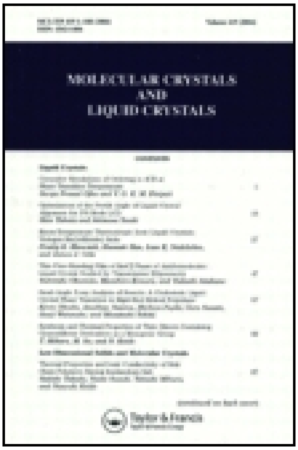

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Birefringence and Order Parameter of Some Alkyl and Alkoxycyanobiphenyl Liquid Crystals

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The refractive indices and densities of six liquid crystals—two alkylcyanobiphenyls, three alkoxycyanobiphenyls and one mixture of two alkylcyanobiphenyls have been reported as a function of temperature. The effective polarizabilities α_e and α_0 in the nematic phase calculated using the methods due to Neugebauer, Vuks and Saupe and Maier, are found to be appreciably different, though the order parameter S evaluated with those values of $(\alpha_e - \alpha_0)$ are in good agreement. The preference of one method over the other two is discussed.

INTRODUCTION

The importance of liquid crystals for their applications in display devices and other manifold scientific uses is now well known. The orientational order parameter, S, of a liquid crystal is an important parameter, the knowledge of which is essential for use in a specific purpose. Various methods are now being used to measure the order-parameter as has been discussed by Saupe and Maier,[1] namely, nuclear magnetic reasonance, dielectric permittivity, diamagnetic susceptibility, refractive indices, X-ray scattering, UV and IR dichroism, etc. In this paper the order parameters of these liquid crystals have been reported from the measurements of refractive indices and densities at different temperatures in the nematic and isotropic phases. Three methods, namely, (1) Neugebauer's anisotropic internal field,[2]

(2) Vuks' isotropic internal field,[3] and (3) Saupe and Maier's anisotropic model[4] with a single internal field parameter, have been used to calculate the effective polarizabilities α_e and α_0. The results have been discussed in this paper.

EXPERIMENTAL

The structural formulae and nematic-isotropic transition temperatures of six liquid crystals which were obtained from BDH, England are given below.

1. K_{15} : 4-Cyano-4′-*n*-Pentylbiphenyl, $C \xleftrightarrow{22.5\,°C} N \xleftrightarrow{35\,°C} I$
2. K_{21} : 4-Cyano-4′-*n*-Heptylbiphenyl, $C \xleftrightarrow{28.5\,°C} N \xleftrightarrow{42\,°C} I$
3. Mixture E_1 composed of components 1 and 2 in proportion of 59 : 41 $C \xleftrightarrow{-2\,°C} N \xleftrightarrow{37\,°C} I$
4. M_{15} : 4-Cyano-4′-*n*-Pentyloxybiphenyl, $C \xleftrightarrow{48\,°C} N \xleftrightarrow{67.5\,°C} I$
5. M_{21} : 4-Cyano-4′-*n*-Heptyloxybiphenyl, $C \xleftrightarrow{53.5\,°C} N \xleftrightarrow{75\,°C} I$
6. M_{24} : 4-Cyano-4′-*n*-Octyloxybiphenyl, $C \xleftrightarrow{67\,°C} N \xleftrightarrow{80\,°C} I$

The liquid crystals were used in the investigations without further purification.

The refractive indices n_e and n_0 for the extraordinary ray and ordinary ray in the nematic phase and refractive index n, in the isotropic phase at different temperatures were measured by means of an Abbe refractometer. The glass prisms of the refractometer were rubbed with a lens paper along the length of the prisms several times. A few drops of the liquid crystal were allowed to fall on the lower prism and were spread with a spatula. These operations helped to align the molecules of the liquid crystal along the prism face. The alignment was complete when the upper prism was clamped in place.

Two positions of a nicol placed over the eyepiece allow distinct separation of the dark and bright space in the eyepiece, corresponding to n_e and n_0 (in the nematic $n_e > n_0$). A second nicol placed between the light source and the lower prism makes the separation between the dark and bright space sharper. The temperature of the refractometer was maintained constant within ±0.2°C by means of a thermostat. When the value of refractive index n_e for extraordinary ray lay outside the range of the refractometer ($n_e > 1.70$), it was calculated from the relation $n^2 = \frac{1}{3}(2n_0^2 + n_e^2)$ where n is the isotropic refractive index extrapolated at the appropriate temperature.

