind as well as tip up and become conical phases. However, the conical phases exist only when $K_2 > K_3$, which is seldom the case experimentally. When $K_2 < K_3$ there is a discontinuous transition from a cholesteric with director in the $xy$-plane to a nematic aligned along the field in the $\hat{z}$ direction. In section 3 I find that hexatic order has the same effect as the magnetic field, although it does not participate in unwinding the helical pitch. Adapting the known consequences of fluctuations and nonlinearities of polymer nematics [9] and cholesterics [10] to the chiral polymer hexatic, I find anomalously large elastic constants which depend on the polymer length. For sufficiently long polymers $K_2$ will eventually grow larger than $K_3$. If liquid crystal phases can still exist for these long polymers, Meyer’s conical phase would appear. This would be evidence of hexatic order. When the nematic director has a conical texture, as in a smectic-$C^*$ texture, and the molecules are chiral, an electric polarization can develop. Thus this phase could exhibit ferroelectric liquid crystalline behavior [11].

Since the bond order is defined in the plane perpendicular to the nematic director $\hat{n}$, its value must be determined in a way which takes into account the texture of the director. Because there is a nematic director $\hat{n}$ and a bond-angle order parameter $\theta_6$, the
non-chiral theory is similar to that of the \( A \) phase of \( ^3\text{He} \). In section 4 I will discuss the geometric constraints relating the nematic director to the bond-order field in the absence of free disclinations. This is known as the Mermin-Ho relation in \( ^3\text{He} \) [12]. It plays an important role in understanding the allowed ground states of the chiral N+6 phase and elucidates the structure of the allowed defects.

With chirality added the N+6 order can exhibit a large variety of phases similar in structure to known phases of smectics. When chiral molecules participate in N+6 order, two kinds of twisting are allowed [7]: twisting of the nematic order, leading to cholesteric states, and twisting of the bond order, leading to braided moiré states. The twisting bond order, with an ever increasing angle, is analogous to the layered order of smectics with the pitch of the bond order equivalent to the layer spacing of the smectic. This allows me to consider defect phases similar to the Renn-Lubensky TGB state in chiral smectics. I discuss this in section 5.

2. Landau Theory and Melting of the Hexagonal Columnar Phase

I first derive the free energy of the chiral N+6 phase as the sum of its constituent parts. Fluctuations in the nematic director are described by the Frank free energy density:

\[
\mathcal{F}_\hat{n} = \frac{K_1}{2} \left( \nabla \cdot \hat{n} \right)^2 + \frac{K_2}{2} \left[ \hat{n} \cdot (\nabla \times \hat{n}) - q_0 \right]^2 + \frac{K_3}{2} [\hat{n} \times (\nabla \times \hat{n})]^2 \tag{2.1}
\]

where \( K_i \) are the Frank elastic constants and \( 2\pi/q_0 \) is the equilibrium cholesteric pitch. In order to define bond-angle order, I must, everywhere in space, define a right-handed, orthonormal triad \( \{ \hat{e}_1, \hat{e}_2, \hat{n} \} \), where \( \hat{e}_2 = \hat{n} \times \hat{e}_1 \). In this case, the usual hexatic order parameter, \( \psi_6 = |\psi_6|e^{i\theta_6} \) is defined by

\[
\psi_6(r) = \sum_{i \in P(\hat{n},r)} e^{i\theta_i(r)} \tag{2.2}
\]

where \( \theta_i(r) \) is the angle between the particle \( i \) and the basis vector \( \hat{e}_1 \) as measured around \( r \) with the sign of the angle determined by \( \hat{n} \), and \( P(\hat{n},r) \) is the set of particles that are in the plane perpendicular to \( \hat{n} \) through the point \( r \). The vector \( \hat{e}_1 \) is chosen unambiguously throughout space (possibly modulo rotations by \( 2\pi/6 \)) and may be thought of as pointing to one of the ground state nearest neighbors. More precisely,

\[
\sin[\theta_i(r)] = \frac{\hat{n} \cdot [\hat{e}_1 \times (r_i - r)]}{\| r_i - r \|}. \tag{2.3}
\]
and the sum (2.2) is taken only over particles at position \( \mathbf{r}_i \) such that \( \hat{n}(\mathbf{r}) \cdot (\mathbf{r}_i - \mathbf{r}) = 0 \). The bond-angle order parameter now depends on the texture of \( \hat{n} \). As a result, one must take into account the nematic when taking derivatives of \( \theta_6 \): a covariant derivative must be employed. For now I will ignore this complication and will return to it in Section 4 and will justify the naïve analysis \textit{a posteriori}.

Because the definition of \( \theta_6 \) depends on the direction of \( \hat{n} \), under the transformation \( \hat{n} \rightarrow -\hat{n} \), (2.3) implies that \( \theta_i \rightarrow -\theta_i \) and hence \( \theta_6 \rightarrow -\theta_6 \) (or equivalently, \( \psi_i \leftrightarrow \psi_i^* \)). When constructing a free energy that includes \( \theta_6 \), the overall nematic symmetry \( \hat{n} \rightarrow -\hat{n} \) must be preserved. Owing to the definition of \( \theta_6 \), the free energy can include any term with even powers of \( \hat{n} \) and \( \theta_6 \) together. The spin wave theory for \( \theta_6 \) includes spin-stiffnesses as well as a new chiral term:

\[
\mathcal{F}_{\theta_6} = \frac{K_A^{||} - K_A^\perp}{2} (\hat{n} \cdot \nabla \theta_6)^2 + \frac{K_A^\perp}{2} (\nabla \theta_6)^2 - K_A^{||} \overline{q}_0 \hat{n} \cdot \nabla \theta_6
\]  
(2.4)

where, since there is a preferred direction \( \hat{n} \), I have included the possibility of anisotropic stiffnesses. The final term is chiral and indicates the tendency for the bond-angle order parameter to rotate around the nematic director with pitch \( 2\pi/\overline{q}_0 \) [7,13]. Finally there are additional non-chiral couplings between \( \hat{n} \) and \( \theta_6 \) [1,14]:

\[
\mathcal{F}_{\hat{n}\theta_6} = \overline{C} (\hat{n} \cdot \nabla \theta_6) [\hat{n} \cdot (\nabla \times \hat{n})] + \overline{C}' \nabla \theta_6 \cdot \nabla \times \hat{n}
\]  
(2.5)

The free energy is the sum of terms \( F = \int d^3x \{ \mathcal{F}_{\hat{n}} + \mathcal{F}_{\theta_6} + \mathcal{F}_{\hat{n}\theta_6} \} \).

