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Comprehensive studies on dielectric properties of p-methoxy benzylidene p-decyl aniline with function of temperature and frequency in planar geometry: A potential nematic liquid crystal for display devices

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The dielectric properties of the nematic mesophase, p-methoxy benzylidene p-decyl aniline (MBDA), measured in planar geometry with a function of frequency and temperature are investigated in detail. The complex dielectric permittivity (ε' and ε'') is also studied at a bias voltage of 10 V for planar aligned sample cell of nematic mesophase. The dielectric permittivity with bias voltage attains a higher (> 2 times) value than that without bias voltage at a temperature of 56 °C, which is due to the fact that the linking group of nematic molecules is internally interacted with an applied bias voltage. This is supported by observing an enhanced dielectric permittivity of nematic liquid crystal (LC) in the presence of bias voltage, which can be fully explained as the increasing of the corresponding dipole moment. The dielectric relaxation behaviors of nematic LC are also demonstrated for planar aligned sample cell. The remarkable results are observed that the relaxation frequency shifts into low frequency region with the increase of the bias voltage applied to the planar aligned sample cells. The dielectric relaxation spectra are fitted by Cole–Cole nonlinear curve fitting for nematic mesophase in order to determine the dielectric strength.

Keywords: dielectric permittivity, dielectric loss, relaxation frequency, nematic mesogen

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1. Introduction

Liquid crystal (LC) with negative dielectric anisotropy is potentially attractive nematogen, and can be used in display and photonics devices. It has been a suitable candidate for fabricating display devices, optical modulators, and its dielectric properties are mainly dominated by the characteristic of the LC materials. The nematic molecules are easily controllable and the response time is high as compared with that of ferroelectric (FLC). The lab-made sample cell is specifically for the nematic LC to improve the alignment and dielectric properties of the novel nematic molecules due to its advantages of high-speed respond, wide viewing angle, high contract ratio, and gray-scale capability. It has received much attention of researchers in the circle of soft condensed matter physics due to its strong dielectric polarization, high structural properties, and efficient viscoelastic properties, thereby increasing the utility of LCs. The LC has unique features, and its tunable properties are important for photonic and display devices and great effort has been made to design display, photonic device, and energy harvesting device.[1–7] For example, spatial light modulators are used in adaptive optics systems to compensate for the wavefront aberration caused by atmospheric turbulence in real time and can be used to improve the image resolution as close to the diffractive limitation of a telescope as possible. The uniaxial structure of nematic LCs and the tunability are provided by applying a voltage for controlling the refractive index of the LC. Nematic LC compound with linking group substitutes along their long molecular axis is studied in order to obtain twisted nematic displays with low threshold voltages, and to realize the practical applications in other electronics or nonlinear optics. The LC is an organic molecule which has and advantage of very high polarizability. The two applications in display and photonic technology affected strongly by LC dielectric permittivity and alignment are considered. The LC materials are used in electro-optical devices due to their anisotropic molecular order which can be controlled by an applied electric field.[8,9] As is well known, the dielectric anisotropy of the LC has an important influence on the image quality of liquid crystal display (LCD).[10,11] However, the presence of impurity ions of mesophase has been considered as a nuisance to LCD industry.[1–7] In the last few years we have tried to reduce the ionic impurity for improving the properties of nematic LC.[12,13] The many researches have demonstrated

