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Refractive Index, Density and Distribution Function in Nematic Mixtures

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Using refractive index and density data for the nematogenic mixtures Octyl benzoic acid (OBA) and Nonyl benzoic acid (NBA), orientational order and hence the distribution function has been determined. From this higher order parameter $\langle P_4 \rangle$ has been computed.

Keywords: Refractive index; density; distribution function

Introduction

Nematic liquid crystals and their mixtures are found to be more useful for display devices due to their exquisite optical and electrical properties [1-11]. Such nematic mixtures are extremely stable, chemically as well as photochemically [12]. By mixing two nematogens one can often obtain a material with a lower melting point. Usually two nematogens exhibit the property of continuous miscibility without crossing any line (first or second order transition line) and possess the same symmetry [1]. Binary systems offer the advantages of frequently exhibiting eutectic behaviours in their solid-mesophase transition, whereas the mesophase-isotropic transition temperature varies linearly with composition. In particular several binary nematic systems have been studied and only small deviations from linearity in the variation of nematic-isotropic transition temperature are noted, even in cases with significant differences in molar volumes and densities of the components. This general behaviour has been satisfactorily accounted for theoretically by Humphries and Luchurst [13]. In this paper, we report the experimental measurements of the refractive indices (n_e , n_o), and

densities (ρ) at different temperatures for the mixtures of two nematic compounds viz., OBA and NBA. Using these data, the orientational order parameter $\langle P_2 \rangle$ has been estimated employing Neugebauer relations. By considering the orientational order parameter corresponding to the nematic-isotropic transition temperature and based on an empirical relation, we have determined the distribution function. From this the higher order parameter $\langle P_4 \rangle$ has been computed.

Experimental

The samples used in this investigation were obtained from M/s Merck Ltd., UK. For use in our experiments, they were purified by recrystallization from their solutions in benzene. Mixtures of OBA in NBA of different concentrations were prepared. The components of mixtures were well mixed in the molten state and allowed to cool very slowly. Its crystal to nematic and nematic to isotropic transition temperatures were determined using a Leitz polarizing microscope equipped with a specially constructed hot stage. The densities of the mixtures at different temperatures in the nematic and isotropic phases were determined using capillary tube technique. The refractive indices of the mixtures were determined at different temperatures using a precision goniometer spectrometer SGO 1.1 (Frieber Prazision mechanik, East Germany) and a hollow glass prism of small angle ($3-5^\circ$) for the wavelength 5780\AA (Hg yellow). The technique of measurement of refractive index and density has been described in an earlier paper [10]. The measurement of the temperature, refractive index and density are estimated to an accuracy of $\pm 0.1^\circ\text{C}$, 0.001 and 0.001 gm/cm^3 respectively.

Results and Discussion

Figures 1 and 2 respectively show the measured densities and the refractive indices of different mixtures (20%, 40% and 60% of OBA in NBA) at various temperatures in the nematic and isotropic phases. Using these data and by employing Neugebauer relations [14-16], effective polarizabilities and hence the orientational order parameter for all the mixtures in their nematic phase have been estimated. Figure 3 shows the variation of the orientational order parameter with temperature in the cases of different mixtures in their nematic phase.

Estimation of distribution function $f(\theta)$ and the higher order parameter $\langle P_4 \rangle$

According to Maier-Saupe model, the distribution function $f(\theta)$ of a nematogenic system can be assumed to form a Gaussian distribution around the director and can be written as [17]

$$f(\theta) = 1/\sigma [\exp(-\theta^2/2\sigma^2) + \exp\{(\theta - \pi)^2/2\sigma^2\}] \quad (1)$$

Where σ is the deviation. The orientational order parameter or the second moment is given by

$$\langle P_2 \rangle = \int_0^{\pi/2} P_2 (\cos(\theta) f(\theta) \sin\theta d\theta) / \int_0^{\pi/2} f(\theta) \sin\theta d\theta \quad (2)$$