Recently the theory of chiral polymer crystals has been formulated [7]. One would imagine that the total free energy resulted from the melting of such a crystal. Indeed, this is the case. Considering only quadratic fluctuations around a ground state with \( \langle \hat{n} \rangle = \hat{z} \), the additional free energy coming from the two-dimensional crystal displacement field \( \mathbf{u} \) is

\[
\mathcal{F}_{\text{crystal}} = \mu (\partial_i u_j - \epsilon_{ij} \theta_6)^2 + \frac{\lambda}{2} u_{ij}^2 + \mu' (\partial_z u_i - \delta n_i)^2
\]  
(2.6)

At long wavelengths \( \theta_6 \) and \( \delta n_i \) are locked into crystal deformations by \( \theta_6 = \frac{1}{2} \epsilon_{ij} \partial_i u_j \) and \( \delta n_i = \partial_z u_i \). Thus \( 2 \partial_z \theta_6 - \epsilon_{ij} \partial_i \delta n_j = 0 \). When dislocations are introduced into the crystal, derivatives do not commute so \( \partial_z \partial_i \neq \partial_i \partial_z \). In this case I introduce \( w_{\gamma i} \) which is equal to \( \partial_\gamma u_i \) away from dislocations. As in [15] I introduce a dislocation density tensor \( \alpha_{\mu i} \), the density of dislocations running in the \( \mu \) direction with Burgers vector in the \( i \) direction.
and $\epsilon_{\mu\nu\gamma}\partial_\nu w_\gamma = -\alpha_\mu_i$. With $\delta n_i = w_{zi}$ and $\theta_6 = \frac{1}{2}\epsilon_{ij}w_{ij}$ the long wavelength constraint between $\theta_6$ and $\delta n_i$ becomes instead [7]

$$2\partial_z \theta_6 - \epsilon_{ij}\partial_i \delta n_j = -\text{Tr}[\alpha]$$

(2.7)

where $\epsilon_{ij}$ is the two-dimensional anti-symmetric tensor. In particular (2.7) constrains the two chiral terms to differ only by the density of screw dislocations.

Since a proliferation of edge dislocations running along the $\hat{z}$ direction would be sufficient to melt a two-dimensional columnar crystal, one might think that the constraint would not be altered and that the free energy of the liquid state would be augmented by it. However, this is not the case. In the usual scenario of two-dimensional melting [2] dislocation pairs unbind, leading to a hexatic liquid. In the case of a columnar crystal, parallel dislocation lines can unbind, leading to a hexatic. In a columnar crystal, defects running parallel to the columnar direction ($\hat{z}$) are edge dislocations. Those defects lying in the perpendicular plane ($xy$) are either edge or screw dislocations, depending on whether the Burgers vector is normal or parallel to the dislocation line, respectively. Thus dislocation loops which contribute to the melting of the crystal through edge dislocations along $\hat{z}$ will contain, as well, screw-like and edge-like dislocations in the plane. Since the energy per unit length of a pair of edge dislocations parallel to $\hat{z}$ is finite, infinite dislocations, ending at the boundaries, will not proliferate. Instead, dislocation loops will unbind [15] leading to a proliferation of screw dislocations. To incorporate the presence of screw dislocations I add to (2.6) a term representing the dislocation free energy density [16]:

$$\delta F = E_{\mu i\nu j}(q)\alpha_{\mu i}(q)\alpha_{\nu j}(q)$$

(2.8)

where $E_{\mu i\nu j}$ represents the core and elastic energy of the defects. In the pure crystal the energy per unit length of an unpaired edge dislocation parallel to the $\hat{z}$-axis diverges logarithmically with the system size. Dislocation loops, however will have a finite energy, scaling like $R \ln R$ where $R$ is the size of the loop. Below the melting transition, where the defect loops are bound, the free energy will favor $\alpha_{\mu i} = 0$ and hence Tr$[\alpha] = 0$ everywhere. Thus $2\partial_z \theta_6 = \nabla \times \delta \vec{n}$. However, when dislocation loops unbind, the effective quadratic energy for $\alpha_{\mu i}$ will be finite. The diagonal part of (2.8) becomes

$$\delta F_{\text{diag}} = E_{\text{screw}} \{\text{Tr}[\alpha]\}^2 = E_{\text{screw}} \{2\partial_z \theta_6 - \nabla \times \delta \vec{n}\}^2$$

(2.9)

Thus the constraint (2.7) will be demoted to merely a preference for configurations with $2\partial_z \theta_6 = \epsilon_{ij}\partial_i \delta n_j$ [17,15] and can be absorbed into shifts in $K_A^\parallel, K_2$ and $\vec{C}$. Thus the melted crystal is precisely described by our liquid crystal free energy.
3. The Uniform Conical Phase

