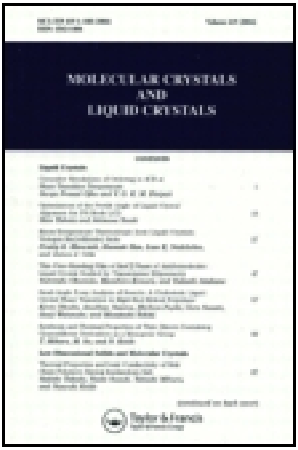

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Orientational Order Parameter - 1 A Birefringence Study

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Orientational Order Parameter – 1 A Birefringence Study

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Many methods are known for the estimation of the orientational order parameter in the nematic phase of a liquid crystal. The order parameter can be calculated directly from a macroscopic quantity measured in the experiment. The birefringence technique has been exploited for the evaluation of order parameter in the nematic phase along with the density measurements which are needed for the calculation of molecular polarizabilities of the molecule. Density and birefringence studies are carried out on four compounds viz., N-(p-n-octyloxybenzylidene)-p-n-propyloxy, butyloxy, hexyloxy and heptyloxy anilines, 8O.O3, 8O.O4, 8O.O6 and 8O.O7. The order parameter is obtained from the molecular polarizabilities obtained using two different internal field models due to Vuks and Neugebauer and the molecular anisotropy calculated using Lippincott δ-function model. The results reveal that both the models agree very near to the isotropic-nematic transition and diverge as nematic field stabilizes. Further, the results are compared with those obtained using Haller's extrapolation technique.

Keywords: birefringence; orientation order parameter; polarizability anisotropy

1. INTRODUCTION

The important parameter of the mesogen which governs nearly all physical properties is its order parameter. According to de Gennes [1], any of the bulk tensorial properties like electric and magnetic

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susceptibilities, elastic constants, refractive indices etc. can be used to determine the order parameter. There are different methods to evaluate order parameter from the birefringence study. One such method to evaluate order parameter is the molecular polarizability and the anisotropy of this property is one of the important properties of liquid crystals because the intermolecular interaction energies, according to several theoretical models [2–6], are based on them.

In this manuscript the study of the dilatometry and the birefringence of four compounds viz., N-(p-n-octyloxybenzylidene)-p-n-propyloxy, butyloxy, hexyloxy and heptyloxy anilines, 8O.O3, 8O.O4, 8O.O6 and 8O.O7 is reported. The manuscript is organized as follows. Section 2 describes the experimental procedures adopted for the determination of the refractive indices of the above compounds with temperature. Section 3 enumerates briefly the molecular theories applied for the evaluation of molecular polarizabilities using the experimental density and birefringence data. Section 4 presents the evaluation of orientational order parameter with temperature in nematic phase.

2. EXPERIMENTAL

The liquid crystals used in the present study are synthesized according to standard procedures [7,8]. The ingredients used are p-octyloxy benzaldehye and the corresponding alkoxy anilines. The synthesized compounds are subjected to repeated crystallization until the transition temperatures are in agreement with the reported data [8].

The refractive indices of the liquid crystal were measured with wedge shaped glass cell, similar to the one used to obtain birefringence by Haller *et al.* [9] and modified spectrometer. A wedge shaped glass cell was formed with two optically flat rectangular glass plates (50 mm × 25 mm) sandwiched with glass plate (0.4 mm) which acts as a wedge spacer. The optical flats are uniformly rubbed along the short edge to get the alignment of the LC molecule. The cell is filled with the LC material. The LC in the cell acts as a uniaxial crystal with its optic axis parallel to the edge of the spacer glass plate. The temperature accuracy of the heating block was ±0.1°C. The accuracy in the measured refractive indices was ±0.0005.

