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Peter J. Collings
Swarthmore College, pcollin1@swarthmore.edu

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Point and Line Disclinations In Models of the Blue Phases

PETER J. COLLINGS*

Department of Physics & Astronomy, Swarthmore College, Swarthmore, PA 19081

The model of the liquid crystalline blue phases proposed by Alfred Saupe in 1969 is examined in light of the recent, successful theoretical models for these phases. Such an analysis demonstrates that Saupe's model captures all of the important features of the recent models, differing only in the density of line disclinations. The fact that Saupe proposed this model over ten years before the more recent work and without the benefit of a significant amout of new experimental evidence is testimony for his keen physical insight. Such models continue to be useful as researchers direct their attention to the less understood third blue phase and the transition between it and the isotropic phase.

Keywords: Liquid crystal; blue phases; disclinations; defects

INTRODUCTION

Observations of the blue phases are as old as liquid crystal research itself. Reinitzer noted the pale blue color of the blue phases in cholesteryl benzoate [1] and Lehmann proposed that this color was due to a thermodynamically stable phase between the chiral nematic and isotropic phases[2]. Friedel mentions it only briefly in his review [3] and no further discussion occurs in the literature until Gray again describes the phenomenon in 1956 [4]. Microscopic observations of the platelet structure of the blue phases brought renewed interest, and for the next fifteen years groups in the United Kingdom, the Soviet Union, the United States, and Germany reported new results [5]. Whether the blue phases were a distinct, thermodynamically stable phase or simply a modification of the chiral nematic phase remained an open question during this period. This changed, however, due to the latent heat and density experiments of Armitage and Price [6],

*Tel: (215) 328-8258; E-mail: Collings@campus.swarthmore.edu

which demonstrated that these were stable phases different from the chiral nematic phase, and due to the optical experiments of Bergmann and Stegemeyer [7], which revealed that there were two distinct blue phases. The last fifteen years has witnessed a flurry of activity in blue phase research, the highlights being (1) the observation of optical Bragg scattering from the blue phases [8], (2) the realization that a third blue phase exists which is quite different from the other two [9], (3) the description of a theoretical structure for two of the blue phases [10], (4) the identification of new phases only stable in an electric field [11], and (5) the discovery of a blue phase – isotropic phase critical point in systems of extremely high chirality [12].

One contribution of immense importance has been omitted from this short historical sketch, and that is an article by Alfred Saupe appearing in 1969 [13]. In tune with other work at the time, Saupe confirmed prior observations on the lack of birefringence of the blue phases, additionally noting that (1) the optical activity is higher than in the isotropic phase, (2) the turbidity is much less than in the chiral nematic phase but more than in the isotropic phase, and (3) birefringence sometimes results when the blue phases are disturbed. But unlike any worker before him, Saupe offered a model for the blue phases over a decade before the definitive theoretical work on the structure of these phases. An examination of Saupe's model in light of all that is now known about the blue phases is a worthwhile exercise, not only because it reveals interesting features of the disclinations present in the blue phases, but also because it testifies to the brilliance of what Saupe proposed at a time when there was precious little evidence to guide his thinking.

SAUPE'S MODEL

In order to explain the lack of birefringence and the presence of optical activity, Saupe knew a chiral, deformable structure of high symmetry was necessary. He therefore put together a three dimensional body-centered cubic structure based on a cylindrically symmetric local director configuration in which the preferred orientation rotates along all directions perpendicular to the symmetry axis. Both of these choices (a cubic structure and a local "double twist" configuration) were right on target, as borne out by the fact that all later models of the blue phases incorporated both of these features.

The local director configuration for a double twist cylinder is shown in Figure 1. In Saupe's model, the director twisted by 90°; in recent models the twist is either 45° or 54.7°, depending on the structure of the phase. Saupe's model using these double twist cylinders is shown in Figure 2. While it was easy to imagine a structure in which parallel double twist cylinders form a square lattice (accompanied by a square lattice of line disclinations of strength −1/2), Saupe knew that cubic symmetry was necessary. He achieved this by placing three sets of such cylinders parallel to the three Cartesian axes and forcing them to deform to produce three additional sets of such cylinders displaced from the first set along the body diagonals. He thought that such a structure possessed point disclinations, and he built up a three dimensional phase by having the structure repeat itself along the three Cartesian axes. The result was the structure shown in Figure 2, in which the point disclinations occupy the sites of a body–centered cubic lattice.

