Thermotropic uniaxial nematic liquid crystal with ±1/2 strength disclinations: defect–antidefect production and correlation

Surajit Dhara[1]**, T Arun Kumar**[1]**, Ken Ishikawa**[2] **and Hideo Takezoe**[2]

[1] School of Physics, University of Hyderabad, Hyderabad 500046, India
[2] Department of Organic and Polymeric Materials, Tokyo Institute of Technology,
2-12-1 O-okayama, Meguro-ku, Tokyo 152-8552, Japan

E-mail: sdsp@uohyd.ernet.in and takezoe.h.aa.@m.titech.ac.jp

Abstract
We report the observation of mostly wedge disclinations of strength $\pm 1/2$ in a uniaxial nematic liquid crystal. The dense network of the disclination is stabilized in cells of thickness ($\sim 6\ \mu$m) treated with perfluoropolymer. The distribution of net defect strength was found to be Gaussian. The scaling exponent obtained from the width of the distribution is $\nu = 0.29 \pm 0.08$ and is in good agreement with the prediction based on the cosmological Kibble mechanism. The exponent and the constant of the scaling relation are compared with the values reported for lyotropic liquid crystal with half-strength disclination by Mukai and co-workers.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Thermotropic liquid crystals exhibit a rich variety of textures under the polarizing microscope [1, 2]. The different phases exhibit their own characteristic defect structures under the polarized optical microscope. For example, nematic liquid crystals are characterized by observing schlieren texture. Smectic and cholesteric liquid crystals are characterized by focal conic and fingerprint textures. Usually the schlieren texture in the nematic case contains point or line defects with the appearance of four and two brashes under crossed polarizers, respectively [2]. These defects belong to a class, called the topological defects, in a system with broken symmetry. The vortices in superfluid helium and in the XY model, the dislocation in periodic crystals, and the cosmic strings in the early Universe also belong to the topological defects and have been a common interest of scientists in various branches in physics. There are several systems in condensed matter reported in which topological defects are formed in the laboratory from the symmetry breaking phase transition, such as superfluid helium [3], thermotropic liquid crystals [4–6] and superconductors [7]. Among these, liquid crystals with the nematic to isotropic (NI) phase transition have proven to be simplest, and are also the systems in which it is easiest to form and study the defect–antidefect pairs. Here antidefects are the defects with opposite sign of the winding number and are generally written with a minus sign. In the nematic case, the tensor order parameter Q_{ij} breaks the rotational symmetry of the isotropic liquid phase and the defect–antidefect pairs are formed spontaneously. There are a few reports on studies of the formation of defect–antidefect pairs and their correlation in thermotropic nematic liquid crystals with defect strength 1 [8] and in lyotropic liquid crystal with half-strength defects [9]. Thus it is important to investigate the correlation in various other systems with different defect strengths. In particular, there are no reports on the observation of mostly half-strength defects in the thermotropic nematic liquid crystals. In this paper we report on a unique system in which mostly half-strength defects in a uniaxial nematic liquid crystal are stabilized in perfluoropolymer treated cells. We also studied the defect–antidefect correlation from the cosmological point of view and a good agreement was found with the theoretical prediction [10].
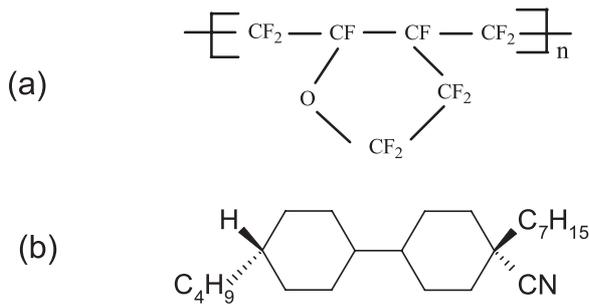
Figure 1. (a) Structure of the perfluoropolymer alignment layer. (b) Schematic of the molecular structure CCN-47 used in the experiment.

