



A method to find the initial temperature range of the short-range order in the isotropic phase of nematic liquid crystals based on the electro-optical Kerr effect

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ABSTRACT

It is well-known that orientational order of nematic liquid crystals (NLCs) is converted during phase transition from the long-range order (LRO) in the nematic phase to the short-range order (SRO) in the isotropic state. Finding the initial temperature range of SRO in the isotropic state can be a challenging issue because most of the physical constants disappear after this phase transition. For the first time, to resolve this issue, we introduced a new method based on the electro-optic Kerr (EOK) effect in NLC mixtures. In this method, the coherence length (ξ), which was qualitatively estimated by employing the pre-transitional temperature (T^*) according to the de Gennes phenomenological theory, and the refractive index (n) were correlated with the Kerr constant (B) at the same temperature in the isotropic phase. Because molecular interaction in the NLCs is one of the essential factors in the investigation of the orientational ordering, we used n and ξ as described intra- and intermolecular interactions, respectively. In other words, n attributes to the polarization and dipole moment as an important parameter of the intramolecular interaction and ξ demonstrates the intermolecular interaction and domains of SRO interaction in the isotropic phase. Finally, the initial temperature range of SRO in the isotropic phase was found from the temperature dependence with B of the normalized values of influential coefficients n and ξ at the similar temperatures. Our proposed method appropriately can provide a suitable approach to distinguish the preservation of the anisotropy effect in the isotropic phase of NLCs.

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1. Introduction

It is well-known that the order parameter changes during the phase transition of liquid crystals (LC) from the nematic to isotropic phase [1–3]. The long-range orientational order (LRO) exists in the nematic phase, whereas local or short-range orientational order (SRO) develops in the isotropic phase with approaching to the phase transition temperature (T_{NI}). In other words, in the isotropic phase of the LC, there is no LRO along the alignment direction of the LC molecules. However, locally, they are still parallel to each other. Indeed, SRO implies that the order is correlated over a finite distance. This order continues over a particular characteristic distance, referred as the coherence length (ξ), which can explain the SRO behavior [4]. It is well-known that the phase transition in the isotropic phase of an NLC is very weak. Therefore, investigations on the characteristics of the development of the ordered and isotropic phases prior to the formation of the LC could provide valuable

information to understand the forces responsible for the LC formation. Although this phenomenon has been approved in most of the theoretical and experimental studies [5–10], finding the initial temperature range of SRO in the isotropic phase of NLCs is considered as a challenging issue, which has been investigated rarely. Because most of the physical constants vanish above the phase transition temperature (T_{NI}) in the isotropic phase, finding a new method which explains the starting temperature range of SRO in that region can be considered as a big achievement.

The electro-optic Kerr (EOK) effect can be a suitable method to resolve this problem. The EOK effect, also referred as the quadratic electro-optic effect, is the change in the refractive index of a material in response to an applied electric field [11–16]. The difference in the refractive index between the two directions, along and perpendicular to the applied electric field, is referred to as the induced birefringence [11–16]. The EOK effect provides valuable information about the geometrical structure and aggregation of molecules in LC solutions as well as in pure nematic and isotropic phases [17,18]. Studies on the temperature dependence of the EOK effect in LCs are useful to investigate molecular anisotropy behavior, inter- and intra-molecular interaction ordering, and pre-transitional temperature (T^*) [19–21]. There is no

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doubt that T^* caused by the thermal fluctuation of the orientational ordering in the near T_{NI} regime is one of the physical evidence to demonstrate SRO in the isotropic phase of an LC. Both theoretical and experimental studies were performed to interpret the pre-translational behavior in isotropic mesogenic liquids at the vicinity of the nematic phase [16–19]. The pre-translational temperature range exists at the vicinity of the clearing temperature. This phenomenon emerges owing to the order-parameter fluctuation [22,23], leading to the formation of pseudo-nematic domains with highly correlated molecules in the isotropic phase [24].

The high Kerr constant (B) of an NLC in the isotropic phase can be attributed to the existence of a molecular association, owing to the strong polar groups and short-range molecular interaction [1–3]. Because molecular interaction is one of the crucial factors in the orientational ordering, correlation of those parameters with B can be reasonable if we can employ suitable parameters from the molecular interactions. For this reason, one of the essential parameters which can describe the SRO interaction is ξ in the isotropic phase. It can be qualitatively estimated from T^* values [4]. Another important parameter which can explain the interactional effect is the refractive index (n). n illustrates the polarization effect as an important factor in the molecular interactions. Therefore, n and ξ possess the interactional properties which can be correlated with B . Consequently, to find the initial temperature range of SRO, n and ξ were used as indications of the molecular interaction in the isotropic phase. The influential coefficients n and ξ were determined by correlating n and ξ values with the B value at temperatures slightly higher than T_{NI} . Then, the initial temperature range of SRO can be recognized obviously from the temperature dependence of the influential coefficients.