3. RESULTS AND DISCUSSION

The refractive indices of the above liquid crystals are measured using the modified spectrometer (the details are described in the experimental section) and a wedge shaped cell. The temperature accuracy is ±0.1°C. The refractive indices n_e and n_o are measured at wavelength 589.3. The refractive index practically shows no change in the isotropic phase (n_{iso}). At the IN phase transformation, the isotropic value splits into two, one value lower and another higher than isotropic value corresponding to extraordinary (n_e) and ordinary refractive (n_o) indices respectively. This is clearly observed in the telescope of the modified spectrometer at the angle of minimum deviation as the line representing the isotropic refractive index splits into two at the IN transition. In the nematic region, the n_e increases while the n_o decreases with the decrease of temperature and both the values attain saturation deep in the nematic region. The refractive indices variation with temperature in the isotropic and nematic phases is illustrated in Figures 1 and 2 for the compounds 8O.O4 and 8O.O7 respectively. The IN transition temperatures observed in the birefringence measurements are in agreement with those observed in density measurements as well as literature data.

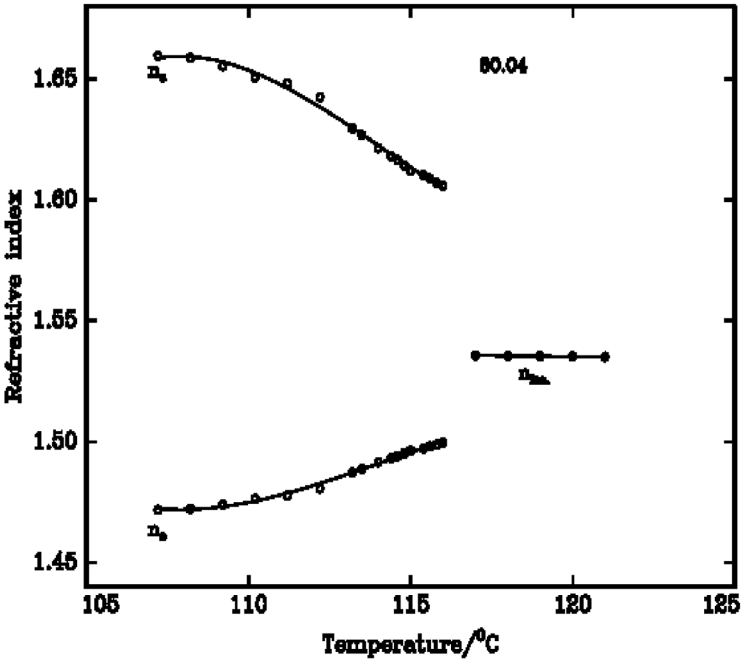

FIGURE 1 Variation of refractive indices with temperature in 8O.O4.

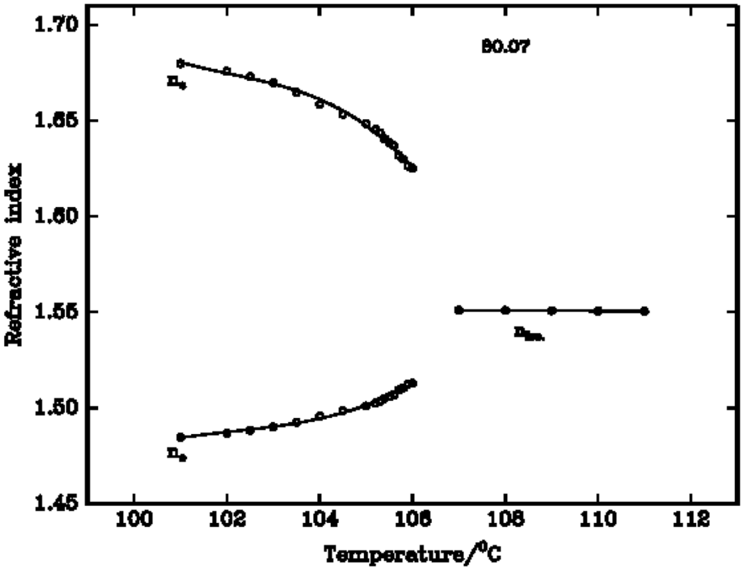

FIGURE 2 Variation of refractive indices with temperature in 8O.O7.

