



Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

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N. V. Madhusudana^a & R. Pratibha^a

^a Raman Research Institute, Bangalore, 560080, India

Version of record first published: 13 Dec 2006.

To cite this article: N. V. Madhusudana & R. Pratibha (1983): Studies on High Strength Defects in Nematic Liquid Crystals?, *Molecular Crystals and Liquid Crystals*, 103:1-4, 31-47

To link to this article: <http://dx.doi.org/10.1080/00268948308071038>

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Studies on High Strength Defects in Nematic Liquid Crystals†

N. V. MADHUSUDANA and R. PRATIBHA

Raman Research Institute, Bangalore 560080, India

(Received February 24, 1983; in final form May 24, 1983)

We recently reported the discovery of stable disclinations of strength $\pm \frac{3}{2}$ and ± 2 in mixtures of several nematogenic compounds with a non-mesomorphic compound having plate-like molecules. We describe here further detailed observations on these defects, particularly on pairs of high strength disclinations.

INTRODUCTION

The schlieren textures of nematic liquid crystals have been studied ever since the discovery of mesophases.^{1,2} For this purpose a sufficiently thin sample ($\sim 20 \mu$ or less) of the nematic sample is taken between a slide and a cover slip which have not been given any special surface treatment. When viewed under a polarising microscope, such a sample usually has several points from which two or four dark brushes are seen to emerge. The points are usually line defects viewed end-on and are called disclinations³ of strength $S = \frac{1}{2}$ and 1 respectively.^{4,5} The sign of the disclination is taken to be positive if the brushes rotate in the same direction as that of the crossed polarisers and negative otherwise.

The director \mathbf{n} changes its orientation continuously around a disclination. This curvature of the director costs elastic energy. Assuming to a first approximation that the elastic constants for splay (K_{11}), twist (K_{22}) and bend (K_{33}) are equal, it is easy to obtain solutions to the director field around disclinations. If Ψ is the angle made by \mathbf{n}

†Presented at the Ninth International Liquid Crystal Conference, Bangalore, December 1982.

with the X -axis, and α the angle made by the radius vector connecting the given point to the centre of the defect with the X -axis, and assuming that \mathbf{n} is confined to the XY plane the solutions take the form

$$\Psi = S\alpha + C \quad (1)$$

where C is a constant.³ The director fields around defects of various strengths are shown schematically in Figure 1. In the present approximation, Ψ becomes multiple valued at the origin and hence leads to a singularity at that point. Assuming that a 'core' region extends from the origin to r_c , the energy per unit length of an isolated disclination line is given by⁶

$$W = W_c + \pi K S^2 \ln(R/r_c) \quad (2)$$

where W_c arises from the core region and R is the size of the sample. It is clear that for a given size R , the elastic energy carried by a defect is proportional to S^2 . This means that all defects of strength $|S| > \frac{1}{2}$ should spontaneously break up to form a number of defects of strength $\pm \frac{1}{2}$ only. However defects of strength ± 1 are very frequently seen and are indeed quite stable. This problem was resolved by Meyer⁷ and Cladis and Kleman⁸ who noted that if the director was allowed to collapse in the third dimension (over a radius R_0) such that it points along the Z -axis at the origin, the energies of defects of strength ± 1 become independent of R_0 and are given by

$$W = 3\pi K \quad \text{for } S = +1$$

and

$$W = \pi K \quad \text{for } S = -1 \quad (3)$$

in the one constant approximation. (In deriving the above relations, the surface contributions to the elastic energy have been ignored.) A simple calculation now shows that defects of strength ± 1 carry lower energy than pairs of defects of strength $\pm \frac{1}{2}$, explaining the stability of the former. Simple topological considerations show that while the director can collapse in the third dimension in defects of integral strength in which \mathbf{n} changes its direction by even multiples of π in going once around the defect, no such collapse can occur for defects of half integral strength in which the director rotates only by odd multiples of π around the director (Figure 1). This has been indeed