I now look for ground states as a function of $K_A$, the amplitude of the hexatic order parameter. The results of this section closely follow those found by Meyer for a cholesteric in a magnetic field parallel to the pitch axis [8]. The analysis here, however, involves the bond order parameter $\theta_6$ and its equilibrium configuration as well as that of the nematic director. For simplicity I take $K_A^+ = K_A || = K_A$ and likewise take $\bar{C} = 0$. I consider as a class of ground states the conical states proposed by Meyer [8]:

$$\hat{n} = [\cos \phi \cos qz, \cos \phi \sin qz, \sin \phi]. \quad (3.1)$$

where $q$ and $\phi$ are free constant parameters. The equations of motion for $\theta_6$ are

$$-K_A \nabla^2 \theta_6 - K_A \tilde{q}_0 \nabla \cdot \hat{n} = 0 \quad (3.2)$$

and so $\nabla^2 \theta_6 = 0$ as $\hat{n}$ is divergence-free. Thus the only solutions for $\theta_6$ are linear functions of the coördinates and so $v \equiv \nabla \theta_6$ is a constant vector. Inserting the ansatz (3.1) and a constant vector $v_0$ into (2.1), (2.4) and (2.5), I have

$$F = \Omega \left\{ \frac{K_A}{2} v_0^2 - K_A \tilde{q}_0 \sin \phi v_0 \cdot \hat{z} + \frac{K_2}{2} \left[ q \cos^2 \phi - q_0 \right]^2 + \frac{K_3}{2} q^2 \sin^2 \phi \cos^2 \phi \right\} \quad (3.3)$$

where $\Omega$ is the volume of space and the oscillating terms drop out upon integration over space. Minimizing with respect to $v_0$ I find

$$v_0 \equiv \nabla \theta_6 = \hat{z} \tilde{q}_0 \sin \phi \quad (3.4)$$

and thus, with $x = \cos^2 \phi$

$$F_{\text{eff}} = \Omega \left\{ \frac{K_2}{2} (qx - q_0)^2 + \frac{K_3}{2} q^2 x (1 - x) - \frac{K_A}{2} \tilde{q}_0^2 (1 - x) \right\} \quad (3.5)$$

This is precisely the energy studied by Meyer for a cholesteric in a magnetic field of strength $H = \tilde{q}_0 \sqrt{K_A}$. In [8] the possibility of the cholesteric unwinding by changing $q$ was considered along with the conical state. In this case the former effect can never happen: because $v_0$ must be constant, the effective field will always be parallel to the pitch axis. Translating Meyer’s results I find that for $K_2 < K_3$ there is a transition with increasing $\sqrt{K_A} \tilde{q}_0$ from the pure cholesteric state ($\phi = 0$, $q = q_0$) to a pure nematic state ($\phi = \pi/2$) at $\sqrt{K_A} \tilde{q}_0 = q_0 \sqrt{K_2}$. For $K_2 > K_3$ there is an intermediate phase as well.
For $\sqrt{K_A}q_0 < q_0\sqrt{K_3}$ the cholesteric state persists, while for $\sqrt{K_A}q_0 > q_0K_2/\sqrt{K_3}$ the nematic state persists. In between

$$q = q_0\sqrt{K_A/K_3}$$

and

$$\cos^2 \phi \equiv x = \frac{K_2\sqrt{K_3/K_A(q_0/\tilde{q}_0) - K_3}{K_2 - K_3}$$

for $q_0\sqrt{K_3} \leq \sqrt{K_A}q_0 \leq q_0K_2/\sqrt{K_3}$. Thus the conical phase should appear continuously as a function of $\tilde{q}_0$, or, alternatively, as a function of $K_A$ for fixed $\tilde{q}_0$. Hence as the hexatic order grows the cholesteric state will become conical and, eventually, nematic. The conical state has, in addition to a smectic-$C^*$-like director texture a rotating bond order as well, with pitch $\tilde{q}_0$. If $\tilde{q}_0/q_0$ is not rational, the fluctuations around this ground state should be akin to those in an incommensurate smectic [16,18].

It is unlikely that $\tilde{q}_0 = 0$ as it is allowed by the same symmetry that allows the cholesteric coupling $q_0$. However, even if this is the case I expect $\tilde{C} \neq 0$. In the case $\tilde{q}_0 = 0$, the $\tilde{C}$ acts as a generated $\tilde{q}_0$, in some sense. I will still have a magnetic field type term, though now it depends on $q$ and $\phi$, which should produce a conical phase. The phase diagram at $\tilde{q}_0 = 0$ will depend on all of $K_2$, $K_3$, $K_A$ and $\tilde{C}$, and mapping it out would be straightforward yet tedious.

Turning back to $\tilde{q}_0 \neq 0$, we have found that the conical phase can persist for $K_2 > K_3$ which is usually not the experimental case. However, in light of recent work on the fluctuation enhancement of elastic constants [9,10] one might hope that this inequality could always be met. Indeed since there is no constraint locking $2\partial_z\theta_6$ with $\nabla_\perp \times \delta\hat{n}$ the non-chiral part of the free energy from [10] would be

$$F = \frac{1}{2} \int d^d x \left\{ E \left[ \nabla_\perp \cdot \bar{\vec{u}} - \frac{1}{2}(\partial_z \bar{\vec{u}})^2 + \omega \right]^2 + G\rho_0^2(\partial_z \omega)^2 + \mathcal{F}_{\hat{n}}[\partial_z \bar{\vec{u}}] \right\}$$

where $E$ is the bulk compression modulus, $\rho_0$ the average polymer density, $\partial_z \bar{\vec{u}} = \delta \hat{n}$, $G = kT\ell/\rho_0$, $\ell$ is the typical polymer length and $\mathcal{F}_{\hat{n}}$ is expanded to quadratic order in $\delta \hat{n}$ where $\hat{n} \approx \hat{z} + \delta \hat{n}$. It was found in [10] that for polymers longer than $\ell_0$, $E$, $K_2$ and $K_3$ took on anomalous, $\ell$-dependent values. For polymers with only steric entropic interactions $\ell_0 = L_P^3/a^2$ where $L_P$ is the polymer persistence length and $a$ is the average areal interpolymer spacing. It was found that $K_2(\ell) \sim (\ell/\ell_0)^{0.20}$ and $K_3(\ell) \sim (\ell/\ell_0)^{0.15}$ to two loops in a $d = 4 - \epsilon$ expansion at $d = 3$.  

