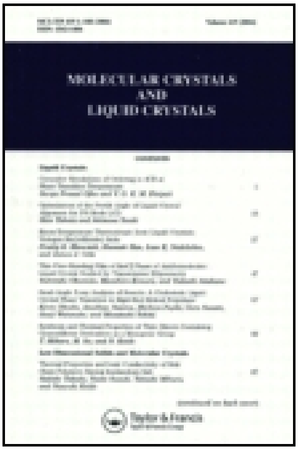

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The Cholesteric Blue Phase: A Progress Report

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The Cholesteric Blue Phase: A Progress Report

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Experimental studies of the blue phase of cholesteric liquid crystals have revealed two cubic phases (BPI and BPII) and a possible amorphous "fog" phase (BPIII). In addition, a number of theoretical models have been proposed which arrive at these structures from the point of view of a Landau theory, a lattice of disclinations, or as an ordered emulsion. We review the present state of knowledge of the blue phase and show the significance of blue phase studies to other problems in condensed matter research.

I. INTRODUCTION

The blue phase,[1] which appears in short pitch cholesteric liquid crystal systems, is currently being intensively investigated. The purpose of this brief review is to bring liquid crystal scientists working in other fields up to date on the main features of blue phase research without getting into too much detail. The material is presented as follows: first, we discuss the unique properties of phase transitions which proceed from a *spatially uniform* upper phase to a *spatially periodic* lower phase and show how the isotropic-cholesteric transition provides a good model of such a system. We then review the main experimental features of the blue phase and show the progress in determining its structure. To explain these results, we next give the general ideas behind three blue phases theories — a Landau theory, a defect theory, and an emulsion theory. Finally, we list a few of the many unsolved questions in blue phase research and suggest some fruitful areas of future study.

II. THE ROLE OF SPATIAL PERIODICITY IN PHASE TRANSITIONS

Many of the phase transitions which have been studied to date involve the transformation of one spatially uniform structure into another. Examples include the liquid-gas, the ferromagnetic-paramagnetic and the nematic-isotropic transitions. A simplest possible Landau free energy for such transitions (here we ignore the details of each system) would take the general form

$$F = \sum_k (a + bk^2)\phi_k^2 \tag{1}$$

where ϕ_k is the Fourier component of the order parameter corresponding to wavevector k, b is a constant, $a = \alpha(T - T^*)$ where T is temperature, and the sum is over all wavevectors of the system. In either phase of this system the free energy is minimized when $\phi_k = 0$ for all $k \neq 0$, that is, when the phases are spatially uniform. It is instructive to plot the location of the minimum of F in k-space; in this case it is just the point at the origin shown in Figure 1a.

Of course, this system also contains thermal fluctuations $\langle\phi_k^2\rangle$ which, for any direction of $\mathbf{k}$, always raise the free energy of the system. These fluctuations are indicated by the arrows in Figure 1a. One can easily calculate the $\langle\phi_k^2\rangle$ from Eq. 1 and then sum them over all k-values to get $\langle\phi(r)^2\rangle$, the mean square fluctuation in $\phi(r)$. The result is

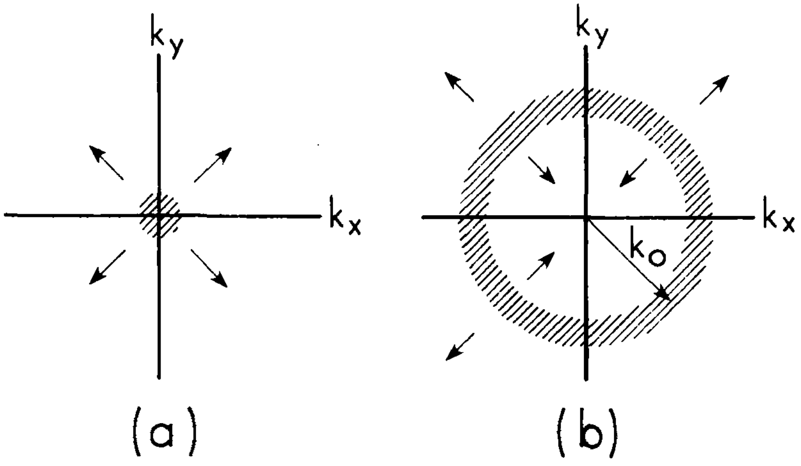

FIGURE 1 Location of the minimum of the free energy in k-space for (a) spatially uniform systems, and (b) systems with a spatially periodic lower phase. In case (a) fluctuations in any direction (arrows) increase the free energy. In case (b) only fluctuations perpendicular to the k-surface increase the energy.

$$\langle\phi(r)^2\rangle = \frac{k_BTV}{(2\pi)^3}\int\frac{d^3k}{a + bk^2} \tag{2}$$

which in the low temperature phase is always smaller than $\langle\phi(r)\rangle^2$ itself. From this we conclude that the fluctuations in $\phi(r)$ do not play a significant role in the phase transition.