In this study, five NLC mixtures, four low-birefringence (Δn) and -dielectric anisotropy ($\Delta\epsilon$) LCs and one high- Δn and - $\Delta\epsilon$ LC, were considered. The high- Δn and - $\Delta\epsilon$ LC was considered to establish a concrete result for our proposed method. In this study, we roughly estimate the actual spatial sizes of the domains in the considered NLCs using the theoretical predictions of de Gennes [1,18]. Finally, after revealing linear correlations of both ξ and n values with the B values, for the first time, we introduced a new method to recognize the initial temperature condition of the SRO in the isotropic phase of the NLCs. This method appropriately reveals the initial distinguishable border of the LRO, which explains the inherent-anisotropy preservation of the NLCs.

2. Theory

The LCs exhibit a long-range orientational order so that a small external field is sufficient to align their directors. The EOK effect can be observed when a high electric field is applied to an isotropic material [25,26]. The application of an electric field to an optically isotropic liquid leads to an anisotropic molecular distribution, which could be attributed to the inherent molecular anisotropy. Some anisotropy may be induced by the applied electric field. Upon application of an external electric field to an LC, the refractive index for the light polarized parallel ($n_{||}$) differs from that of the light polarized perpendicular (n_{\perp}) with respect to the field direction [1,27]. The phase difference (δ) between the parallel and perpendicular light components after passing through the LC can be expressed as [16]:

$$\delta = 2\pi l B E^2 = \frac{2\pi l \Delta n_{induced}}{\lambda} \quad (1)$$

where l is the length of the Kerr cell as an optical path length, B is referred as the Kerr constant representing the magnitude of the Kerr effect, E is the external electric field strength, λ is the wavelength of the light passing through the LC, and $\Delta n_{induced}$ is the induced

birefringence, i.e., the difference of the refractive indices. The magnitude of the Kerr effect can be measured by the Kerr constant [13,19]:

$$B = \frac{\Delta n_{induced}}{\lambda E^2} \quad (2)$$

Upon application of an external electric field to the Kerr cell, the rotation of the polarization plane, α , is related to the phase difference (δ):

$$\alpha = \frac{\delta}{2} \quad (3)$$

Using Eqs. (1) and (3), we obtain:

$$B = \frac{2\alpha}{\pi l E^2} \quad (4)$$

Using Eq. (4), the EOK constant (B) can be determined by plotting α against the square of the external electric field (E^2) and determining the gradient of the straight line.

With the increase of the temperature, thermotropic LCs undergo a transition between different mesophases, owing to the decrease of the order parameter until the normal isotropic phase can be formed. However, the isotropic phase of the LCs differs from that of conventional isotropic liquids owing to the formation of the SRO at the pre-translational temperature at the vicinity of T_{NI} . This feature considerably changes the B value of the isotropic phase in the pre-translational region. Therefore, a giant optical non-linearity can be observed in LCs. Studies on the behavior prior to the formation of the isotropic phase in LCs reveal very valuable information to understand the forces responsible for the formation of the LCs. They can also reveal temperature dependence in some physical parameters, such as the heat capacity and dielectric permittivity [28]. The pre-translational behavior based on the temperature dependence of B in LCs can be expressed using the Landau-de Gennes theory [1,18] with a linear relationship between B and temperature:

$$B = \frac{\epsilon \cdot \Delta n \cdot \Delta\epsilon}{4a\lambda\sqrt{\epsilon}} (T - T^*)^{-\gamma} \quad (5)$$

where $\Delta\epsilon$ and Δn are the low-frequency dielectric anisotropy and high-frequency optical birefringence corresponding to perfectly aligned LCs, respectively. The parameters a and T^* are the temperature-independent coefficient and pre-translational temperature, respectively. The parameters ϵ , ϵ_0 , λ , and γ are the optical permittivity of the phase of the LC, dielectric constant of vacuum, wavelength of the light used in the characterization of the Kerr effect, and critical exponent, respectively.