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To the free energy in (3.8) I add the hexatic stiffness $\mathcal{F}_{\theta_6}$. Expanding around $\mathbf{n} = \hat{z}$ I can consider the possible corrections to the hexatic stiffness $K_A$ as well as the chiral couplings $q_0$ and $\tilde{q}_0$. Following the analysis in [10], I find that $K_A$ and $\tilde{q}_0$ do not anomalously renormalize and suffer only finite shifts in their values. To illustrate, I consider adding $\mathcal{F}_{\theta_6}$ to (3.8) with $K_A^\perp = K_A^\parallel = K_A$. I am interested in fluctuation-induced changes to $K_A$ and $\tilde{q}_0$. As in the argument of [10] for the non-renormalization of $q_0$, I consider the $\tilde{q}_0 \approx 0$ limit. Thus I do not consider corrections to quadratic propagators arising from the coupling $K_A \tilde{q}_0 \partial_z \mathbf{u} \cdot \nabla_\perp \theta_6$. It is easy to see that, since I consider only corrections linear in $\tilde{q}_0$, $K_A$ does not acquire any infinite renormalization, and, in fact, even if I consider $K_A^\perp \neq K_A^\parallel$ the corrections from fluctuations in $\delta \mathbf{n}$ only lead to finite renormalizations. The corrections to $\tilde{q}_0$ must come from non-linearities in the chiral term itself; no other term can generate the appropriate chiral coupling. The tadpole graph arising from this is proportional to $\langle \delta \mathbf{n}^2(\mathbf{r}) \rangle$ and thus only contributes to a finite shift in $K_A \tilde{q}_0$ in three dimensions, where nematic order persists. Alternatively, by a judicious choice of rescalings [19], one can show that the longitudinal part of $\mathbf{u}$ is irrelevant to the renormalization of the Frank constants, and thus the new coupling, which only involves the longitudinal part of $\mathbf{u}$, will not change the results in [10]. Thus the mean field analysis of the uniform conical state holds when I replace the constant Frank constants with $\ell$-dependent Frank constants. Hence, for sufficiently long polymers, it may be possible to have the unusual situation of $K_2 > K_3$, though since $K_2/K_3 \sim (\ell/\ell_0)^{0.05}$, a twofold increase in the ratio of Frank constants implies the polymer must be $10^6 \ell_0$. For instance, in DNA $L_P = 600 \text{Å}$ and $a = 35 \text{Å}$ lead to $\ell_0 \approx 18 \mu m$. With typical chain lengths on the order of centimeters, this can lead to a 37% increase in the ratio of $K_2$ to $K_3$. This increase leads one to hope that the conical phase could persist and could be observed.

Aside from the light scattering behavior of this phase due to its N+6 character [1], it should be possible to see this phase through crossed polarizers. As hexatic order grows past the cholesteric-to-conical point, the extinction directions (perpendicular to the director) will continuously change from the nematic direction to the (perpendicular) cholesteric plane. This will happen without a density modulation forming, as in the smectic-$C^*$ phase which would have a similar behavior. The density modulation could be detected (or not) via electron microscopy and x-ray scattering, thus distinguishing this new phase from the smectic. As in the moiré phase of chiral columnar crystals [7], in each constant $z$ cross section the structure function will be, in Fourier space, that of a hexatic – six broad spots. When looking at the structure of the bulk sample, however, the rotation of the bond order
will merge these spots into a ring in the $q_x-q_y$ plane. The periodicity of the rotation will lead to additional rings at $q_z = 6n\tilde{q}_0 \sin \phi$ where $n = \pm 1, \pm 2, \ldots$.

In addition, as in a smectic-$C^*$, this uniform conical texture can exhibit ferroelectric behavior through a term in the free energy

$$F_{\text{flc}}^* = \mu (\mathbf{v} \cdot \hat{n}) \mathbf{E} \cdot (\mathbf{v} \times \hat{n}) = \mu \tilde{q}_0 \sin \phi \cos \phi [E_y \cos qz - E_x \sin qz]$$ (3.9)

This extra interaction preserves the nematic symmetry and is chiral, since under parity, $\mathbf{E} \rightarrow -\mathbf{E}$. I have taken the “layer normal” to be $\mathbf{v}$ in constructing (3.9). There is, however, a term that is allowed in these chiral phases that has no analogue in the smectic-$C^*$. Since the direction of $\mathbf{v}$ is determined by the chiral coupling $\tilde{q}_0$ and $\hat{n}$, a non-chiral term is allowed

$$F_{\text{flc}} = \mu \mathbf{E} \cdot (\mathbf{v} \times \hat{n})$$ (3.10)

which preserves the nematic symmetry ($\hat{n}$ and $\mathbf{v}$ both change sign) and is not chiral. Under spatial reflection $\mathbf{v} \times \hat{n}$ unambiguously changes sign, as does $\mathbf{E}$. This term is allowed because there is a difference, for fixed $\hat{n}$, between a configuration with $\mathbf{v}$ and $-\mathbf{v}$. They differ by the handedness of the hexatic helix. In a smectic-$C^*$ the layer normal does not have an unambiguously defined direction because of the up-down symmetry of the layered structure. Unlike a smectic, this ferroelectric liquid crystal does not have a density wave as in the usual ferroelectric smectic liquid crystals. The wave that supports the conical texture is one of twisting hexatic order along $\mathbf{v}$. In section 5 I will make the analogy between smectics and chiral hexatics more precise. Note that $F_{\text{flc}}$ also implies that an applied electric field will favor configurations in which the hexatic order twists in planes perpendicular to $\hat{n}$.