The densities of the liquid crystals in the nematic and isotropic phases were determined by introducing a weighed sample of liquid crystal inside a glass capillary tube in a thermostat. The length of the column in the capillary was measured at different temperatures with a travelling microscope. The density was calculated after correcting for the expansion of the glass tube.

RESULTS

The values of the refractive indices of the liquid crystals in the isotropic phase n, and in the nematic phase, n_e and n_0 together with the densities of the liquid crystals at different temperatures are given in Tables I and II. The refractive index anisotropy is shown in Figures 1 and 2. The effective polarizabilities α_e and α_0 of the liquid crystals were calculated using three methods: 1) Neugebauer's[2] method of anisotropic internal field, given by

$$n_e^2 - 1 = 4\pi N\alpha_e(1 - N\alpha_e\gamma_e)^{-1} \tag{1}$$

and

$$n_0^2 - 1 = 4\pi N\alpha_0(1 - N\alpha_0\gamma_0)^{-1} \tag{2}$$

where N is the number of molecules per c.c and γ_i's are the internal field constants. The relevant equations for calculating the polarizabilities α_e and α_0 obtained from Eqs. (1) and (2), are

$$\frac{1}{\alpha_e} + \frac{2}{\alpha_0} = \frac{4\pi N}{3}\left[\frac{n_e^2 + 2}{n_e^2 - 1} + \frac{2(n_0^2 + 2)}{n_0^2 - 1}\right] \tag{3}$$

and

$$\alpha_e + 2\alpha_0 = \alpha_{\parallel} + 2\alpha_{\perp} = \frac{9}{4\pi N}\left[\frac{n^2 - 1}{n^2 + 2}\right] \tag{4}$$

TABLE I

Values of density, refractive indices, n_e, n_0 and n of Pentylcyanobiphenyl, Heptylcyanobiphenyl, Mixture E_1

Pentylcyanobiphenyl				Heptylcyanobiphenyl				Mixture E_1			
Temp° C	Density gm/cc	n_0	n_e	Temp° C	Density gm/cc	n_0	n_e	Temp° C	Density gm/cc	n_0	n_e
24 nem	1.0220	1.5350	1.6975	29 nem	0.9874	1.5200	1.6852	28 nem	1.0059	1.5300	1.7080
28	1.0185	1.5365	1.6891	32	0.9845	1.5217	1.6788	30	1.0037	1.5302	1.7040
30	1.0160	1.5380	1.6836	35	0.9813	1.5220	1.6740	32	1.0018	1.5321	1.6960
32	1.0144	1.5381	1.6827	38	0.9777	1.5248	1.6638	34	1.0001	1.5330	1.6880
34	1.0143	1.5450	1.6672	45 Iso	0.9680	1.5700		36	0.9964	1.5370	1.6798
37.5 Iso	1.0051	1.5860		50	0.9632	1.5665		38 Iso	0.9931	1.5804	
41	1.0024	1.5849		55	0.9589	1.5650		42	0.9886	1.5800	
43	0.9999	1.5835						45	0.9863	1.5770	

TABLE II

Values of density, refractive indices n_e, n_0 and n of Pentyloxycyanobiphenyl, Heptyloxycyanobiphenyl, Octyloxycyanobiphenyl

Pentyloxycyanobiphenyl				Heptyloxycyanobiphenyl				Octyloxycyanobiphenyl			
Temp° C	Density gm/cc	n_0	n_e	Temp° C	Density gm/cc	n_0	n_e	Temp° C	Density gm/cc	n_0	n_e
49 nem	1.0455	1.5260	1.7058	55 nem	1.0127	1.5140	1.6820	69 nem	0.9956	1.5076	1.6513
52	1.0426	1.5270	1.7018	56	1.0115	1.5142	1.6802	71	0.9934	1.5090	1.6465
55	1.0399	1.5276	1.6968	58	1.0097	1.5150	1.6774	73	0.9914	1.5110	1.6423
58	1.0370	1.5280	1.6910	60	1.0078	1.5155	1.6734	75	0.9896	1.5156	1.6332
61	1.0339	1.5318	1.6807	63	1.0051	1.5161	1.6698	77	0.9876	1.5240	1.6169
65	1.0297	1.5368	1.6687	67	1.0012	1.5174	1.6623	78	0.9867	1.5300	1.6050
66	1.0284	1.5390	1.6601	70.5	0.9981	1.5200	1.6536	79	0.9854	1.5370	1.5910
67	1.0274	1.5430	1.6521	72	0.9966	1.5245	1.6418	80	0.9842	1.5450	1.5748
68	1.0262	1.5560	1.6270	73	0.9954	1.5262	1.6376	81 Iso	0.9810	1.5550	
69 Iso	1.0229	1.5802		74	0.9938	1.5400	1.6109	82	0.9796	1.5549	
70	1.0225	1.5790		75 Iso	0.9906	1.5635		83	0.9785	1.5542	
72	1.0205	1.5788		76	0.9901	1.5630		85	0.9765	1.5540	
74	1.0192	1.5778		77	0.9895	1.5620		87	0.9746	1.5530	
76	1.0170	1.5774		79.5	0.9869	1.5610					