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that the dielectric and electro-optical properties of nematic LC are a function of frequency, temperature, voltage along with external stimuli. Fei *et al.*[14] have studied the electro-optical properties of FLC without applying dc voltage to remove the degeneracy in the layer structure of the molecules. They have found the polarity of self-assembled monolayer is the formation of mono-domain alignment of FLC. The monodomain alignment of FLC molecules occurred in the asymmetric boundary cell without applying any voltage. The interaction between LC molecules and electromagnet wave have been investigated theoretically and experimentally[15,16] for enhancing the nonlinear optical properties of nematic LC. It is found that the interaction becomes larger when the incident intensity increases at particular pretilt angle. Peng *et al.*[17] studied the electro-optical properties of a series of LC compounds with isothiocyanate and naphthyl group for the application of optical devices. They have found that the enthalpy and birefringence values of these LC compounds are higher than those of corresponding compounds with the phenyl group. They have also investigated the figure-of-merit (FoM) of LC compound with the naphthyl group for high response to external stimuli. In such conditions, the dielectric and electro-optical properties of LC device are dominated by the dielectric permittivity, dielectric strength and relaxation behavior of nematic molecules. The dielectric permittivity of LC should be designed with a long conjugation molecular structure for improving the high dielectric anisotropy of functional groups *e.g.*, biphenyl, terphenyl, tolane, phenyl-tolane, naphthalene group, double-bond and triple-bond structures. The nematic molecule containing the linking (cynao) group possesses a large dipole moment along the long axis of the molecule. This leads to large value of parallel component of dielectric permittivity and consequently to a large positive dielectric anisotropy.[18] The linking group of nematic molecules exhibits a dipole moment which can influence the dielectric permittivity and anisotropic properties. In this order, de Jeu *et al.* have systematically studied the dielectric permittivities of some p, p′–n–alkyl, and n–alkoxy substituted azobenzenes and azoxybenzenes in the nematic and in the isotropic phase. They explained the influence of the molecular structure on the dielectric property.[19] Furthermore, in the uniaxial structure of nematic LC the tunability was provided by applying a voltage controlling the refractive index of the LC. Ren *et al.*[20] have studied the C_{60}-doped homeotropically aligned nematic LC for the application of dynamic phase holographic grating. They have studied the dynamic phase grating with an asymmetric profile in a C_{60}-doped homeotropically aligned nematic LC under a direct current (DC) voltage applied to the sample cell. The investigations of phase dynamic gratings in LCs have been concentrated mainly on their diffraction characteristics. The dynamic gratings have shown great promise in applications of optical devices and optical information processing. It should be noted that the dielectric and electro-optical parameters of nematic LC materials make it possible to use them in dynamic holography, which opens the way for creating new devices for optical information processing. In the nematic phase, the orientation of the molecules along a preferred direction is denoted by director n. The dielectric relaxation when studied in the frequency range from 100 Hz to 10 MHz, provides the information about orientational polarizability influenced molecular rotation.[21] The dielectric relaxation is chiefly observed due to ionic space-charge polarization whereas the rotation of LC molecules along the short molecular axis gives rise to dielectric relaxation polarization.[22] The electrode polarization[3,4] of LC is found to be beyond the 10 MHz.[1,2] The study of this frequency regime reflects the impurity ions in nematic LC and the fabrication of active matrix addressed display. There have been numerous attempts to study the dielectric properties of nematic LCs properties in the frequency range from 100 Hz to 10 MHz in recent years. It is important in such a frequency range to understand the electric or space charge polarization, orientational or dipolar polarization, ionic or atomic polarization and electronic polarization. Each polarization of LC has its own relaxation frequency which may be different in different mesophase. Atomic and electronic contribution occur at higher frequencies (more than MHz). In a frequency range from Hz to MHz, space charge polarization and orientational polarization are effective. In our case it is important for the nematic LC to understand the dielectric relaxation behavior which is remarkable structural dynamic property of nematic phase. In the past years, we have published many papers on dielectric relaxation in LC materials.[23]

Dielectric measurements reflect the electronic and orientational polarizations of the mesophase. The knowledge of dielectric properties gives the fruitful information about the structure of the LC. In order to determine the dielectric relaxation of neamtic LC, Cole–Cole plot, *i.e.*, the complex dielectric permittivity (ε' and ε'') as a function of frequency is presented. The dielectric relaxation of nematic LC empirical Cole–Cole circular arc law in the Cole–Cole plot is well known.[24] This study focuses on the temperature-dependent dielectric dynamic properties of a nematic meosgen, p-methoxy benzylidene p-decyl aniline (MBDA) as a function of frequency and temperature. The temperature effect and bias voltage effect of nematic mesogen on the dielectric properties of planar geometry are examined. In addition, by expanding the frequency range from 100 Hz to 10 MHz, the ionic transport behaviors ane analysed in a low frequency range region.