4. Topological Constraints: The Mermin-Ho Relation and Saddle-Splay

Up to this point I have ignored the topological relation between $\theta_6$ and $\hat{n}$. From the definition of $\theta_6$ arising from (2.3), it is clear that textures in $\hat{n}$ can influence the local value of $\theta_6$. The two orientational order parameters are related to each other in much the same way that the two order parameters of $^3$He-A are related, namely, through the Mermin-Ho relation [12]. I will sketch here a brief derivation, following [20]. Let $\{\hat{\mathbf{e}}_1, \hat{\mathbf{e}}_2, \hat{n}\}$ be a right-handed, orthonormal triad. If I consider a rotation $\delta\theta_6$ about $\hat{n}$, then

$$\hat{\mathbf{e}}'_1 + i\hat{\mathbf{e}}'_2 = e^{i\delta\theta_6} (\hat{\mathbf{e}}_1 + i\hat{\mathbf{e}}_2)$$ (4.1)
and so
\begin{equation}
\hat{e}_1' - \hat{e}_1 \approx -\delta \theta \hat{e}_2
\end{equation}
\begin{equation}
\hat{e}_2' - \hat{e}_2 \approx \delta \theta \hat{e}_1
\end{equation}
Thus \( \delta \theta \approx \hat{e}_1 \cdot \hat{e}_2 \), or
\begin{equation}
v_\mu \equiv \nabla_\mu \theta = e^\alpha_1 \nabla_\mu e^\alpha_2
\end{equation}
Taking the curl of both sides and using \( \hat{e}_1 \times \hat{e}_2 = \hat{n} \), we have
\begin{equation}
[\nabla \times v]_\mu = \frac{1}{2} \epsilon_{\mu \nu \rho} e^\alpha_\nu n^\alpha \partial_\nu n^\beta \partial_\rho n^\gamma,
\end{equation}
the celebrated Mermin-Ho relation. The content of this relation is that in the ground state of the N+6 phase, disclinations in \( \theta_6 \) are forced in by the nematic texture. It quantifies a simple topological consequence of non-cylindrically-symmetric molecules. For concreteness, take a biaxial molecule in a cylindrical geometry with normal boundary conditions. One might imagine a +1 disclination line will run up the center of the cylinder. However, it is well known that such a geometry will “escape into the third dimension” along the disclination. However, if the molecules have the same (biaxial) orientation at the cylinder wall, when they escape they will be compelled to force a disclination into the biaxial order parameter in the center. This is illustrated in Figure 1. This frustration is summed up in (4.4). Note that in the previous section, I took \( \nabla \times v = 0 \), which, in the case of the Meyer ansatz, is justified by simply plugging (3.1) into (4.4).

When I look for ground state configurations I must take the constraint into account. The excited states differ from the ground state by the energy of a free disclination. In the hexatic phase, this energy diverges with the system size, so the Mermin-Ho relation should be enforced. This is not unlike hexatic phases on flexible membranes. There disclinations were forced in by gaussian curvature [21]. In particular, if I take as the the hexatic director \( \mathbf{\hat{N}} = \cos \theta_6 \hat{e}_1 + \sin \theta_6 \hat{e}_2 \) and compute the covariant derivatives of \( \mathbf{\hat{N}} \) in the local basis determined by \( \{ \hat{e}_1, \hat{e}_2, \hat{n} \} \), I find
\begin{equation}
D_\mu N_1 = -\hat{e}_\mu \cdot [\nabla \theta_6 - \Omega] \sin \theta_6
D_\mu N_2 = \hat{e}_\mu \cdot [\nabla \theta_6 - \Omega] \cos \theta_6
\end{equation}
where \( \hat{n} \equiv \hat{e}_3 \) and \( \Omega_\mu = e^\alpha_1 \partial_\mu e^\alpha_2 \). The appropriate elastic free energy would be
\begin{equation}
F_\Omega = \frac{k_{\perp}}{2} [D_1 N_j]^2 + \frac{k_{||}}{2} [D_3 N_j]^2 = \frac{k_{\perp}}{2} [\nabla \theta_6 - \Omega]^2 + \frac{k_{||} - k_{\perp}}{2} [\hat{n} \cdot (\nabla \theta_6 - \Omega)]^2
\end{equation}
In terms of $\hat{N}$, the term $\hat{n} \cdot \nabla \theta_6$, for instance, becomes $n_\mu \epsilon_{ij} N_i D_\mu N_j \equiv \hat{n} \cdot (\nabla \theta_6 - \Omega)$. Thus, in general, one should always include $\Omega$ with derivatives of $\theta_6$ as a consequence of the wobbling about of $\hat{n}$. Because a change of basis vectors $\hat{e}_1(\mathbf{r})$ and $\hat{e}_2(\mathbf{r})$ should not change the physics of $\theta_6$, I will consider only “gauge-invariant” correlations of $\theta_6$ through

$$G_{\theta_6 \theta_6}(\mathbf{r}; \Gamma) = \langle e^{i \theta_6(\mathbf{r})} \exp \{ i \int_\Gamma \Omega \cdot d\mathbf{\ell} \} e^{-i \theta_6(\mathbf{0})} \rangle \quad (4.7)$$

where $\Gamma$ is a path connecting $\mathbf{r}$ and $\mathbf{0}$. When this Greens function is non-vanishing for large separations $\mathbf{r}$, there is long-range hexatic order. This correlation function will be invariant under shifts of $\Omega$ by $\nabla \omega$ and $\theta_6$ by $\omega$, where $\omega$ is an arbitrary function of space. These transformations are precisely those which do not change $\nabla \times \Omega$ and hence do not change the disclinations structure of $\theta_6$. When $\nabla \times \Omega = 0$ everywhere, there are no obstructions to choosing $\hat{e}_i(\mathbf{r})$ to be constant in space, leading to $\Omega = 0$ and to the usual definition and interpretation of $\theta_6$. Note that when the nematic is highly disordered, with a rich wobbling texture, the contribution to (4.7) from the nematic curvature can greatly reduce, and possibly destroy, long-range hexatic order.

