# HIGH STRENGTH DEFECTS IN NEMATIC LIQUID CRYSTALS

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### ABSTRACT

After giving a brief introduction to the present state of knowledge of defects in nematic liquid crystals, we report the first observation of disclinations of strength $\pm 3/2$ and $\pm 2$ in mixtures of several nematogenic materials with a nonmesomorphic compound having plate like molecules.

NEMATIC liquid crystals are characterized by a long range *orientational* order of anisotropic molecules[1,2]. The simplest method of identifying the nematic state is by making observations under a polarizing microscope on a thin sample of the material taken between a slide and a coverslip. If the glass plates have not been specially treated to get a uniform alignment of the medium, one usually observes a typical 'texture' or a pattern of characteristic defects in the alignment of the medium. For sufficiently thin samples ($\sim 20$ $\mu$ or less), a schlieren texture is obtained, in which two or four dark brushes are seen to emerge from some points in the field of view (figure 1). The points which are usually line defects viewed end-on are called disclinations[3].

The *local direction of orientation* of the molecules at any given point is represented by a unit vector **n**, called the 'director'. Assuming that n is in the plane of the sample it is clear that the dark brushes occur in areas in which n is either parallel or perpendicular to the plane of polarization of the incident beam. n changes its orientation continuously around any disclination, and for ensuring continuity in the medium, it is obvious that the change in the orientation on going once around any given defect should be a multi-

ple of $\pi$, assuming that the nematic director is *apolar*. If this angle is $\pi$, we get two dark brushes, while if it is $2\pi$, we get four brushes, etc. The observation of defects with two brushes is indeed a proof that the director is apolar, *i.e.*, n and $-$ n are physically equivalent. The 'strength' of the defect $S$ is defined as $1/4$ times the number of brushes emerging from it. The sign of the defect is taken to be positive if the brushes rotate in the same direction as that of the crossed polaroids and negative if the brushes rotate in the opposite direction. The Schlieren textures were studied soon after the discovery of liquid crystals, and detailed descriptions were given[4,5]. Upto the present, only defects of strengths $\pm 1/2$ and $\pm 1$ appear to have been found in nematic liquid crystals[1,2,6,7].

The curvature of the director around a given defect costs elastic energy. The nematic is characterized by three curvature elastic constants $k_{11}$, $k_{22}$ and $k_{33}$ corresponding to splay, twist and bend.[1] They are generally of the same order of magniture, $\sim 10^{-6}$ dyne. In the one constant approximation, it is assumed that $k_{11} = k_{22} = k_{33} = K$, say. It is then particularly simple to apply the theory of curavature elasticity to get solutions for the director field around disclinations. If $\Psi$ is the angle made by n with the x-axis, and $\alpha$ the angle made by the radius vector connecting the given point to the centre of the defect with the x-axis, the solutions take the simple form

$$\Psi = S\alpha + C \tag{1}$$

where C is a constant.[3] The director fields around disclinations of various strengths are shown schematically in figure 2. Except in the case of $S = 1$ defect, changing the value of $C$ merely rotates the entire pattern. In the case of $S = 1$, the pattern itself changes with $C$ (see figure 2). In the present approximation, n becomes multiple valued at the origin, and hence leads to a singularity. Assuming that a 'core' region extends from the orgin to $r_c$, the energy per unit length of an isolated disclination line is given by[1,8]