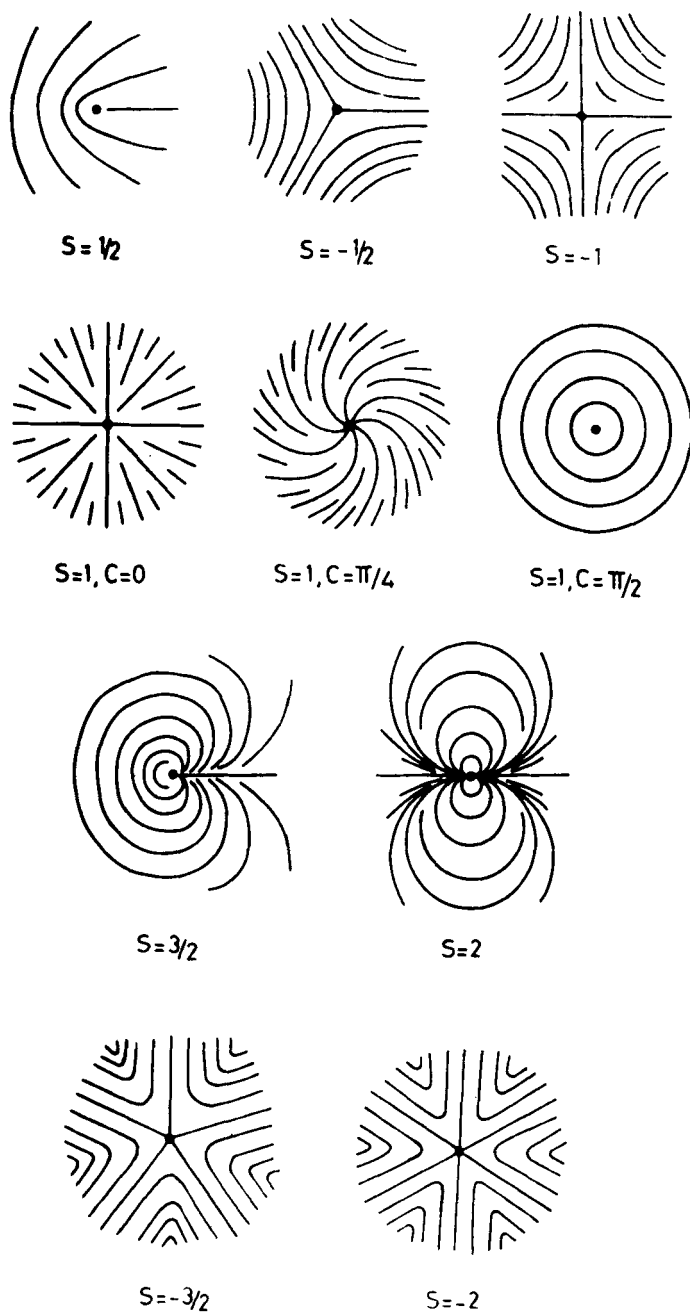


FIGURE 1 Schematic diagram of the director fields around disclinations of various strengths, according to Eq. (1).

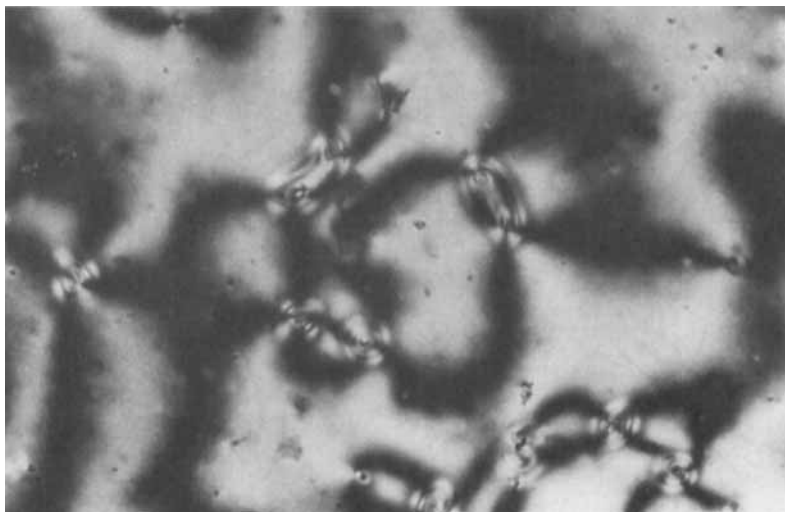


FIGURE 2 Disclinations of strengths $\pm \frac{1}{2}$ and ± 1 in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate. Crossed linear polarizers and illumination with sodium light ($\times 540$). The dark rings around disclinations of strength ± 1 are due to the collapse of the director in the 3rd dimension.

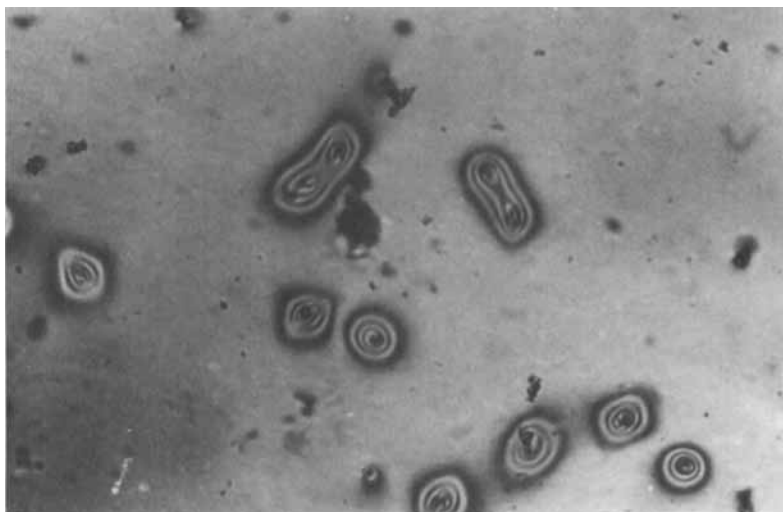


FIGURE 3 Same sample as in Figure 2 between crossed circular polarisers ($\times 540$).

confirmed in many observations (see for example, Figures 2 and 3; however see ref. 9).