Since unbound disclinations cost a logarithmically-divergent energy they will not appear in equilibrium. However regions where $\nabla \times \Omega \neq 0$ have precisely the same divergent energy as a free disclination. Writing $\theta_6 = \theta_6^{\text{smooth}} + \theta_6^{\text{sing}}$ we can expand around a background of disclinations chosen to screen these regions with $\nabla \theta_6^{\text{sing}} = \Omega$, leaving only smooth variations in $\theta_6$. The dislocations are locked to $\nabla \times \Omega$ according to the Mermin-Ho relation (4.4). Notice that in the case that $\hat{n}$ corresponds to a field of normal vectors to surfaces, $\hat{n}(\mathbf{r}) \cdot [\nabla \times \Omega(\mathbf{r})]$ is the Gaussian curvature of the surface at $\mathbf{r}$ to which $\hat{n}(\mathbf{r})$ is normal. Rewriting the curvature, I find that $\hat{n} \cdot (\nabla \times \Omega) \equiv -\nabla \cdot [(\hat{n} \cdot \nabla) \hat{n} - \hat{n} (\nabla \cdot \hat{n})]$, the well known saddle-splay term of nematic liquid crystals. Thus I can identify $\bar{C}'$ with $K_{24}$ the saddle-splay elastic constant.

This can be understood geometrically: to lowest order in $\delta \tilde{n}$

$$[\nabla \times \nabla \theta_6]_i = \frac{1}{2} \epsilon_{ijk} \epsilon_{\alpha \beta \gamma} n^\alpha \partial_j n^\beta \partial_k n^\gamma \approx \epsilon_{ijk} \partial_j \delta n_x \partial_k \delta n_y \quad (4.8)$$

Disclinations pointing along $\hat{z}$ are constrained by

$$[\nabla \times \nabla \theta_6]_z = \partial_x \delta n_x \partial_y \delta n_y - \partial_x \delta n_y \partial_y \delta n_x \quad (4.9)$$

in other words, disclinations must appear if the directors are the normals to saddle-surface. The saddle-splay term measures the amount of saddle-like deformations in the director.
Since $\theta_6$ is single-valued on a surface, the Mermin-Ho relation tells us that the saddles must be such that the integral of their saddle-splay around their boundary is a multiple of $2\pi/6$. However, in the N+6 case, the director $\hat{n}$ need not be the collection of normals to a surface. Indeed, in the double twist texture of liquid crystal blue phases there is no surface to which the directors are normal. If such a texture were present in the chiral N+6 system, (4.9) implies that as a circuit is traversed around a central molecule of a double twist cylinder, there would be fewer than $z = 6$ nearest neighbors since the saddle-splay is positive. Considering the director texture around the center of a double twist core, Figure 2 shows 19 braided polymers with the center one being straight. It is not necessary that these be polymers, just that the nematic director be tangent to the lines. This texture has double twist in the nematic field as found in blue phases of cholesteric liquid crystals [22,23]. The integrated trajectories are also known to be the projections of great circles of $S_3$, the surface of a 4-dimensional sphere, onto flat 3-dimensional space. The curvature of $S_3$ implies that the coordination number is less than $z = 6$ [23,24] which is consistent with (4.9). This is illustrated in Figure 3. Here the molecular centers (represented by dots) lie on a local triangular lattice, as in the hexatic phase. When the polymers are tipped over, the distance between their centers is necessarily increased. This reduces the allowed packing density leading to coordination numbers less than $z = 6$.

5. The Equivalence of Smectics and Chiral Hexatics

5.1. Derivation of Chiral Hexatic Free Energy

I have shown that for $K_2 > K_3$ a uniform state with a smectic-$C^*$ texture can persist for sufficiently strong hexatic ordering. A different scenario is also possible, leading to a defect state, analogous to the Abrikosov flux lattice in superconductors and isomorphic to the twist grain boundary phase of chiral smectic-$A$ [6]. Consider first the Landau theory for an isotropic hexatic in a background nematic field

$$\mathcal{F}_{\text{hex}} = \frac{1}{2}|\mathbf{D}\psi_6|^2 + r|\psi_6|^2 + u|\psi_6|^4$$

(5.1)

where $\psi_6$ is the hexatic order parameter, which, in the broken phase, is approximately $\psi_6 \approx (\sqrt{K_A}/6)e^{i\theta_6}$, and, as usual, $r \propto (T - T_c)$. $\mathbf{D}$ refers to the covariant derivative, which subtracts $\Omega$ from the gradient of $\theta_6$.
Adding to this the chiral term (covariantly modified) in $F_{\theta_6}$, I have

$$F_{\text{hex}}^* = 6i\tilde{q}_0 \hat{n} \cdot \left[ \psi_6^* D \psi_6 - \psi_6 D \psi_6^* \right] + \frac{36}{2} \tilde{q}_0^2 |\psi_6|^2 (\hat{n}^2 - 1) \quad (5.2)$$

Recall that under $\hat{n} \to -\hat{n}$, $\psi_6 \leftrightarrow \psi_6^*$ and so the first term in (5.2) respects the nematic symmetry. The second term, though identically zero, will be split up so that I may write the total free energy as

$$F_{\text{hex}} = \int d^3x \left\{ \frac{1}{2} |(D - 6i\tilde{q}_0 \hat{n}) \psi_6|^2 + \left[ r - 18\tilde{q}_0^2 \right] |\psi_6|^2 + u|\psi_6|^4 + F_{\hat{n}} \right\} \quad (5.3)$$