2. Experimental details

The LC sample cell employed in this study consists of highly conductive ($\approx$ 20 Ω/square and the visible light trans-

mission is more than 90%) indium tin oxide (ITO) coated glass plates. The ITO patterned glass plates with an active area of 5 cm were achieved by photolithographic technique.[25,26] Mylar spacer is used to maintain uniform thickness between the two glass plates. Now the masked plates were treated with polymer nylon 6/6 and rubbed unidirectionally in order to attain planar alignment. The calibration of the empty cell was carried out by using AR grade CCl_4 or benzene (C_6H_6). The LC material was filled into the cell through capillary action at a temperature slightly higher than its isotropic temperature. A planar aligned cell of 12 μm was finally fabricated. The LC compound MBDA had been obtained from Frinton Laboratories Inc. USA and were used as such without further purification. The molecular structure and phase sequence of the compound MBDA is shown in Fig. 1. The dielectric spectra were recorded by impedance/gain phase analyzer HP- 4194A in a frequency range from 100 Hz to 10 MHz. The dielectric response consists of capacitance value and the dissipation factor. Therefore, we subtracted the value of stray capacitance. The contributions from sheet resistance, ITO coatings and lead inductance of the cell were removed by the least square nonlinear fitting of experimental data of relative dielectric permittivity.[25,26] The temperature of the sample was maintained with the help of Instec Hot Plate HCS 302.

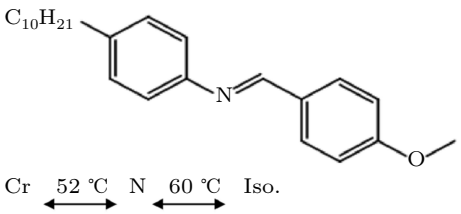


Fig. 1. Molecular structure and phase transition sequence of nematic LC, p-MBDA.

Figure 2 shows the polarized optical micrograph (POM) of nematic mesogen MBDA for planar aligned cell. It has been found that nematic molecules are uniformly filled in planar aligned sample cell. From this figure, we observed no variation of nematic molecules in planar anchoring strength. In contrast, we observed a net increase of the order parameter for the nematic molecules in aligned sample cell, which drives the orientation of the planar molecules anchored on the substrates.

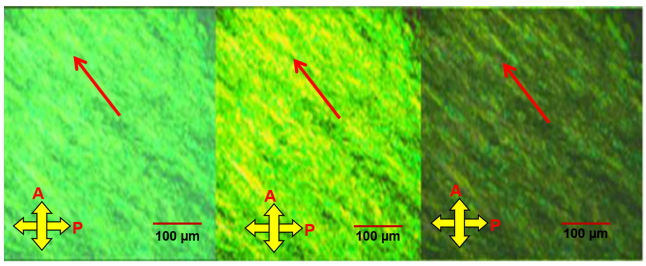


Fig. 2. Polarized optical micrographs for nematic MBDA in planar geometry at temperature 56 °C, with arrow showing rubbing direction and crossed arrows indicating crossed polarizer (P) and analyser (A).

3. Results and discussion

The capacitance and conductance measured as a function of frequency are used to calculate relative dielectric permittivity (ε') and dielectric loss (ε'') of nematic meosgen. The value of relative dielectric permittivity is determined from the following equations:

$$\varepsilon' = \frac{C_{\rm m} - C_0}{C_L} + 1, \tag{1}$$

$$\varepsilon'' = \frac{G_{\rm m} - G_0}{2\pi f C_{\rm L}}, \tag{2}$$

where $C_{\rm m}$ and $G_{\rm m}$ are the capacitance and conductance of the filled sample, whereas C_0 and G_0 are capacitance and conductance of empty sample holder. The dielectric spectrum results are obtained by plotting the real component and imaginary component of the complex dielectric permittivity function, which is expressed as

$$\varepsilon^*(\omega) = \varepsilon'(\omega) - {\rm i}\varepsilon''(\omega). \tag{3}$$

Here, $\varepsilon^*(\omega)$ is the dielectric permittivity, the real part, $\varepsilon'(\omega)$, is the dielectric constant, and the imaginary part, ${\rm i}\varepsilon''(\omega)$, is the dielectric loss.[27]

Figure 3(a) shows the frequency-dependent relative dielectric permittivity for the nematic mesogen at temperature 56 °C. The value of relative dielectric permittivity is constant in a frequency range from 100 Hz to 2 MHz in the case without bias voltage and in a frequency range from 100 Hz to 390 kHz in the case with bias voltage after that, its value decreases with frequency increasing. The dielectric polarization relaxation occurs in an MHz region for nematic mesogen due to the interaction between the electric dipole moment of nematic molecules and applied electric field. The dynamical behavior of nematic mesogen gives the information about its applications in display device. The dipole moment of nematic molecules arises from the linking group of the material. The nature of relative dielectric permittivity varying with frequency is similar to those reported in other cases of LCs.[28–30]