Recently Brazovskii[2] has considered systems which condense from a spatially uniform phase to a spatially periodic phase. If the ordering direction is arbitrary, a simplest free energy would take the form

$$F = \sum_k [a + b(k - k_0)^2]\phi_k^2 \tag{3}$$

where k_0 is the wavevector corresponding to the periodic length in the lower phase. Eq. 3 is the same as Eq. 1 with the replacement of $k - k_0$ for k. This free energy is minimized at k_0 rather than at $k = 0$; the minima of F in k-space now lie on a sphere of radius k_0 as shown in Figure 1b. Looking again at the fluctuations in ϕ_k, note that the new topology of the minimal surface has a profound effect on the fluctuation amplitudes. Thus any fluctuations which preserve the length but not the direction of k_0—that is, fluctuations along the surface of the k_0-sphere—do not change the free energy and may be arbitrarily large. Only in a direction perpendicular to the k_0-sphere will the free energy be raised by a fluctuation. Calculating $\langle\phi_k^2\rangle$ as before and summing to get $\langle\phi(r)^2\rangle$, we find

$$\langle\phi(r)^2\rangle = \frac{k_BTV}{(2\pi)^3}\int\frac{d^3k}{a + b(k - k_0)^2} \tag{4}$$

This integral behaves quite differently from Eq. 2; in particular, it diverges as the temperature T^* is approached. Brazovskii[2] has shown that the net effect of the large fluctuations is to disrupt the onset of the lower phase and, through a renormalization of the coefficient a in Eq. 3, to move the phase transition temperature from T^* to $T = 0$. Given such a situation, the free energy of the upper phase must now remain lower than that of other competing structures if it is to survive. If, however, a new structure with a lower free energy does appear, a first order phase transition to the new structure occurs. Note that this transition has nothing to do with any cubic term in the free energy. It is purely a consequence of the presence of spatial periodicity in the lower phase.

The above arguments are entirely general and have been applied to a number of problems such as the nematic-smectic C transition[3] and the Rayleigh-Benard instability in fluids.[4] Their application to the isotropic-cholesteric transition[5] leads to the prediction that this transition must have fundamentally different behavior from the isotropic-nematic transition, despite the prevailing view at one time that nematics and cholesterics were thermodynamically identical.

We now know from experiment that cholesteric systems with short pitch generally exhibit an intermediate phase — the blue phase — at the isotropic transition. Since blue phases do not occur in nematics or even in long pitch cholesterics, it is evident that periodicity in the lower phase can indeed have a profound effect on a phase transition. The Brazovskii scenario gives a theoretical basis to this experimental fact.

III. EXPERIMENTAL FEATURES OF THE BLUE PHASE

Although the blue phase has been noticed for years, not until recently has it been accepted as a legitimate thermodynamic phase. Figure 2 shows differential scanning calorimetry measurements[6] of cholesteryl myristate (CM) and cholesteryl nonanoate (CN) as one passes from the cholesteric to the isotropic phase. Two peaks and a shoulder appear; these mark the (first order) phase boundaries between the cholesteric, isotropic, and *two* intermediate blue phases (BPI and BPII). Actually a third amorphous phase called the "fog phase" has been observed by optical microscopy between the BPII and isotropic phases.[7] Since, however, no manifestation of this has been detected thermally it is not clear whether the "fog" represents a true thermodynamic phase.

Additional evidence appears in optical reflection studies. Just as Bragg scattering of X-rays is caused by the lattice periodicities of an ordinary crystal, the long wavelength periodicities of a liquid crystal can cause Bragg scattering (selective reflection) of visible light. Thus periodicity in the cholesteric phase causes the well known cholesteric selective reflection. As the temperature of the cholesteric is increased the cholesteric phase transforms into BPI, which exhibits several reflections, each decreasing at the same rate with increasing temperature. At the BPI-BPII transition a discrete wavelength jump occurs and BPII exhibits a similar set of wavelengths all shifted with respect to those in BPI.

