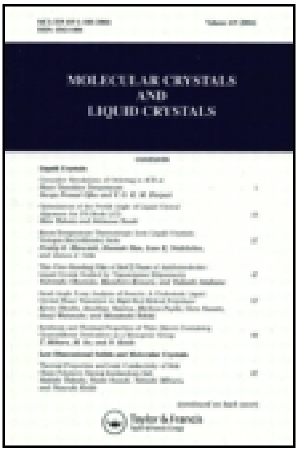

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Sudhaker Dixit [a], Abhishek Kr. Misra [a], Rajiv Manohar [a] & A. K. Arora [b]

[a] Liquid Crystal Research Laboratory, Physics Department, University of Lucknow, Lucknow, India

[b] IGCAR, Department of Atomic Energy, Kalpakkam, Tamilnadu, India

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Enhancement in Dielectric Properties of Nematic Liquid Crystal by Gamma Irradiation

SUDHAKER DIXIT,[1] ABHISHEK KR. MISRA,[1] RAJIV MANOHAR,[1,*] AND A. K. ARORA[2]

[1]Liquid Crystal Research Laboratory, Physics Department, University of Lucknow, Lucknow, India
[2]IGCAR, Department of Atomic Energy, Kalpakkam, Tamilnadu, India

A gamma irradiation technique has been used to alter the dielectric properties of a liquid crystal system. The present paper reports a comparative dielectric study of a gamma-irradiated and an unirradiated nematic liquid crystal. This study is an attempt to see the irradiation effect in the alignment and dielectric parameters of a nematic liquid crystal. The dielectric measurements have been done with the temperature variation in the frequency range 100 Hz to 10 MHz. The dielectric data show relaxation modes that follow Cole-Cole theory. The Cole-Cole plots have been used to determine the dielectric parameters such as relaxation frequency and relaxation strength. The dielectric permittivity is found to increase for the gamma-irradiated sample as compared with the unirradiated sample with variation in temperature and frequency and well explained in this paper.

Keywords Dielectric parameter; electrical conductivity; gamma radiation; relaxation frequency

1. Introduction

Nematic liquid crystals (NLCs) consist of asymmetric molecules that tend to align in a common local direction described by the director. Smectic liquid crystals (LCs) usually do not respond to applied electric and magnetic fields as easily as NLCs, and so attempts are continuously being made to enhance the properties of these LCs from application point of view [1–3]. In addition to this, NLCs have long-range orientational order, which gives rise to many properties important for LC displays [4–8]. It is known that dielectric studies of liquid crystalline materials are a valuable source of information on their molecular arrangement, molecular dynamics, and specific intermolecular interactions, in both mesomorphic and isotropic phases [9,10].

Heavy ions of various energies are being used for material modifications. These induced modifications depend on defect in the material during interaction of ions with the target material [11]. Absorption of gamma rays by LC may cause physical conformational changes due to thermal and thermomechanical effects. These physical changes may cause scattering of light, changes in transmission and reflection properties of filters and coatings. Early

*Address correspondence to Rajiv Manohar, Liquid Crystal Research Laboratory, Physics Department, University of Lucknow, Lucknow 226007, India. Tel: +91-9415000687. E-mail: rajiv.manohar@gmail.com

LC phase retarders were screened at Raytheon for radiation sensitivity circa 1989 under the Airv Force Research Lab (AFRL) beam agility technique program (F33615-87-1507). Phase retarders were used as surrogates for optical phased arrays and subjected to increasing gamma ray dose from a cobalt-60 source, up to a total dose of 9.5 Mrad. It was the first-known radiation testing of LC [12,13].

Dielectric studies of the LCs are important as they provide useful information about molecular structure, molecular dynamics, phase transition, and display performance of LCs [14]. Most of the dielectric studies on LCs are concentrated in the nematic phase and usually examine the nematic to isotropic phase transition. The optical characteristics, threshold voltages, and switching times of LCs are strongly dependent on the absolute value of their dielectric permittivity. Dielectric relaxation studies provide one of the few techniques for finding the nature of molecular reorientation within a system. Dielectric studies of a large number of thermotropic liquid crystalline substances have been conducted, and thus dielectric spectroscopy is found to be one of the best techniques for measuring dielectric permittivity and dielectric losses with high accuracy and sensitivity [15–17].