2. Experimental details

We used perfluoropolymer, poly[perfluoro(4-vinyloxy-1-butene)], known as CYTOP (Asahi Glass Co., Ltd), as an alignment layer. The chemical structure is shown in figure 1(a). The polymer was dissolved in CT-Solv-180 at a weight ratio of 1:2 and the solution was spin coated on ITO (indium tin oxide) coated glass substrates by using a conventional spin coating method. Then it was baked at 100 °C for 30 min. The typical polymer film thickness was ∼100 nm. The chemical structure of the compound 4′-butyl-4-heptyl-bicyclohexyl-4-carbonitrile (CCN-47) is shown in figure 1(b). The compound exhibits the following phase transition temperatures under cooling: Cr 25.6 °C SmA 28.2 °C N 57.3 °C I. It shows a large negative dielectric anisotropy ($\Delta\epsilon = -5.7$ at 30 °C). The chemical structure and some physical properties of the present compound have already been reported [11]. CCN-47 in CYTOP coated cells shows a planar texture in the nematic phase. It exhibits a discontinuous anchoring transition from planar to homeotropic at ∼45 °C with a large hysteresis (∼5°) [12] in heating and cooling. Exploiting this large

hysteresis, a bistable device has also been proposed by us [13]. Very recently we also reported on the shock free homeotropic alignment of SmA and SmC* phases in this alignment layer [14]. In the present experiment the typical cell thickness used was ∼6 μm. All the observations were carried out while cooling the sample. The temperature of the sample was controlled with a Mettler Hotstage and a controller to an accuracy of 0.1 °C. Texture observation was carried out with the help of an optical polarizing microscope.

3. Results and discussion

In figure 2, we show the textures obtained for some common liquid crystals with positive and negative dielectric anisotropies in CYTOP coated cells. 8CB (octyl-cyanobiphenyl), a liquid crystal with a positive dielectric anisotropy ($\Delta\epsilon = \sim8$ at 30 °C), exhibits a homeotropic alignment (figure 2(a)). MBBA (methoxybenzylidene butylaniline) liquid crystal with a negative dielectric anisotropy ($\Delta\epsilon = -0.6$ at 30 °C) shows homeotropic alignment just below the nematic–isotropic transition temperature (figure 2(b)) and the director gradually tilts from the vertical direction as the temperature is lowered [12]. In the case of MLC-6608, a large negative dielectric anisotropy ($\Delta\epsilon = -10$ at 25 °C) liquid crystal mixture, we noticed a planar texture with mostly ±1 defects (figure 2(c)). However, the single-component liquid crystal CCN-47, with a similar large negative dielectric anisotropy, exhibits mostly ±1/2 defects and the defects are stable (figure 2(d)).

The temporal variation of the typical textures of CCN-47 on CYTOP just after the transition is shown in figure 3. The sample is cooled at the rate of 0.2 °C s⁻¹ in a 6 μm thick cell. We noticed that mostly ±1/2 and a few ±1 disclinations appeared during the phase transition. After 5 s, some of the ±1 defects disappeared from the field of view (figure 3(b)). After 15 s, no textural change occurs in the
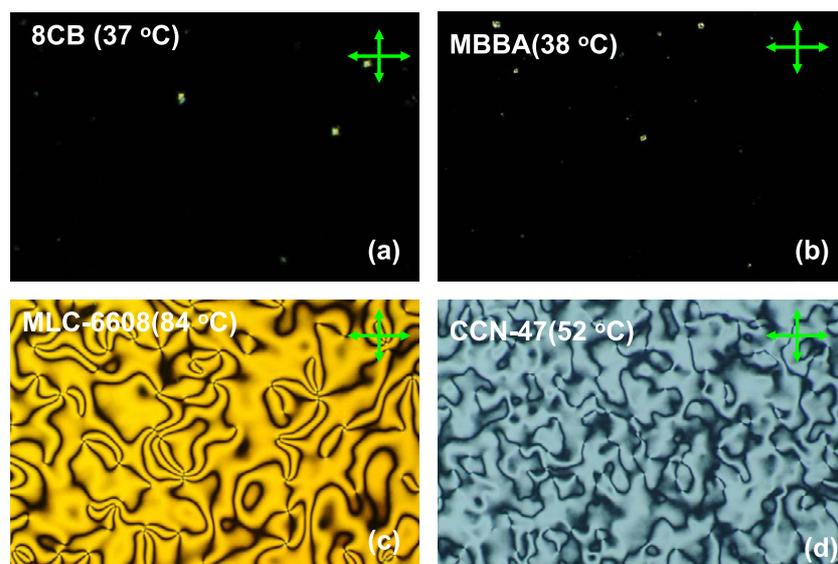