We recently discovered defects of strength > 1 in some thermotropic nematic mixtures.¹⁰ Interestingly enough, at about the same time, Lee and Labes¹¹ found 'apparent high strength singularities' in a rather different system, viz., in emulsified mixtures of 5CB or MBBA with the lyotropic liquid crystal system (di-sodium cromoglycate + ethylene glycol). After a special preparation of the sample and removing the coverslip, they found defects of strength up to 4 which were unstable as the microemulsion itself broke up after several minutes.

On the other hand, the systems that we have studied are quite stable. In the following, we describe our observations on not only single defects of high strength but also on pairs of high strength defects ($\pm S$) which we found very recently.

OBSERVATIONS AND RESULTS

The high strength defects were observed in mixtures of various nematogens with the non-mesomorphic compound leucoquinizarin (LQ for short) which has platelike molecules. LQ was obtained from Aldrich Chemical Co. and has a melting point of $\sim 147^\circ\text{C}$. The mixtures typically contained ~ 10 – 15 weight per cent of LQ and consequently the nematic-isotropic transition temperature was drastically reduced by $\sim 30^\circ\text{C}$ or more, compared to that of the pure nematogen. Further, there was a considerable range ($\sim 20^\circ\text{C}$) of co-existence of the nematic and isotropic phases. All the observations were made using a Leitz Ortholux Polarizing Microscope (Model II POL-BK) in conjunction with a Mettler FP-52 hot stage. As the sample was cooled below T_{NI} , nematic droplets formed and coalesced in the sample and there was a strong tendency to exhibit the schlieren texture (see Figures 2 and 3 for example). In many cases, we could observe defects of strength $\frac{3}{2}$ (i.e., with 6 brushes, Figure 4). On rare occasions, we could also see defects of strength 2 (Figure 5) which usually had a blurred region near the centre. Defects with both positive and negative signs were found in both the cases. Usually the $\frac{3}{2}$ defects occurred in an isolated configuration (Figure 6), but sometimes these defects occurred in combination with others as, following the description of Nehring and Saupe,⁶ inversion walls of the first kind (Figures 7 and 8).

Just as in the case of $\frac{1}{2}$ defects, we cannot expect any extensive collapse of the director in the third dimension around defects of

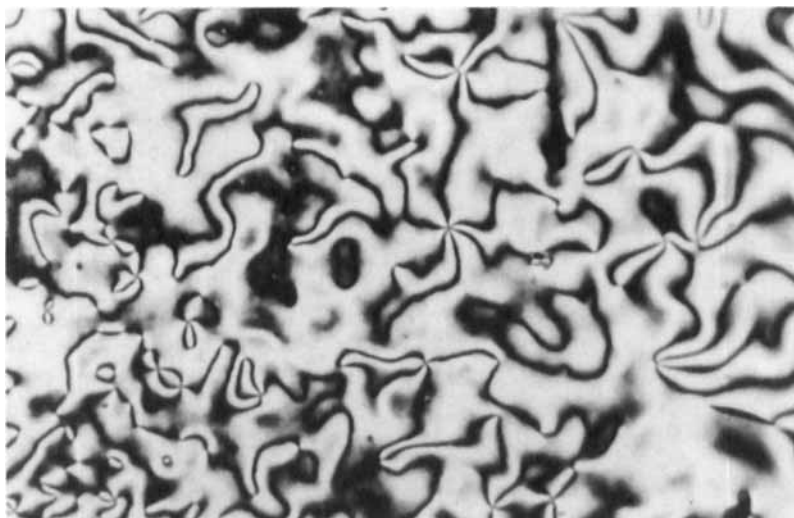


FIGURE 4 A $-\frac{1}{2}$ defect in a mixture of 20 weight per cent LQ with *p*-octyloxy-*p*'-cyanobiphenyl at 53°C. Illumination with white light ($\times 270$).



FIGURE 5 A -2 defect in a mixture of 20 weight per cent of LQ with *p*-octyloxy-*p*'-cyanobiphenyl at 50°C. Illumination with white light ($\times 540$).