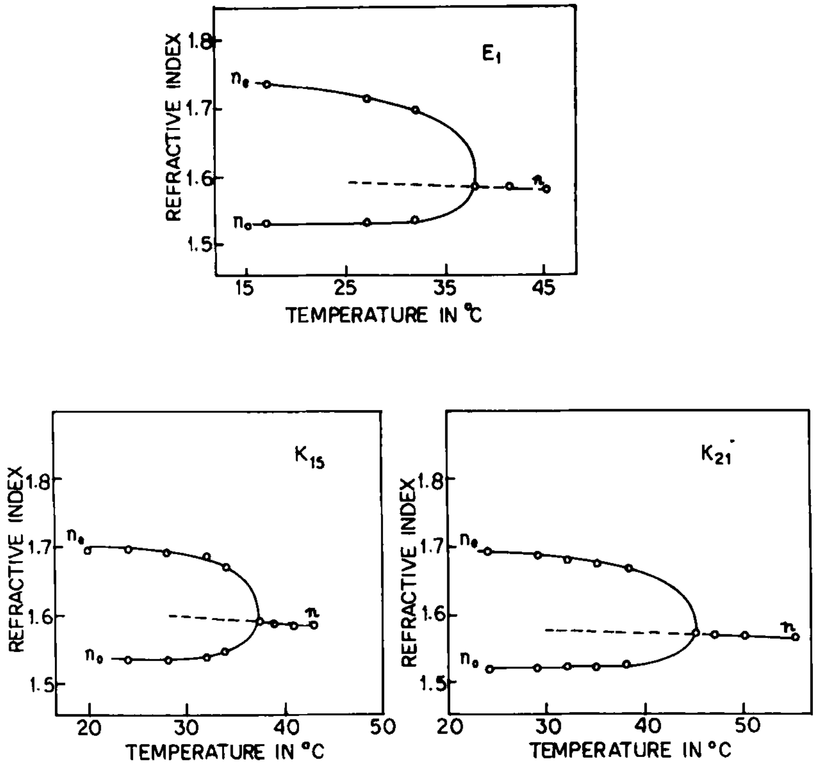

FIGURE 1 Refractive index anisotropy plots for E_1, K_{15}, K_{21}

2) Vuks[3] assumed the local field to be isotropic and his modifications of the Lorentz–Lorenz equation are:

$$\frac{n_e^2 - 1}{\overline{n^2} + 2} = \frac{4\pi N\alpha_e}{3} \tag{5}$$

and

$$\frac{n_0^2 - 1}{\overline{n^2} + 2} = \frac{4\pi N\alpha_0}{3} \tag{6}$$

where

$$\overline{n^2} = \tfrac{1}{3}\left(n_e^2 + 2n_0^2\right)$$

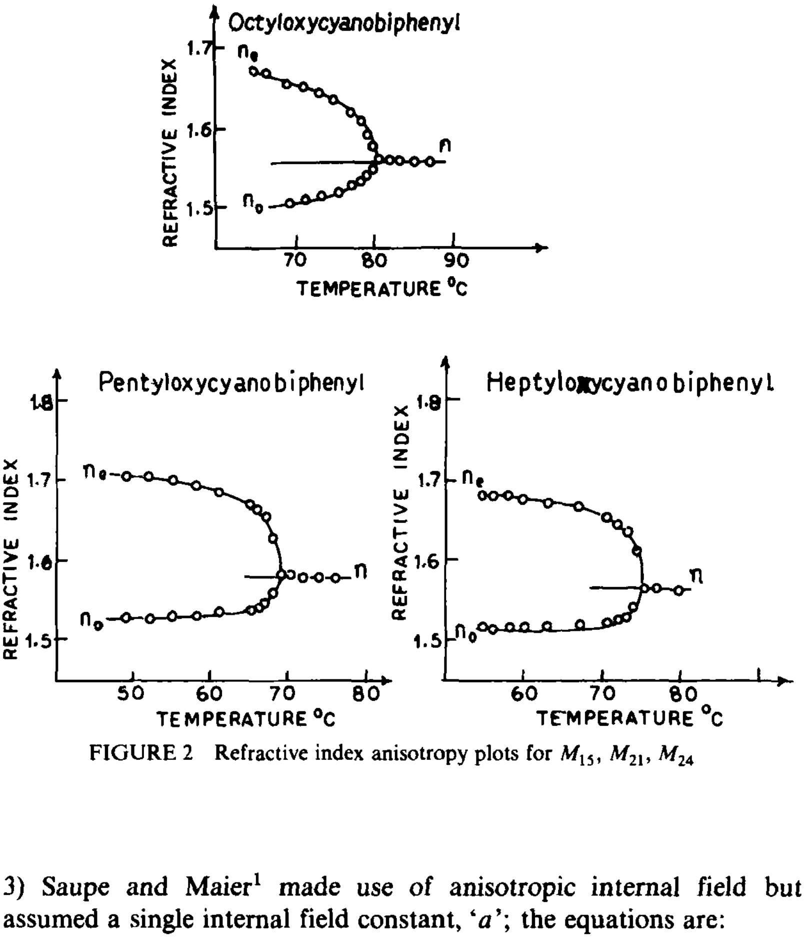

FIGURE 2 Refractive index anisotropy plots for M_{15}, M_{21}, M_{24}

3) Saupe and Maier[1] made use of anisotropic internal field but assumed a single internal field constant, '*a*'; the equations are:

$$\frac{n_e^2 - 1}{n_e^2 + 2 - 2a\left(n_e^2 - 1\right)} = \frac{4\pi N}{3}\alpha_e \tag{7}$$

and

$$\frac{n_0^2 - 1}{n_0^2 + 2 + a\left(n_0^2 - 1\right)} = \frac{4\pi N}{3}\alpha_0 \tag{8}$$

The value of the internal field parameter '*a*' was obtained from the

TABLE III*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ Å and order parameter S of Pentylcyanobiphenyl

	Neugebauer's Method			Vuks' Method			Saupe and Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
24	38.67	29.62	0.62	40.10	28.90	0.64	40.77	29.07	0.67
28	38.35	29.81	0.59	39.68	29.14	0.60	40.39	29.24	0.64
30	38.13	29.98	0.56	39.41	29.33	0.57	39.99	29.43	0.61
32	37.80	30.19	0.52	39.34	29.34	0.56	39.68	29.50	0.58
34	37.26	30.40	0.47	38.32	29.87	0.48	39.31	29.76	0.54
	$\alpha_\parallel = 42.58, \alpha_\perp = 28.13$			$\alpha_\parallel = 44.60, \alpha_\perp = 27.12$			$\alpha_\parallel = 44.55, \alpha_\perp = 27.15$		

*Polarizabilities in units of 10^{-24} cm^3

TABLE IV*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ Å and order parameter S of Heptylcyanobiphenyl

	Neugebauer's Method			Vuks' Method			Saupe and Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
29	43.97	33.29	0.64	45.73	32.57	0.65	46.45	32.76	0.62
32	43.68	33.51	0.61	45.38	32.83	0.62	45.85	33.05	0.58
35	43.52	33.64	0.59	45.19	33.01	0.60	45.59	33.23	0.56
38	43.02	33.96	0.54	44.83	33.59	0.55	45.17	33.77	0.52
	$\alpha_\parallel = 48.26, \alpha_\perp = 31.67$			$\alpha_\parallel = 50.74, \alpha_\perp = 30.43$			$\alpha_\parallel = 51.89, \alpha_\perp = 29.86$		

*Polarizabilities in units of 10^{-24} cm^3

TABLE V*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ Å and order parameter S of Mixture E_1