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

where $W_c$ is the energy/unit length from the core region and $R$ is the size of the sample. Assuming that
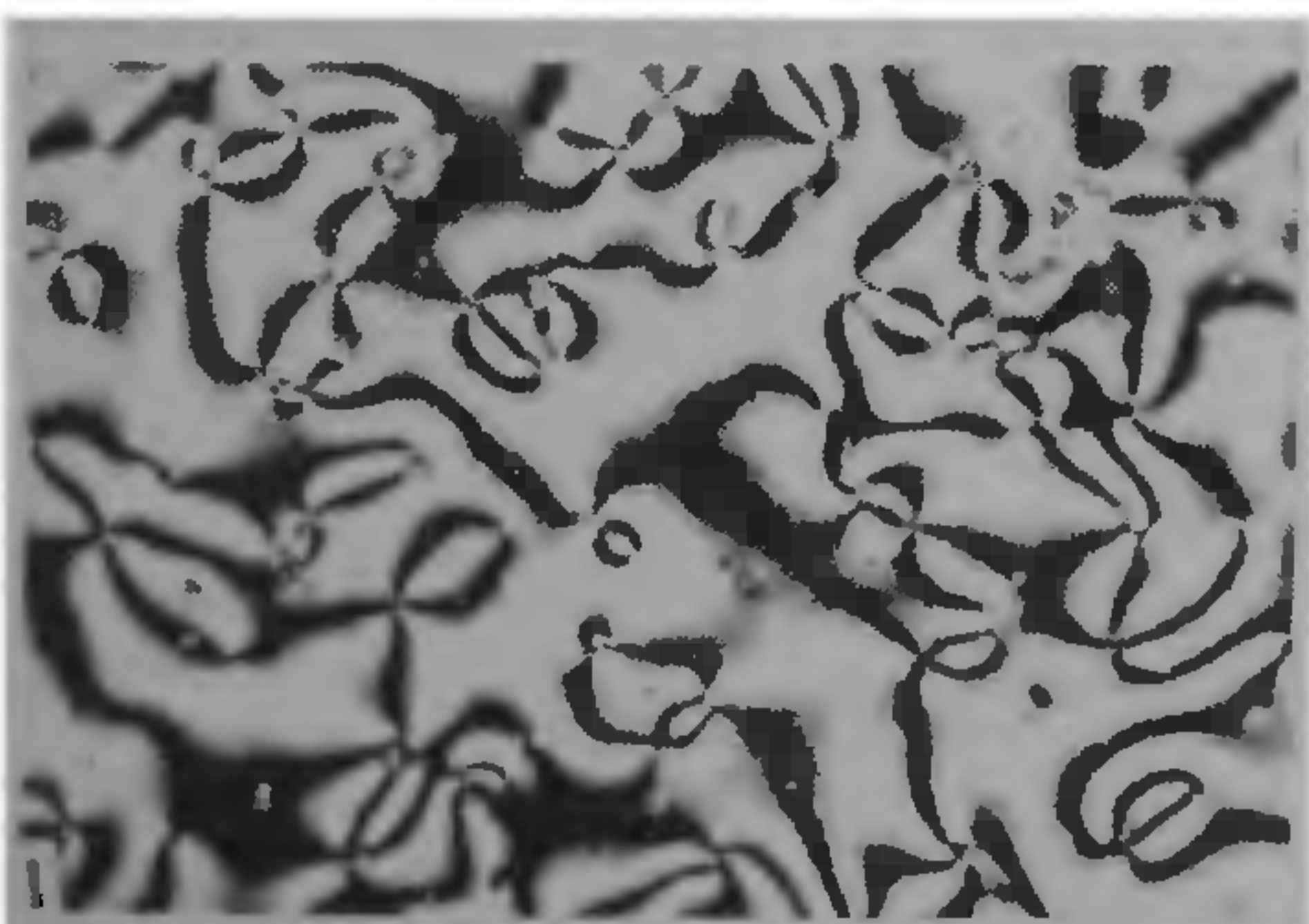