3a. Molecular Polarizabilities and Order Parameter S

In uniaxial LC the extraordinary (α_e) and ordinary (α_o) polarizabilities corresponding to the electric vector parallel and perpendicular to the optic axis are given as

$$\begin{aligned} \alpha_e &= \bar{\alpha} + 2(\alpha_{\parallel} - \alpha_{\perp})S/3 \\ \alpha_o &= \bar{\alpha} - (\alpha_{\parallel} - \alpha_{\perp})S/3 \end{aligned} \tag{1}$$

where S is the order parameter $\alpha_{\parallel}$ and $\alpha_{\perp}$ are polarizabilities of the molecule parallel and perpendicular to the long molecular axis of the LC molecule. The average molecular polarizability $\bar{\alpha}$ is given as

$$\bar{\alpha} = (\alpha_e + 2\alpha_o)/3 = (\alpha_{\parallel} + 2\alpha_{\perp})/3 \tag{2}$$

Combining the above two equations S, the order parameter is

$$S = (\alpha_e - \alpha_o)/(\alpha_{\parallel} - \alpha_{\perp}) \tag{3}$$

For the estimation of the molecular polarizabilities of LC molecules, the authors have considered Vuks model [2] which considers the local field of the molecule is isotropic and Neugebauer model which considers the local field as anisotropic. The relevant equations of the two models for the calculation of molecular polarizabilities are given below.

3b. Vuks Method

This model was first applied to LC molecules by Chandrasekhar *et al.* [10] assuming the internal field is isotropic even in anisotropic crystal. These assumptions lead to the following equations.

$$\begin{aligned} \alpha_e &= [3/(4\pi N)][(n_e^2 - 1)/(n^{-2} + 2)] \\ \alpha_o &= [3/(4\pi \mathrm{N})][(n_o^2 - 1)/(n^{-2} + 2)] \end{aligned} \tag{4}$$

where N is the number of molecules per unit volume, n_e and n_o are the extraordinary and ordinary refractive indices of the LC molecule.

$$n^{-2} = [(n_e^2 + 2n_o^2)/3]$$

and $N = N_{A\rho}/M$ where N_A is the Avogadro number, ρ is the density and M is the molecular weight.

3c. Neugebauer Method

Saupe and Maier [11] and Subramanyam *et al.* [12] applied this method to LC molecule. According to this method the molecular polarizabilities are

$$\alpha_e = \left(AB - 3 \pm \sqrt{(AB - 3)^2 - 4AB}\right)/2A \tag{5}$$

$$\alpha_o = \left(AB + 3 \pm \sqrt{(AB + 3)^2 - 16AB}\right)/4A \tag{6}$$

where

$$\begin{aligned} A = ((1/\alpha_e) + (2/\alpha_o)) = (4\pi N/3)[(n_e^2 + 2)/(n_e^2 - 1)] \\ + [(2((n_o^2 + 2))/((n_o^2 - 1))] \end{aligned} \tag{7}$$

$$B = \left(\alpha_{\parallel} + 2\alpha_{\perp}\right) = (\alpha_e + 2\alpha_o) = 3\alpha = 9(n^{-2} - 1)/\left[(4\pi N_i)(n^{-2} + 2)\right] \tag{8}$$

N_i is the number of molecules per unit volume in the isotropic phase.

Using the two models the molecular polarizabilities and the polarizability anisotropy ($\alpha_e - \alpha_o$) of the liquid crystals are calculated at different temperatures.

3d. Molecular Polarizability Anisotropy and Mean Polarizability

The modified Lippincott δ-function model is used for the evaluation of the above parameters in the case of all compounds. The detailed description is given in references [13,14]. For ready reference a brief description of the method employed for the liquid crystals is given below.

In crystalline state, there will be only the crystalline field acting on the system whereas in liquid phase, pure Brownian field only acts on the system. However, in liquid crystalline state, both these fields will be acting as this state will have the flow property like a liquid and anisotropic property like pure crystal. The resultant effect is to increase the potential on the electron (system). In other words, the shielding on the electrons will be less thus contributing to more polarization. This behaviour can be expressed empirically as

$$A_{LC} = A \exp[T - T_C]/T_C \tag{9}$$

where T is the temperature pertinent to the study of the liquid crystal property and T_C is the liquid crystalline transition temperature (clearing temperature). A and A_{LC} are the reduced electro negativities (REN) values in isotropic and LC phases.

$$\begin{aligned}\sum \alpha_{\|_P} = {} & [(4nA[\exp(T - T_C)/T_C])/a_0][(R^2/4) \\ & - (1/2C_R^2)]\exp[(x_1 - x_2)^2/4]\end{aligned} \tag{10}$$