The reason Saupe's model was not utilized by workers in the field was that the orientation of the director was not specified between the point disclinations. Yet armed with some of the more recent work into such structures, it is not difficult to gain some insight into what Saupe's model actually entails. For example, as outlined by Meiboom, Sammon, and Brinkman [14], one can search for line disclinations by choosing closed

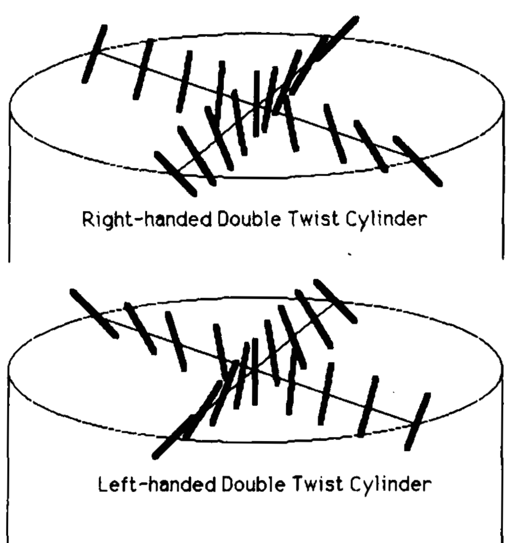

FIGURE 1 Right-and left-handed double twist cylinders (after Ref. 17). The director is parallel to the cylinder axis in the center of the cylinder and rotates about any radius. There is no variation in the director parallel to the cylinder axis. In these examples the director at the edge of the cylinder makes an angle of 45° with the cylinder axis.

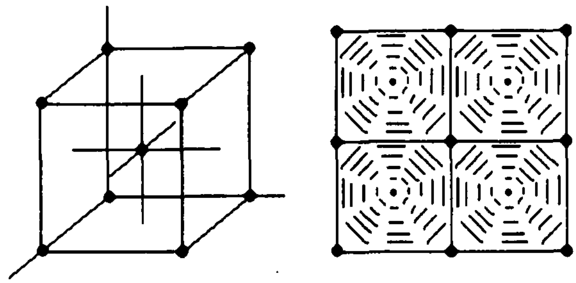

FIGURE 2 Saupe's model of the blue phases (after Ref. 13). The three dimensional structure is shown in (a) and a cross-section parallel to one of the orthogonal axes is shown in (b). The dots represent points where the director is undefined.

paths in the structure and seeing whether the director rotates by some multiple of π for a trip around the closed path. This can be done with the help of Figure 3, where the director orientation is shown at various points in the unit cell. If one forms a loop around an edge or face diagonal of the unit cell (away from the corners of the unit cell), moving around these loops results in no change in the orientation of the director. However, if one forms a loop around a body diagonal of the unit cell (away from the center and corners of the unit cell), moving around the loop results in a director change of π. Thus line disclinations of strength $-1/2$ must run along the four body diagonals, so Saupe's model contains line disclinations and not point disclinations [14]. The places where Saupe thought point disclinations

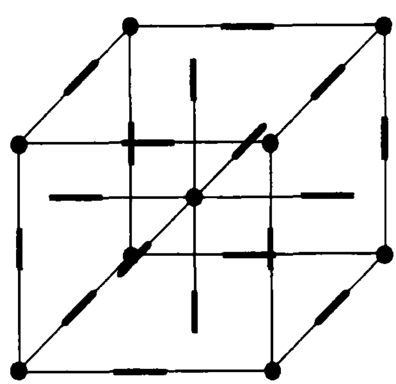

FIGURE 3 The director orientation at various points in Saupe's model of the blue phases. The dots represent points where the director is undefined.

occurred, however, represent locations where four line disclinations meet. These line disclinations are shown in Figure 4(a). The fact that the model contains line and not point disclinations actually makes sense, as point disclinations are usually found on surfaces and in restricted geometries rather than in bulk samples. More recent models for the blue phases, at least in the low chirality limit, also possess line disclinations of strength $-1/2$.