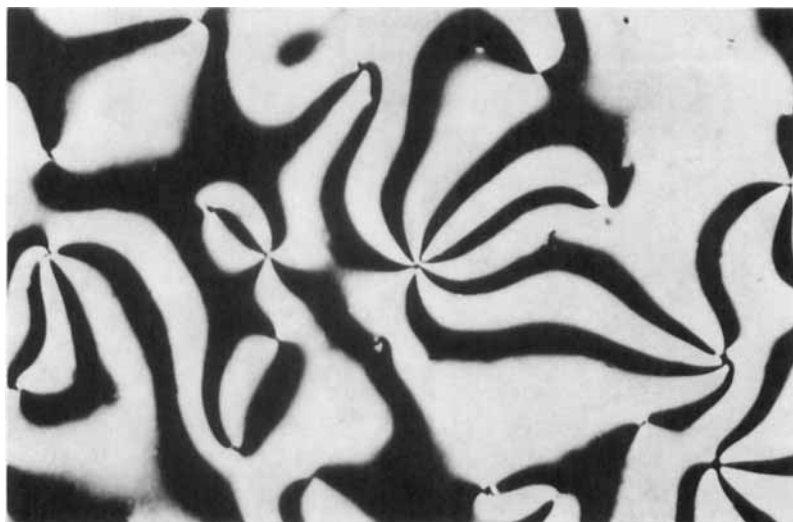


FIGURE 6 A $+ \frac{3}{2}$ defect in a mixture of 20 weight per cent of LQ with 8 OCB at 53.2°C . Illumination with white light ($\times 540$).

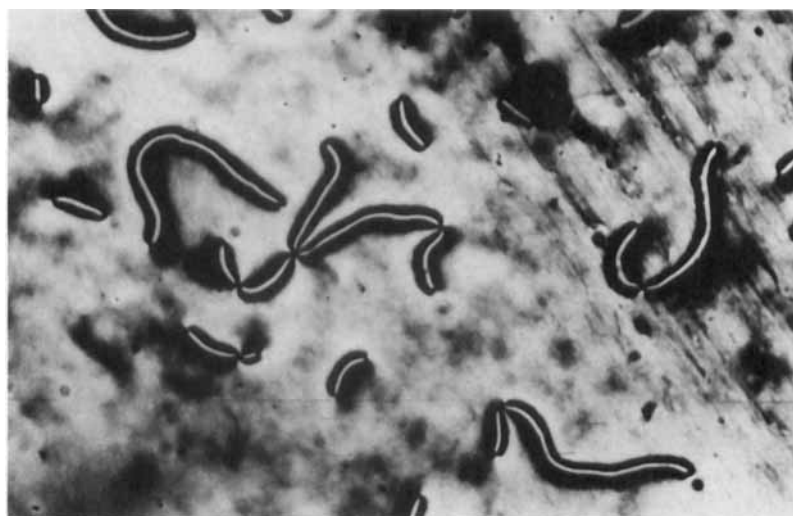


FIGURE 7 An inversion wall of the first kind with a $- \frac{3}{2}$ disclination in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate at 28°C ($\times 540$).

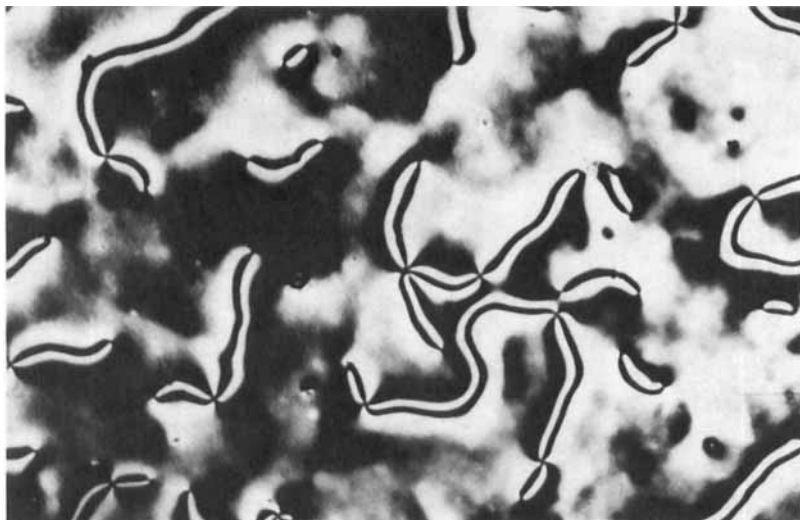


FIGURE 8 An inversion wall of the first kind with a $+ \frac{3}{2}$ disclination in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate at 34 °C ($\times 540$).