Figure 3 is a graph of this wavelength variation in mixtures of chiral CB15 and nematic E9[8] corrected for refractive index dispersion.[9] The ratios of wavelengths ($1 : 1/\sqrt{2} : 1/\sqrt{3} : \ldots$) in both BPI and BPII are just those for Bragg scattering from either a simple cubic (SC) or a body centered cubic (BCC) lattice. We conclude that the blue phase consists of some kind of a cubic arrangement of the liquid crystalline order.

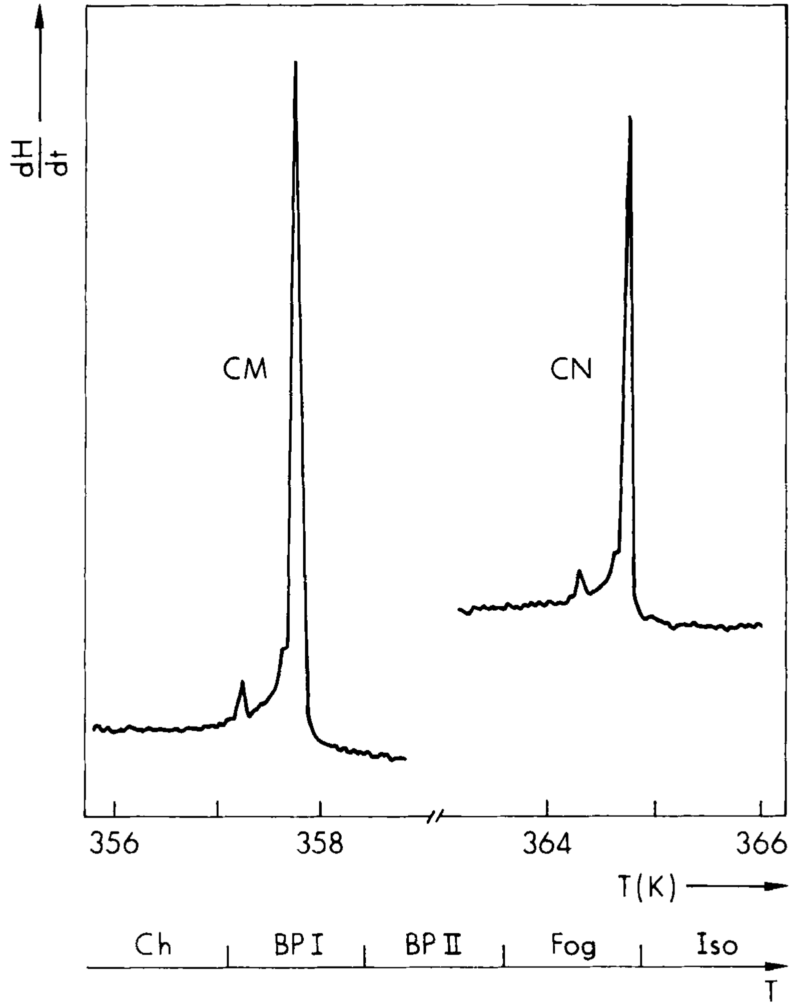

FIGURE 2 DSC traces of cholesteryl myristate (CM) and cholesteryl nonanoate (CN). The sequence of phases shown includes an optically observed "fog" phase which is not detected by thermal measurements. After Bergmann and Stegemeyer, Ref. 6.

Further elucidation of the lattice symmetry consists of determining the space group, within the BCC or SC framework, of the crystal lattice. Hornreich and Shtrikman[10] have extended the International Crystallographic X-ray tables to include the polarization behavior of chiral cubic space groups, and these results have been used to interpret the measured