Using the above Eq. (10), we can find out $\sum \alpha_{\|_P}$, and $\sum \alpha_{\|_n}$ and $\sum 2\alpha_\perp$ can be calculated using other formulae. Now the mean polarizability

$$\alpha = \left[\left(\sum \alpha_{\|_P} + \sum \alpha_{\|_n} + \sum 2\alpha_\perp\right)/3\right] \tag{11}$$

The mean polarizability determined from the above equation refers to the isotropic state and the calculation of mean polarizability for the LC state needs the following considerations. If N and N_{LC} are the number of molecules per unit volume in the isotropic and LC phases and α and α_{LC} are the respective polarizabilities, they can be related by the following equation.

$$(\mathrm{n}_{\mathrm{L}}^2 - 1)/(\mathrm{n}_{\mathrm{L}}^2 + 2) = (4\pi \mathrm{N}_{\mathrm{L}}/3)\alpha_{\mathrm{L}} \tag{12}$$

and

$$\begin{aligned}(1/3)\big[\big((\mathrm{n}_{\mathrm{e}}^2 - 1)/(\mathrm{n}_{\mathrm{e}}^2 + 2)\big) + \big(2\big(\mathrm{n}_{\mathrm{o}}^2 - 1\big)/\big(\mathrm{n}_{\mathrm{o}}^2 + 2\big)\big)\big] \\ = (4\pi \mathrm{N}_{\mathrm{LC}}/3)\alpha_{\mathrm{LC}}\end{aligned} \tag{13}$$

From the observation of many LCs, the refractivity of many LC compound so that does not depend very much on temperature the right side of the above two equations can be equated. Therefore,

$$\begin{aligned}&(4\pi N_L/3)\alpha_L = (4\pi N_{LC}/3)\alpha_{LC} \\ &\text{or} \\ &(\mathrm{N}_{\mathrm{L}}/\mathrm{N}_{\mathrm{LC}}) = (\alpha_L/\alpha_{LC})\end{aligned} \tag{14}$$

N_{L} and N_{LC} are related by the expressions

$$\mathrm{N}_{\mathrm{L}} = \mathrm{N}\rho_L/\mathrm{M} \text{ and } \mathrm{N}_{\mathrm{LC}} = \mathrm{N}\rho_{LC}/\mathrm{M}$$

Hence,

$$\mathrm{N}_{\mathrm{L}}/\mathrm{N}_{\mathrm{LC}} = \rho_L/\rho_{LC} \tag{15}$$

ρ_L and ρ_{LC} are the densities of the material in isotropic and LC states respectively and M is the molecular weight. From the density measurements ρ_{LC} is related to ρ_L by

$$\rho_{LC}/\rho_L = 1 + [(\mathrm{m}/\rho_L) - (T_C - T)] \tag{16}$$

m is the slope of the temperature vs density graph. Substituting Eq. (15) into Eq. (16) the mean polarizability of the LC compound is

$$\bar{\alpha} = \alpha_{\mathrm{L}}[1 - ((\mathrm{m}/\rho_{\mathrm{L}})(\mathrm{T}_{\mathrm{c}} - \mathrm{T}))] \tag{17}$$

where α_{L} is the mean polarizability in the isotropic state obtained from Lippincott δ-function model. Using above equation $\bar{\alpha}$ can be obtained.

3e. Estimation of Orientational Order Parameter in the Nematic Phase

The orientational order parameter $\mathrm{S} = 1/2\ (3\cos^2\theta - 1)$, where θ is the angle between the long axis of the molecule and the director 'n'.

