



Homeotropic alignment of nematic liquid crystals with negative dielectric anisotropy

G. Singh^a, G. Vijaya Prakash^a, A. Choudhary^b, A.M. Biradar^{b,*}

^a Nanophotonics Laboratory, Department of Physics, Indian Institute of Technology, New Delhi 110016, India

^b National Physical Laboratory, Dr. K. S. Krishnan Road, New Delhi 110012, India

ARTICLE INFO

Article history:

Received 5 October 2009

Received in revised form

5 January 2010

Accepted 22 January 2010

Keywords:

Liquid crystals

Negative dielectric anisotropy

Homeotropic alignment

Optical transmission

ABSTRACT

We report here uniform and defect-free homeotropic alignment of nematic liquid crystal having negative dielectric anisotropy ($\Delta\epsilon < 0$) on cleaned substrates, which can be changed to bright state (homogeneous alignment) by applying an electric field. However, non-uniformity in the bright state would not be desirable for display devices. Therefore, silane treated and gently rubbed substrates have been utilised to obtain uniform bright state. Several textural, dielectric, and optical transmission studies have been carried out to explore the applicability of studied material in the liquid crystal display applications. Furthermore, such materials could also be useful for making colourful displays without colour filters.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Since last few decades, nematic liquid crystals (NLCs) have shown profound impact on versatile display applications. The key point for all possible applications is the alignment of liquid crystal molecules (i.e. the director) on the substrates [1,2]. Mainly three types of alignments such as homogeneous, homeotropic and hybrid are being used for various liquid crystal electro-optical applications. Recently homeotropic alignment of liquid crystals has got worldwide applications in liquid crystal displays such as high information display devices, large area LCD TVs, and digital display devices in the medical field like digital medical imaging [3], due to their unprecedented contrast ratio and wide viewing angle characteristics. Moreover, the contrast ratio is insensitive to the incident light wavelength, liquid crystal layer thickness, and operating temperature of the device [4,5].

Silanes or long alkyl side chain alcohols [6] and oblique evaporation of silicon monoxides [7] have been extensively utilised for homeotropic alignment of NLCs. Moreover, nanoparticle induced homeotropic alignment studies [8,9] has also been recently reported. Although several studies [10–14] have been carried out to understand the mechanism of such alignment it is rather unclear. In the recent past, few reports [15,16] have explored the possibility of homeotropic alignment of nematic liquid crystals in simply cleaned substrates such as glass, oxides,

and metals. But poor reproducibility and uniformity of such alignments have limited their applications to provide reliable liquid crystal electro-optic devices. To the best of our knowledge, no further study has been reported regarding homeotropic alignment with better reproducibility and uniformity in simply cleaned substrates.

Here, we report the achievement of reproducible uniform and defect-free homeotropic alignment of negative dielectric anisotropic NLC on cleaned bare indium tin oxide (ITO) patterned glass substrates. Further, silane treated and gently rubbed substrates have been used for the homeotropic alignment, where uniformity of alignment is preserved even in the bright state after the bias field application unlike untreated substrates. Several textural, dielectric, and optical transmission studies have been carried out to evaluate the monodomain nature of the fabricated NLC cells from treated substrates and to reveal the applicability for display applications. Further, we also proposed that such materials could also be exploited in the fabrication of colourful displays devoid of colour filters.

2. Experimental details

In this study, nematic liquid crystal (MLC6608, Merck) has been used having physical parameters $\Delta\epsilon = -4.2$, optical anisotropy (Δn) = 0.0830, and nematic to isotropic transition temperature = 90 °C. Highly conducting (10–18 Ω/\square) and optically transparent $\lambda/2$ sputtered ITO glass substrates have been utilised to fabricate treated and untreated type of liquid crystal cells.

* Corresponding author. Tel.: +91 11 45608569.

E-mail address: abiradar@mail.nplindia.ernet.in (A.M. Biradar).