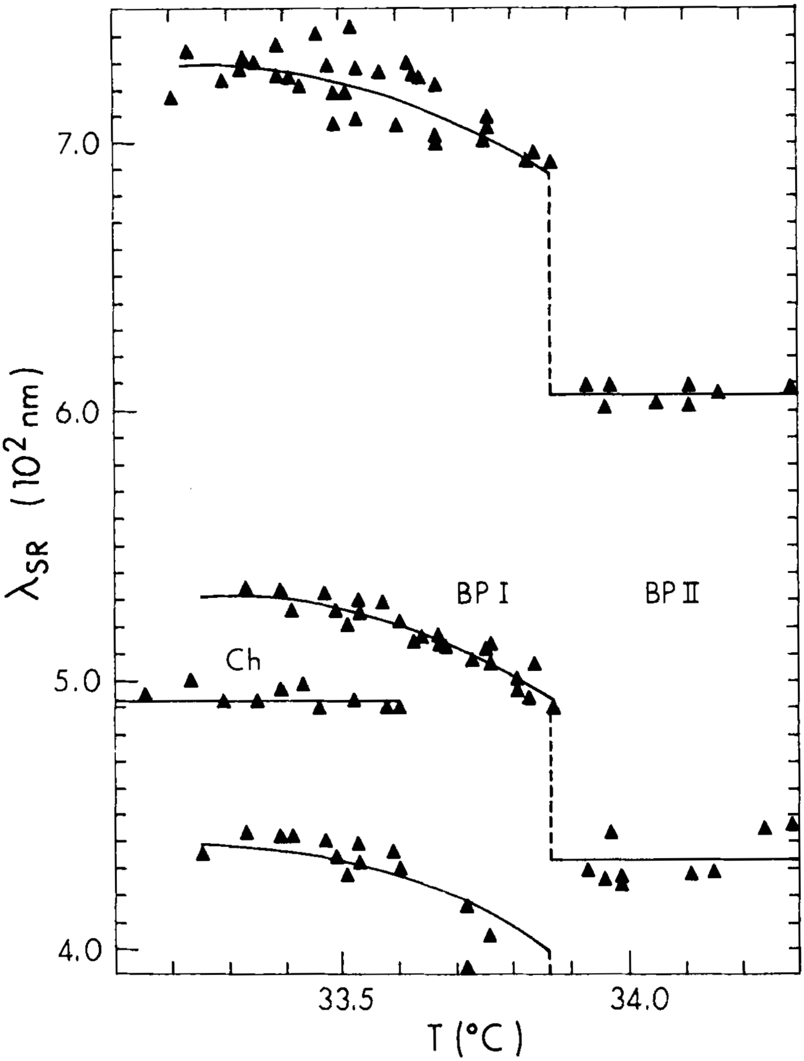

FIGURE 3 Selective reflection wavelengths λ_{SR} for a CB15–E9 mixture, corrected for index of refraction dispersion. The dashed lines show the BPII–BPI wavelength jump. Note that BPI may be supercooled into the Ch region. After Johnson, Flack, and Crooker, Ref. 9.

polarizations of the Bragg lines. Thus in CB15-E9 mixtures, all the lines observed so far (4 lines in BPI, 3 lines in BPII have reflected circularly polarized light.[11] The conclusion is that BPI and BPII can only have the symmetry of the BCC 0^8 ($I4_132$), T^3($I23$), or T^5($I2_13$) space groups. Within the framework of Hornreich and Shtrikman's tables these results are unequivocal; however, other workers have argued that BPII in other materials is SC from the appearance of novel square platelet textures.[7,12] Measurements of the polarization of light scattered at an angle to the normal (the above measurements were all in back reflection) should provide additional information; these are already underway.[13]

In summary, the blue phase is seen to consist of two cubic phases and a possible amorphous phase. The allowed symmetries of the cubic phases have been narrowed down, but are not yet known conclusively. Proposals for the molecular structure of these phases do exist, as we shall see below, but have yet to be confirmed experimentally.

IV. THEORIES OF THE BLUE PHASE

The questions of first, how a fluid with cubic symmetry could possibly occur, and second, what its microscopic structure might be are highly interesting and certainly unique in condensed matter physics. There are, in fact, three theories for the blue phase, the main ideas of which we shall present below.

A. Landau theory

A Landau theory for the isotropic-blue phase transition has been proposed by Brazovskii, *et al.*[5] and by Hornreich and Shtrikman (HS).[14,15] Like all Landau theories, it consists of first writing down an order parameter containing the symmetries of the upper and lower phase, then constructing a Landau free energy from the lowest powers of the order parameter allowed by symmetry, and finally minimizing the free energy to obtain the structure. In the HS theory, the order parameter $\phi(r)$ is a tensor closely related to the dielectric tensor. It has the form (where we have dropped the tensor subscripts for simplicity):

$$\phi(r) = \sum_{\mathbf{q},m} [\epsilon_{\mathbf{q},m} Y_2^m(\theta, \phi)] e^{i\mathbf{q}\cdot\mathbf{r}} \tag{5}$$

Here $\phi(r)$ is a sum of Fourier components having wavevector $\mathbf{q}$ and coefficients given by the quantities in brackets. There are two parts to these coefficients. The Y_2^m is a spherical harmonic which contains the *director symmetry* of that particular Fourier component. Thus for any particular $\mathbf{q}$ vector there may be as many as five different waves of different symmetry corresponding to the different m values ($m = 0, \pm 1, \pm 2$) of the spherical harmonic Y_2^m. The *amplitude* of each Fourier component is given by the number $\epsilon_{q,m}$. The sum over **q** includes all those wavevectors allowed by the symmetry of the system. Thus in the isotropic phase all **q**-fluctuations are allowed, while in the blue phase only those **q**'s corresponding to the reciprocal lattice vectors (*hkl*) of the cubic structure appear.