Thus, after shifting $T_c$ upwards by $18\tilde{q}_0^2$, the free energy of a chiral hexatic is precisely that of a chiral smectic [25] where $2\pi/6\tilde{q}_0$, the spacing between surfaces with the same bond-order orientation, is the equilibrium smectic layer spacing. There is an important difference between the smectic and the chiral hexatic which is what allowed the non-chiral ferroelectric term (3.10) in the latter. In a smectic, the physics is invariant under $\tilde{q}_0 \to -\tilde{q}_0$ and $\theta_6 \to -\theta_6$ ($\psi_6 \leftrightarrow \psi_6^*$). While that appears to be a symmetry of (5.3) recall that $\theta_6 \to -\theta_6$ must be accompanied by $\hat{n} \to -\hat{n}$. In other words, there is a difference between $\tilde{q}_0$ and $-\tilde{q}_0$ in the hexatic that does not exist in the analogous smectic. It is also important to note the distinction between $\Omega$ and $\hat{n}$. Shifts of $\theta_6$ by $\omega$ accompanied by shifts of $\Omega$ by $\partial \omega$ are true gauge transformations. The invariance under these transformations is not the sign of a symmetry, but rather a redundancy of identical physical descriptions. Rigid rotations in space, on the other hand, are responsible for the gauge-like coupling of $\theta_6$ to $\hat{n}$. Under rotation by angle $\Phi$, $\hat{n} \to \hat{n} + \Phi \times \hat{n}$ and $\theta_6 \to \theta_6 + [\Phi \times \hat{n}] \cdot \mathbf{r}$, so that, to lowest order in fluctuations of $\hat{n}$ around a spatially-uniform, ordered state (e.g., $\hat{n} = \hat{z}$), the rotation is a symmetry of the theory. A rotation is physical and corresponds to different but equivalent physical systems [26]. In the highly aligned limit, when $\delta \vec{n}$ is small and $n_z \approx 1$, $\nabla \times \Omega = O(\delta \vec{n}^2)$ and hence the contribution from the covariant derivative is much smaller than that from the chiral term in the derivative $\tilde{q}_0 \delta \vec{n}$. Thus I expect that the energetics of the chiral hexatic phases should be similar to that of the analogous smectic phases.

It is now clear that the uniform conical state is just one possible smectic phase. In the nematic state ($\sin \phi = 1$), there is pure chiral hexatic order, with $\theta_6 = \tilde{q}_0 z$. This is equivalent to a smectic-A phase as shown in Figure 4. Upon reduction of the nematic order, the nematic director tips out of the $\hat{z}$ direction and the phase is equivalent to a smectic-C* phase. The chiral order acquires a longer pitch, which is equivalent to a larger smectic layer spacing. Finally, as the hexatic order is reduced further, the director lies down into a nematic texture and there is no chiral hexatic order. This is just a pure cholesteric. The smectic layer spacing has gone to infinity and there is no smectic order.
5.2. Defect Phase: The Renn-Lubensky State

Ignoring the complications of the Mermin-Ho relation and the covariant derivative, this model has been studied extensively [6] and, in the type-II regime (where \( u \) is sufficiently large) one expects a ground state with a proliferation of defects. I propose then a new twist grain boundary state of *hexatics*. It will consist of regions of twisted N+6 separated by grain boundaries made up of hexatic disclinations. Within in each region the bond order will twist with pitch \( 2\pi/\tilde{q}_0 \) and the director will be well aligned in a nematic state. As one moves along a pitch axis perpendicular to the nematic director, across a grain boundary, the nematic direction will jump by some finite angle \( \alpha \). This will lead to a state which, at long distances, will appear to be pure cholesteric, but will in fact have regions with rotating hexatic order.

Since the possibility of the defect phase is independent of the existence of the uniform conical phase, it is possible that the uniform conical phase could be punctuated itself by defects. In this case \( \phi_\infty \) would be the equilibrium conical angle and the director would relax to this cone. This exotic phase would be similar to the TGB\(_C\) phase [27] in which the defect-free regions are not smectic-A, but rather smectic-C, though in this case it may be more appropriate to think of the clean regions as being smectic-C* instead.

One might speculate on other possible phases. Since the bond order field \( \theta_6 \) has much the same behavior as \( \theta_2 \), a biaxial order parameter, structures of chiral N+6 phases should be in close analogy with the myriad of blue phase textures: in other words an N+6 phase is closely related to a biaxial nematic. Owing to the connection with smectics it is a savory proposition to think of blue phases made of smectic layers, where now the phase \( \theta_2 \) present in the high-chirality limit of a blue phase [22] could be interpreted as ticking off the lamellar layers of the smectic in space. This prospect is under investigation [28].

5.3. The Structure of Topological Defects

In light of the Mermin-Ho relation, the structure of the defects requires some clarification. Configurations which satisfy the Mermin-Ho relation can have disclinations in \( \theta_6 \) without a logarithmically divergent energy per unit length. However, configurations are not required to be so innocuous. The defects necessary for a TGB state are still possible. The energy of the core will be modified, of course, by the extra energy associated with the “curvature” of the nematic field. Considering a screw dislocation in the hexatic order, \( \theta_6 \)
will wind around in each $xy$-plane, independent of $z$, and so $\theta_6 = \theta$, where $\theta$ is the azimuthal coördinate. The equations of motion imply that at radii larger than $\sqrt{K_1/K_A}/\tilde{q}_0$ the nematic field will lock to the “superfluid velocity” $\vec{v} = \nabla_\perp \theta_6/\tilde{q}_0$, and thus

$$\delta \vec{n} \approx \frac{1}{\tilde{q}_0 r} \hat{\theta}$$

(5.4)

where $\hat{\theta} \equiv (x \hat{y} - y \hat{x})/r$ is the unit azimuthal vector in cylindrical coördinates. Note that since $\delta \vec{n}$ does not depend on $z$ the only component of the Mermin-Ho relation that is non-zero is the $z$ component. Using (4.9) I find, to leading order in $\delta \vec{n}$, that

$$[\nabla \times \Omega]_z = -\frac{1}{\tilde{q}_0^2} \frac{1}{r^4}$$

(5.5)

Thus for large $r$ the gauge field $\Omega$ falls off like $r^{-3}$, which is much faster than the rate at which the vorticity falls off ($r^{-1}$) and equal to the rate at which the difference ($\nabla_\perp \theta_6 - \tilde{q}_0 \delta \vec{n}$) falls off at infinity. Thus, ignoring the effect of the nematic curvature adds an energy comparable to the energy of the defect itself, and certainly does not cause the defect to become energetically prohibited.