The substrates simply cleaned with soap solution followed by acetone, have been used for making untreated cells. However, treated cells have been prepared by dipping substrates in silane solution (phenyl trichlorosilane:toluene: 1:100) for 6–7 min, washing in isopropanol to remove the adsorbed silane molecules, baking at 90 °C for 20 min and then gentle rubbing (~ 10 – 12 strokes) with velvet cloth. To cross check for the homeotropic alignment in an untreated cell, we have also experimented with optically flat, $\lambda/2$ glass substrates and fluorinated SnO_2 coated glass substrates without any further treatment. The thickness of liquid crystal cells has been maintained by using spacers of thickness of 6–12 μm . The filling of cells has been done at isotropic temperature of the material through capillary action. The optical micrographs (i.e. texture) observations of both (treated/untreated) type cells with different bias voltages under high-resolution crossed polarizing microscope (Carl Zeiss, Axioskop 40) have been recorded. The complex dielectric permittivity measurements have been carried out by using computer controlled HP 4192A impedance analyzer in the frequency range (100 Hz–1 MHz). For optical transmission measurements, NLC cell attached with sample holder has been kept under crossed polarizing system. The light from He–Ne laser ($\lambda=633$ nm, 15 mW) incident normally to the sample and transmitted intensity has been monitored with Si-detector attached with Optometer (UDT-S370) and an Oscilloscope (Tetronix TDS 1001B) for steady-state as well as time-resolved transmissions, respectively. As an AC signal (0–10Vpp @ 1 Hz) bias source, function generator (GWINSTEK GFG-3015) has been used. For spectral domain ($\lambda=300$ – 800 nm) measurements, halogen lamp and spectrophotometer have been employed.

3. Results and discussions

The optical micrographs of homeotropically aligned untreated liquid crystal cell of thickness 6 μm at different bias voltages at room temperature are shown in Fig. 1. From Fig. 1(a), it is clear that cell is perfectly homeotropically aligned, since it shows perfect dark state under crossed polarizers at 0V bias and even

after rotating the cell. When we apply the bias voltage ($\geq \sim 2.5$ V, threshold value), the cell switches from dark (i.e. OFF) state to bright (i.e. ON) state due to the negative dielectric anisotropy of the material and after removing the bias voltage, it quickly goes back to its original dark state, i.e. switching of the cell from ON to OFF state. The contrast of the cell seems to be quite noticeable because of its perfect dark state in both cases, i.e. at zero bias and after removal of the bias also. However, the cell does not retain its monodomain orientation in the bright state, which could be hindrance for its use in good contrast display applications. The reason for getting homeotropic alignment in the untreated substrates could be that surface energy (due to the intermolecular interaction among liquid crystal molecules) is more than the substrate surface energy. However, after the bias field application the non-uniformity in the bright state might be due to the reduction of liquid crystal surface energy as compared to substrate surface energy unlike unbiased case.

To overcome this problem, we have used silane treated and gently rubbed substrates for the homeotropic alignment. Optical micrographs of such treated cell of thickness 6 μm with different bias voltages at room temperature are shown in Fig. 2. Here, one can see clearly that such sample retains its monodomain nature in both dark/bright states. In addition to the remarkable optical contrast, the cell also shows a particular colour at particular bias voltage and which could be due to electronically controlled birefringence [4]. The generation of colour may replace the colour filter used in colourful displays by applying an individual voltage to each pixel corresponding to the desired colour.

Dielectric relaxation spectroscopy [17] has also been considered to be one of the prominent characterizing tools in liquid crystals that reveal molecular information such as molecular and collective relaxations depending on alignment. Therefore, we have carried out complex dielectric permittivity measurements at room temperature to show the OFF and ON mechanism in these treated NLC cells. Fig. 3(a) shows the room temperature frequency dispersion of real part of dielectric permittivity (ϵ') at different bias voltages (0–10V). At 0V bias the magnitude of permittivity is very small and after the application of bias more than or equal to threshold value, its magnitude increases due to the molecular

