Quasicrystalline Textures of Cholesteric Liquid Crystals: Blue Phase III?

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The energetics of icosahedral ordering in cholesteric liquid crystals are studied within the Landau mean-field approximation, in an attempt to describe the blue fog (blue phase III). We expand the free energy of a quasicrystalline phase in the high-chirality limit (coherence length much greater than pitch), and find that although it is energetically competitive with the cubic blue phases, no vertex-edge icosahedral phase is ever favored in this regime.

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Cholesteric liquid crystals are fluids composed of chiral, rodlike molecules. At high temperatures, the molecular orientations are isotropic; at low temperatures the system remains fluid, but the molecular orientations "crystallize" into the conventional cholesteric or "helical" phase. In short-pitch cholesteric liquid-crystal systems (pitch less than 2500 Å), the transition from the isotropic to the helical phase can be interrupted by up to three "blue phases"[1] which occur in a narrow range of temperatures (≈ 0.1–2 K) near the isotropic-helical transition. Measurements on the two lower-temperature phases (blue phases I and II) are consistent with cubic structures (O^8 and O^2, respectively), based on the wave numbers and intensities of observed Bragg peaks,[2] and the growth of single crystals.[3] The highest-temperature blue phase, known as the "blue fog"[4] or blue phase III,[2] appears to exhibit neither Bragg peaks nor single crystals, and has been assumed to be amorphous.

Heat-capacity and elastic-moduli measurements[5] have shown that the fog, which is optically active and appears optically isotropic, is a distinct thermodynamically stable phase and not merely a pretransitional effect. Most of the isotropic-helical latent heat is released in the transition to the blue fog, indicating that it is a substantially ordered phase. The fog cannot be a traditional glass because it persists on both heating and cooling; liquidlike correlations between the ordered regions are precluded by the fog's nonzero shear modulus. Inspired by parallels between the Landau theories of quasicrystals and the blue phases, and by their common ties to metallic glasses, we present below a calculation of the free energy of a quasicrystalline blue-phase texture.

The blue phases have been extensively studied via Landau theory,[6] in which the free energy is written as a power series in the order parameter $Q_{ij}(\mathbf{x})$, a symmetric traceless matrix parametrizing the local distribution of molecular orientations. The expansion is typically taken to be a sum of bulk and gradient terms, truncating the bulk energy at the quartic term (the lowest order which provides stability) and retaining only the lowest-order gradient terms[7]:

$$f_{\text{grad}} = (\kappa^2/4)\langle \mathrm{Tr}(\nabla \times \mathbf{Q} - \mathbf{Q})^2 + (1+\sigma)(\nabla \cdot \mathbf{Q})^2 \rangle, \tag{1a}$$

$$f_{\text{bulk}} = \tau \langle \mathrm{Tr}\mathbf{Q}^2 \rangle - \sqrt{6}\langle \mathrm{Tr}\mathbf{Q}^3 \rangle + 2\langle \mathrm{Tr}\mathbf{Q}^4 \rangle, \tag{1b}$$

where $\langle \ldots \rangle$ denotes a spatial average, $(\nabla \times \mathbf{Q})_{rs} = \epsilon_{ijr}\partial_i Q_{js}$ and $(\nabla \cdot \mathbf{Q})_j = \partial_i Q_{ij}$. In (1) the free energy and the order parameter have been scaled and lengths measured in units of the pitch; the "chirality" κ is then the correlation length, σ is a ratio of elastic constants, and τ is proportional to the temperature measured from the second-order transition which would occur in the absence of the cubic term.

Minimizing even this highly simplified free energy is enormously complicated, and progress has only been made in the "high-chirality limit," $\kappa \to \infty$. For large κ the free energy is dominated by the gradient terms (1a), allowing only textures $Q_{ij}(\mathbf{x})$ which minimize them, i.e., a sum of plane waves

$$\tfrac{1}{2}(\hat{\mathbf{a}} + i\hat{\mathbf{b}})_i^n (\hat{\mathbf{a}} + i\hat{\mathbf{b}})_j^n \exp(ik_n \hat{\mathbf{c}}^n \cdot \mathbf{x}), \tag{2}$$

where $\{(\hat{\mathbf{a}}^n, \hat{\mathbf{b}}^n, \hat{\mathbf{c}}^n)\}$ is a set of right-handed orthonormal triads and $k_n \approx 1$. To minimize the bulk free energy within this restricted set of order parameters, one must choose wave vectors which can take maximal advantage of the stabilizing influence of the cubic term. A large cubic contribution requires a set $\{k_n \hat{\mathbf{c}}^n\}$ with many triples of wave vectors which sum to zero ("triangles").