In order to investigate the temperature-dependent SRO behavior, the use of a new parameter with a quantitative value can be helpful. The coherence length (ξ) is one of the most important parameters in the pre-translational regime, which can be predicted by the de Gennes phenomenological theory [1,18]. ξ characterizes the extension of the local nematic order in the isotropic phase, which can increase to limited values of approximately 10–12 nm at T_{NI} [4,24]. However, immediately above T_{NI} , where the LRO is extinct, the value of ξ can correspond to the SRO behavior. With the increase of the temperature, the value of ξ is expected to decrease, and then it is leveled off at a high temperature in the isotropic phase [1,18]. The ξ value can be qualitatively estimated by the relationship introduced by Gulari et al. [4] as:

$$\xi = 5.5 \pm 0.2 \left(\frac{T}{T^*} - 1 \right)^{-0.5 \pm 0.01} \quad (6)$$

where T^* is the pre-translational temperature. By employing the temperature-dependent ξ value, the decrease of the SRO in the NLCs can be properly explained.

3. Materials and experiments

3.1. Liquid crystals

Unlike in LCs which consist of a single type of molecules, five NLC mixtures (Merck Ltd.) with unknown structures were selected in our investigation. In this study, one NLC with high Δn and $\Delta\epsilon$ values, MAT-160968 ($\Delta n = 0.1874$, $\Delta\epsilon = 15.8$) is compared with the other four NLCs, ZGS-8045-LA ($\Delta n = 0.119$, $\Delta\epsilon = 5.1$), ML-1309 ($\Delta n = 0.1096$, $\Delta\epsilon = -3.9$), ML-0648 ($\Delta n = 0.1029$, $\Delta\epsilon = 10.3$), and ML-1407 ($\Delta n = 0.1011$, $\Delta\epsilon = -4.1$), as low- $\Delta\epsilon$ and - Δn LCs. Detailed information on the investigated NLCs with their T_{NI} values are listed in Table 1. Among them, LC-I has the highest Δn value, compared with low Δn values of other samples. Two of the selected NLCs (i.e., LC-III and LC-V) have negative and similar $\Delta\epsilon$ magnitudes, which can be used to compare their EOK effect based on the different Δn values.

3.2. Birefringence measurements

An Abbe's refractometer with a precision of 0.00001 (Bellingham Stanley Abbe 60ED) was used to measure the birefringence (Δn) of the selected NLC mixtures. The temperature of the Abbe's refractometer was precisely controlled with a circulating glycerol bath. A thermometer with an accuracy of ± 0.1 °C was placed at the vicinity of the sample to check the temperature.

3.3. Kerr effect equipment

The B value was measured by nulling the optical response from an electric field across the Kerr cell [13,15]. The Kerr cell was fabricated using a Teflon spacer for insulation between stainless-steel electrodes with an electrode gap of 1.8 mm. The cell gap (i.e., 1.8 mm) of our Kerr cell is higher than that of a conventional LC cell. This helps to decrease the effect of the surface boundary conditions. Moreover, no treatment was made for initial orientation of LCs in our Kerr cell. This feature may decrease the interface and the surface effects. The electrodes, with an active measuring volume of $2 \times 1.8 \times 10 \text{ mm}^3$, were inserted into a quartz spectrophotometer cell with a light path length of 10 mm. A 5-mW He-Ne laser beam, as a probe beam with a wavelength of 632.8 nm, was passed through the Kerr cell. A thermocouple and PT 100 sensor were used to control the temperature with an accuracy of ± 0.1 °C. To stabilize the temperature precisely, the temperature of the test sample was kept in equilibrium for at least one hour. Glan-Thompson double-refraction-type prisms (polarizer and analyzer) were employed, crossed to each other at an angle of 45° with respect to an applied AC field (1 kHz). A high-voltage power supply was used to apply an electric field to the Kerr cell. A plate with quarter-wave retardation at 632.8 nm was placed between glass disks of the Kerr setup. The light intensity passed through the analyzer was detected by a photomultiplier tube (E.M.I. 9816B). The optical signal from the photomultiplier was sent to a digital storage oscilloscope and personal computer. Fig. 1 shows a schematic of the thermo-optical setup used to measure the Kerr effect.

Table 1

Material parameters of the investigated nematic LCs: phase transition temperature (T_{NI}), birefringence (Δn), and the dielectric anisotropy ($\Delta\epsilon$).