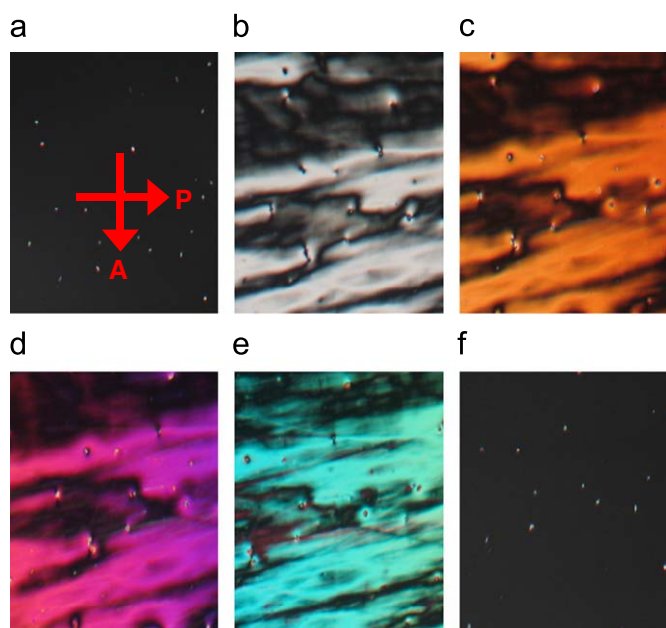


Fig. 1. (Colour online) Optical micrograph (magnification of $50\times$) of homeotropically aligned nematic liquid crystal cell (without any surface treatment) at room temperature at (a) 0V, (b) 3V, (c) 4V, (d) 4.5V, (e) 10V, and (f) 0V after 10V.

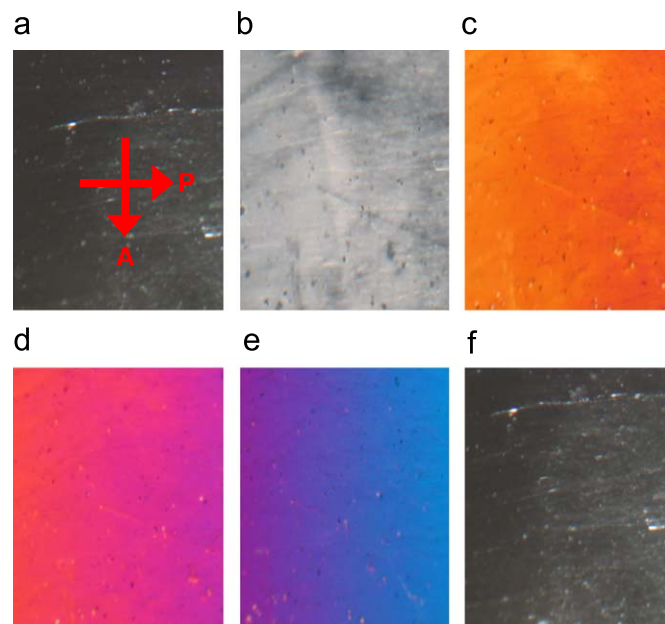


Fig. 2. (Colour online) Optical micrograph (magnification of $50\times$) of homeotropically aligned nematic liquid crystal cell (silane treatment with gentle rubbing) at room temperature at (a) 0V, (b) 2.5V, (c) 4.5V, (d) 6V, (e) 10V, and (f) 0V after 10V.