Using this expression and the order parameter obtained from the refractive index data we have estimated the parameter σ by employing the one dimensional minimization programme of SIMPLEX [18] at various temperatures for different mixtures. Now using the computed σ value we have determined the distribution function $f(\theta)$. We have plotted in Figure 4 the variation of the distribution function $f(\theta)$ with θ for a 60% of OBA mixture at various T_c -T. The distribution function $f(\theta)$ is more fundamental than the parameters $\langle P_2 \rangle$ and $\langle P_4 \rangle$. In fact the orientational order parameter $\langle P_2 \rangle$ is directly related to the measure of variance (or width) of $f(\theta)$. Higher order parameter $\langle P_4 \rangle$ which is a measure of Peakedness of $f(\theta)$. It is evident from Figure 4 that, with increase in temperature $\langle P_2 \rangle$ as well as $\langle P_4 \rangle$ decrease showing that there is a decrease in the ordering of the molecules in the nematic phase. Higher order parameter $\langle P_4 \rangle$ is given by

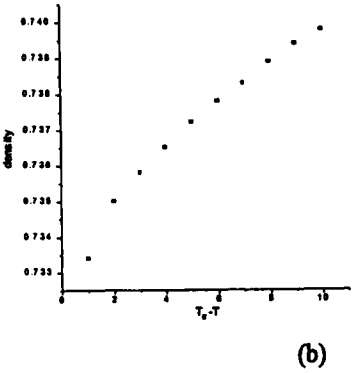
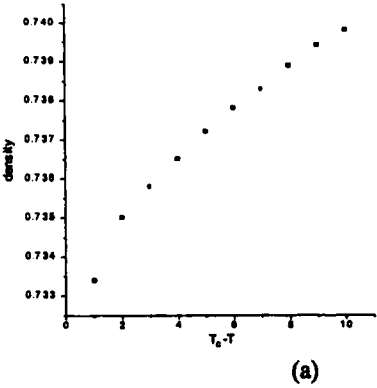
$$\langle P_4 \rangle = \int_0^{\pi/2} P_4 (\cos(\theta) f(\theta) \sin\theta d\theta) / \int_0^{\pi/2} f(\theta) \sin\theta d\theta \quad (3)$$

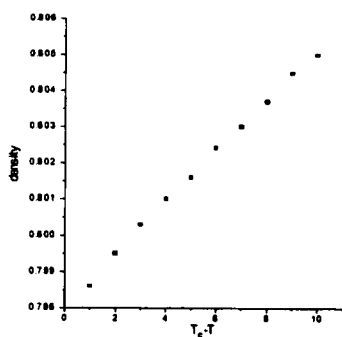
In Figure 5, we have plotted $\langle P_4 \rangle$ versus $\langle P_2 \rangle$ for a 60% of OBA mixture, along with the results of Maier-Saupe, Hamphries-James-Luckhurst (HJL) and the limit, which arises from Schwartz inequality [Ref.1, p56],

$$P_4 \geq (35 P_2^2 - 10 P_2 - 7)/18 \quad (4)$$

The computed values of $\langle P_4 \rangle$ are in agreement with Hamphries-James-Luckhurst model which suggests that the pair correlations are quite important in these systems and they can have a profound influence on the interpretation of data based on a one-particle

approximation. The fact that $\langle P_4 \rangle$ is smaller than predicted by Maier-Saupe theory suggests that ϕ fluctuations are not large as expected from the theory.





(c)

Figure 1 variation of density with $T_c - T$.