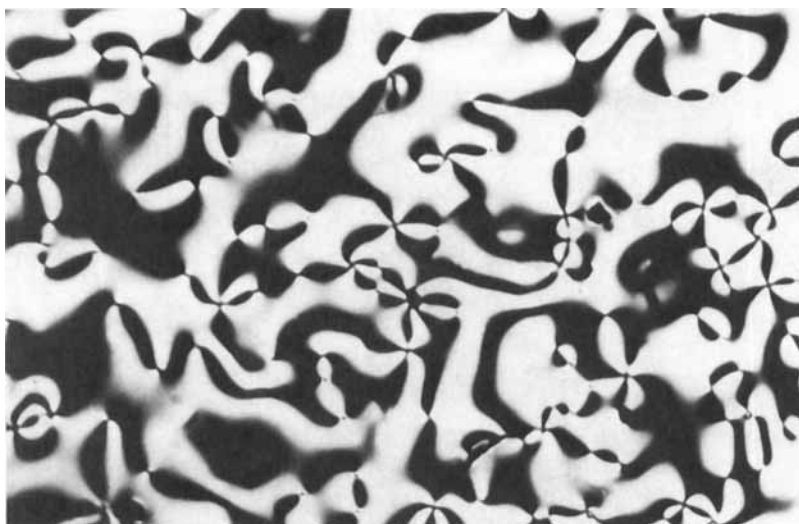


FIGURE 9 A $+ \frac{3}{2}$ defect (near the centre of the photograph) just becoming isotropic at the centre in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate at 46 °C ($\times 270$).

strength $\pm \frac{3}{2}$. Indeed observations in monochromatic light did not reveal any extensive change in birefringence as one approached the centre of the disclination from the outer regions. Consequently the director pattern probably corresponds to that given in Figure 1, or, at any rate, to a configuration which can be continuously deformed to a planar configuration. For the planar configuration, the line energy is given by Eq. (2). Hence one would expect the $\pm \frac{3}{2}$ defects to be unstable and break-up into defects of lower strength. However, this was generally not the case and the $\frac{3}{2}$ defects could be observed for several hours even when the sample was cooled by $\sim 30^\circ\text{C}$ below the temperature at which the entire sample was covered with the nematic phase. Indeed, as the sample was heated, in most cases, the $\pm \frac{3}{2}$ defects remained intact and one could see the isotropic phase starting to form at the centre of the defect and get enlarged as the temperature was raised further (Figure 9). This most probably signifies a concentration gradient around the defect. However on rare occasions, on warming the sample a $\frac{3}{2}$ defect split into a 1 and a $\frac{1}{2}$ defect (Figures 10 and 11).

In some samples as the temperature was lowered, a large number of brushes (up to 14) could be seen emerging from an isotropic droplet. For example, Figure 12 shows a drop from which 12 brushes are found to emerge, i.e., it is topologically equivalent to a defect of strength 3. If we assume that the director makes a constant angle with the nematic-isotropic interface it is easy to see^{12,13} that such a drop should be topologically equivalent to a defect of strength +1. However, Ericksen¹⁴ has pointed out the possibility that the angle that the director makes with the interface need not necessarily be constant. Such a non-uniform orientation at the nematic isotropic interface has also been considered by Bouligand.¹⁵ If the elastic anisotropy is also taken into account, the cross-section of the isotropic region is not circular. In such a case, the isotropic drop is no longer topologically equivalent to a defect of strength +1.

Often, we could also see pairs of $\pm \frac{3}{2}$ defects. Figures 13 and 14 show the brush patterns around one such pair as the crossed polarisers are rotated through 45° . The separation between the two defects is $\sim 10\ \mu\text{m}$ and when the sample was heated close to the N-I transition point, the two moved closer and annihilated each other. The solution for the director pattern around a pair of defects of strength $\pm S$ in the one-constant approximation can be written as

$$\Psi = S(\alpha_1 - \alpha_2) + C \quad (4)$$

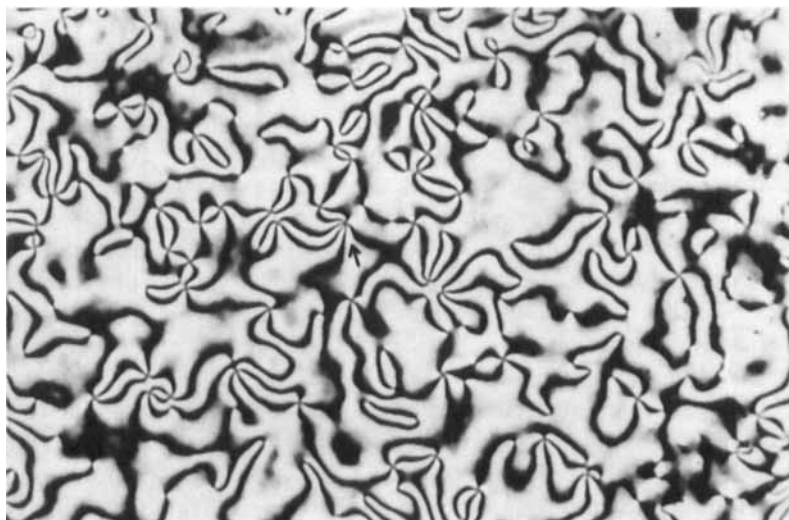


FIGURE 10 A $+\frac{3}{2}$ defect (slightly above and to the left of the centre of the photograph, indicated by an arrow), along with ± 1 and $\pm \frac{1}{2}$ defects in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate at 36.2°C. Illumination with white light ($\times 270$).