Therefore, to observe the effect of gamma radiation on NLC materials, we have applied gamma irradiation technique. This is a powerful technique for investigations of condensed matter. Relaxation of different physical origin such as molecular reorientation, dynamics of collective or surface polarization modes, and conductivity can be investigated in different LC systems. Detailed qualitative and quantitative information characterizing these processes has been obtained by this technique. In fact, we have attempted to investigate how this irradiation affects in the alignment of the sample and consequently how it affects the dielectric parameters.

2. Experimental Details

2.1. Material

The LC sample under investigation is a rod-shaped molecule with a cyano group at one end, which makes it a highly polar molecule. The NLC is 4 Cyano-4′-pentylbiphenyl (5CB), and the molecular structure and transition temperatures are shown in Fig. 1.

2.2. Preparation of Cells

Two similar cells having active areas of 25 mm^2 (sheet resistance and the visible light transmission are 10 Ω/mm^2 and more than 90%, respectively) were prepared by using transparent and highly conducting ITO (indium tin oxide, Diamonds Coating UK) coated optically flat glass substrates used as electrodes. These electrodes give a base to the LC sample to align. Planar alignment is obtained by treating with both an adhesion promoter and

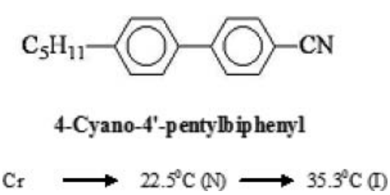

Figure 1. Chemical structure of 5CB.

a polymer (Nylon 6/6) and then rubbed unidirectionally with a velvet cloth. The thickness of the cell was maintained at 5 μm by means of mylar spacers. The complete preparation of cells has been given in our earlier papers [18]. The correct and proper alignment of the LC molecules is extremely important for precise measurement of electrical properties, which in turn influences the dielectric parameters and thus plays an extremely important role in molecular geometry.

2.3. Gamma Ray Treatment

We irradiated the 5CB LC as well as a blank planar cell. The irradiation used a ^{60}Co source, at the dose rate of 2.9 kGy/h, up to a total dose for 34.5 h is 100 kGy.

2.4. Dielectric Permittivity Study

The dielectric behavior of the material has been studied by using a computer-controlled impedance/gain phase analyzer (Hewlett Packard HP 4194 A). The dielectric parameters have been measured as a function of temperature and frequency. In order to vary the temperature of the sample holder, a microprocessor-based heating device Instec hot plate (HCS-302, INSTEC U.S.A.) with an accuracy of $\pm 0.1^\circ$C has been used. Before taking measurements, the sample was left for 15 min at a particular temperature.

3. Results and Discussion

The live capacitance value of empty irradiated planar cell was measured, and it was found to be flippantly lower (approximately 2%) than that of the unirradiated empty sample cell. The difference of live capacitance is very small for gamma-irradiated and unirradiated cell within the scope of experimental error. It is not expected to affect the dielectric properties of the LC material. The dielectric data have been analyzed by well-known Cole-Cole dispersion Equation (1) given by:

$$\varepsilon^* = \varepsilon'(\infty) + \frac{\delta\varepsilon'}{1 + (j\omega\tau)^{(1-\alpha)}} \tag{1}$$

Here $\delta\varepsilon'$ is the dielectric strength of the material, $\varepsilon'(\infty)$ is the high frequency limit of dielectric permittivity, ω $(= 2\pi f)$ is the angular frequency,τ is the relaxation time, and α is the distribution parameter. A value of the distribution parameter of more than 0.5 suggests the existence of more than one relaxation process.

The Cole-Cole plot of the sample, which exhibits nematic phase, has been drawn for both gamma-irradiated and unirradiated sample cell, filled with 5CB, at a specific temperature of 29°C as shown in Fig. 2; the black legend shows experimental data, while the solid red line shows the best theoretical fitting of the Cole-Cole equation into the experimental data. Using such plots, dielectric parameters such as relaxation frequency and relaxation strength were calculated, and their temperature dependence has been discussed in the later part of the paper.