Liquid crystal	T_{NI}/K	Δn (589 nm, 298 K)	$\Delta\epsilon$ (1.0 kHz, 298 K)
LC-I (MAT-160968)	345.85	0.1874	15.8
LC-II (ZGS-8045-LA)	348.95	0.1190	5.1
LC-III (ML-1309)	353.50	0.1096	-3.9
LC-IV (ML-0648)	354.15	0.1029	10.3
LC-V (ML-1407)	348.65	0.1011	-4.1

4. Results and discussion

The temperature-dependent B value of each NLC was measured at temperatures above T_{NI} , which was obtained from the field-induced birefringence ($\Delta n_{\text{induced}}$) values, as shown in Figs. 2 and 3. The electrically induced $\Delta n_{\text{induced}}$ was a linear function of the square of the applied electric field at different temperatures, as shown in Fig. 2. Fig. 3 shows the temperature dependence of B for our NLCs. With the increase of the temperature, the B value of all the NLCs decreased owing to the increased thermal fluctuation and decreased intermolecular interaction at elevated temperatures. The highest B values, measured at 0.1 °C above T_{NI} , of LC-I, -II, -III, -IV, and -V, were $B (10^{11} \text{ m} \cdot \text{V}^{-2}) = 9.86 \pm 0.04$, 8.17 ± 0.03 , 7.36 ± 0.01 , 6.27 ± 0.02 , and 4.3 ± 0.05 , respectively. Among the investigated NLCs, the highest B value was obtained for LC-I, which is easily predictable based on Eq. (1), considering its relatively high Δn and $\Delta\epsilon$ values. The large Δn and $\Delta\epsilon$ values imply that highly polar groups exist in the structure of this NLC, which remarkably increase the conjugation length and subsequently induce dipole moment and polarizability effects. On the other hand, comparison among other NLCs, LC-II, -III, -IV, and -V, with low Δn values, reveals that the highest and lowest B values are obtained for LC-II and LC-V, respectively, over the entire temperature range in our experiments. This can be attributed to the magnitude of Δn because a slight increase of Δn can considerably influence the B value. Therefore, the minimum B value is obtained for LC-V with the lowest magnitude of Δn . To confirm this claim, we compared LC-III with LC-V. They have similar negative $\Delta\epsilon$ magnitudes but different Δn values. Comparison between these two NLCs confirms that the B magnitude significantly increases with a slight increase of the Δn value.

The delocalized π electronic structures of the NLCs provide potential sources of fast and large third-order optical non-linearities [1,2,27]. Therefore, the NLCs are of significance in non-linear optical and electro-optical devices for various applications in electro-optic tunable notch filters, logic devices, switching, bistable elements, modulators, etc. [2,3]. This important feature can be determined with the B value in the isotropic phase of the NLCs.

By precise measurement of the EOK effect, we can determine the pre-translational behavior. The Landau-de Gennes theory [18] successfully illustrates the pre-translational behavior in the isotropic phase of LCs based on the assumption that short-range interaction predominates at the vicinity of T_{NI} . The application of an electric field simply disturbs the pattern of pre-translational fluctuation under the continuous or even weakly discontinuous phase transition in the LCs. The pre-translational behavior based on the temperature dependence of the Kerr constant in the LCs can be expressed by the Landau-de Gennes theory [15] as:

$$B \propto (T - T^*)^{-\gamma} \quad (7)$$

where T^* is a fictitious second-order transition temperature close to the macroscopic phase transition temperature T_{NI} , referred as the pre-translational temperature. It can be obtained by a linear extrapolation of the temperature dependence on the inverse value of the Kerr constant (B^{-1}) [13,15,16]. γ is a numerical factor equal to unity based on the Maier-Saupe theory for NLCs [13,15,16]. Dependences of the inverse of the Kerr constant (B^{-1}) on the temperature for the investigated NLCs are shown in Fig. 3. The T^* values just below T_{NI} for our NLCs, determined using the linearity of B^{-1} with the temperature, were 345.45 ± 0.03 , 348.61 ± 0.01 , 353.05 ± 0.02 , 353.75 ± 0.02 , and 348.55 ± 0.01 K for LC-I, -II, -III, -IV, and -V, respectively.

