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# Anomalous density and refractive index variation of a nematic liquid crystal

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# Anomalous density and refractive index variation of a nematic liquid crystal

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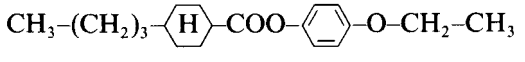
This paper reports the results of the measurement of refractive indices and densities of *p*-ethoxyphenyl trans-4-butyl cyclohexane carboxylate at different temperatures. The molecular polarizabilities have been calculated from refractive indices using Vuks' formula and orientational order parameters are determined from the polarizability values. The order parameter values have been compared with those obtained from Maier–Saupe theory. The compound shows anomalous behaviour regarding variation of density, ordinary refractive index and order parameter values with temperature. This is consistent with our previous X-ray study of this compound. The possible reason for this peculiar behaviour has been discussed.



## 1. Introduction

The birefringence of liquid crystals is the visible result of their long-range order and is defined only for a uniformly ordered domain. Therefore, by studying the birefringence we can estimate the degree of ordering of molecules at different temperatures. In this paper we have reported the results of the measurement of refractive indices, densities and order parameters of *p*-ethoxyphenyl trans-4-butyl cyclohexane carboxylate (EPBCC) at different temperatures. The structural formula of the compound is given below.



## 2. Experimental

A purified and recrystallized sample was donated to us by Hoffmann La-Roche and Co., which was used without further purification. The transition temperatures found by us from texture studies using a Metler hot stage, agree well with those mentioned in the literature. However, we found that the compound possesses a supercooled nematic phase confirmed by X-ray diffraction studies [1] and magnetic anisotropy measurements [2]. The transition temperatures of the compound obtained by us are given below; the transition temperatures reported by Beens and de Jeu [3] are slightly different.

$$\text{Solid} \underset{\sim 11^{\circ}\text{C}}{\overset{36{\cdot}3^{\circ}\text{C}}{\rightleftharpoons}} \text{Nematic} \overset{74{\cdot}6^{\circ}\text{C}}{\rightleftharpoons} \text{Isotropic.}$$

The refractive indices have been determined using the thin prism method. The details of the experimental procedure have already been reported by Zeminder *et al.* [4]. A magnetic field of 6·0 kgauss was applied to align the optic axis along the refracting edge

of the prism. The temperature during the experiment was controlled to $\pm 0{\cdot}5°C$. The densities of the sample at different temperatures were determined by putting the weighed sample inside a glass capillary tube which was placed in a water bath heated electrically with a temperature controller. Sufficient time was allowed for obtaining the desired temperature. The length of the column was measured by a travelling microscope. The densities were calculated after correcting for the expansion of the glass tube.

## 3. Results and discussions

The values of the experimental refractive indices for ordinary and extraordinary rays ($n_o$, $n_e$) are shown in figure 1. We used Vuk's formula [5] to calculate the principal polarizabilities ($\alpha_o$, $\alpha_e$). The table contains our polarizability values for four different wavelengths and the densities of the compound at different temperatures.

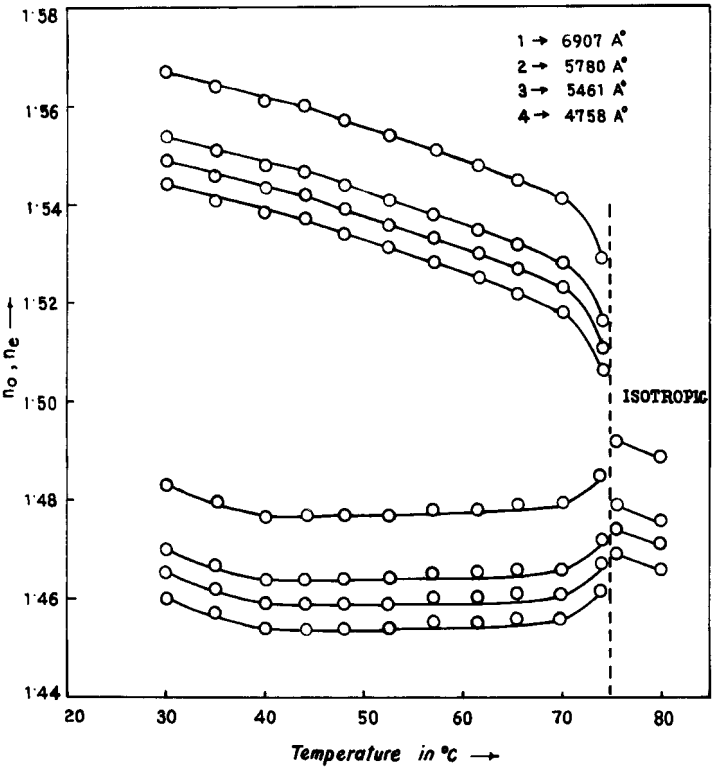



Figure 1. Variation of the refractive indices of EPBCC with temperature.

It is evident from figure 1 that the refractive index of the ordinary ray first decreases with temperature and then rises after reaching a minimum value. Measurement of the density at different temperatures shows that it reaches a maximum at the temperature where the minimum value of the refractive index ($n_o$) occurs and then begins to decrease in the conventional way. We repeated these measurements several times and always found the same variation of $n_o$ and density with temperature. The variation of the density with temperature is shown in figure 2. Our X-ray study [1] of the compound shows anomalous behaviour in the value of the intermolecular distance, $D$ (figure 2).