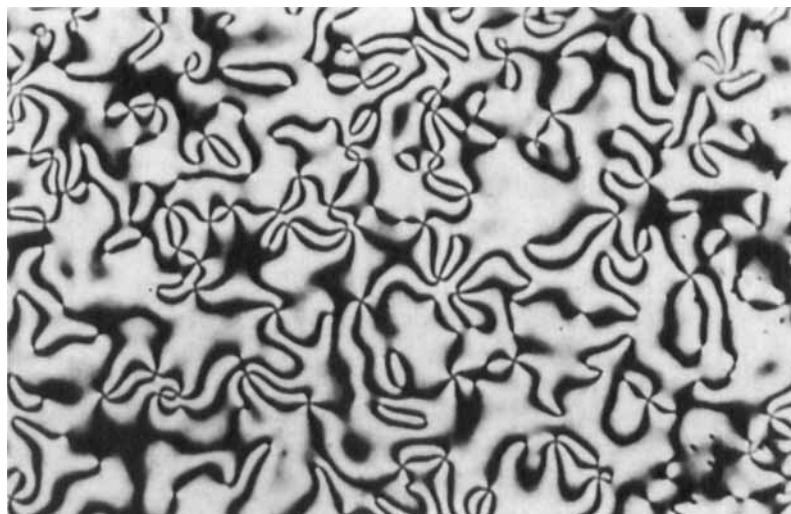


FIGURE 11 Same sample as in Figure 10 at 38.7°C. Note that the $+\frac{3}{2}$ defect has split into a $+1$ and a $+\frac{1}{2}$ defects ($\times 270$).

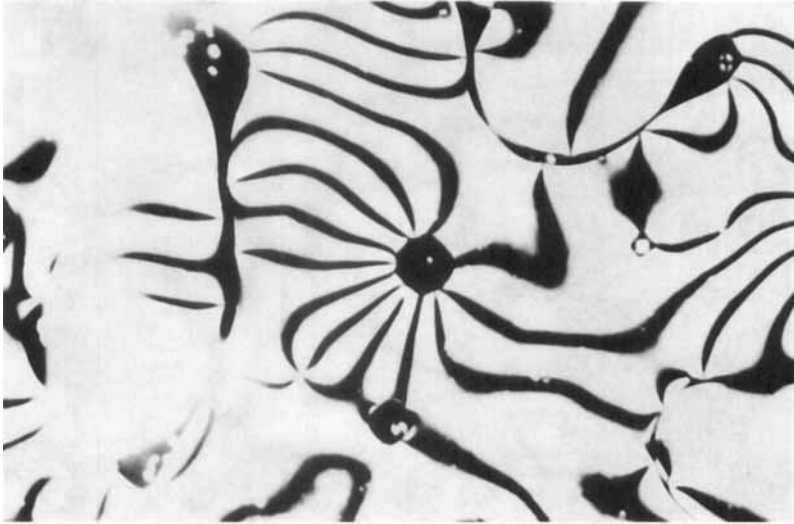


FIGURE 12 12 brushes around an isotropic droplet in a mixture of 25 weight per cent of LQ with CBOOA at 57°C ($\times 270$).

where α_1 and α_2 are the angles as in Eq. (1) with respect to the centres of the two defects.⁶ The constant C determines the orientation of the director far away from the defects. We show the director patterns for various values of C in Figure 15. In the one constant approximation, the interaction energy between a pair of defects $\pm S$ should be independent of C .⁶ Anisotropy of elastic constants ($K_{11} \neq K_{22} \neq K_{33}$) changes the solution for the director field around a single defect⁶ as also the configuration and the energy of interaction between the pair $\pm S$. In particular, Ranganath¹⁶⁻¹⁸ has shown that depending on the anisotropy, specific values of C should be favoured. Most of the $\pm \frac{3}{2}$ pairs that we have studied appear to favour values of $C = 0$ or $\approx 45^{\circ}$.