T^* is the critical temperature at which the nematic coherence length deviates to infinity. The T^* value attributes to the manifestation of the SRO in the isotropic phase of the NLCs, where this phenomenon differs from the behavior in conventional isotropic liquids. Therefore, it is reasonable to discuss the domain of the SRO by considering T^* as an important physical parameter. Effects from other physical parameters are

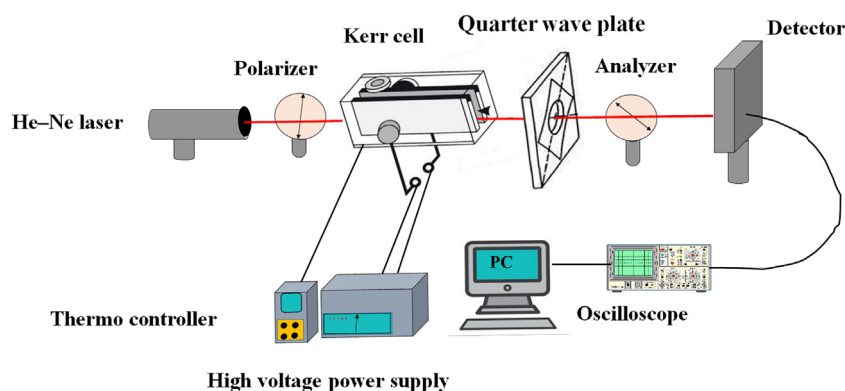


Fig. 1. Optical apparatus used to determine the EOK effect.

absent in this region. In the pre-translational regime, de Gennes extended the theory of the order parameter as a function of the spatial position and introduced gradient terms to the free-energy expansion, showing that ξ varies with temperature [1,18]. The temperature dependence of ξ can be expressed by the Landau-de Gennes theory [1,18,29–32] as:

$$\xi^2 \propto (T - T^*)^{-1}, (T > T^*) \quad (8)$$

Fig. 4(a) shows the temperature dependence of the correlation length. The rapid decrease of the qualitatively estimated ξ values was observed with the increase of the temperature for the investigated NLCs. In other words, the ξ values reveal that the SRO decreases with the increase of the temperature. As shown in Fig. 4(a), very similar values of the estimated ξ can be observed for LC-II and LC-V owing to their similar T^* values. However, in order to obtain the precise values of ξ , the light-scattering method is highly recommended, which have been already employed for NLCs including MBBA ($\xi = 1.2 \times 10^2$ at $T - T^* \sim 11$ °C). According to the $\xi(T)$ results,

the relationship between the determined ξ values and B magnitudes can provide useful insights into the SRO. As shown in Fig. 4(b), a linear correlation is observed between the ξ and B values. With the increase of the temperature, the ξ and B values decreased. Fig. 5(b) describes the decrease of the domain size with the increase of the temperature in all the investigated NLCs. As confirmed by Fig. 4(b), owing to the LRO, the B values at the vicinity of T_{NI} are higher than in the high-temperature region where SRO is formed. Therefore, the decrease of the ξ and B values can be attributed to the conversion of the LRO to the SRO. This conversion can be attributed to the weak intermolecular interaction and high thermal fluctuation at a high temperature.

It is well-known that NLC molecules within a small distance from each other are correlated by intermolecular interaction. The SRO intermolecular interaction in the isotropic phase of the NLCs was already explained by ξ . Furthermore, it is obvious that the refractive index (n) can be attributed to the dipole moment as an important parameter of the intermolecular interaction. As a relationship of the ξ and n with the intermolecular interaction originating from the polarizability of the LC, they