A SIMPLE CUBIC MODEL

A recent model with simple cubic symmetry seems to describe the second blue phase [10]. Like Saupe's model, it is composed of double twist cylinders, but these involve a twist of only 45°. The double twist cylinders are placed together in a simple cubic lattice, with cylinders running along three orthogonal directions. This structure is shown in Figure 5, where it can be seen that, just as in Saupe's model, the director is continuous where two double twist cylinders touch. To uncover where line disclinations are present, the same technique of moving around a closed path to see if the director rotates by some multiple of π can be used. For this structure, line disclinations are found only along four half-diagonals as shown in Figure 4(b), and the points where two line disclinations meet form a simple cubic lattice [14]. Thus the paramount difference between this model and Saupe's model is that the density of line disclinations and the number of line disclinations that meet at points in the structure are both reduced by a factor of two.

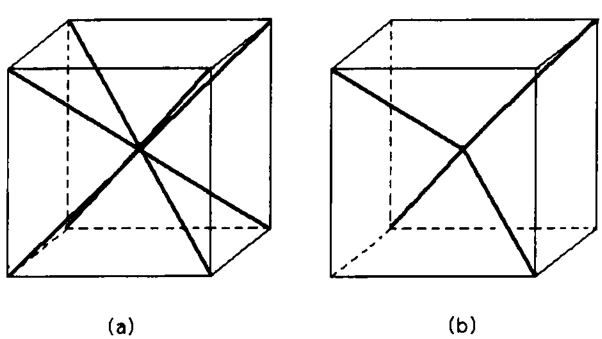

FIGURE 4 The location of line disclinations in the unit cell of two models of the blue phase (after Ref. 14). (a) Saupe's model, and (b) a simple cubic model.

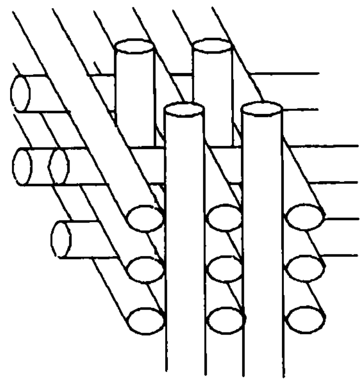

FIGURE 5 The arrangement of double twist cylinders in a simple cubic model of the blue phases (after Ref. 14). The line disclinations connecting points where three cylinders meet in this structure are shown in Figure 4(b).

DISCUSSION

This analysis demonstrates just how close Alfred Saupe came to producing a successful model for the blue phases long before much was known about them. His model incorporated double twist local order in a cubic structure with a lattice spacing on the order of the chiral nematic pitch. The only thing lacking was a free energy calculation, an exercise that would have required a careful analysis of the director configuration and probably would have led to alternative models.

These models are still important as new observations are made on the blue phases. For example, what is the structure of the third blue phase (BP_{III}), the one that shows no long range cubic symmetry? One possibility is that it is a phase in which the cubic network of line disclinations has melted, creating a random network of inter-connected line disclinations [15]. The symmetry of such a structure is isotropic, suggesting that it can convert to the isotropic liquid phase without a phase transition. This has been confirmed very recently by both theoretical and experimental work showing that the BP_{III}-isotropic transition line ends at a critical point in the temperature—chirality plane [12,16].

Thus traditional ideas concerning the structures of both BP_{III} and the isotropic phase must be revised. Again, the work with models similar to Saupe's is useful. Calculations show that in order for these structures to have a free energy lower than the chiral nematic phase, the amplitude of the orientational order parameter must vary throughout the unit cell, vanishing at the line disclinations. Thus it is more proper to think of BP_{III} as a random arrangement of more orientationally ordered regions separated by less orientationally ordered regions. The proximity of an extremely weak phase transition also indicates that spatial and temporal fluctuations are large. Once this is realized, it is clear that the usual description of the isotropic phase near a phase transition to a liquid crystal phase, namely a spatially and temporally fluctuating random collection of regions of short range orientaion order separated by regions of no orientational order, is really the same description as for BP_{III}. With this in mind, the BP_{III} and isotropic phases are identical from a symmetry standpoint (just as the liquid and gas phases possess the same symmetry), and therefore a property that is non-zero in both phases (like the density in the liquid-gas system) should be useful in defining an order parameter. Very recent theoretical work has chosen an order parameter related to the optical activity and finds that, depending on the chirality of the system, it can change discontinuously at a phase transition or continuously as one phase converts to the other [16]. Such a critical point has been found experimentally [12].

CONCLUSION

Although Alfred Saupe contributed only a few ideas to research on the blue phases, one of his ideas captured most of the important aspects of the first successful theoretical models of these phases. The fact that his contributions came over ten years earlier testifies to his great creativity and deep insight.

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