Figure 2. (a) Homeotropic texture of 8CB at 37 °C. (b) Homeotropic texture of MBBA at 38 °C. (c) Schlieren texture with 1 defects in MLC-6608 at 85 °C. (d) Schlieren texture with 1/2 defects in CCN-47 at 50 °C. Crossed arrows denote the crossed polarizers.
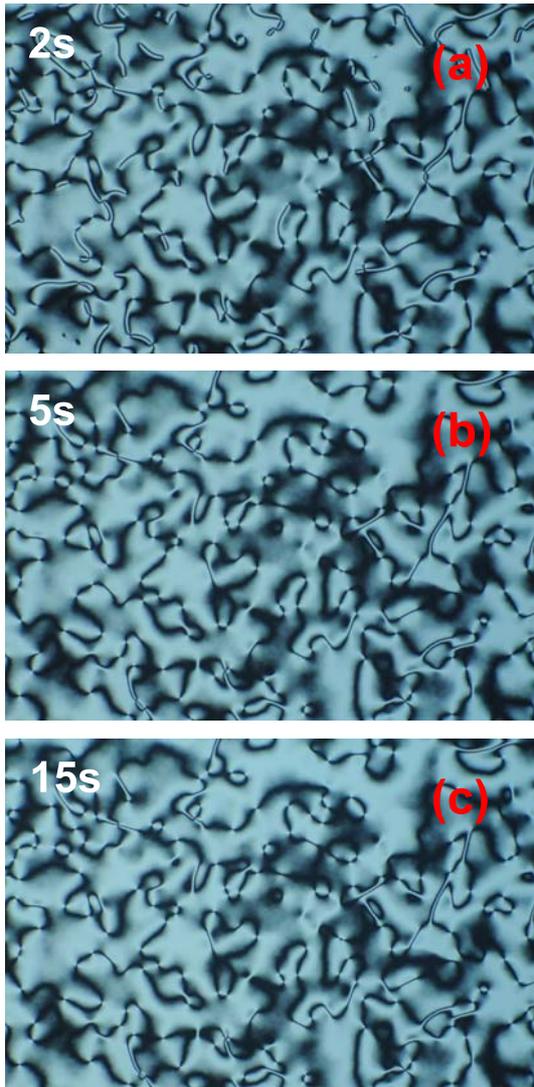
Figure 3. Textures observed under crossed polarizers in a 6 μm thick cell at (a) 2 s, (b) 5 s and (c) 15 s after the $T_{NI} = 57.3\,^{\circ}$C transition (cooling rate 0.2 °C).

cell (figure 3(c)). No noticeable change was observed even after several hours of waiting. This shows that the disclination network is very stable. The abundance of $\pm 1/2$ strength disclinations is sometimes regarded as evidence of a biaxial system [15], but we confirmed that the present medium is definitely uniaxial, as is clear from the completely dark view in the homeotropically aligned state. In the nematic phase of a lyotropic liquid crystal, similar texture was also observed [9]. The energy per unit length of an isolated disclination, from the simple one-elastic-constant approximation, can be written as $W = W_c + \pi K S^2 \ln(R/r_c)$ [16], where W_c is the energy per unit length of the core, R is the sample size, r_c is the radius of the core and S is the strength of the defect. Thus for a given sample size R, the disclination with strength ± 1 will cost four times the energy of those with strength $\pm 1/2$. However, experimentally, in the nematic phase ± 1 defects are more often seen in the literature [8, 16–19] and to our knowledge this is the first example of a case of thermotropic nematic liquid crystal exhibiting mostly $\pm 1/2$ strength defects. The physical

implication is that the director is in the plane, as any tilt of the director in such a case is topologically forbidden [19].