	Neugebauer's Method			Vuks' Method			Saupe and Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
28	41.73	31.20	0.69	43.42	30.36	0.68	41.09	31.39	0.64
30	41.60	31.29	0.68	43.24	30.47	0.67	40.84	31.51	0.62
32	41.20	31.46	0.64	42.75	30.70	0.63	40.51	31.67	0.58
34	40.85	31.58	0.61	42.27	30.86	0.60	40.48	31.69	0.57
	$\alpha_\parallel = 44.75, \alpha_\perp = 29.69$			$\alpha_\parallel = 47.38, \alpha_\perp = 28.22$			$\alpha_\parallel = 44.73, \alpha_\perp = 29.55$		

*Polarizabilities in units of 10^{-24} cm^3

TABLE VI*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ Å and order parameter S of Pentyloxy cyanobiphenyl

	Neugebauer's Method			Vuks' Method			Saupe Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
49	40.79	30.34	0.66	42.43	29.52	0.67	43.03	29.76	0.67
52	40.67	30.48	0.64	42.26	29.68	0.65	42.62	29.97	0.64
55	40.49	30.61	0.63	42.04	29.83	0.63	42.33	30.11	0.62
58	40.28	30.72	0.60	41.76	29.98	0.61	42.13	30.22	0.60
61	39.79	31.04	0.55	41.14	30.36	0.56	41.41	30.57	0.55
65	39.23	31.45	0.49	40.43	30.85	0.49	40.33	31.11	0.46
66	38.78	31.63	0.45	39.87	31.08	0.45	40.20	31.18	0.45
67	38.33	31.89	0.41	39.32	31.39	0.41	39.65	31.45	0.41
68	36.98	32.62	0.27	37.50	32.35	0.26	38.25	32.15	0.31
	$\alpha_{\parallel} = 44.64$, $\alpha_{\perp} = 28.96$			$\alpha_{\parallel} = 47.03$, $\alpha_{\perp} = 27.76$			$\alpha_{\parallel} = 47.32$, $\alpha_{\perp} = 27.61$		

*Polarizabilities in units of 10^{-24} cm^3

TABLE VII*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ Å and order parameter S of Heptyloxycyanobiphenyl

	Neugebauer's Method			Vuks' Method			Saupe and Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
55	45.20	33.99	0.63	46.92	33.14	0.67	47.56	33.38	0.66
56	45.15	34.05	0.62	46.83	33.21	0.66	47.42	33.45	0.65
58	45.04	34.17	0.61	46.69	33.34	0.64	47.05	33.62	0.63
60	44.87	34.27	0.59	46.47	33.47	0.63	46.86	33.73	0.62
63	44.75	34.41	0.58	46.31	33.63	0.61	46.45	33.93	0.59
67	44.43	34.64	0.55	45.90	33.91	0.58	45.86	34.22	0.54
70.5	43.98	34.93	0.51	45.33	34.25	0.53	45.19	34.56	0.50
72	43.25	35.28	0.45	44.42	34.69	0.47	44.63	34.84	0.46
73	42.99	35.44	0.42	44.12	34.88	0.44	44.26	35.03	0.43
74	41.22	36.40	0.27	41.92	36.05	0.28	42.37	35.97	0.30
	$\alpha_{\parallel} = 49.89$, $\alpha_{\perp} = 32.21$			$\alpha_{\parallel} = 51.84$, $\alpha_{\perp} = 31.24$			$\alpha_{\parallel} = 52.23$, $\alpha_{\perp} = 31.04$		

*Polarizabilities in units of 10^{-24} cm^3

relation

$$\frac{n^2-1}{n^2+2}V_{\text{iso}} = \frac{1}{3}\left[\frac{n_e^2-1}{n_e^2+2-2a\left(n_e^2-1\right)} + \frac{2\left(n_0^2-1\right)}{n_0^2+2+a\left(n_0^2-1\right)}\right]V_n \tag{9}$$

and α_e and α_0 were obtained from Eqs. (7) and (8).

TABLE VIII*

Values of the effective polarizabilities α_e, α_0 for $\lambda = 5890$ A and order parameter S of Octyloxycyanobiphenyl