(a) 20% OBA+80%NBA

(b) 40%OBA+60%NBA

(c) 60%OBA+40%NBA

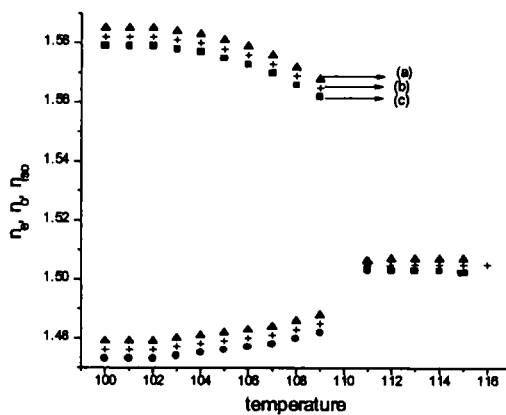


Figure 2 variation of refractive indices with temperature.

(a) 60%OBA+40%NBA

(b) 40%OBA+60%NBA

(c) 20%OBA+80%NBA

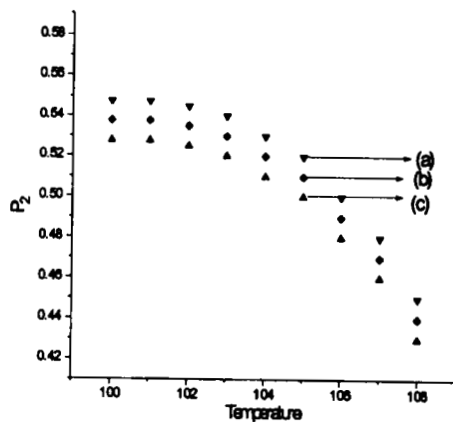


Figure 3 variation of $\langle P_2 \rangle$ with temperature.

(a) 60%OBA+40%NBA

(b) 40%OBA+60%NBA

(c) 20%OBA+80%NBA

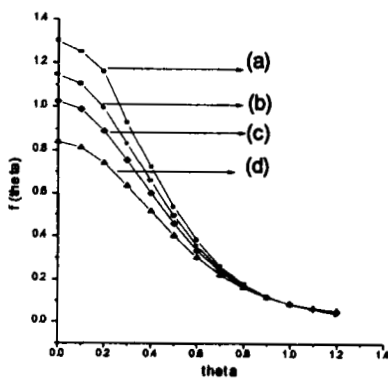


Figure 4: Variation of $f(\theta)$ with θ for a case of 60%OBA+40%NBA at various T_c-T .

(a) $T_c-T=10^\circ\text{C}$ (b) $T_c-T=8^\circ\text{C}$ (c) $T_c-T=6^\circ\text{C}$ (d) $T_c-T=4^\circ\text{C}$

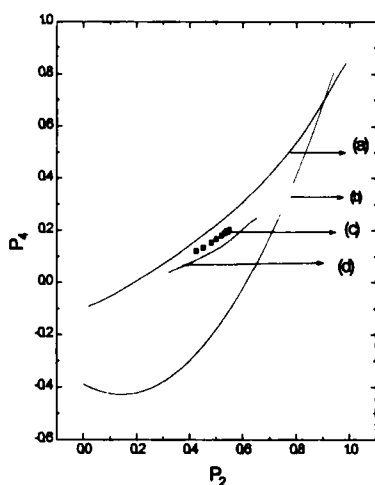


FIGURE 5: The variation of P_4 with respect to P_2
 (a) Maier-Saupe Model
 (b) $P_4 \geq (35 P_2^2 - 10 P_2 - 7)/18$
 (c) 60% of OBA+ 40% NBA
 (d) Humphries-James-Luckhurst Model

Conclusion

Using refractive index and density data for the mixtures of OBA and NBA we have determined the orientational order parameter $\langle P_2 \rangle$ and hence distribution function has been determined. From this the higher order parameter $\langle P_4 \rangle$ has been computed using Humphries-James-Luckhurst model estimated for these mixtures. With increase in temperature $\langle P_2 \rangle$ as well as $\langle P_4 \rangle$ decreases showing that there is a decrease in the ordering of the molecules in the nematic phase. Also optical method gives a better agreement with the Humphries-James-Luckhurst for the plot of $\langle P_4 \rangle$ versus $\langle P_2 \rangle$ indicating that the pair correlations are quite important.

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