Figure 1 Schlieren texture of a mixture of 15 weight % of THA with p-cyanobenzylidene-p'-octyloxyaniline (CBOOA) at 55.8°C. Illumination with sodium light ($\times$ 300). Note the dark rings around disclinations of strength $\pm 1$.
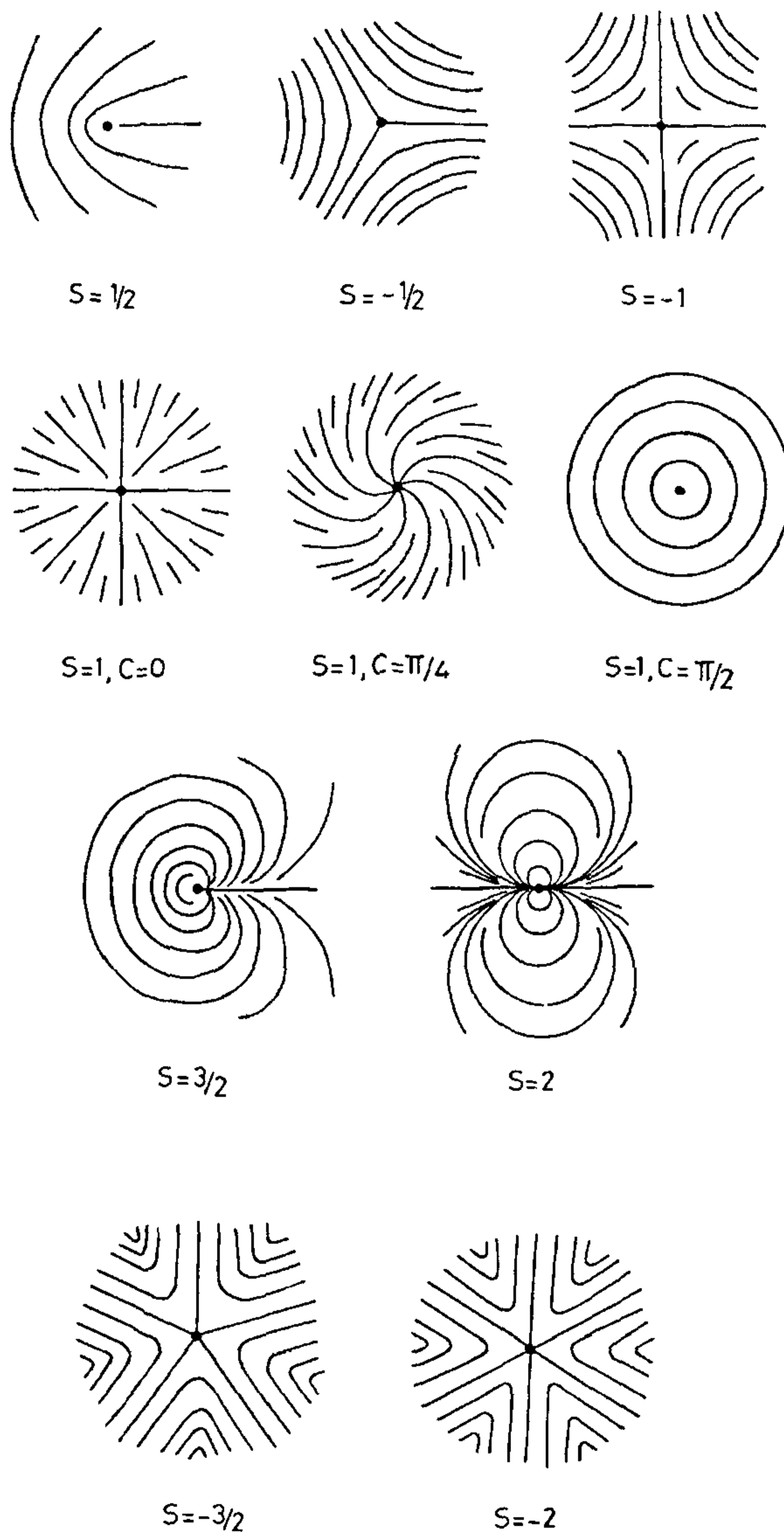
**Figure 2.** Schematic diagram of the director fields around disclinations of various strengths according to the solution given by Eq. (1).

$W_c$ makes a relatively small contribution, $W$ has a logarithmic divergence with the size of the sample or as $r_c \to 0$. For a given size $R$, it is clear that disclinations with $S = \pm 1$ cost 4 times the energy of those with $S = \pm 1/2$ in this approximation. However, experimentally $S = \pm 1$ disclinations are rather easily obtained (see figure 1) and are quite stable.

At this stage, we can point out that n need not be confined to a plane normal to a disclination line if its strength has an integral value ($S = \pm 1, \pm 2$ etc.). As we can easily verify with reference to the patterns shown in figure 2, in such cases a tilted n is a topologically allowed solution. However, in the case of disclinations with half integral strength ($S = \pm 1/2, \pm 3/2$, etc.) any tilting of n is *not* allowed topologically. Going in a closed circuit around a disclination of half -integral strength, a tilted director whose projection in the plane of the paper (see figure 2) has changed its angle $\Psi$ by $\pi$ (or $3\pi$) *cannot* match with the initial director. The physical implication of this difference in the topological structure of integral and half-integral disclinations is that the director can collapse in the third dimension (normal to the plane of the paper in figure 2) for defects with integral strength. n is then along the z-axis at the centre and is no longer a multiple-valued function, and there is no singularity. Indeed calculations based on the theory of elasticity[9,10] yield in such a case