The variation of dielectric loss with frequency is shown in Fig. 3(b). Its value is high in a low frequency region of both planar geometry with and without bias voltage. In a low frequency region, more ions are generated in the case without bias voltage of nematic mesogen and the ionic impurity is reduced in the case with bias voltage. The ionic conductivity of nematic mesogen is larger in case without bias voltage, in contrast, it is small in the case with bias voltage. The ionic contribution hence is found to decrease with frequency increasing. However, the contribution from ionic entities is significantly less at low frequencies than from nematic counterpart. At lower frequencies the impurity ions have enough time to reach the electrodes, thus forming dipole moments, but the scenario changes at high frequencies: the space-charge polarization lags behind the polarity alteration of applied electric

field, thereby leading to dielectric relaxation. The relaxation frequencies are observed to be 2.3 MHz and 7.3 MHz for nematic mesogen with and without bias voltage at temperature 56 °C, respectively. Figure 3(b) shows the pronounced influence of bias voltage on nematic mesogen in a low-frequency regime. One can see from this figure that, with a bias voltage applied, the relaxation frequency shifts toward lower frequency.

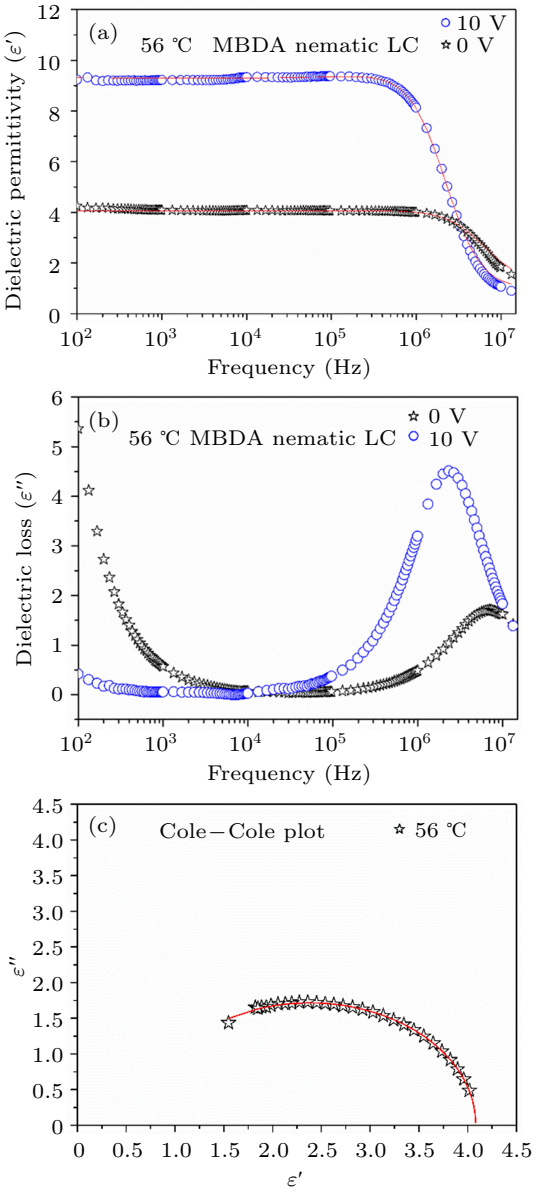


Fig. 3. (a) Dielectric permittivity and (b) dielectric loss varying with frequency, (c) Cole–Cole plot (ε' *versus* ε'') for nematic MBDA in planar geometry at temperature 56 °C.

For the demonstration of the dielectric properties, Cole–Cole plots are very instructive.[24,31,32] In order to investigate the high frequency dielectric response and single relaxation mechanism, *i.e.* the data of ε'' and ε', the high frequency dielectric dispersion is fitted by Cole–Cole plots in Fig. 3(c). The data of high frequency dielectric parameters corresponding to the relaxation modes, *viz.*, relaxation frequency f_R, loss maximum ε''_{max}, dielectric strength $\Delta\varepsilon$, distribution parameter α are estimated from the Cole–Cole plots.