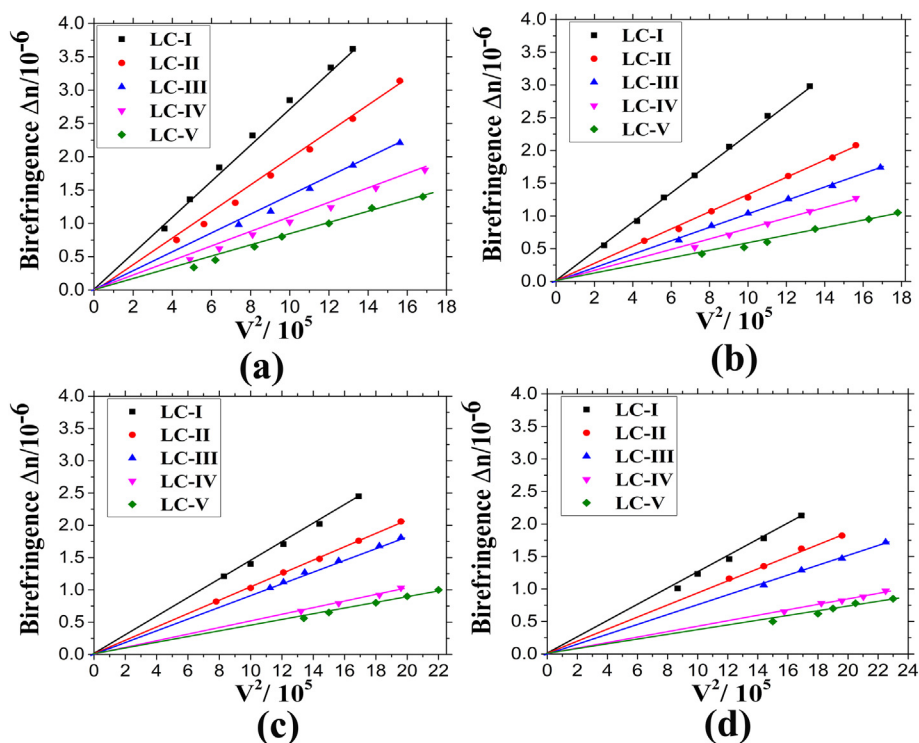


Fig. 2. Field-induced birefringence ($\Delta n_{induced}$) as a function of a square of the applied electric field for the investigated NLCs measured under different isotropic temperature conditions: (a) $T_{NI} + 0.5$ K, (b) $T_{NI} + 1.5$ K, (c) $T_{NI} + 2.5$ K, and (d) $T_{NI} + 3.5$ K.