HS next utilizes the Landau free energy of de Gennes[16] and Brozovskii,[5] which contain the lowest significant powers of $\phi(r)$ and the gradients compatible with the symmetry of the system. Very schematically, this has the form (again dropping tensor subscripts)

$$F = \int [a\phi^2 + c(\nabla\phi)^2 - d\phi\nabla \times \phi + 0(\phi^3, \phi^4)]\, d^3r \qquad (6)$$

where $a = \alpha(T - T^*)$ as usual and c and d are constants. The $(\nabla\phi)^2$ term represents terms quadratic in the gradients; it and the ϕ^2 term alone would give the quadratic part of F the form of Eq. 1. In materials without an inversion center, however, the chiral term represented by $\phi\nabla \times \phi$ is also allowed. When added to the previous two terms, the form of the quadratic part of F is changed from that of Eq. 1 to that of Eq. 3, which exhibits all the unusual behavior discussed in Section II above.

The remaining task is to put the order parameter in the free energy expression and minimize. This can be computationally tedious, however, unless one arbitrarily limits the number of (*hkl*) reciprocal lattice vectors used to describe a cubic lower phase. Thus, if the only nonzero $\varepsilon_{hkl,m}$ coefficients allowed are those lying along the various BCC 110 directions, HS find that a structure with 0^5 space group symmetry appears between the isotropic and cholesteric phases. Unfortunately, this structure doesn't agree with the observed polarization data (so far) so by adding other (*hkl*) vectors HS can derive more compatible BCC structures such as 0^8, T^3, and T^5. They can also derive some SC groups and have generated phase diagrams such as that shown in Figure 4.[15] Figures such as these show how the various blue phases can be created by increasing chirality.

To date the HS theory has shown that a cubic intermediate phase is theoretically attainable and many of the experimentally observed features can be explained. On the other hand their phase diagrams do not yet look like the experimental phase diagrams on chiral-racemic mixtures. This is not entirely the theory's fault since more experimental work is certainly in order; however, the addition of a larger set of reciprocal lattice vectors to the lower phase (as is observed experimentally), although algebraically messy, may be a necessary ingredient.

B. Defect theory

Defect theories of melting have been around for a while[17] but the blue phase defect theory of Meiboom, Sethna, Anderson, and Brinkman (MSAB)[18] is

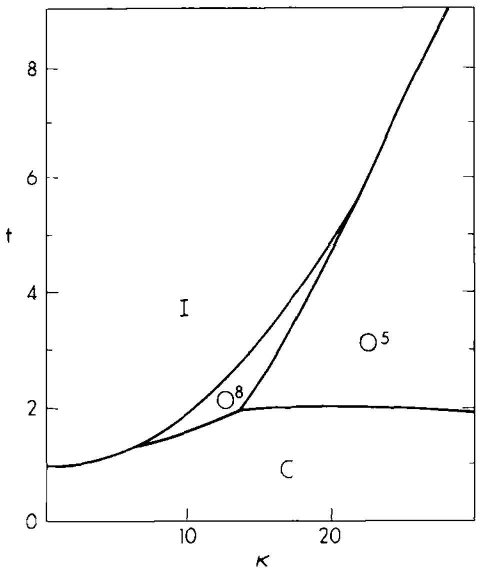

FIGURE 4 Representative phase diagram calculated from the Landau theory. The normalized ordinate t is proportional to $T - T^*$ as described in the text. The normalized abscissa K is proportional to the chirality $1/P$, where P is the pitch of the cholesteric phase. This particular diagram includes $[hkl;m]$ order parameter components corresponding to variations of [110;2] and [200;2]. After Hornreich and Shtrikman, Ref. 15.

an interesting variation on this theme. In the usual three dimensional defect melting theory, the free energy F_{def} for production of a defect out of a perfect crystal is calculated and is shown to be larger than zero below some temperature T_c. Thus, below T_c, the material is a defectless solid. At T_c, however, the free energy of a defect passes smoothly through zero and becomes negative (Figure 5a). For $T > T_c$, therefore, defects become energetically favorable, a catastrophic onset of defects occurs, and the resulting liquid is taken to be a solid saturated with defects.