On separating real and imaginary parts of Equation (1), one may get after adding high and low frequency correction parameters

$$\varepsilon'(\omega) = \varepsilon'(dc)f^{-n} + \varepsilon'(\infty) + \frac{\delta\varepsilon'[1 + (\omega\tau)^{(1-\alpha)}\sin(\alpha\pi/2)]}{1 + (\omega\tau)^{2(1-\alpha)} + 2(\omega\tau)^{(1-\alpha)}\sin(\alpha\pi/2)} \tag{2}$$

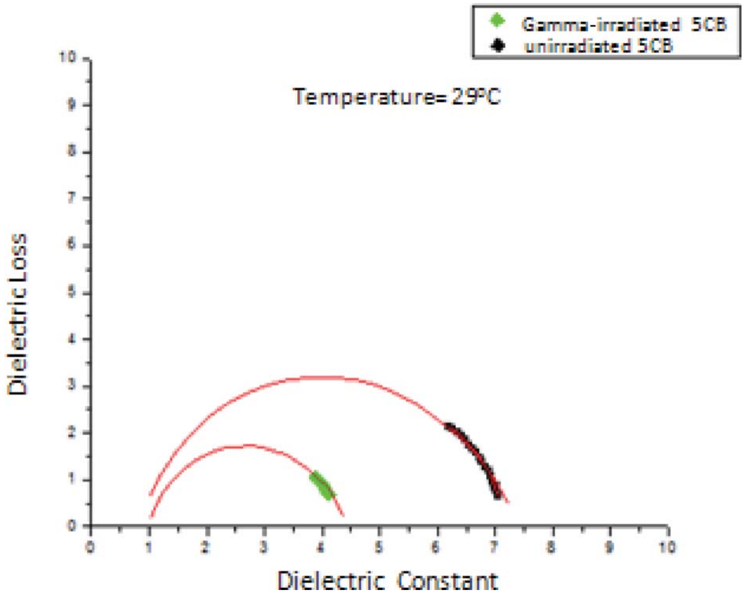

Figure 2. Cole-Cole plot for gamma-irradiated and unirradiated 5CB. (Color figure available online).

and

$$\varepsilon''(\omega) = \frac{\sigma(dc)}{\varepsilon_0 \omega^k} + \frac{\delta\varepsilon'(f\tau)^{(1-\alpha)}\cos(\alpha\pi/2)}{1 + (\omega)^{2(1-\alpha)} + 2(\omega)^{(1-\alpha)}\sin(\alpha\pi/2)} + Af^m \tag{3}$$

where σ (dc) is the ionic conductance, ε_0 is the free space permittivity and f is the frequency, while n, m, and k are the fitting parameters. The term $\varepsilon'(dc)f^{-n}$ and $\sigma(dc)/\varepsilon_o 2\pi f^k$ are added in equations for low frequency effect due to the electrode polarization, capacitance, and ionic conductance. The term Af^m is added in Equation (3) for high frequency effect due to the ITO sheet resistance and lead inductance of the cell. By the least square fitting of above equation into experimental data, we have removed the low and high frequency errors. After adding correction terms, the complex dielectric permittivity has been plotted against natural log of frequency as shown in the Fig. 3.

The observed value of complex dielectric permittivity for both irradiated and unirradiated 5CB sample, nature of variation with frequency is same [19], but the values have increased for gamma-irradiated 5CB sample. This increment in the values of dielectric permittivity for gamma-irradiated sample could be explained on the basis of physicochemical [20] change in the LCs. Actually, irradiation causes a chemical change in LCs; it may include cross linking, chain scission, formation of alkyl groups, and depletion of hetero atoms. The effect of gamma irradiation or such other ionizing radiation is primarily chain scission. Therefore, many physical and chemical properties can show modification with gamma irradiation. Radiation mainly affects in two basic ways, both resulting with excitation or ionization of atoms. In this fashion, ionic conductivity of LC material has also been investigated. The ionic conductivity for the irradiated 5CB sample is comparatively higher than that of the unirradiated 5CB sample. This increment in ionic conductivity between irradiated and unirradiated samples has been shown with the variation of temperature in Fig. 4. When the 5CB cell is gamma irradiated, the irradiation causes a chemical