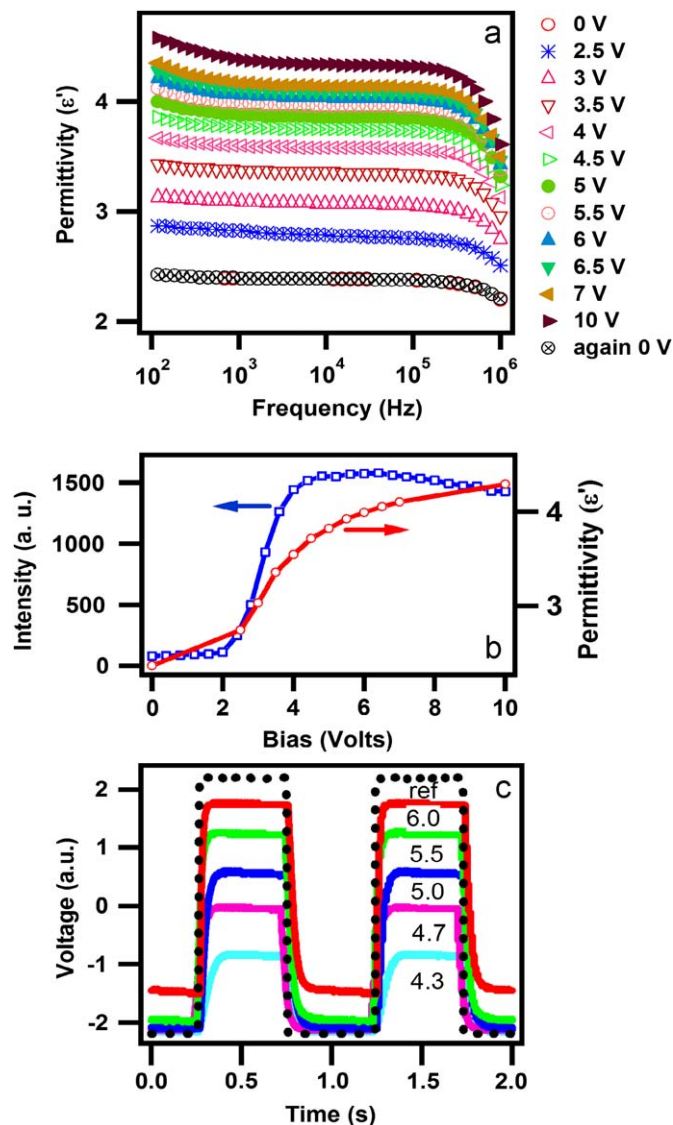


Fig. 3. (Colour online) At room temperature (a) frequency dependent dielectric permittivity (ϵ') at different bias voltages, (b) dielectric permittivity (ϵ') versus bias voltages at 20 KHz and steady-state (square wave 0–10 V @ 1 KHz) optical transmission at $\lambda=633$ nm, and (c) time-resolved (ac bias 4.3–6.0 Vpp @ 1 Hz) optical transmission with white light.

realignment from homeotropic to homogeneous geometry, i.e. switching from OFF to ON state. The permittivity at 10 V bias (higher than threshold value) overlaps with 0 V bias permittivity just after removing the bias field, which confirms the switching from ON to OFF state. The variation of permittivity with bias voltages (0–10 V) at 20 KHz frequency is shown in Fig. 3(b). The monotonous increment in the permittivity with bias voltage again confirms the switching of liquid crystal cell from homeotropic to homogeneous alignment. Thus the dielectric spectroscopy also gives supportive evidences for its use in display applications.

Furthermore, we have carried out voltage dependent optical transmission measurements of the same cell at room temperature to study its switching threshold voltage and the optical contrast. Steady-state optical transmission at $\lambda=633$ nm is shown in Fig. 3(b). It is obvious from Fig. 3(b) that the transmission intensity remains almost constant with minimum value up to bias field less than threshold voltage (~ 2.1 V), i.e. perfect dark state and after that increases continuously up to certain bias and then become almost stagnant. As we know that one can also compute the elastic constant (k_{33}) with known experimental threshold

voltage (V_{th}) value of investigated NLC by using the following equation:

$$E_{th} = \frac{\pi}{d} * \sqrt{\frac{k_{33}}{\epsilon_0 |\Delta\epsilon|}} \quad (1)$$

where E_{th} is the threshold electric field, d is the sample thickness, k_{33} is the elastic constant, ϵ_0 is the electric permittivity of the free space and $\Delta\epsilon$ is the dielectric anisotropy.

The above equation can also be rewritten as

$$V_{th} = \pi * \sqrt{\frac{k_{33}}{\epsilon_0 |\Delta\epsilon|}} \quad (2)$$

The V_{th} values have been computed in untreated cell (~ 2.5 V) and in surface treated cell (~ 2.1 V) from Figs. 1 and 3(b), respectively, and found that threshold voltage in case of untreated cell is slightly higher than the surface treated cell. Further, the elastic constant (k_{33}) has also been computed in both untreated as well as surface treated cells from Eq. (2) by using $\epsilon_0=8.86*10^{-12} C^2/N\cdot m^2$, $\Delta\epsilon=-4.2$, and computed V_{th} values. The computed value of elastic constant (k_{33}) in untreated cell (~ 74 pN) is found more than the surface treated cell (~ 52 pN). Moreover, elastic constant (k_{33}) follows the same trend as threshold voltage (V_{th}) in both untreated as well as surface treated cells.

The time-resolved optical transmission with white light is shown in Fig. 3(c), for qualitative description of optical contrast of the liquid crystal cell. As is seen in figure, the magnitude of electro-optical response increases with bias voltages but at the same time after certain bias the contrast of the cell starts to diminish. In other words, the amount of light leakage in the dark state of the cell under crossed polarizers after certain bias field (~ 5.5 Vpp) increases in comparison with the light leakage below this field. The reason could be that when we apply higher bias voltage pulse, the director tilt angle has been increased from minimum to a certain maximum value but as the pulse goes to zero then not necessarily all the bulk molecules will come to their minimum tilt position in the time up to which pulse remains zero. Therefore, in device applications the maximum contrast of the device can be achieved by selecting appropriate voltage range. Overall, such optical transmission studies also confirm for switching of the cell OFF–ON–OFF mechanism with good optical contrast for selected range of voltages.