Carrying this idea over into the blue phase problem, one must use disclinations—topological singularities in the director field of a liquid crystal—instead of crystal defects. Calculating the free energy of a disclination, we again find that it is greater than zero below the cholesteric-blue phase transition temperature T_1 (Figure 5b). Below T_1 we then expect a disclination-free cholesteric. At T_1, the free energy again passes through zero, but only for a regular lattice of disclination lines. Thus the blue phase is envisioned to be a regular network of disclination lines, possessing the same sorts of space group symmetries allowed for crystals.

Although it is not part of the MSAB theory, one can envision a higher temperature T_2 where the free energy of a *lattice* of disclinations becomes

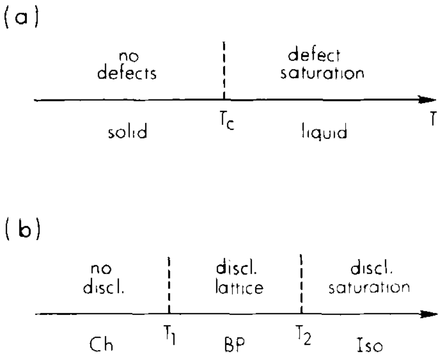

FIGURE 5 (a) The solid-liquid transition as caused by a catastrophic onset of defects at T_c. The free energy for formation of defects is positive for $T < T_c$ and negative for $T > T_c$. (b) The Ch-BP-Iso transitions as caused by the sudden onset of a disclination lattice at T_1 followed by a saturation of disclinations at T_2.

unstable with respect to that for a random collection of disclinations. At this point a total "melting" of the blue phase to the isotropic phase would occur. A nice feature of this model is that it has allowed us to separate the usual highly discontinuous one-step melting process into two progressive transitions. Each is still discontinuous but, speaking loosely, perhaps not so much as before, so that a theoretical treatment of each step is mathematically more tractable. In any case the defect theory is attractive since it seems difficult to fit together any continuous arrangement of long molecules possessing both cubic and chiral symmetry without singularities *somewhere*. The MSAB theory makes these singularities and a consideration of their energies mandatory.

The calculation of the free energy of a lattice of defects is, in general, a difficult task. After first intelligently guessing at a possible structure, the free energy F_{discl} is found to consist of four additive contributions. Figure 6 shows an $m = -\frac{1}{2}$ disclination line in cross-section. It consists of an isotropic cylindrical core of radius R_c surrounded by a distorted director field. The first contribution to F_{discl} is F_{el}, the Frank elastic energy of the distorted director field which, if integrated over a region from an inner radius R_c to some outer cutoff radius R_0 takes the form $F_{el} \sim \log(R_0/R_c)$. The next term F_{surf} comes from additional elastic terms in the derivation of the Frank elastic energy which now survive due to the presence of the disclination. $F_{\text{surf}} \sim -K$ where K is a Frank elastic constant; notice that this is the term which tends to pull the free energy below zero. The core term F_{core} is the energy to convert an essentially nematic core into isotropic material; $F_{\text{core}} \sim (T_2 - T)R^2$ where T_2 is the blue phase–isotropic transition

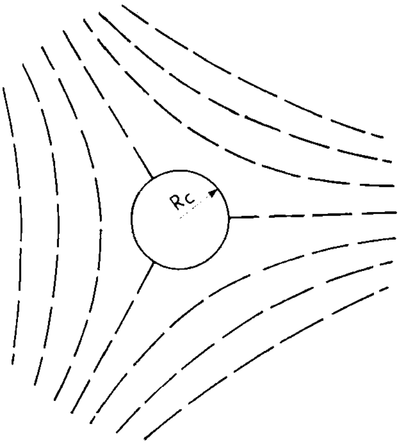

FIGURE 6 Cross-sectional view of an $m = -\frac{1}{2}$ disclination. Disclination consists of an isotropic core of radius R surrounded by a distorted director field.

temperature previously defined. Finally there is a surface tension term $F_{\rm int} \sim R_c$ representing the interfacial energy at the surface of the core.

Putting all these terms into the model, it is possible, using prodigious amount of computer time, to show that a number of cubic disclination lattices, including those with SC 0^2 and BCC 0^5 and 0^8 space group symmetries, have lower free energies than that of the cholesteric phase. The appearance of the unit cell for each of these symmetries (including two possibilities for 0^8) is shown in Figure 7.[19] Actual structures may be derived in principal by this method; however, the results are sensitive to specific material parameters which are not always known. Nevertheless the results are encouraging so far.