Turning back to the Meyer ansatz, one might consider defects in the uniform conical state. I can solve the Mermin-Ho relation to find a family of defects in a conical state. Using a radially dependent ansatz, I take

$$\hat{n} = \begin{bmatrix} \cos \phi(r) \cos \tilde{q}_0 z, & \cos \phi(r) \sin \tilde{q}_0 z, & \sin \phi(r) \end{bmatrix}$$

(5.6)

In the center of the defect, $\phi(0) = 0$ and I have a pure cholesteric region – a region which does not support hexatic order. Moreover, at infinity, the director relaxes to a conical state, i.e. $\phi(\infty) = \phi_\infty$. In this case, it is straightforward to compute

$$[\nabla \times \vec{v}]_x = -\frac{yq_0 \cos \phi \partial \phi}{r} \frac{\partial \phi}{\partial r}$$

$$[\nabla \times \vec{v}]_y = \frac{xq_0 \cos \phi \partial \phi}{r} \frac{\partial \phi}{\partial r}$$

$$[\nabla \times \vec{v}]_z = \frac{\cos \phi \partial \phi}{r} \frac{\partial \phi}{\partial r}$$

(5.7)

solving for $\vec{v}$ I have

$$\vec{v} = \frac{\sin \phi(r)}{r} \hat{\phi} + \tilde{q}_0 (\sin \phi_\infty - \sin \phi(r)) \hat{z} + \nabla \theta_6$$

(5.8)
where \( \hat{\theta} \equiv \frac{(x\hat{y} - y\hat{x})}{r} \) is the unit azimuthal vector in cylindrical coordinates, and \( \nabla \theta_6 \) is a non-singular variation of the bond-angle direction, with equilibrium value \( \tilde{q}_0 \sin \phi_\infty \hat{z} \) in a pure conical state. Far from the core, as \( r \to \infty \), \( \mathbf{v} \to \nabla \theta_6 = \tilde{q}_0 \sin \phi_\infty \hat{z} \) and is not singular. Inside the core, \( \sin \phi \approx 0 \) and, minimizing the free energy for \( \nabla \theta_6 \), the bond order is constant. To be more precise, in order to have nonsingular field configurations, \( \nabla \times \mathbf{v} \) must not diverge at the origin. Since \( \cos \phi(r \to 0) = 1 \), (5.7) implies that \( \partial_r \phi \) must go to 0 no slower than linearly in \( r \). Hence \( \sin \phi(r)/r \) is well behaved at \( r = 0 \). In the core of this defect there is pure cholesteric order of the nematic field, while outside the core there is a pure uniform conical state.

In considering configurations (5.8), it is essential to note that, when integrated around a closed loop of radius \( R \) in an arbitrary constant-\( z \) slice, \( \theta_6 \) must be single-valued up to shifts by \( 2\pi/6 \) and hence

\[
\frac{2\pi n}{6} = \int_{\partial M} d\mathbf{\ell} \cdot \mathbf{v} = \int_M dx dy \left[ \nabla \times \mathbf{v} \right]_z = 2\pi \left[ \sin \phi(R) - \sin \phi(0) \right] \tag{5.9}
\]

Thus, if I consider configurations without any free disclinations, then \( \sin \phi(r) \) must take on values which are integer multiples of \( 1/6 \). This would prevent \( \mathbf{n} \) from relaxing back to a nematic configuration on a longer length scale than the disappearance of \( |\psi_6| \). In other words, the quantization of the circulation of \( \theta_6 \) enforces a quantization in the tilt of \( \mathbf{n} \). Therefore, if there were a core in which \( \psi_6 \) vanished (of radius \( \sim \xi \)), outside that core the nematic would have to have a constant tilt. This is as if the London penetration depth \( \lambda \) were smaller than \( \xi \). This prevents the usual type II defects of the TGB state. This sort of restriction is reminiscent of the disclination buckling transition in hexatic membranes [29]. This type of defect has a core in which there is not hexatic order, or perhaps an annular region in which the tilt of the nematic changes gradually by \( 2\pi m/6 \), for integer \( m \). Outside the core there is hexatic order as well as nematic order. Inside there is a gain in cholesteric energy as the nematic tips over and exploits its chirality, while there is a loss of hexatic energy as its order is destroyed. A detailed energetic calculation, using the entire rotationally invariant theory must be performed in order to determine whether this non topological defect is stable.

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Figure Captions

Fig. 1. Escape into the third dimension of a biaxial object. As a +1 nematic disclination tries to escape into the third dimension, the small axis director is forced to have a +1 disclination. The long thicker line is the nematic axis while the shorter, thinner line is the small director. At the boundary the nematic is normal and the small director is uniform and normal to the plane of the boundary.

Fig. 2. A braided state with a double twist texture. The center line is straight and has fewer than $z = 6$ nearest neighbors. The lines may represent long polymers or merely curves with tangent vectors equal to the local nematic director (the integral curves).

Fig. 3. Illustration of the frustrated packing of tilted molecules. Note that when the molecules are tipped over, the distance between their centers increases. This reduces the allowed packing density and leads to coördination numbers less than $z = 6$.

Fig. 4. Model of a chiral hexatic. In each plane the bond-order parameter is $\theta_6 = \theta_6^0 \mod 2\pi/6$. Between the planes the bond order uniformly precesses along the average nematic director, $\hat{n} = \hat{z}$. The planes are analogous to smectic planes, though there is no density wave in this liquid crystalline phase.
Figure 3
Figure 4