	Neugebauer's Method			Vuks' Method			Saupe and Maier's Method		
Temp° C	α_e	α_0	S	α_e	α_0	S	α_e	α_0	S
69	46.20	35.92	0.45	47.71	35.17	0.48	49.33	35.06	0.50
71	45.96	36.10	0.43	47.40	35.38	0.46	48.91	35.27	0.48
73	45.73	36.31	0.41	47.12	35.62	0.44	48.33	35.56	0.45
75	45.15	36.70	0.37	46.38	36.08	0.40	47.48	36.00	0.40
77	44.04	37.36	0.29	45.01	36.87	0.31	46.15	36.65	0.33
78	43.20	37.81	0.23	43.98	37.42	0.25	45.36	37.04	0.29
79	42.23	38.36	0.16	42.80	38.07	0.18	44.38	37.53	0.24
80	41.17	38.97	0.09	41.42	38.82	0.10	43.41	38.02	0.19
	$\alpha_\parallel = 55.05, \alpha_\perp = 32.20$			$\alpha_\parallel = 56.90, \alpha_\perp = 31.27$			$\alpha_\parallel = 58.62, \alpha_\perp = 30.41$		

*Polarizabilities in units of 10^{-24} cm^3

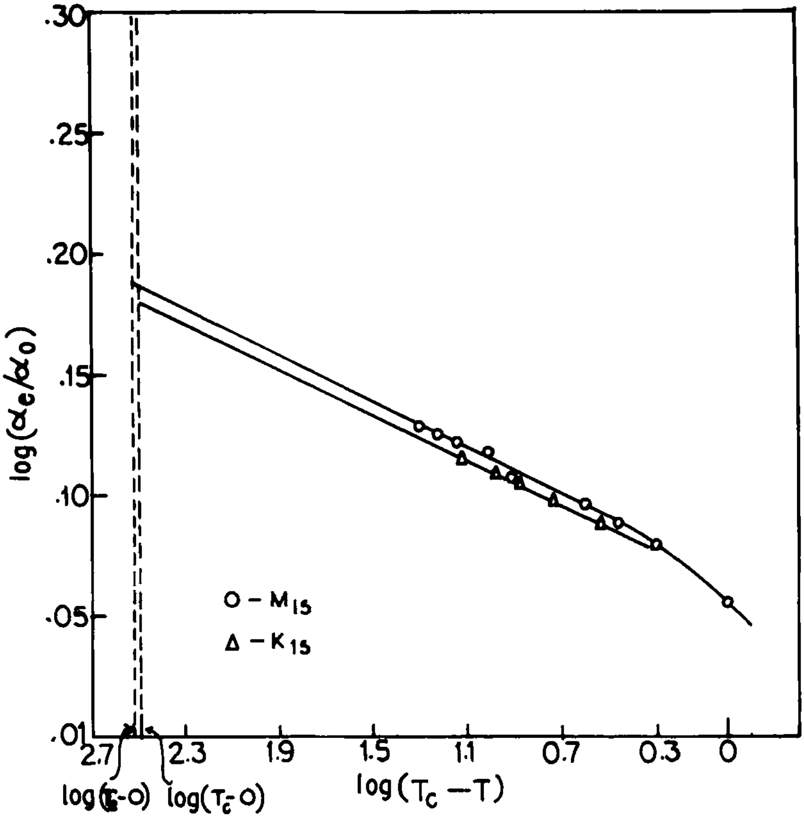

FIGURE 3 Plots of $\log(\alpha_e/\alpha_0)$ vs $\log(T_c - T)$ for M_{15} and K_{15}

The values of the polarizabilities α_e and α_0 obtained from the three methods are given in Tables (III–VIII). The principal polarizabilities $\alpha_{\parallel}$ and $\alpha_{\perp}$, parallel and perpendicular to the long axis of the molecules being not available, the method due to Haller *et al.*[4] was adopted. Graphs (Figure 3) were plotted with $\log \alpha_e/\alpha_0$ vs $\log(T_C - T)$, where T_C is the nematic–isotropic temperature. The plots which were straight lines at lower temperatures intersect the $\log \alpha_e/\alpha_0$ axis at 0 °K, assumed to correspond to the $\alpha_{\parallel}/\alpha_{\perp}$ in the crystalline state. From equation (4) the values of $\alpha_{\parallel}$ and $\alpha_{\perp}$ are determined and included in Tables (III–VIII). The order parameter, S, was then calculated from the relation $S = (\alpha_e - \alpha_0)/(\alpha_{\parallel} - \alpha_{\perp})$. The values of S obtained from the different methods are given in Tables (III–VIII). The plots of S vs T/T_C are shown in Figures 4a and 4b.