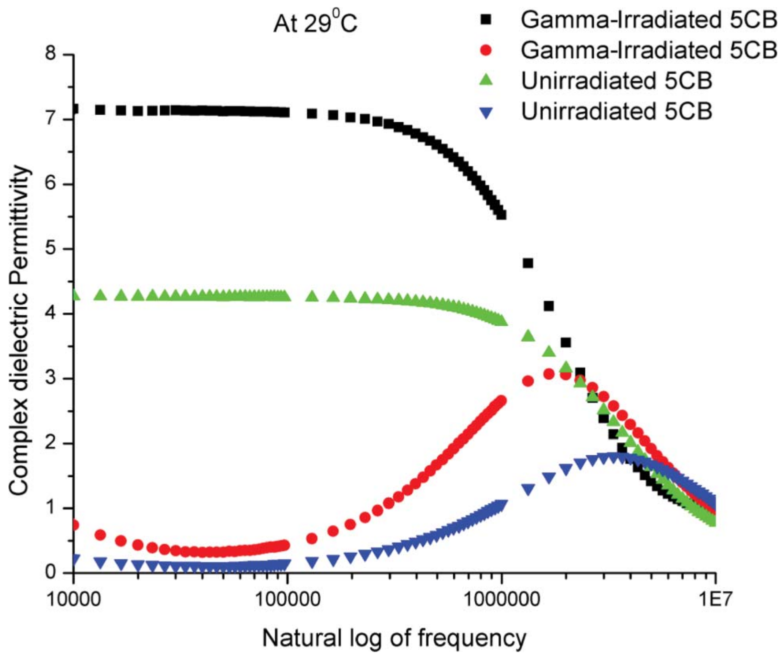

Figure 3. Complex dielectric permittivity with respect to natural log of frequency for gamma-irradiated and unirradiated 5CB. (Color figure available online).

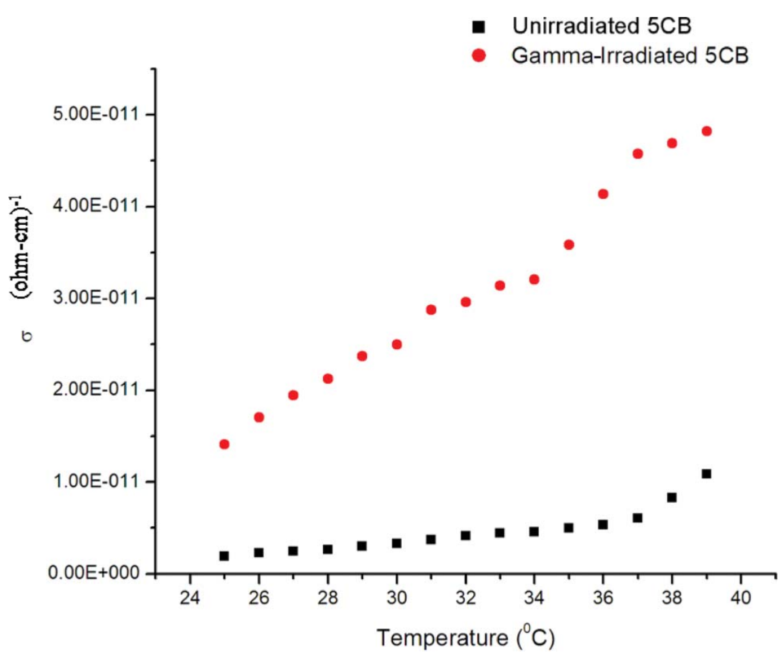

Figure 4. Ionic conductivity with respect to temperature for gamma-irradiated and unirradiated 5CB. (Color figure available online).