TABLE 1 Polarizability Anisotropy from Lippincott δ-Function Method

Compound	Polarizability anisotropy ($10^{-24}\,\mathrm{cm}^3$) $\alpha_{\parallel}$	$\alpha_{\parallel_a}$	$\alpha_{\perp}$	Mean polarizability ($10^{-24}\,\mathrm{cm}^3$) α_M	$(\alpha_{\parallel}-\alpha_{\perp})$
8O.O3	84.22	1.09	54.27	46.52	31.04
8O.O4	88.02	1.09	56.41	48.50	32.71
8O.O6	95.31	1.09	60.60	52.36	33.70
8O.O7	99.11	1.09	62.83	54.34	37.27

For the estimation of the order parameter S the Eq. (3) is used. The α_e and α_o are estimated (using Vuks and Neugebauer equations) from the refractive index and density data. The principle polarizabilities viz., the $\alpha_{\parallel}$ and $\alpha_{\perp}$ are estimated from the Lippincott δ-function model (the particulars of the calculation is given above, and the scaling factors are obtained from both methods (Vuks and Neugebauer) and the estimation particulars are described below. The principle polarizabilities estimated from Lippincott δ-function model are given in Table 1. Further, the principal polarizabilities $\alpha_{\parallel}$ and $\alpha_{\perp}$ are also obtained from Haller extrapolation technique for the case of both the model and the values are depicted in Table 2.

In the Vuks method the order parameter is given by [15,16]

$$S = \left((\alpha)/\left(\alpha_{\parallel}-\alpha_{\perp}\right)\right)\left(\left(n_e^2-n_o^2\right)/\left(n^{-2}-1\right)\right) \tag{18}$$

where

$$n^{-2} = (n_e^2 + 2n_o^2)/3$$

In the Neugebauer method the order parameter S is given [3]

$$S = ((\alpha)/(\alpha_{\parallel}-\alpha_{\perp}))f(B) \tag{19}$$

TABLE 2 Value of Different Parameters Used for the Evaluation of Order Parameter Through Different Methods Obtained by Log–Log Graphs Extrapolated to Absolute Zero

		Haller method $(\alpha_{\parallel}-\alpha_{\perp})$		Scaling factors	
Compound	Δn	Vuks	Neugebauer	Vuks	Neugebauer
8O.O3	0.265	30.71	30.19	1.488	1.600
8O.O4	0.335	32.80	32.80	1.501	1.501
8O.O6	0.318	36.20	33.78	1.635	1.616
8O.O7	0.285	37.83	37.67	1.490	1.600

where

$$f(B) = (9/4AB)[(B^2 - (10/3)B + 1)^{1/2} + B/3 - 1]$$

and

$$B = [(n^2 - 1)/(n^2 + 1)][(n_e^2 + 2)/(n_e^2 - 1) + 2((n_o^2 + 2)/(n_o^2 - 1))]$$

The scaling factors for the determination of order parameter are obtained in both the cases by plotting log–log plots between $(n_e^2 - n_o^2)/(n^{-2} - 1)$ and f(B) in Vuks and Neugebauer cases respectively against $(T_C - T)/(T_C - T_{NC/NK})$ i.e., the reduced temperature. These factors are given in Table 2.

3f. Order Parameter S from Birefringence Studies

It is well known that any physical property of a nematic liquid crystal and the orientational order parameter S is closely connected to one another. Further, de Gennes [17] pointed out the anisotropy of any physical quantity can be a measure of orientational order. In the case of uniaxial liquid crystal, this parameter can be defined as

$$Q = \delta A/\Delta A \tag{20}$$

where $\delta A(A_{\|} - A_{\perp})$ is the anisotropy of any arbitrary physical quantity A and ΔA is the hypothetical anisotropy of A in the case of perfect order. Among many anisotropic physical quantities which could be used for the determination of the order parameter Q, the dielectric anisotropy for optical frequencies $\delta\varepsilon = n_e^2 - n_o^2$ is useful. Thus $\delta\varepsilon$ can be used [17] for the determination of S if a particular local field is applied for the liquid crystal molecule. Further, de Jeu [18] showed that birefringence, δn can be used for this purpose. Based on this, Kuczynski *et al.* [19,20] proposed a simple procedure for the determination of order parameter S from the birefringence measurements $(n_e - n_o)$ without considering the local field experienced by the molecule in a liquid crystal phase. The birefringence δn which is a function of temperature is fitted to the following equation:

$$\delta n = \Delta n \cdot (1 - (T/T^*))^{\beta} \tag{21}$$

where T is the absolute temperature, T^* and β are constants. (T^* is about 1–4 K higher than the clearing temperature and the exponent β is close to 0.20). This procedure enables one to extrapolate δn to the absolute zero temperature. In practice, the three adjustable parameters T*, Δn and β were obtained by fitting the experimental data for δn to the following equation written in the logarithmic form:

$$\log \delta \mathrm{n} = \log \Delta \mathrm{n} + \beta \cdot \log((T^* - T)/T^*) \tag{22}$$