It is also clear from figure 3 that the interdefect separation is sufficiently large and we can identify the strength of the defect by the rotation of the polarizer. If the brashes rotate in the same direction as the rotation of the polarizer then it is assigned a positive defect; otherwise it is assigned a negative defect. In figure 4 we show a small part of the texture before and after rotating the polarizer by 20°. The open squares and open circles denote the defect with positive sign and negative sign respectively. We have taken about 25 images (textures) from the different parts of the sample at the same temperature $(T_{NI} - T = 0.5°)$. Each texture was divided into several small square regions of equal size. In each square, suppose there are n_+ of the $+1/2$ defects and n_- of the $-1/2$ defects. Then the total number of defects is $N = n_+ + n_-$ and the net defect strength is $\Delta n = n_+ - n_-$. The net defect strength in each square region was determined for the statistical analysis of all the 25 textures. For the present size of the square region we can get three different average values for the total of defect–antidefect pairs (N), namely $N = 8, 12$, and 16. The numbers of regions obtained in each case were about 96, 52 and 32 respectively. For the present size of the square, not enough data were available for statistical analysis with $N < 8$ or $N > 16$. In order to find the correlation we followed the procedure mentioned in [9]. For each square region the net defect strength (Δn), i.e. the number of defects minus the number of antidefects, is determined. Then the occurrence distribution, $f(\Delta n)$, for each Δn was determined. In figure 5, we show the variation of $f(\Delta n)$ versus Δn for $N = 8, 12$, and 16. A nonlinear least squares program is used to fit the following equation [8]:

$$f(\Delta n) = a \exp[-(\Delta n - \overline{\Delta n})^2/2\sigma^2] \qquad (1)$$

where σ is the variance of the distribution. Good Gaussian fittings are obtained for various values of N. The fit parameters with standard error are summarized in table 1. The maximum of the Gaussian is centered at zero and consistent with the fit parameter $\overline{\Delta n}$. The widths of the Gaussian distribution σ for $N = 8, 12$, and 16 are significantly different to those for the system with ± 1 defects as measured by Digal *et al* [8]. The σ values for our system are comparable to the values reported for $N = 10, 14$, and 18 in the case of lyotropic liquid crystals with $\pm 1/2$ by Mukai *et al* [9]. Interestingly our measured σ values are almost half the values predicted for $N = 10, 20$, and 30 for a square shaped elementary region. In order to find the correlation, the σ values were fitted with the following equation:

$$\sigma = CN^{\nu} \qquad (2)$$

where C is a constant and the exponent ν determines the correlation. The theoretical value of exponent ν is 1/2 and 1/4 in cases where the defects and antidefects are uncorrelated and correlated respectively. The width of the distribution (σ) for different N values and the fitting with equation (2) is shown in figure (6). For the present system we find $\nu = 0.29 \pm 0.08$ and $C = 0.35$. This is similar to the value reported by Mukai *et al* [9] and almost half of the value reported by Digal *et al* [8]. This difference with respect to the case of ± 1 defects may be attributed to the strength of the disclination.
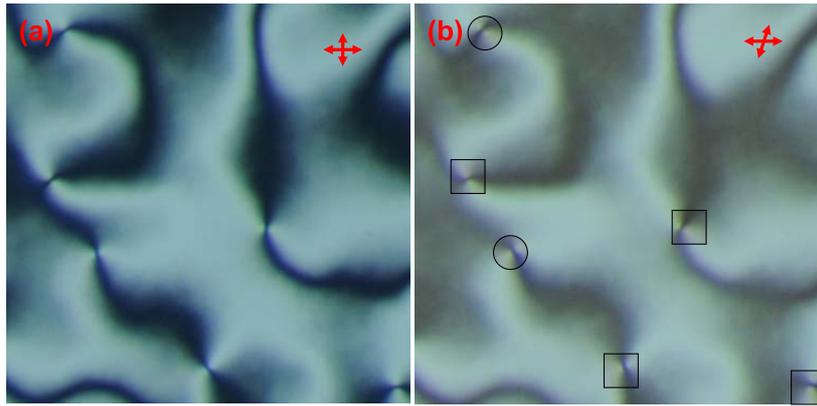
Figure 4. (a) Part of the texture obtained from figure 2 between the crossed polarizers at 53 °C. (b) Texture after rotating the crossed polarizers by an angle of 20°. Squares and circles represent the $+1/2$ and $-1/2$ defects, respectively.
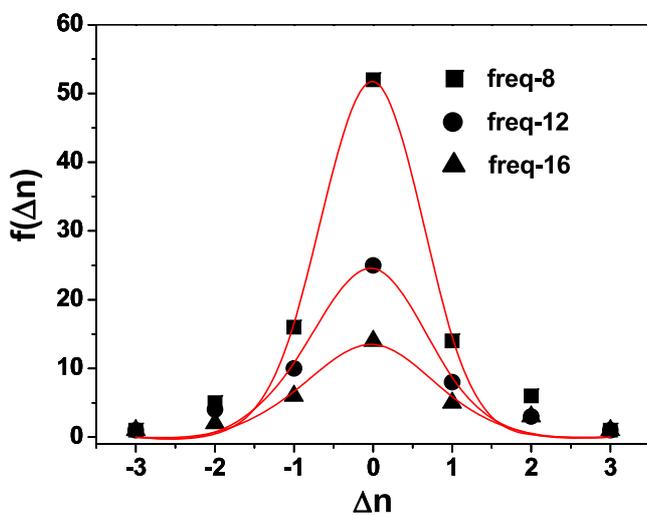

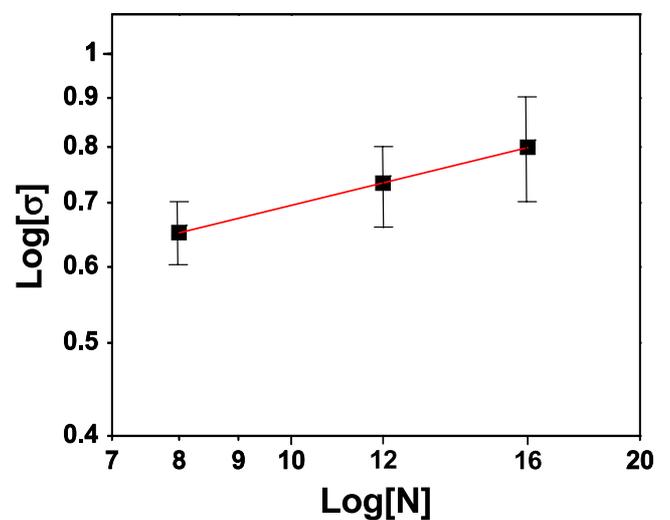**Figure 5.** Plot of the frequency $f(\Delta n)$ versus Δn.

Figure 6. A log–log plot for determining the critical exponent ν.

4. Conclusions

In conclusion, in the present work we reported that mostly $\pm 1/2$ defects can be stabilized over a large area in uniaxial nematic liquid crystal on surfaces treated with perfluoropolymer. We described the evolution of defects from the nematic–isotropic phase transition. We found that the defect–antidefect correlation exponent $\nu = 0.29$ and this is consistent with the Kibble mechanism [10] for the defect–antidefect correlation in the early Universe. This value is also consistent with the previous reports on uniaxial thermotropic liquid crystal with ± 1 and lyotropic liquid crystal with $\pm 1/2$ strength defects. The present study together with the results of [8] and [9] clearly suggests that irrespective of the strength of the defects and the experimental systems, the critical exponent of the defect–antidefect correlation is universal in nature. Commonly ± 1 defects are observed in NI quench studies. In the present case it is not yet clearly understood why mostly $\pm 1/2$ defects are observed in the present compound in cells treated with perfluoropolymers. However, the molecular dynamics studied by Billert *et al* [20] and recent computer simulations by Bradac *et al* [21] on the kinetics of the NI quench suggest that the point defects are

Table 1. Fit parameters obtained by fitting equation (1).