Although the Landau and defect theories seem at first quite different, the results have many similarities. In particular they both incorporate "double twist", i.e. if the director at some point lies along the z axis, it tends to rotate spatially about *any* axis lying in the x–y plane. Both theories fit such structures together three dimensionally by allowing the order parameter to go to zero at various locations—either suddenly as in the defect theory or gradually as in the case of the Landau theory. The former requires an order parameter with many spatial Fourier harmonics, while in the latter case only a few or even only one of the possible Fourier components is sufficient.

Thus the main difference between the Landau and defect theories may just be the number of Fourier components included in the order parameter rather than any truly fundamental difference in the structure.

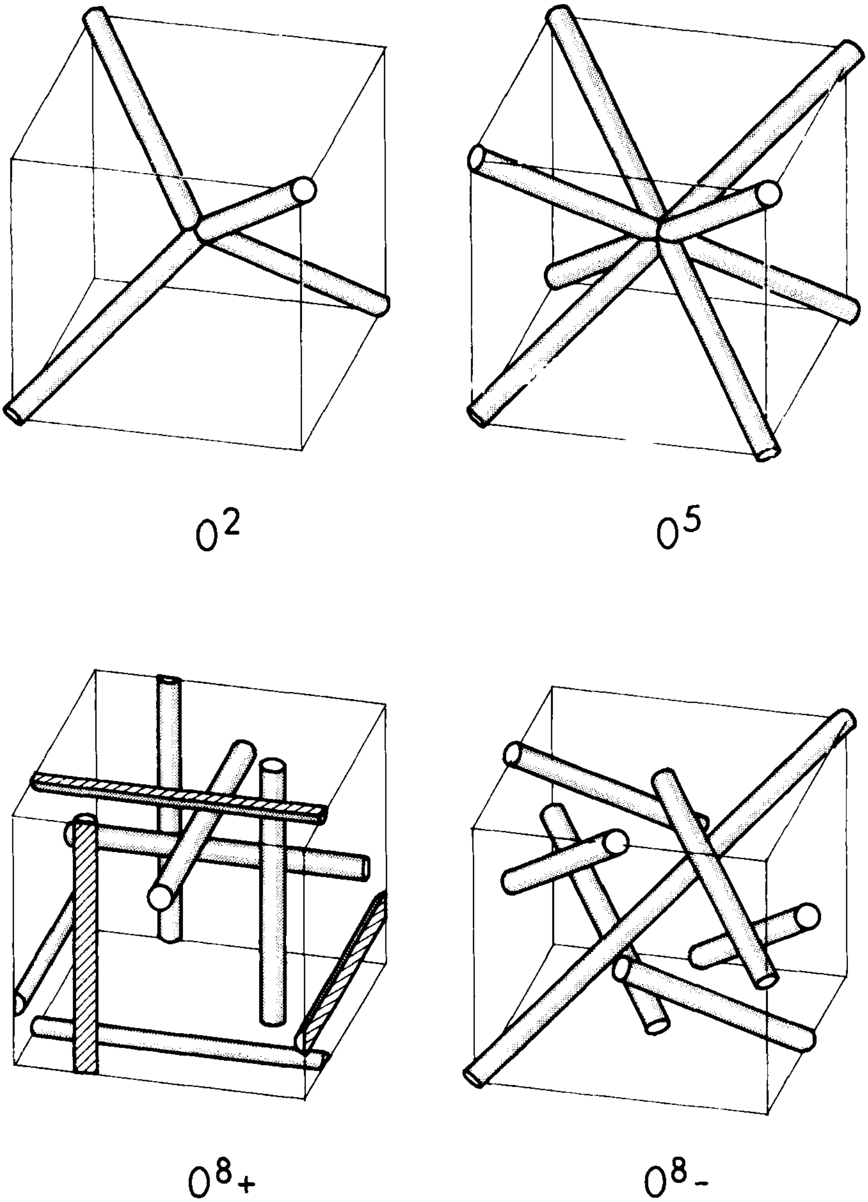

FIGURE 7 Examples of cubic unit cells for lattices of disclinations of various space group symmetries. 0^2 is simple cubic while 0^5, 0^8+ and 0^8- are body centered cubic. 0^8+ and 0^8- are two versions of the 0^8 space group symmetry. After Berreman, Ref. 19.