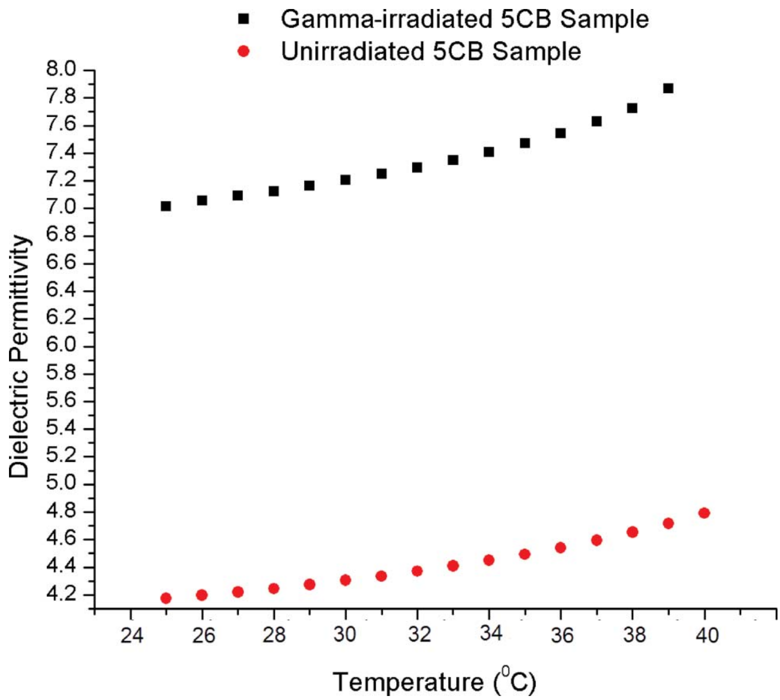

Figure 5. Dielectric permittivity with respect to temperature for gamma-irradiated and unirradiated 5CB. (Color figure available online).

change in the 5CB, which results is the generation of new ions. The influence of the ions on the dielectric properties of NLCs has been well explained by many authors because of their technological importance. The theoretical analysis of the problem usually contains only a group of cations and anions, with equal or different diffusion coefficients. The extension of theory to the case where several groups of ions are dissolved in NLC is important from the practical point of view because more than one type of impurities is usually present in the NLCs, which intern affects the creation of ions that are responsible for the electrical parameter. These ions play an important role in the formation of charge carriers in the nematic LC. The presence of positive ions due to gamma irradiation causes the change in the conductivity of LC cell. It can be said that such a trend in the dielectric permittivity is due to the modification in 5CB molecule as well as substrate because of high-energy gamma irradiation. This increase in permittivity for the irradiated sample has also been observed at all temperatures under investigation, as shown in Fig. 5.

Variation of different dielectric parameters (distribution parameter, relaxation time, and relaxation strength) for gamma-irradiated and unirradiated 5CB sample at constant temperature 29°C has been presented in Table 1. The value of relaxation time and relaxation strength is increased, while value of distribution parameter is decreased for gamma-irradiated sample as compared with unirradiated sample.

Another important dielectric parameter on which we have concentrated is relaxation frequency with variation in temperature shown in Fig. 6. The relaxation frequency has been extracted from the theoretical fittings of the Cole-Cole equation to experimental results, and its temperature dependence has been plotted for both irradiated and unirradiated samples.

Table 1. Variation of different dielectric parameters for gamma-irradiated and unirradiated 5CB sample at constant temperature (29°C)

Sample (5CB)	Distribution parameter (α)	Relaxation time (τ in s)	Relaxation strength ($\delta\varepsilon$)
Irradiated	0.00721	5.37E-07	6.10605
Unirradiated	0.00866	3.42E-07	3.3358

It is clear from the figure that the relaxation frequency shifts toward the lower side for the gamma-irradiated sample, and this relevant decrease occurring in the nematic phase and the relaxation process can be interpreted in the terms of gamma-irradiation effect on the sample. As discussed earlier that as soon as the cell is irradiated, this results in creation of ions that are responsible for currents through the LC. The presence of positive ions in 5CB that govern conductivity in the LC cell gradually affects the molecular reorientations. Due to this reason, each molecule moves in time as a sequence of small angular steps caused by collisions with its surroundings and under the influence of a potential of mean-torque setup by these molecules, decreases the relaxation frequency. Figure 7 shows variation of relaxation strength with temperature for both gamma-irradiated and unirradiated 5CB sample. From the figure, we can see that after gamma irradiation on 5CB, the value of relaxation strength increases as compared with the unirradiated 5CB sample. The reason is same as earlier discussion of relaxation frequency with variation in frequency.