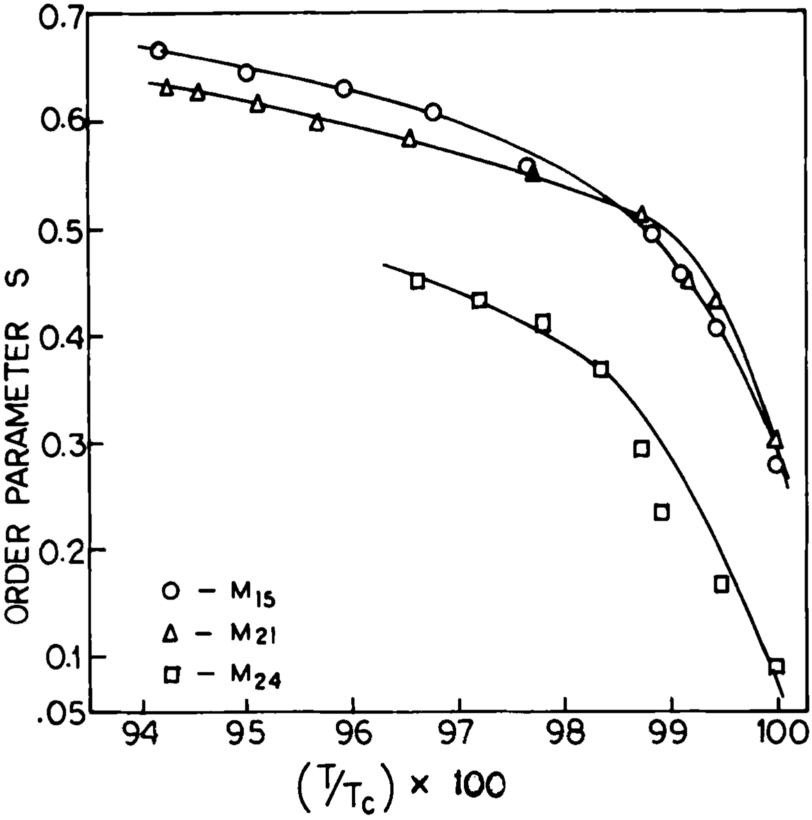

FIGURE 4(a) Plots of Order Parameter S vs T/T_c for M_{15}, M_{21}, and M_{24}

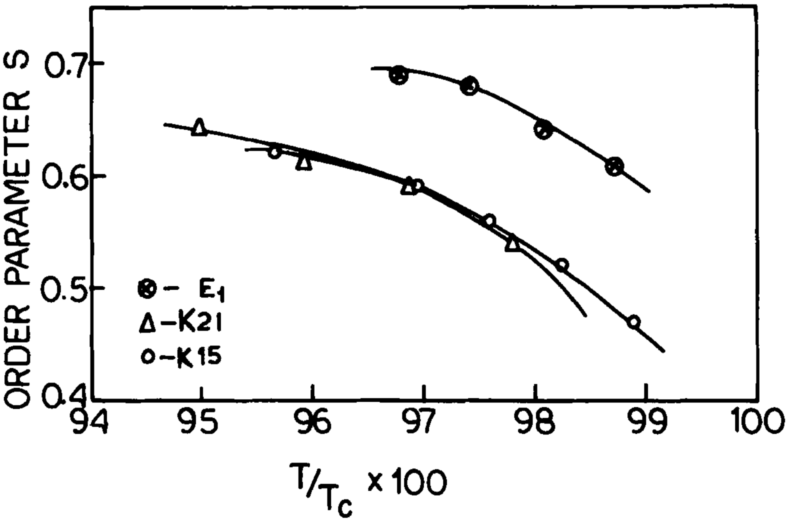

FIGURE 4(b) Plots of Order Parameter S vs T/T_c for E_1, K_{21}, K_{15}

DISCUSSION

It can be seen from Tables (III–VIII) that the order parameter S obtained from the three methods namely—Neugebauer[2], Vuks[3] and Saupe and Maier[1] are almost the same, though the anisotropy of the effective polarizabilities ($\Delta\alpha = \alpha_e - \alpha_0$) in the nematic phase obtained from the three methods are appreciably different. (For example, in pentyloxy cyanobiphenyl in the nematic state at 49 °C the values are 10.45, 12.91 and 14.27 × 10^{-24} cm^3 from Neugebauer, Vuks and Saupe and Maier method respectively.) The reason for similar values of the order parameter from the three methods is that the anisotropy of the principal polarizabilities ($\alpha_{\parallel} - \alpha_{\perp}$) increases in the same proportion, when one uses Vuks' or Saupe and Maier's method. It was shown by Subramhanyam *et al.*,[5] that the polarizability values obtained from Neugebauer's relations agree well with the calculated values of isotropic polarizabilities calculated from bond polarizability data. Moreover, the assumptions of anisotropic internal field constants in liquid crystals, by Neugebauer are logical. The internal field constants γ_e and γ_0 along and perpendicular to long molecular axis are expected to be different: the former should be smaller than the latter. The values γ_e and γ_0 calculated from the Eqs. (1) and (2) given in Tables IX and X show that γ_e is always less than γ_0. Since the effective length of the molecules and their density variation in the nematic phase of the liquid crystals are comparable (neglecting the alkoxy group) the internal field constants are also expected to be comparable as can be seen from Tables IX and X.