C. The emulsion theory

The emulsion theory, by Finn and Cladis,[20] is an attempt to explain the blue phase as an emulsion of cholesteric spheres in a background of either nematic or isotropic material. The basic ideas are illustrated in Figure 8. At high temperature the isotropic phase has the usual uniform disordered structure. As the temperature is lowered there appear small droplets of cholesteric order randomly dispersed throughout the isotropic background; this is just the fog phase. As the temperature is lowered further, the droplets grow larger and eventually order into a cubic pattern to form BPII. Finally, at a still lower temperature the emulsion "turns itself inside out", the cholesteric spheres touching and forming the backgound material with small isotropic blobs in the interstices.

A free energy argument can be constructed for this model and it can be shown that stable emulsions occur over a temperature range of about .2°C for pitches below 500 nm as observed. On the other hand, there are still features of the model to be worked out, not the least of which is how an emulsion of one phase in another can occur in a pure material.

IV. FUTURE RESEARCH

Within the context of condensed matter theory in general, the greatest utility of the blue phase is in whether it demonstrates the validity of Brazovskii's ideas on transitions from a uniform to a periodic phase. By studying chiral-racemic mixtures of the same materials, the periodic length

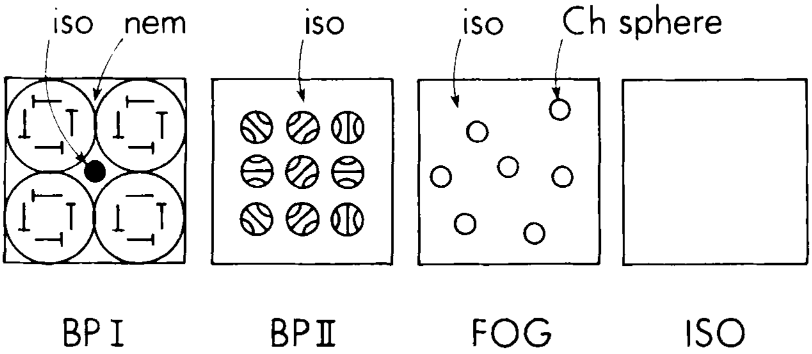

FIGURE 8 Pictorial representation of the blue phases in the emulsion theory. See text for explanation.

alone can be varied and the resulting behavior studied. The behavior of various quantities at the Iso–BP transition is particularly important insofar as they can be related directly to the Brazovskii theory. Also relevant is the understanding of the Ch–BP–Iso system phase diagrams. These should display the blue phase transition temperatures as a function of the *chirality*, which is assumed by the Brazovskii theory to be the relevant independent thermodynamic variable. As mentioned earlier, both improved experiment and theory are necessary here.

An important question concerning the blue phase itself is a general agreement on the nature of its properties. What are the universal features of the blue phase, if any? Are two blue phases always observed? Does the fog phase always exist? Will the structures of all BPI's, for example, be the same? To date we don't really know.

Determination of the various BP structures is still important. So far the possible symmetries have been limited to a few choices, and models are producing detailed director configurations. We now need to match these models to experiments and determine which ones are correct. Useful experimental techniques here are the optical probes: the polarizations and relative intensities of the Bragg reflections, and possibly the rotatory power. In particular more data and theory is needed on the fog phase, the nature of which is still quite unknown.

The fact that the blue phase may be a regular lattice of disclinations makes this structure rather unique in condensed matter physics. It is known, for example, that the blue phase viscosity as measured by simple falling ball experiments is orders of magnitude larger than that of either the cholesteric or isotropic.[21] Is a lattice of disclinations liquid or solid? What, in particular, are its viscoelastic properties? More detailed experiments and a general theory of the viscoelastic properties of a regular lattice of disclinations are necessary here.

Biaxiality seems to play an important role in determining the Ch–BP transition temperature.[22] Does the blue phase always result when the biaxiality rises to a certain threshold value? If so, what is the underlying mechanism?

Finally, there are a number of questions that are just now beginning to be explored. First, what are the effects of electric and magnetic fields on the blue phase?[20] Second, is the blue phase only limited to thermotropic materials or can it occur in lyotropics and polymeric liquid crystals? The former problem may lead to new liquid crystal applications. The latter may lead to new and interesting liquid crystal materials.

Blue phase research is still in its infancy. As the field develops, new insights will be gained not only on the nature of liquid crystals themselves, but also on some of the larger questions of condensed matter physics. In short, the best is yet to come.

Acknowledgment

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