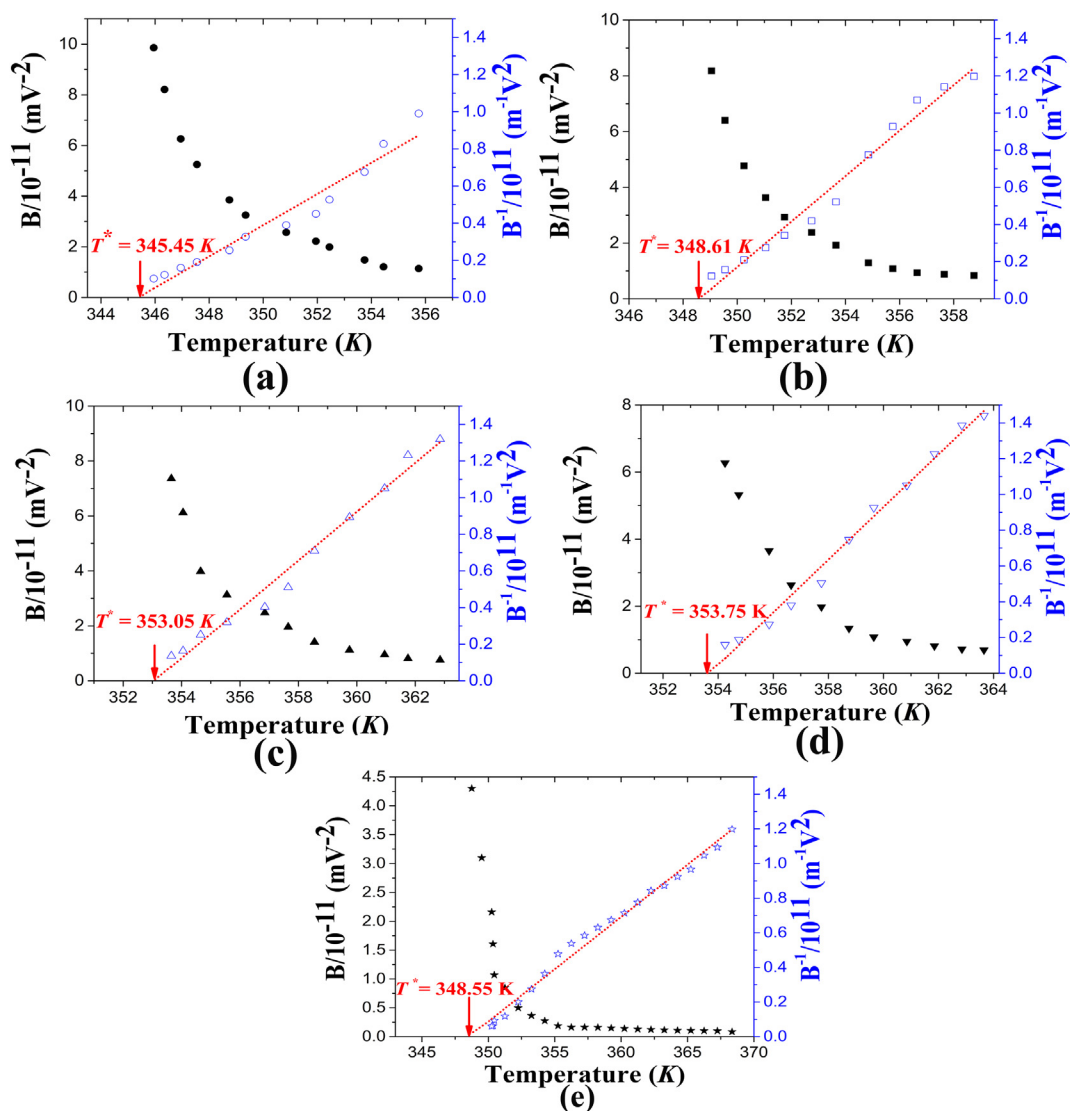


Fig. 3. Temperature-dependence of the Kerr constant (B) and its inverse (B^{-1}) used to determine the pre-transitional temperatures using the Landau–De Gennes' theory for the investigated NLCs. (a) LC-I, (b) LC-II, (c) LC-III, (d) LC-IV, and (e) LC-V. Filled and empty symbols represent B and B^{-1} , respectively.

are expected to exhibit similar behaviors. A relationship of n with B , especially under the same temperature conditions as those for ξ , can expand the SRO concept in the isotropic phase of the NLCs. As shown in

Fig. 5, the refractive index as a function of B exhibits a behavior similar to that of ξ (Fig. 4(b)). n and B near T_{NI} are higher than those at high temperatures.

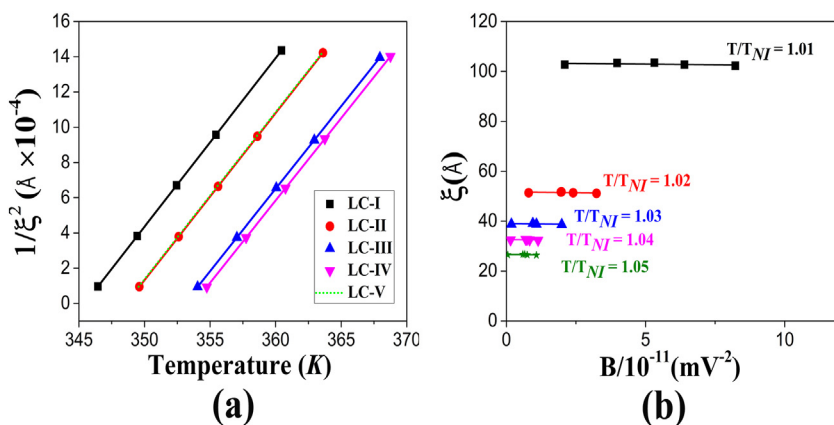


Fig. 4. (a) Calculated temperature dependence of the coherence length (ξ), (b) coherence length (ξ) as functions of the Kerr constant (B) at different temperatures for the investigated NLCs.

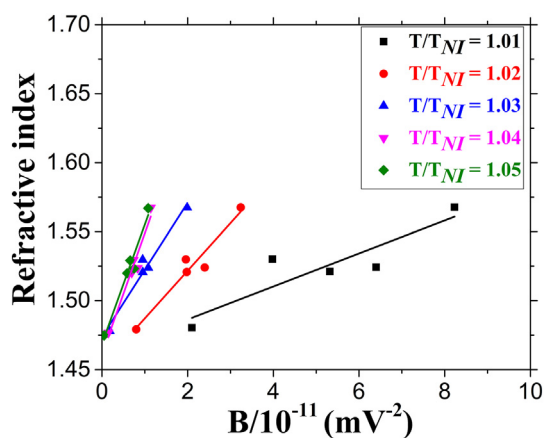


Fig. 5. Dependence of the refractive index as a function of the Kerr constant (B) for temperatures above the phase transition (T_{NI}) in our investigated NLCs.