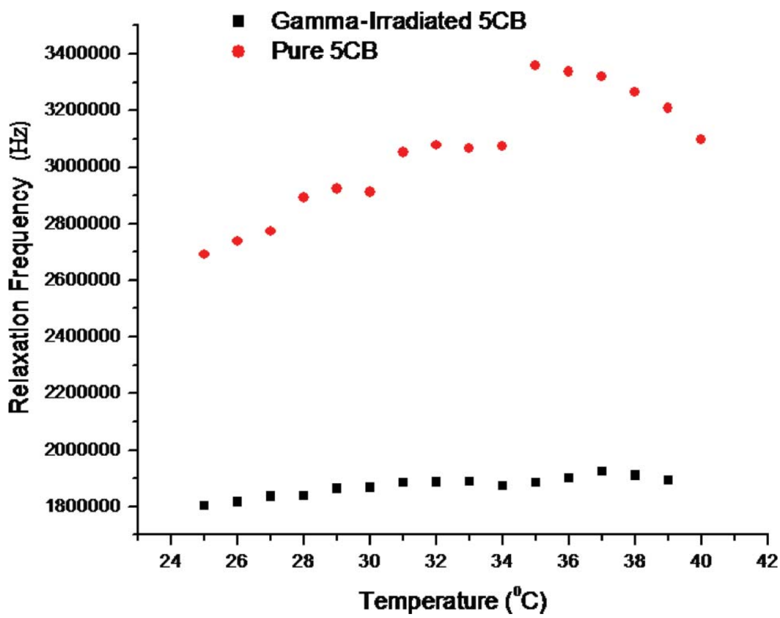

Figure 6. Relaxation frequency with respect to temperature for gamma-irradiated and unirradiated 5CB liquid crystal. (Color figure available online).

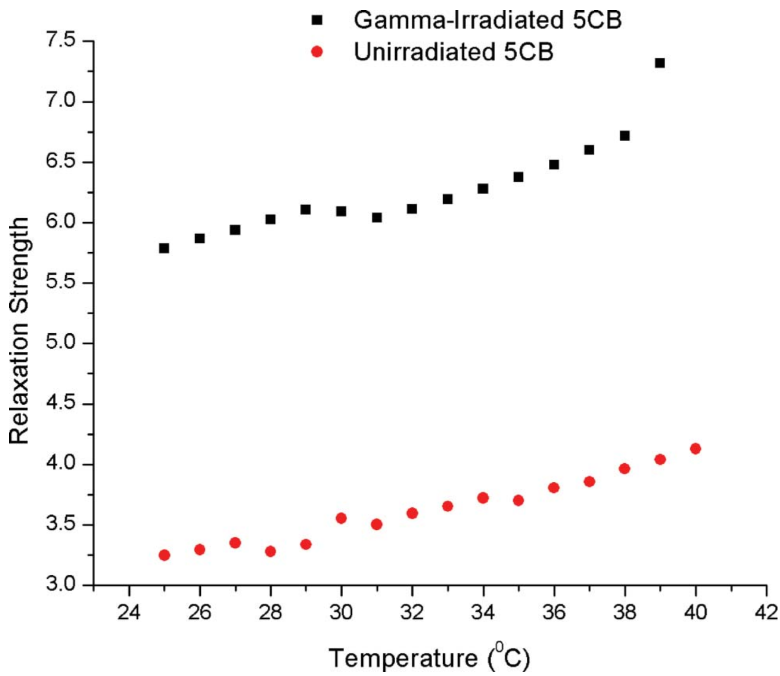

Figure 7. Relaxation strength with respect to temperature for gamma-irradiated and unirradiated 5CB liquid crystal. (Color figure available online).