The variations of dielectric permittivity of nematic mesogen with temperature are shown in Fig. 4 for three different frequencies, specifically, 1 kHz, 5 kHz, and 10 kHz, indicating that with the increase of temperature, the values of the three dielectric permittivities decrease, but at isotropic temperature they are constant. This can be inferred from the values of ε' decreasing with the increase of temperature. The dipole moment of nematic molecules decreases with the increase of temperature because of the rigidity of aromatic chain and interaction of linking group decreasing. It means that nematic molecules have acquired compatibility in planar geometry which reduces the dipole moment of MBDA nematic mesogen.

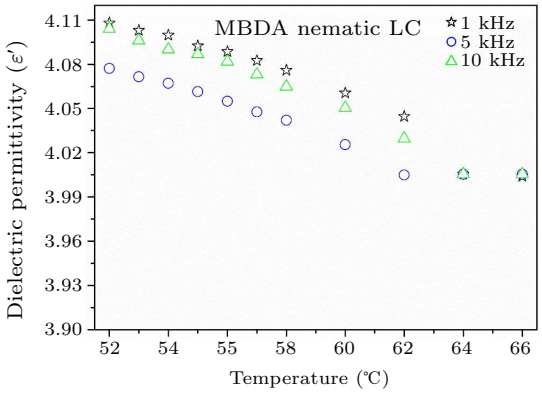


Fig. 4. Variations of dielectric permittivity with temperature for nematic MBDA in planar geometry at three different frequencies and temperature 56 °C.

The coupling between an electric field and the dipole moment is known as electroclinic effect. The electroclinic effect also exists in the nematogen: an electric field enhances the increase of the polarizability. Study of dielectric parameters play a major role in analyzing the nematic mesogen and using it for technical application. Thus, the dipole moment of nematic molecules decreases with temperature increasing because of the rigidity of chain of aromatic linking group of nematic mesogen.

The variation of dielectric loss with temperature for nematic mesogen MBDA in planar geometry for different frequencies, specifically, 1 kHz, 5 kHz, and 10 kHz are shown in Fig. 5. The value of dielectric loss increases with temperature increasing for nematic mesogen. From this figure we can see that the value of dielectric loss decreases separately at a frequency of 5 kHz and 10 kHz. The dielectric loss is also related to the energy dissipation of the system. The dielectric loss is also dependent on the energy dissipation for breaking and forming hydrogen bonds to neighboring molecules and is affected by the chain length of the rotating whole molecules. From these investigations, the dielectric loss is reduced in the case of bias voltage in a low frequency range and the strong dielectric loss occurs in a high frequency range. The completed molecular rotation is considered to occur in the high

frequency range because of the small molecular chain. Therefore, the whole molecules rotate in the high frequency range. The experimental results of the investigated nematogen which has the property of negative dielectric anisotropy, reflect the structural symmetry and the planar alignment of the sample and can be employed in the display.

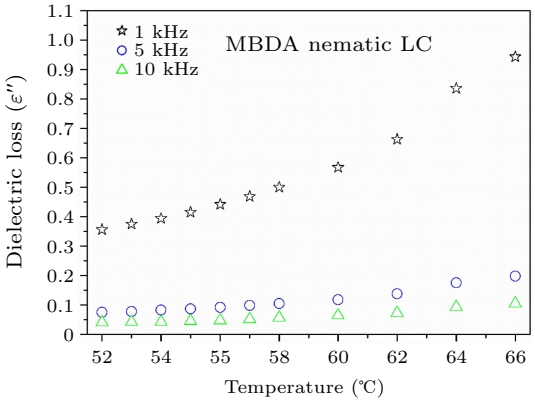


Fig. 5. Variation of dielectric loss with temperature at three different frequencies for nematic MBDA.

4. Conclusions

The dielectric permittivity plays a major role in the dipolar interaction of nematic molecules due to the linking group of meosgen, p-methoxy benzylidene p-decyl aniline (MBDA). Based on results revealing the dielectric relaxation frequency shift in a low frequency range for the case of with bias voltage, it is concluded that the MBDA is useful in economical display technology. The Cole–Cole plot is also analyzed to determine the dielectric strength and single relaxation behavior of nematic mesogen. The Cole–Cole functions of the complex dielectric permittivity curve resulting from a set of dielectric strength can be related to implicit dielectric activated phenomena. The significant functional dependence of the dielectric properties for the nematic mesogen is highly exploitable in display technology.

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