In the present investigations, the values of log Δn and β are calculated by the linear regression method. The parameter T* is adjusted to get the best correlation coefficient of the linear regression. Thus, S is given by

$$Q = \delta \mathrm{n}/\Delta \mathrm{n} \tag{23}$$

The linear fit for all the compounds is presented in Figure 3. For the rest of the compounds, the Δn values, birefringence in perfect order are given in Table 2. The order parameter Q determined in this way (Eq. (23)) describes well the nematic order parameter.

The orientational order parameter S is evaluated using all these seven methods viz., 1. S calculated using polarizability anisotropy calculated by Lippincott δ-function method and the molecular polarizabilities obtained from Vuks model, 2. S calculated using polarizability anisotropy calculated by Lippincott δ-function method and the

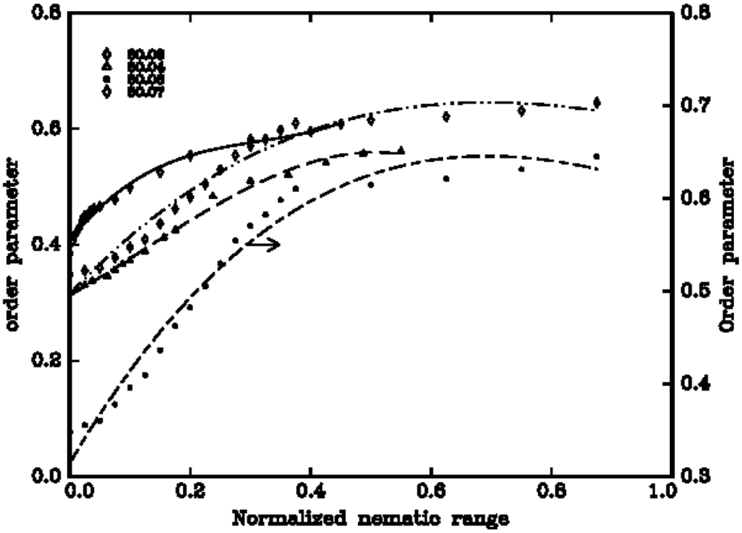

FIGURE 3 The log-log plot of birefringence δn, verses reduced temperature for the Compounds 8O.O3, 8O.O4, 8O.O6 and 8O.O7.

molecular polarizabilities obtained from Neugebauer model, 3. S calculated using Δn from Eq. (23), 4. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Vuks model, 5. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Neugebauer model, 6. S calculated using Vuks scaling factor (Eq. (18)) and 7. S calculated using Neugebauer F(B) parameter (Eq. (19)). The variation of order parameter with nematic normalized range for the case of 8O.O4 and 8O.O7 are shown in Figures 4 and 5.