The transition sequence for the free energy (1) in the high-chirality limit is believed to be[6e] helical → hexagonal → O^5 → isotropic, in poor agreement with the experimental sequence helical → O^8 → O^2 → fog → isotropic. The discrepancy is not surprising, since for real systems $\kappa \approx 0.1$–0.5.[6e] The large-chirality regime is interesting as a simple tractable model and a supplement to calculations which examine the low-chirality limit directly.[8]

It is well known that pentagonal and icosahedral point groups are incompatible with 2D and 3D translational order; until recently, however, it was not widely

appreciated that point diffraction patterns with other than twofold, threefold, fourfold, and sixfold axes of symmetry *are* possible. Icosahedral ("quasicrystalline") diffraction patterns[9] in rapidly quenched $Al_{0.84}Mn_{0.16}$ have been interpreted[10] as diffraction from systems with long-range orientational order and quasiperiodic translational order.

A variety of approaches based on Landau's theory of crystallization[11] have been applied to the problem of quasicrystal stability.[12] In dimensionless form, the free energy for a scalar order parameter $\rho(\mathbf{x})$ $(=\int d^3k\rho_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{x}})$ is taken to be

$$F=\sum_{\mathbf{k}}A(k)\rho_{\mathbf{k}}\rho_{-\mathbf{k}}-\sum_{\mathbf{k}_1,\mathbf{k}_2}\rho_{\mathbf{k}_1}\rho_{\mathbf{k}_2}\rho_{-\mathbf{k}_1-\mathbf{k}_2}+\cdots, \tag{3}$$

with $A(k)=\tau+\kappa^2(k-1)^2+\ldots$; this is the scalar analog of the cholesteric free energy (1). Landau's original treatment only considers orderings with $|k|=1$, corresponding to the "one wave number" approximation $\kappa=\infty$. The phase with the lowest barrier to formation is then body-centered cubic (wave vectors in octahedral directions).[13] It might be expected that a phase with wave vectors parallel to the edges of an icosahedron centered in reciprocal space would be energetically favored because of a large cubic term (an icosahedron has twenty equilateral triangular faces), but such a phase is never globally stable because of an increased quadratic term.[12b]

Kalugin, Kitaev, and Levitov[12c] extended this calculation to large but finite κ, noting that the center-vertex distance of an icosahedron, k_V, is only 5% less than the edge length k_E (an icosahedron is nearly a packing of twelve tetrahedra). For κ not too large, both k_E and k_V can be accommodated near the minimum of $A(k)$ without a prohibitive quadratic energy, and the cubic term is enhanced by thirty more triangles. This enables a vertex-edge (k_E and k_V) phase to have a lower barrier to formation than body-centered cubic for $1 \ll \kappa^2/\tau \lesssim 69.5$. The inclusion of a local quartic term $\langle\rho^4\rangle$ analogous to the quartic term in (1) raises the energy of the vertex-edge phase above that of body-centered cubic for all κ.

We have completed the analogous calculation for a cholesteric liquid crystal near the limit $\kappa\to\infty$, and find that for the tensor order parameter even the barrier height to quasicrystal formation is larger than that of the cubic phases composed of wave vectors in octahedral directions. As will be described below, for a scalar order parameter all "triangles" can contribute coherently to lower the free-energy barrier. However, for a tensor order parameter the contributions compete with each other, reducing how effectively the additional triangles can increase the cubic term. While the barrier height may be relevant to the scalar quasicrystal, which appears on rapid cooling, we require a globally stable phase to describe the equilibrium "blue fog." We find that no phase composed of plane waves along vertex and edge directions of an icosahedron is ever stable, with respect to O^5, even if κ is set equal to zero.