Density ($\varrho$), polarizability ($\alpha$) and orientational order parameters ($S$) of EPBCC.

| Temperature (°C) | $\varrho$ (g/cm$^3$) | $\lambda$ 6907 Å | | | 5780 Å | | | 5461 Å | | | 4758 Å | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | $\alpha_o$ | $\alpha_e$ | $\langle P_2 \rangle$ | $\alpha_o$ | $\alpha_e$ | $\langle P_2 \rangle$ | $\alpha_o$ | $\alpha_e$ | $\langle P_2 \rangle$ | $\alpha_o$ | $\alpha_e$ | $\langle P_2 \rangle$ |
| 30 | 1·0565 | 30·63 | 37·46 | 0·6481 | 30·92 | 37·75 | 0·6480 | 31·20 | 38·03 | 0·6482 | 31·94 | 38·76 | 0·6483 |
| 35 | 1·0580 | 30·41 | 37·24 | 0·6472 | 30·70 | 37·52 | 0·6471 | 30·99 | 37·81 | 0·6472 | 31·72 | 38·54 | 0·6471 |
| 40 | 1·0590 | 30·21 | 37·03 | 0·6467 | 30·51 | 37·24 | 0·6467 | 30·79 | 37·52 | 0·6468 | 31·53 | 38·26 | 0·6466 |
| 44 | 1·070 | 29·91 | 36·57 | 0·6324 | 30·19 | 36·86 | 0·6325 | 30·48 | 37·14 | 0·6325 | 31·21 | 37·87 | 0·6323 |
| 48 | 1·069 | 29·96 | 36·39 | 0·6099 | 30·24 | 36·67 | 0·6098 | 30·53 | 36·95 | 0·6099 | 31·26 | 37·68 | 0·6096 |
| 52·5 | 1·066 | 30·07 | 36·27 | 0·5885 | 30·35 | 36·55 | 0·5884 | 30·64 | 36·84 | 0·5886 | 31·37 | 37·57 | 0·5884 |
| 57 | 1·053 | 30·52 | 36·48 | 0·5646 | 30·81 | 36·76 | 0·5446 | 31·10 | 37·05 | 0·5447 | 31·85 | 37·79 | 0·5448 |
| 61·5 | 1·0498 | 30·64 | 36·36 | 0·5429 | 30·93 | 36·66 | 0·5430 | 31·22 | 36·94 | 0·5428 | 31·97 | 37·68 | 0·5426 |
| 65·5 | 1·0462 | 30·83 | 36·25 | 0·5135 | 31·13 | 36·54 | 0·5136 | 31·42 | 36·83 | 0·5134 | 32·17 | 37·57 | 0·5132 |
| 70 | 1·039 | 31·08 | 36·19 | 0·4855 | 31·37 | 36·49 | 0·4856 | 31·66 | 36·78 | 0·4854 | 32·42 | 37·53 | 0·4852 |
| 74 | 1·0275 | 31·92 | 35·58 | 0·3477 | 32·22 | 35·88 | 0·3478 | 32·51 | 36·18 | 0·3478 | 33·28 | 36·94 | 0·3476 |

$\alpha_o$ and $\alpha_e$ are in $10^{-24}$ cm$^3$.

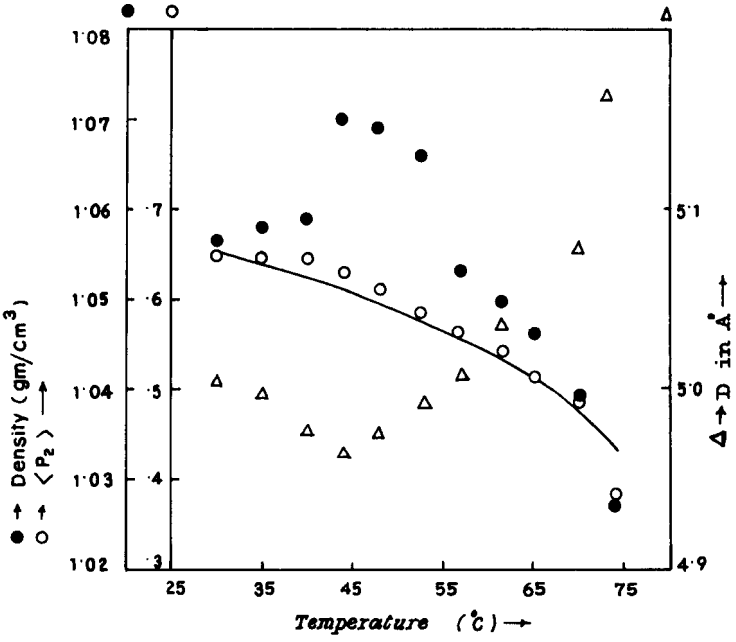



Figure 2. Variation of the order parameter (○), density (●) and intermolecular distance (△) with temperature. The continuous curve is obtained from the Maier–Saupe theory [8].

The orientational order parameter $\langle P_2 \rangle$ was calculated from molecular polarizabilities by using the relation [6]

$$\langle P_2 \rangle = (\alpha_e - \alpha_o)/(\alpha_\parallel - \alpha_\perp),$$

where $\alpha_\parallel$ and $\alpha_\perp$ are the molecular polarizabilities parallel and perpendicular to the long axis of the molecule. We used the extrapolation procedure given by Haller *et al.* [7] to calculate $(\alpha_\parallel - \alpha_\perp)$. Figure 2 shows the variation of the order parameters of the sample with temperature. Anomalous behaviour is also observed in the $\langle P_2 \rangle$ values.

In summary, it would appear that the minimum in $n_o$ and maximum in $\langle P_2 \rangle$ can be attributed to the minimum in the nearest neighbour distances derived from X-ray studies.

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