4. Conclusion

We have demonstrated here the achievement of homeotropic alignment of studied negative dielectric anisotropic liquid crystal on substrates without any surface treatment and speculated that it would be material's intrinsic behaviour. To exploit intrinsic homeotropic aligning nature of such material, silane treated and gently rubbed substrates have been utilised instead of bare substrates, i.e. to freeze the monodomain even in the bright state. The stronger surface energy due to intermolecular interaction between liquid crystal molecules than substrate surface energy may foster the homeotropic alignment in the untreated substrates. However, the exact mechanism for such alignment is still unclear. Several textural, dielectric, and electro-optical transmission studies reveal the applicability of such material in the liquid crystal display devices. Furthermore, such material may also replace colour filters used in colourful displays due to its capability to generate a particular colour at a particular bias voltage. This study would also be very much inspiring for liquid crystal chemists to synthesize such intrinsic homeotropic aligning negative dielectric anisotropic materials. Future investigations to understand the reasons for getting homeotropic alignment with-

out any surface treatment and how to freeze the monodomain of alignment even in the bright state are under way.

Acknowledgements

The authors sincerely thank Dr. Vikram Kumar, Director, National Physical Laboratory, for continuous encouragement and interest in this work. The authors are also thankful to Mr. K. Pradeesh, J. Prakash, and A. Kumar for fruitful discussions. The authors (G.S. and A.C.) are thankful to Council of Scientific and Industrial Research (CSIR), New Delhi, India, for financial assistance.

References

- [1] P.G. de Gennes, J. Prost, *The Physics of Liquid Crystals*, Clarendon, Oxford, 1993.
- [2] K. Takato, M. Hasegawa, M. Koden, N. Itoh, R. Hasegawa, M. Sakamoto, *Alignment Technologies and Applications of Liquid Crystal Devices*, Taylor & Francis, New York, 2004.
- [3] K.P. Andriole, *J. Amer. College Radiol.* 2 (2005) 543.
- [4] E. Lueder, *Liquid Crystal Displays*, Wiley, New York, 2001.
- [5] S.T. Wu, D.K. Yang, *Reflective Liquid Crystal Displays*, Wiley, New York, 2001.
- [6] J.L. Janning, *Appl. Phys. Lett.* 21 (1972) 173.
- [7] K. Hiroshima, *Japan. J. Appl. Phys.* 21 (1982) L761.
- [8] S.C. Jeng, C.W. Kuo, H.L. Wang, C.C. Liao, *Appl. Phys. Lett.* 91 (2007) 061112/1.
- [9] C.W. Kuo, S.C. Jeng, H.L. Wang, C.C. Liao, *Appl. Phys. Lett.* 91 (2007) 141103/1.
- [10] I. Haller, *Appl. Phys. Lett.* 24 (1974) 349.
- [11] S. Naemura, *J. Appl. Phys.* 51 (1980) 6149.
- [12] K. Okano, N. Matsuura, S. Kobayashi, *Japan. J. Appl. Phys.* 21 (1982) L109.
- [13] E.F. Luk'yanchenko, V.A. Kozunov, V.I. Griogs, *Russian Chem. Rev.* 54 (1985) 214.
- [14] T. Beica, R. Moldovan, I. Zgura, S. Frunza, M. Poterasu, *J. Optoelectron. Adv. Mater.* 8 (2006) 1512.
- [15] J. Cognard, *Molecular Cryst. Liq. Cryst. Suppl. Ser. 1* (1982) 1–78.
- [16] M. Macchione, G.D. Filpo, F. Iemma, F.P. Nicoletta, N. Picci, G. Chidichimo, *Molecular Cryst. Liq. Cryst.* 363 (2001) 137.
- [17] W. Haase, S. Wrobel, *Relaxation Phenomena, Liquid Crystals, Magnetic Systems, Polymers, High-Tc Superconductors, Metallic Glasses*, Springer, Germany, 2003.