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

independent of $R$, the length over which the collapse takes place. If the structure has a singularity and the core radius is of the order of a few molecular dimensions ($r_c \sim 50$ Å say) and the size of the sample $R$ is $\sim 50$ microns, we find from eq. (2) that the line energies for $S = +1$ are definitely greater than those given by the non-singular solutions (Eq. 3). Experimental results [8-10] generally confirm this conclusion: for example, in figure 1, which was taken in monochromatic radiation ($\lambda = 5893$ Å), several dark rings can be seen around $S = 1$ disclinations. The bands arise from a smooth reduction in the effective birefringence of the medium as the director collapses towards the z-axis as the centre is approached. On the other hand, the changes in the birefringence near $S = \pm 1/2$ singularities are confined to a rather small region around the centre. Indeed often the singular line corresponding to $S = \pm 1/2$ can be seen side-on as a 'thread' lying in the plane of the sample.
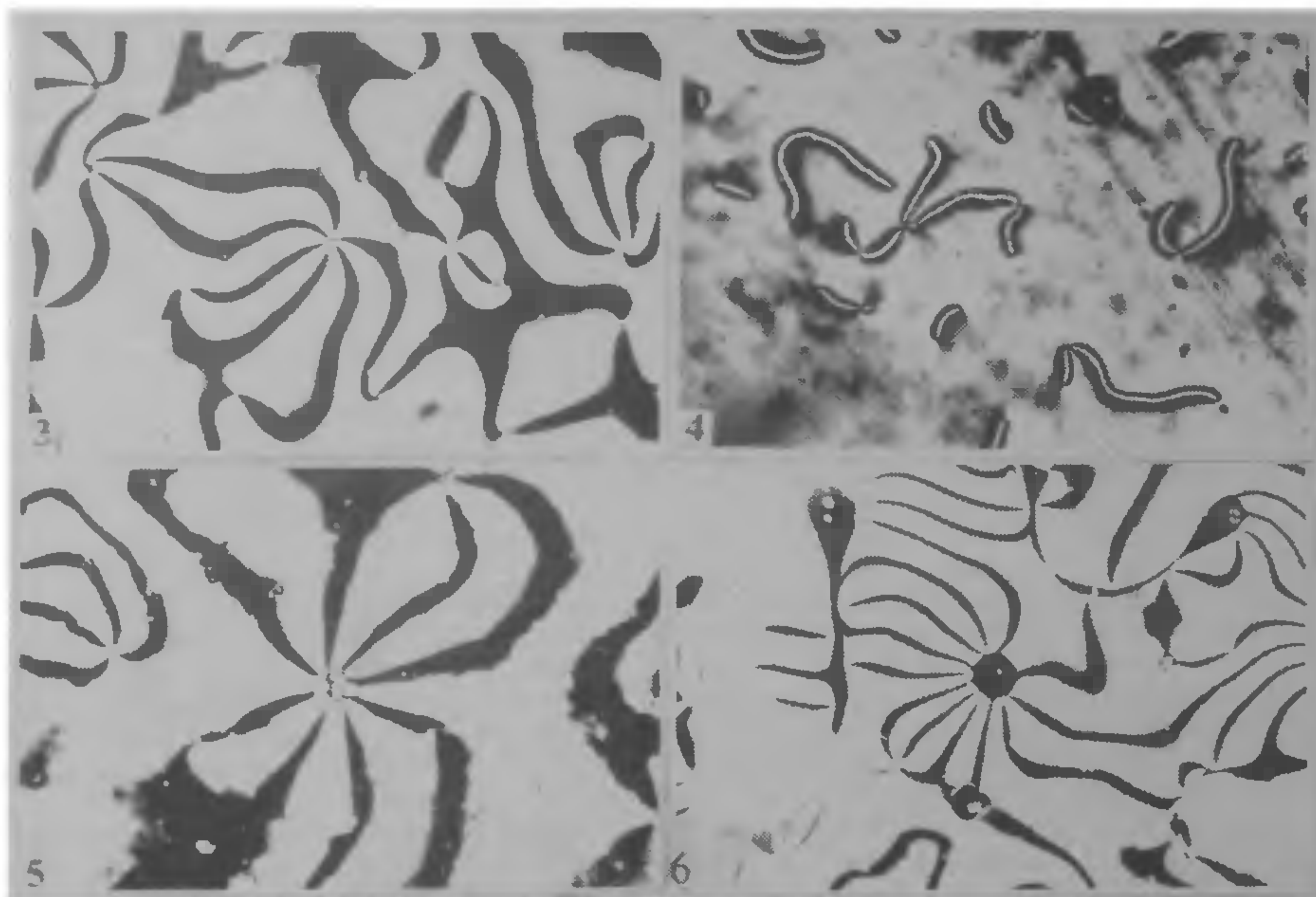
Real nematics have anisotropic elasticity ($k_{11} \neq k_{22} \neq k_3$.) but an analysis including the anisotropy[8,10,11] will not alter the above conclusions, even though it affects the magnitudes of the energies. (The anisotropic elasticity has important consequences for the interactions between defects[12,13] which we do not consider here.) Another interesting result of the non-singular solution for $S = \pm 1$ is that the corresponding

energy (Eq. 3) can be *lower* than the sum of energies of two-singularities of the same sign (Eq. 2) Indeed it is sometimes observed[9,10] that two half singularities of the same sign merge together to give rise to a 1 disclination, especially if there is a three-dimensional distortion in the director field in the region between the two half singularities.[14]