It was pointed out by Haller *et al.*,[4] and Subramhanyam *et al.*,[5] that the value of γ_e shows a maximum at a temperature about 3° below the nematic-isotropic transition. But in the present case as the Table IX shows that γ_e values increase monotonically with increase of temperature up to the nematic-isotropic transition.

Vuks[3] assumed an isotropic internal field in liquid crystalline media, but it is difficult to visualise how in a medium of anisotropic molecular distribution, as in liquid crystals, the local field would be isotropic.

Saupe and Maier[1] assumed an anisotropic molecular distribution similar to Neugebauer but used a single internal field parameter '*a*'. The idea is similar to the isotropic field model of Vuks. The internal

TABLE IX

Values of the internal field constants γ_e, γ_0 and '*a*'

Sample	$(T_c - T)$°C	γ_e	γ_0	'*a*'	$\left(1 - \frac{3\gamma_e}{4\pi}\right)$
Pentyloxy cyanobiphenyl	1	3.96	4.30	0.072	0.055
	2	3.91	4.33	0.078	0.067
	3	3.89	4.34	0.084	0.071
	4	3.85	4.36	0.076	0.080
Heptyloxycyanobiphenyl	1	3.99	4.28	0.061	0.048
	2	3.89	4.34	0.074	0.071
	3	3.88	4.35	0.079	0.074
	4.5	3.84	4.36	0.077	0.083
Octyloxycyanobiphenyl	1	4.09	4.23	0.089	0.020
	2	4.04	4.26	0.088	0.036
	3	4.00	4.29	0.092	0.045
	4	3.94	4.31	0.094	0.060

TABLE X

Values of the internal field constants γ_e, γ_0 and '*a*'

Sample	$(T_c - T)$°C	γ_e	γ_0	'*a*'	$\left(1 - \frac{3\gamma_e}{4\pi}\right)$
Pentylcyanobiphenyl	3.5	3.88	4.35	0.10	0.07
Heptylcyanobiphenyl	7	3.96	4.61	0.03	0.11
Mixture E_1	4	3.73	4.38	0.03	0.05

field parameter was calculated using Saupe and Maier's relations. The internal field parameter '*a*' can be roughly related to the γ_e values of Neugebauer by an expression '*a*' = $(1 - 3\gamma_e/4\pi)$ as shown in Tables IX & X. In an anisotropic molecular distribution as in liquid crystals the internal field parameter cannot be the same along the long molecular axis and perpendicular to it as proposed by Saupe and Maier.

Though the determination of the order parameter of liquid crystals by Neugebauer's,[2] Vuks[3] and Saupe and Maier's[1] methods give nearly the same value, Neugebauer's method is to be preferred as it is theoretically sound in having internal field constants different along and perpendicular to molecular axis. Also, the principal polarizabilities obtained by it agree to the values of isotropic polarizabilities obtained from bond polarizability calculations.

It can be seen from Tables (VI–VIII) that the order parameters of pentyloxy cyanobiphenyl and heptyloxy cyanobiphenyl at the lowest temperature in the nematic phase are 0.66 and 0.63 respectively, but in the octyloxy cyanobiphenyl it is only 0.45 which is much lower than the former two liquid crystals. Though the structures of the three liquid crystals are similar, the increase in the alkyl chain length seems to reduce the order parameter rapidly.

The order parameter for pentyl and heptyl cyanobiphenyl at the lowest temperature in the nematic state are .62 and .64 respectively Tables (III–IV), but the order parameter of the mixture E_1 composed of pentyl and heptyl cyanobiphenyl (59 : 41) is about .70 at 28°C which is larger than the S-values of each of the components.

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