As shown in Figs. 4 and 5, n and ξ have linear relationships with B , respectively. These results encourage to relate these two parameters with the B value at the same temperature conditions. Because both n and ξ illustrate the interactional effects, they can show a mutual relationship with the B value. In addition, by increasing temperature in the isotropic phase, similar reduced values can be observed for n , ξ , and B . For this reason, if we correlate n and ξ with B in the same temperature condition, we can obtain equal effects of these parameters. However, because of huge magnitude differences of n and ξ , employing the influential coefficients of these parameters in correlation with B can be a justifiable method. Then, to determine the coefficients of the correlations of n and ξ with B , for the correlation coefficients of the two parameters a normalization process was performed based on the maximum values because the values of ξ are significantly larger than those of the refractive index. The temperature dependence of the correlation coefficients on the two parameters are shown in Fig. 6. Each point in Fig. 6 illustrates the normalized influential coefficients of ξ and n for all the investigated NLCs at the same temperature condition. As shown in Fig. 6, the impact factors of ξ and n have negative and positive values, respectively. This is a reasonable result owing to the linear multi-parameter correlations and large ξ values in high dimensions, compared with the dimensionless n values. As shown in Fig. 6, it is obvious that, for $1 < T/T_{NI} < 1.02$, the normalized coefficients of ξ and n show variations in both values and trends. This can be attributed to the different n and ξ behaviors with respect to the B value at above T_{NI} . In other words, at near T_{NI} , n and ξ have different values. On the other hand, for $T/T_{NI} > 1.02$, the impact factors of the correlation coefficients for n and ξ have similar

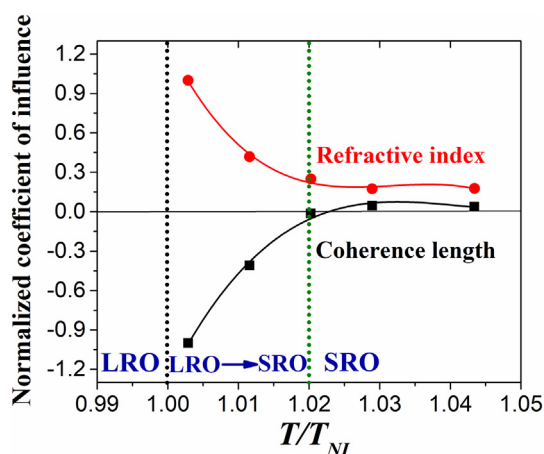


Fig. 6. Temperature dependence of the normalized refractive index and coherence length, correlated with the Kerr constant for the investigated NLCs.

values with the same trend. It means that, for $T/T_{NI} > 1.02$, there is no interaction which might be considered as an absolute isotropic liquid. In other words, for $T/T_{NI} > 1.02$, there is no inherent anisotropy in the isotropic phase of the NLCs. Therefore, the temperature dependence of influential coefficients for n and ξ are equalized. In addition, it can be concluded that the LROs or quasi-long-range orders (QLROs), and anisotropies of the investigated NLCs are maintained up to $T/T_{NI} = 1.02$. Beyond this temperature, the SRO can be primarily formed. However, we firmly believe that more investigation is still necessary to find the optimized temperature range.

5. Conclusions

It is well-known that after phase transitions from the nematic to isotropic phase, most of macroscopic physical constants which demonstrate LRO disappear above the phase transition temperature (T_{NI}). However, the SRO behavior exists between the highly polar NLC molecules in the isotropic phase which leads to the formation of pseudonematic microscopic NLC domains. Finding the initial temperature range of SRO in the isotropic phase by using the EOK effect was aimed in this work. We used NLCs with different Δn to find the general initial temperature range to distinguish the temperature range of SRO. For a sensitivity comparison of the range of the EOK effect among the NLCs, four NLC mixtures with very close Δn values and one high- Δn NLC mixture were selected. By measuring the Kerr-constant (B), the pretransitional temperature (T^*) was determined for these NLCs. By determining the T^* value, the coherence length (ξ) was measured as a useful parameter to show the presence of the SRO domains at temperatures slightly higher than T_{NI} . The ξ value was qualitatively estimated according to the de Gennes phenomenological theory. The temperature dependence of the ξ value demonstrated the decrease of the SRO domains with the increase of the temperature in the isotropic phase of the NLCs. Finally, a new approach to distinguish the initial temperature range of SRO was introduced for the first time based on the n and ξ values correlated with the B magnitudes at different temperatures in the isotropic phase. We found that the temperature dependence of influential coefficients n and ξ were almost the same at temperatures slightly higher than T_{NI} , where we established the initial temperature range of SRO. Therefore, our results confirm that there is no evidence of ordering and interaction at the higher temperature condition in the isotropic phase of NLCs, which was already approved in the phase transitional studies. In conclusion, our method revealed the preservation of the anisotropy in the isotropic phase of the NLCs where effects from other physical parameters were absent.

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