4. Conclusion

The changes due to gamma irradiation on NLCs have been investigated using a dielectric spectroscopy technique. Some of the dielectric parameters like complex dielectric permittivity, relaxation frequency have been evaluated for the gamma-irradiated sample and compared with that of the unirradiated sample. We observe that the nature of variation of dielectric permittivity with frequency and temperature remains the same for both the gamma-irradiated and the unirradiated 5CB samples, but the value of the dielectric permittivity for the gamma-irradiated sample is higher than the unirradiated 5CB sample. This increment in the gamma-irradiated sample can be explained on the basis of physicochemical change in the 5CB molecules due to irradiation. The relaxation frequency for gamma-irradiated sample is shifted toward the lower side as compared with the unirradiated sample. The value of dielectric strength for gamma-irradiated sample is found to be higher than the unirradiated sample.

Acknowledgments

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References

[1] Klysubun, P., & Indebetouw, G. (2002). *J. Appl. Phys.*, *91*, 897.
[2] Takeda, M., Kitazume, T., Yamazaki, T., & Koden, M. (1995). *J. Mater. Sci.*, *30*, 5199.

[3] Khoo, I. C., Ping, Z., Yu, L., & Hong, Li. (1993). *IEEE J. Quantum Electron.*, *29*, 2972.
[4] Bahadur, B. (1991). *Liquid Crystals Applications and Uses*, World Sci.: Singapore.
[5] Heilmeier, G. H., & Zanoni, L. A. (1968). *Appl. Phys. Lett. 13*, 91.
[6] Marino, S., Castriota, M., Bruno, V., Cazzanelli, E., Strangi, G., Versace, C., & Scaramuzza, N. (2005). *J. Appl. Phys. 97*, 013523.
[7] Oh-e, M., & Kondo, K. (1996). *Appl. Phys. Lett. 69*, 623.
[8] Muenster, R., Jarasch, M., Zhuang, X., & Shen, Y. R. (1997). *Phys. Rev. Lett.*, *78*, 42.
[9] Meier, G., Sackmann, E., & Grabmaier, J. G. (1975). *Application of Liquid Crystals*, Springer-Verlag: Berlin.
[10] Kresse, H. (1983). In: G. H. Brown (Ed.), *Advances in Liquid Crystals*, Academic: New York.
[11] Dixit, G., Singh, J. P., Srivastava, R. C., Agrawal, H. M., & Asokan, K. (2012). *Radiat. Eff. Def. Solids*, *167*, 307.
[12] Stockley, J., Serati, S., Dauwe, D., Deaton, T., & Nonnast, J. (2006). In: In situ measurement of liquid crystal spatial light modulators' beam steering characteristics during gamma irradiation. *Proceedings of the SPIE*, p. 6308, SPIE: Washington.
[13] De, W. S., Miller, K., & Stockley, J. (2004). In: Nematic liquid crystal spatial light modulator's response to total-dose irradiation. *Proceedings of the SPIE*, p. 5554, SPIE: Washington.
[14] Date, R. W., Imrie, C. T., Luckhurst, G. R., & Seddon, J. W. (1992). *Liq. Cryst.*, *12*, 203.
[15] Srivastava, A. K., Misra, A. K., Chand, P. B., Shukla, J. P., & Manohar, R. (2008). *Phys. Lett. A*, *372*, 6254.
[16] Madhsudana, N. V., Srikanta, B. S., & Urs M. S. R. (1984). *Mol. Cryst. Liq. Cryst.*, *108*, 19.
[17] Gennes, P. G. D., & Prost, J. (1993). *Physics of Liquid Crystals,* 2nd ed.; Clarendon Press: Oxford.
[18] Manohar, R., Pandey, K. K., Srivastava, A. K., Misra, A. K., & Yadav, S. P. (2010). *J. Phys. Chem. Solids*, *71*, 1311.
[19] Manohar, R., Misra, A. K., Singh, D. P., Yadav, S. P., Tripathi, P., Prajapati, A. K., & Varia, M. C. (2010). *J. Phys. Chem. Solids*, *71*, 1684.
[20] Graham, A., Kopp, G., Vargas-Aburto, C., & Uribe, R. (1996). In: E. W. Taylor (Ed.), *Photonics for Space Environments IV*, Proceedings of the SPIE, 2811.