Very occasionally we could observe pairs of defects of strength ± 2 . Figures 16-18 show the brush patterns around such a pair as the crossed polarisers are set at various angles. It is clear from the photographs that while the director collapses in the third dimension near ± 1 defects, strangely no such collapse is seen near ± 2 defects. The director patterns based on Eq. (4) are shown in Figure 19. From the configuration of the dark brushes for various settings of the polarisers, the pair shown in Figures 16-18 has a $C \approx 45^{\circ}$.

It is interesting to speculate about the reason for the stability of the high strength defects. As we mentioned earlier, a concentration gradient close to the defect is a possible mechanism and we are working out



FIGURE 13



FIGURE 14

FIGURES 13–14 $A + \frac{3}{2}$ and $-\frac{3}{2}$ pair of defects in a mixture of 12 weight per cent of LQ with *p*-cyanophenyl-*p*-*n*-heptylbenzoate. Illumination with sodium light ($\times 540$). Polarisers are rotated by 45° on going from Figure 13 to Figure 14 in the clockwise direction.

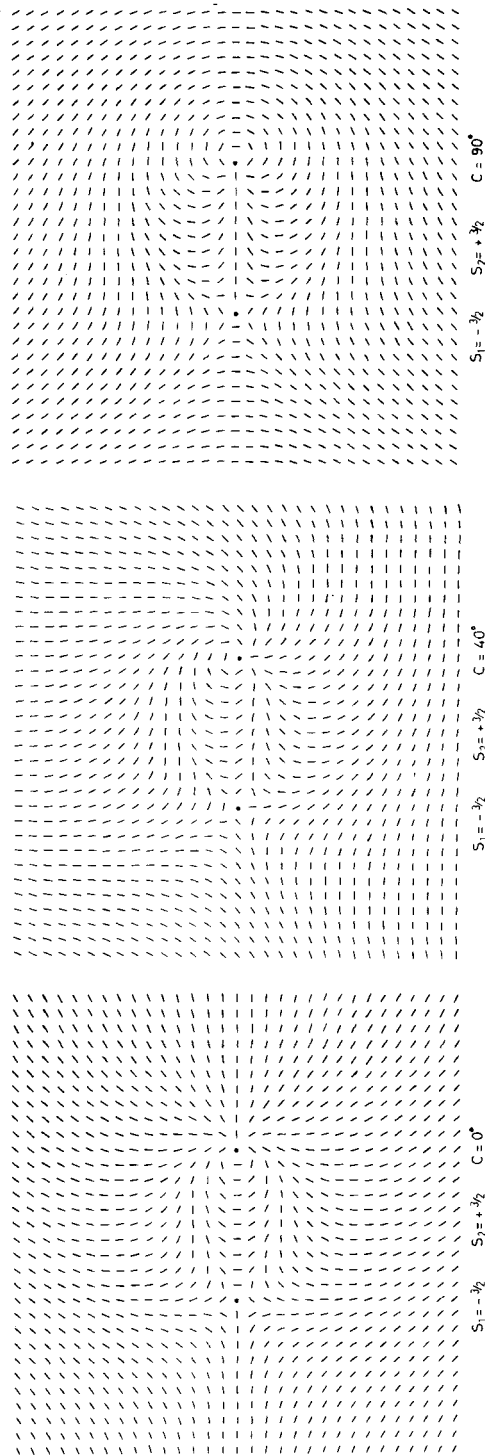


FIGURE 15 The director fields in the neighbourhood of a pair of $\frac{1}{2}$ defects of opposite sign, according to Eq. (4) for various values of C .