N	a	Δn	σ
8	51.7 ± 3.7	-0.02 ± 0.07	0.65 ± 0.05
12	24.6 ± 2.2	-0.06 ± 0.08	0.73 ± 0.07
16	13.5 ± 1.6	-0.04 ± 0.011	0.80 ± 0.10

much more mobile compared to the line defects, and easier to annihilate than the lines. It may be mentioned that the surface energy of perfluoropolymer is much smaller compared to the other surfaces [14], hence, the point defects are highly mobile on a perfluoropolymer surface and as a result they are annihilated easily. Therefore, more $\pm 1/2$ strength defects are expected in NI quench systems. A systematic measurement of the anchoring energy and the knowledge of some physical properties are needed for further understanding of this specific system. Finally we expect that the present study will inspire various other experiments on defect–antidefect formation and correlation.

References

[1] de Gennes P G 1993 *The Physics of Liquid Crystals* 2nd edn (Oxford: Clarendon)

[2] Dierking I 2003 *Textures of Liquid Crystals* (Weinhein: Wiley–VCH)

[3] Zurek W H 1985 *Nature* **317** 505
Dendry P C, Lawson N S, Lee R A, McClintock P V E and Williams C D H 1994 *Nature* **368** 315

[4] Chuang I, Durrer R, Turok N and Yurk B 1991 *Science* **251** 1336

[5] Bowick M J, Chandar L, Schiff E A and Srivastava A M 1994 *Science* **263** 943 (arXiv:hep-ph/9208233)

[6] Durrer R arXiv:astro-ph/0003363 e-print
Ray R and Srivastava A M 2004 *Phys. Rev.* D **69** 103525 (arXiv:hep-ph/0110165)

[7] Monaco R, Mygind J, Aaroe M, Rivers R J and Koshelets V P 2006 *Phys. Rev. Lett.* **96** 180604

[8] Digal S, Ray R and Srivastava A 1999 *Phys. Rev. Lett.* **83** 5030

[9] Mukai H, Fernandes P R G, de Oliveira B F and Dias G S 2007 *Phys. Rev.* E **75** 061704

[10] Kibble T W B 1976 *J. Phys. A: Math. Gen.* **9** 1387

[11] Dhara S and Madhusudana N V 2008 *Phase Transit.* **81** 561

[12] Dhara S, Kim J K, Jeong S M, Kogo R, Araoka F, Ishikawa K and Takezoe H 2009 *Phys. Rev.* E **79** 060701R

[13] Kim J K, Araoka F, Jeong S M, Dhara S, Ishikawa K and Takezoe H 2009 *Appl. Phys. Lett.* **95** 063505

[14] Jeong S M, Kim J K, Shimbo Y, Araoka F, Dhara S, Ha N Y, Ishikawa K and Takezoe H 2009 *Adv. Mater.* **21** 1

[15] Chandrasekhar S *et al* 1998 *Liq. Cryst.* **24** 67

[16] Chaikin P M and Lubensky T C 1998 *Principles of Condensed Matter Physics* (Cambridge: Cambridge University Press)

[17] Chandrasekhar S 1977 *Liquid Crystals* (Cambridge: Cambridge University Press)

[18] Brown G H (ed) 1975 *Advances in Liquid Crystals* vol 1 (New York: Academic) p 267

[19] Madhusudana N V and Pratibha R 1982 *Curr. Sci.* **57** 877

[20] Billert J L, Smondyrev A M, Loriot G B and Pelcovits R A 1999 *Phys. Rev.* E **60** 6831

[21] Bradac Z, Kralj S and Zumer S 2002 *Phys. Rev.* E **65** 021705