We have computed the free energy of textures of the form

$$Q_{ij}(\mathbf{x})=\sum_{|n|=1}^{15}E^n(\hat{\mathbf{d}}_E^n)_i(\hat{\mathbf{d}}_E^n)_j\exp[i(k_E\hat{\mathbf{c}}_E^n\cdot\mathbf{x}+\psi_E^n)]+\sum_{|m|=1}^{6}V^m(\hat{\mathbf{d}}_V^m)_i(\hat{\mathbf{d}}_V^m)_j\exp[i(k_V\hat{\mathbf{c}}_V^m\cdot\mathbf{x}+\psi_V^m)], \tag{4}$$

where $\{\hat{\mathbf{c}}_E^n\}$ are unit vectors parallel to the thirty edges of an icosahedron and $\{\hat{\mathbf{c}}_V^m\}$ are unit vectors pointing from the center to its twelve vertices. The vectors are labeled so the $\hat{\mathbf{c}}^{-n}\equiv-\hat{\mathbf{c}}^n$ and the sum in (4) is over both positive and negative n and m. We use the shorthand notation $\hat{\mathbf{d}}\equiv(\hat{\mathbf{a}}+i\hat{\mathbf{b}})/\sqrt{2}$, where $(\hat{\mathbf{a}},\hat{\mathbf{b}},\hat{\mathbf{c}})$ are right-handed orthonormal triads; reality of $\mathbf{Q}$ is assured by $\hat{\mathbf{d}}^{-n}=[\hat{\mathbf{d}}^n]^*$, $\psi^{-n}=-\psi^n$, $E^{-n}=E^n$, and $V^{-n}=V^n$, with E and V real.

Consider first the infinite-chirality limit in which only edge vectors are included[6d]: $V^m\equiv0$. To compute the stabilizing cubic term, we cube the order parameter (4), take the trace, and spatially average. What remains can be written as a sum over triangles (distinct triples of wave vectors which sum to zero) and the permutations of their sides. The triangle indexed by t is the triple (t_1,t_2,t_3). Trivially summing over the permutations (which all contribute equally) gives

$$\langle\mathrm{Tr}\mathbf{Q}^3\rangle_{EEE}=12\sum_{t=1}^{10}\mathrm{Re}[E^{t_1}E^{t_2}E^{t_3}\exp(i\phi_{EEE}^t)\Pi_{EEE}(t)], \tag{5}$$

where $\Pi(t)\equiv(\hat{\mathbf{d}}^{t_1}\cdot\hat{\mathbf{d}}^{t_2})(\hat{\mathbf{d}}^{t_2}\cdot\hat{\mathbf{d}}^{t_3})(\hat{\mathbf{d}}^{t_3}\cdot\hat{\mathbf{d}}^{t_1})$ and $\phi_{EEE}^t\equiv\psi_E^{t_1}+\psi_E^{t_2}+\psi_E^{t_3}$. The sum over triangles includes one member from each of the ten pairs $[t,-t\equiv(-t_1,-t_2,-t_3)]$ of EEE triangles formed by the edge vectors; we have used $\Pi(-t)=[\Pi(t)]^*$ to sum over negative t.

Similarly, the quartic term simplifies to a sum over quadruples q of wave vectors $(\hat{\mathbf{c}}^{q_1}\hat{\mathbf{c}}^{q_2},\hat{\mathbf{c}}^{q_3},\hat{\mathbf{c}}^{q_4})$ which sum to zero. Note that degenerate quadruples of the form $(\hat{\mathbf{c}}^n,\hat{\mathbf{c}}^n,\hat{\mathbf{c}}^{-n},\hat{\mathbf{c}}^{-n})$ and $(\hat{\mathbf{c}}^n,\hat{\mathbf{c}}^{-n},\hat{\mathbf{c}}^m,\hat{\mathbf{c}}^{-m})$ must be considered, as well as those quadruples which contain no conjugate pairs of wave vectors. We do not discuss the details of these tedious calculations here, but the quartic terms have been computed and included in our evaluation of the free energy (1).