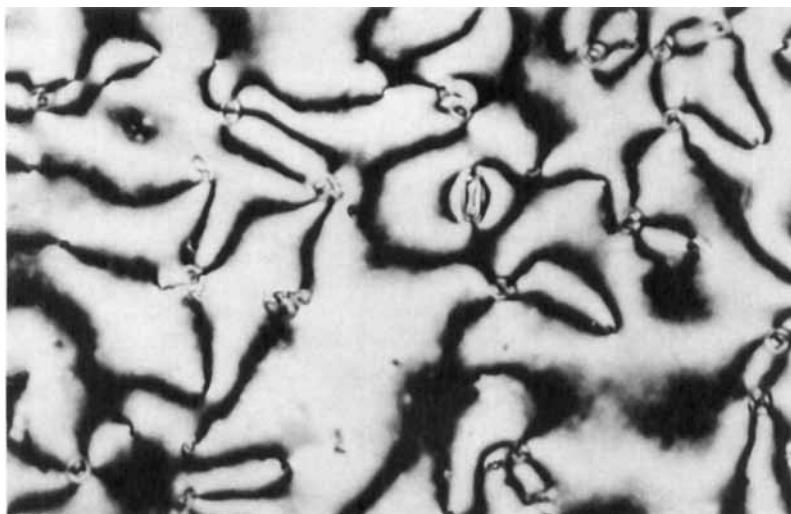


FIGURE 16

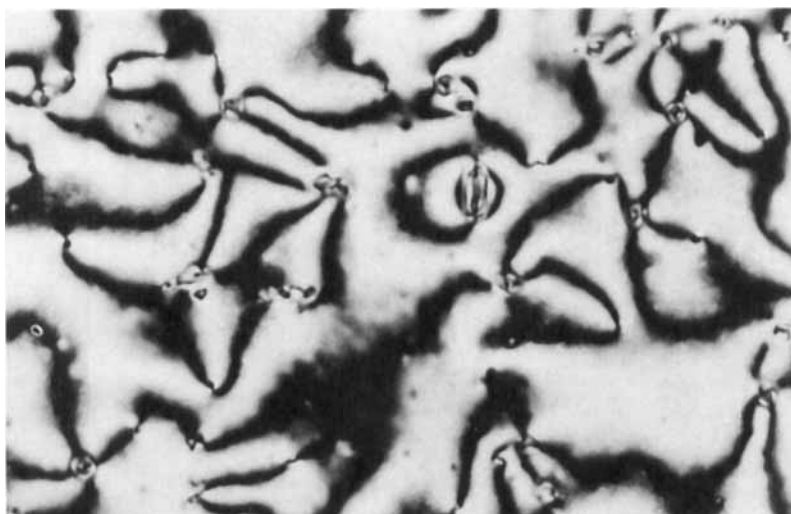


FIGURE 17



FIGURE 18

FIGURES 16–18 A +2 and -2 pair of defects in a mixture of 12 weight per cent of *p*-cyanophenyl-*p*-*n*-heptylbenzoate. Illumination with sodium light ($\times 540$). Polarisers rotated by 22.5° on going from Figure 16 to Figure 18 in anticlockwise direction.

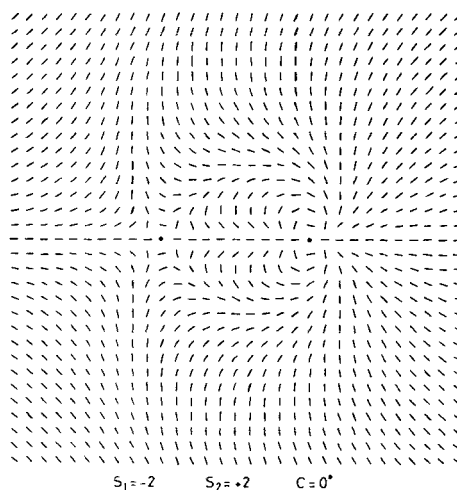
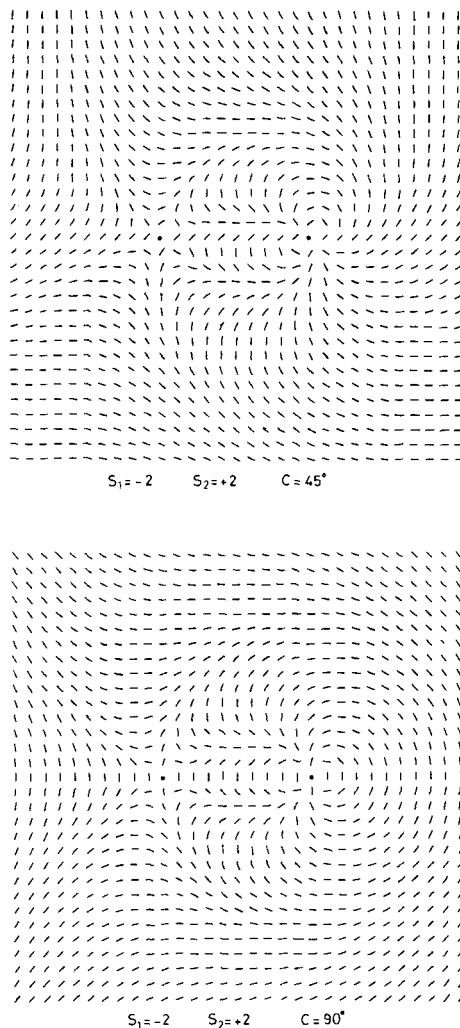


FIGURE 19 The director fields in the neighbourhood of a pair of defects of strength 2 of opposite sign, according to Eq. (4) for various values of C .

FIGURE 19 *Continued*

the implications of this idea. Further, it is possible that the director may collapse in the third dimension *close* to the disclination even for defects of $\frac{1}{2}$ integral strength by acquiring a twist deformation.^{8,15} This may lower the energy by relaxing the strong bend which would otherwise be present close to the centre of the defect.

Further studies on these mixtures are in progress.

Acknowledgments

The authors are grateful to Prof. S. Chandrasekhar for useful discussions. Thanks are also due to Dr. G. S. Ranganath for helpful comments.

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