Various other details of the defect structure in nematics have been discussed in review articles by Kleman.[6,7] In particular, the nature of the core around the singularity or $S = \pm 1/2$ defects is not known. It is sometimes speculated that the core region is in the isotropic phase.[2,10]

With this background, we now report our observation of disclinations of strength $> 1$ in some nematic mixtures. These are probably the first observations of such defects in nematic liquid crystals.

The high strength defects were observed in mixtures of the nonmesomorphic compound 1,4,9,10-tetrahydroxy-anthracene (THA for short, also called leucoquinizarin) with various nematogenic compounds. THA was purchased from Messrs. Aldrich Chemical Co. and has a melting point of $\sim 147^\circ$C and was mostly used without further purification. The molecules of this red-coloured compound are rigid and flat. The mixtures typically contained $\sim 10$-15 weight precent of the nonmesomorphic compound. As a result of adding THA to the nematogenic compound, the nematic-isotropic transition temperature ($T_{NI}$) was drastically reduced, by $\sim 30^\circ$C or more. Further, there was a considerable range ($\sim 20^\circ$C) of coexistence 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. Nematic droplets formed as the sample was cooled from the isotropic phase, and slowly grew in size as the temperature was further lowered. There was a strong tendency to form the schlieren texture (see figures 1,3-6). Further, in many samples one could see defects with six brushes as the isotropic phase disappeared on cooling. They were quite stable, and remained intact even when the sample was cooled by $\sim 30^\circ$C below the temperature at which the entire sample was transformed to the nematic phase. We have observed both positive and negative disclinations with six brushes, *i.e.*, with strength $\pm 3/2$. Observations with sodium light did not reveal any dark bands around the centre, *i.e.*, there was no visible collapse of the director in the 3 rd dimension. This is in conformity with the expectation from topological arguments which we mentioned earlier. Hence one could expect that these defects also have singularities at the centre and the director pattern around such defects would be as shown in figure 2. In that case the line energy would be given by Eq. (2), in the one-constant approximation. It is clear that the elastic energy carried by a 3/2 defect would be consid-

erably reduced if it splits into defects of lower strength. Sometimes we did observe such a splitting (usually to a pair of defects of strength 1 and 1/2) at high temperatures. More often, however, the 3/2 defects were stable. They could occur either in a relatively isolated configuration (figure 3) or as a part of an inversion wall of the first kind (figure 4) according to the description of Nehring and Saupe.[8] Such a wall is a relatively narrow region which separates two areas in which the director has rotated by an angle $\pi$. On heating the sample, we observed that the isotropic phase starts to form at the centres of the 3/2 defects. This could be a confirmation of the speculation that the core region of singularities are in the isotropic phase. However, we must mention that we could not observe a similar phenomenon in the core regions of $S = \pm 1/2$ defects occurring in the same sample.

On some rare occasions, we saw defects with eight brushes, or disclinations with strength 2 (figure 5), of both positive and negative signs. However, usually the pattern close to the centre of the defect was not very clear.

We could sometimes see a large number (up to 14) of dark brushes emerging from an isotropic droplet surrounded by a nematic region. Figure 6 shows a photograph with 12 brushes. It is topologically equivalent to a defect of strength 3, even though it has been argued that such drops should correspond to a defect of strength 1.[15,16]

At this stage, we would prefer not to speculate on the origin of the high strength defects in the systems that we have studied. We have taken up a detailed study of the structure of these defects, and the determination of the elastic constants of the mixtures. The results will be published elsewhere.

**Figures 3-6. 3.** A relatively isolated disclination of strength 3/2 in a mixture of 20 weight % of THA in p-octyloxy-p'-cyanobiphenyl (8 OCB) at 53°C. Illumination with white light ( × 800). **4.** An inversion wall of the first kind with a 3/2 disclination in a mixture with 12 weight % of THA in p-cyanophenyl-p'-heptyl benzoate at 28°C ( × 800). **5** A disclination of strength 2. The mixture is the same as the one used for obtaining figure 3. ( × 800). **6** Twelve dark brushes emerging from an isotropic droplet in the 2 phase region of a mixture with 25 weight % THA in CBOOA at 57°C ( × 300).

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