While the assignment of triads $(\hat{\mathbf{a}}_E,\hat{\mathbf{b}}_E,\hat{\mathbf{c}}_E)$ to each wave vector is arbitrary, it is convenient to select triads which respect the rotational symmetry of the icosahedron. We choose $\hat{\mathbf{a}}_E$ directed radially outward from the center of the icosahedron bisecting the edge

$\hat{\mathbf{c}}_E$; $\hat{\mathbf{b}}_E$ is given by $\hat{\mathbf{c}}_E \times \hat{\mathbf{a}}_E$, satisfying $\hat{\mathbf{d}}_{\bar{E}}^{\bar{n}} = (\hat{\mathbf{d}}_E^n)^*$. All ten pairs of triangles are related by symmetry, with

$$\Pi_{EEE} = \tfrac{27}{64} e^{-i\Phi_{EEE}}, \tag{6}$$

where $\Phi_{EEE} = \tan^{-1}(22\sqrt{5}/35) = +54.5690\ldots^\circ$; the cubic term (5) becomes

$$\langle \mathrm{Tr}\mathbf{Q}^3 \rangle_{EEE} = \tfrac{81}{16} \sum_{t=1}^{10} E^{t_1} E^{t_2} E^{t_3} \cos(\phi_{EEE}^t - \Phi_{EEE}). \tag{7}$$

Numerically minimizing the free energy (1) for order parameters of the form (4) with $V^m = 0$ and variational parameters $\{E^n\}$ and $\{\phi_{EEE}^t\}$, we find[14] that the lowest-energy icosahedral phase has equal amplitudes $|E^n|$, and equal triangular phases $\phi_{EEE}^t = 2\pi/5$. This icosahedral phase has higher free energy than the O^5 phase for all temperatures.

In the hope of lowering the energy of the icosahedral phase below that of the cubic phases, we extend the above calculation to include vertex vectors,[12c] which are admitted for large but finite κ. In addition to the ten pairs of *EEE* triangles described above, there are now fifteen pairs of vertex-vertex edge (*VVE*) triangles which contribute to $\langle \mathrm{Tr}\mathbf{Q}^3 \rangle$, one for each pair of edge vectors. We show below that, even if we ignore the extra gradient energy due to the two wave numbers [i.e., use the vertex-edge approximation (4) and set $\kappa = 0$ in (1)], the cubic energy is insufficient to make the vertex-edge phase a global minimum.

It is impossible to define triads $(\hat{\mathbf{a}}_V, \hat{\mathbf{b}}_V, \hat{\mathbf{c}}_V)$ which respect the icosahedral symmetry of the $\{\hat{\mathbf{c}}_V^m\}$, reflecting the impossibility of constructing an order parameter (4) with icosahedral point symmetry and nonzero V^m. Triads *can* be assigned in a way that respects the tetrahedral symmetry of an icosahedron, as follows. Inscribe the icosahedron in a cube, with six of the edge vectors lying along the face bisectors of the six cube faces. Each vertex $\hat{\mathbf{c}}_V^m$ connects to one of the six edge vectors lying in the cube faces, $\hat{\mathbf{c}}_E^n$. Define $\hat{\mathbf{a}}_V^m$ as the unit vector perpendicular to $\hat{\mathbf{c}}_V^m$ and in the plane defined by $\hat{\mathbf{c}}_V^m$ and $\hat{\mathbf{c}}_E^n$; $\hat{\mathbf{b}}_V^m$ is determined by $\hat{\mathbf{c}}_V^m \times \hat{\mathbf{a}}_V^m$.

The fifteen pairs of *VVE* triangles then divide naturally into two types: three pairs whose edge vectors lie in cube faces [*VVE*(1)] and twelve whose edge vectors do not [*VVE*(2)]. We find

$$\Pi_{VVE(\alpha)} = [\cos\theta(1+\sin\theta)/2]^2 e^{-i\Phi_{VVE(\alpha)}}, \tag{8}$$

with $\theta = \tan^{-1}[(\sqrt{5}-1)/2]$, $\Phi_{VVE(1)} = 0°$, and $\Phi_{VVE(2)} = 144°$, yielding

$$\langle \mathrm{Tr}\mathbf{Q}^3 \rangle_{VVE} = 3(1+\sin\theta)^2 \cos^2\theta \sum_{s=1}^{15} V^{s_1} V^{s_2} E^{s_3} \cos(\phi_{VVE}^s - \Phi_{VVE(\alpha(s))}); \tag{9}$$

$\phi_{VVE}^s = \psi_V^{s_1} + \psi_V^{s_2} + \psi_E^{s_3}$ and $\alpha(s)$ is 1 or 2 depending on where the edge vector of triangle s lies.