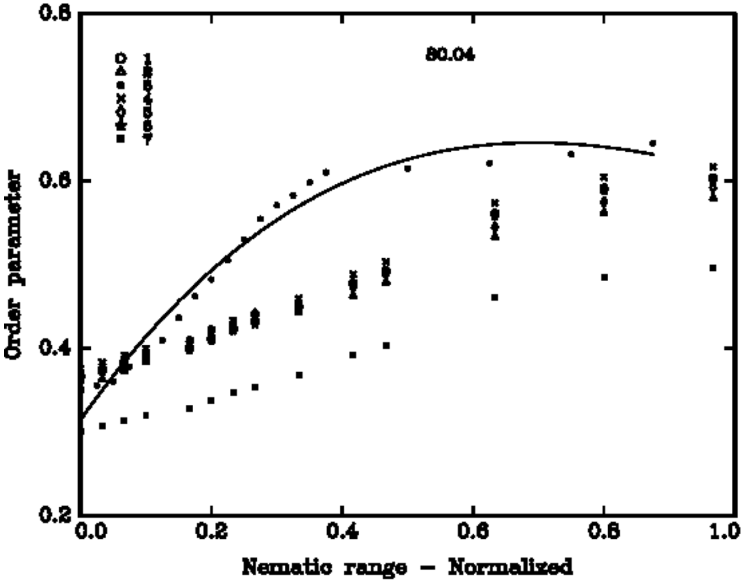

FIGURE 4 Order parameter verses the normalized nematic range for the compound 8O.O4. 1. S calculated using polarizability anisotropy calculated using Lippincott δ-function method and the molecular polarizabilities obtained using Vuks model, 2. S calculated using polarizability anisotropy calculated using Lippincott δ-function method and the molecular polarizabilities obtained using Neugebauer model, 3. S calculated using Δn from Eq. (23), 4. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Vuks model, 5. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Neugebauer model, 6. S calculated using Vuks scaling factor (Eq. (18)) and 7. S calculated using Neugebauer F(B) parameter (Eq. (19)).

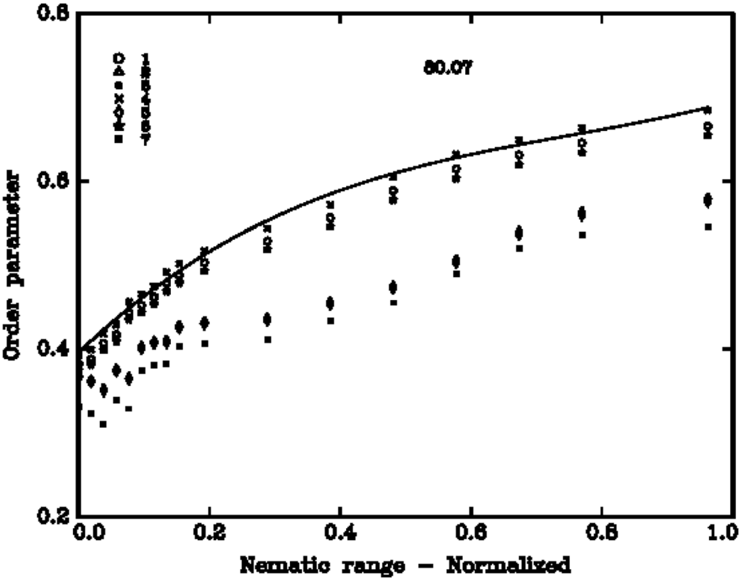

FIGURE 5 Order parameter verses the normalized nematic range for the compound 8O.O7. 1. S calculated using polarizability anisotropy calculated using Lippincott δ-function method and the molecular polarizabilities obtained using Vuks model, 2. S calculated using polarizability anisotropy calculated using Lippincott δ-function method and the molecular polarizabilities obtained using Neugebauer model, 3. S calculated using Δn from Eq. (23), 4. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Vuks model, 5. S calculated using polarizability anisotropy calculated using Haller's extrapolation method and the molecular polarizabilities obtained using Neugebauer model, 6. S calculated using Vuks scaling factor (Eq. (18)) and 7. S calculated using Neugebauer F(B) parametesr (Eq. (19)).

The Salient Features of the Study are

- the birefringence observed in all the compounds is around 0.25 to 0.30,
- the birefringence could not be observed in the smectic-C and smectic-G phases of compounds 8O.O4, 8O.O6 and 8O.O7 as the splitting has blurred and this may be due to absorption and diffusion of the light,
- the extraordinary and ordinary refractive indices (n_e and n_o) attain saturation deep in the nematic phase as expected like the orientational order parameter,

- the mean polarizability obtained using modified Lippincott δ-function model is reasonably good agreement with the mean polarizability obtained from birefringence data using both isotropic and anisotropic internal field models due to Vuks and Neugebauer respectively,
- the order parameter evaluated using both the models show increase with the decrease of temperature and attains saturation deep in the nematic region,
- there is divergence in the values of S from one method to the other. In the compounds 8O.O4 and 8O.O7, it has been found that that value calculated using Δn is found to be higher when compared to the others. From our systematic study on number of similar compounds common rigid core with different end chain length, in most of the cases it has been found that the S value calculated falls in middle of the plots due to other methods,
- depending on the compounds there is close agreement between some methods. However, there exists no commonality,
- One important observation which applies to all compounds is that the S value calculated using Neugebauer F(B) parameter exhibits low value compared to the others.

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