The order parameter depends on 21 phases, while the free energy can be expressed in terms of the fifteen *VVE* triangular phases (all other relevant phases can be computed from these fifteen); the icosahedral phase therefore exhibits six hydrodynamic modes.[15] The fifteen *VVE* phases can be chosen equal to their optimal phases $\Phi_{VVE(\alpha)}$, maximizing the cubic contribution of each *VVE* triangle. Optimal *VVE* phases correspond to all *EEE* phases equal to 288°, which gives an *EEE* contribution (7) of opposite sign: *The two terms oppose each other.* This is in contrast with the scalar case where all Π are equal to one ($\Phi_{VVE(\alpha)} = \Phi_{EEE} = 0$); choosing all triangular phases equal to zero then optimizes both cubic contributions simultaneously.

Comparing the magnitudes of the dimensionless cubic parameter $\beta = \sqrt{6}\langle \mathrm{Tr}\mathbf{Q}^3 \rangle / \langle \mathrm{Tr}\mathbf{Q}^2 \rangle^{3/2}$, we find $\beta_{\mathrm{icos}}(\kappa = \infty) = 0.720$ and $\beta_{\mathrm{icos}}(\text{vertex-edge}, \kappa = 0) = 1.010$, compared with $\beta_{O^5} = 1.017$. The free-energy barrier to formation is proportional to $1/\beta^2$, so that although the vertex-edge phase has a lower barrier than the one-wave-number phase, the competition between the *EEE* and *VVE* contributions prevents the vertex-edge phase from lowering its barrier below that of the O^5 phase.

We have minimized numerically the free energy (1) (including the local quartic $\langle \mathrm{Tr}\mathbf{Q}^4 \rangle$)[16] in the vertex-edge, $\kappa = 0$ approximation (with $\{E^n\}$, $\{V^m\}$, and $\{\phi_{VVE}^s\}$ variable) and find that, although there are many local minima in the space of vertex-edge textures (4), none has lower energy than O^5. The lowest-energy minimum has *VVE* phases chosen optimally as described above. All these minima have equal amplitudes among the $\{E^n\}$ and $\{V^m\}$ and equal phases for the *EEE*, *VVE*(1), and *VVE*(2) triangles, respectively. Their order parameters have tetrahedral point symmetry at the origin (icosahedral for $V^m = 0$ and $\phi_{EEE}^t = 0$).

For physical κ, the coherence length is comparable to the pitch, corresponding to a shallow $A(k)$. A proper calculation therefore requires many harmonics, including order parameters not of the form (2). While many more triangles utilizing higher harmonics can be formed for the icosahedral case than for the cubic structures, we have seen that for a tensor order parameter, interference between triangular phases may prevent the free energy from taking full advantage of these terms. To compare the cubic and icosahedral phases would require a detailed calculation to assess the effects of the higher-order wave numbers. Furthermore, for κ small, we have moved away from the

weak first-order limit, and the higher-order bulk terms can no longer be ignored; even if we disregard fluctuations, Landau theory as described above is no longer valid.

An alternative approach to the low-chirality regime[8] approximates the unit cells of the cubic blue phases by a lattice with directors $\mathbf{n}$ at each grid point. The directors are relaxed to minimize (1), with $Q_{ij} \equiv n_i n_j - (\frac{1}{3}) n_k n_k \delta_{ij}$. In the quasicrystalline case, the lack of a unit cell hampers such an approach, but we are considering the analogous calculation using the two three-dimensional Penrose tiles.

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Note added.—There are preliminary indications[17] that blue phase II (*not* the fog) in 2-methylbutyl-*p*-[N-(*p*-methoxybenzylidene)-amino] cinnamate may have icosahedral order. In particular, selective reflection along a threefold axis is observed. The threefold axes in our phase can be shown to be equivalent within a phason[15]; since some are true threefold axes, none can exhibit selective reflection. The 2-methylbutyl-*p*-[N-(*p*-methoxybenzylidene)-amino] cinnamate phase therefore cannot have the symmetry of the phase described above.

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