



Trade Science Inc.

April 2008

Volume 4 Issue 1

# Macromolecules

*An Indian Journal*

*Full Paper*

MMALJ, 4(1), 2008 [65-72]

## Optical birefringence and order parameter of pure cholesteric liquid crystals and their mixture

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Index classification (PACS): 61.30.-v, 42.70.Df.

Received: 14<sup>th</sup> February, 2008 ; Accepted: 19<sup>th</sup> February, 2008

### ABSTRACT

The refractive indices and densities of liquid crystals Cholesteryl Pelargonate and Cholesteryl Butyrate and their mixtures of different ratio of 25:75, 50:50 and 75:25 (by weight) have been reported as a function of temperature. The effective polarizabilities  $\alpha_e$  and  $\alpha_o$  in the cholesteric phase has been calculated by using the anisotropic internal field model of Neugebauer, Saupe and Maier and isotropic field model of Vuks, from  $n_e$  and  $n_o$  values. The order parameters as evaluated by using different local field models for both the pure samples and their mixture of different concentration have been compared. The values of principal polarizabilities are found to be slightly different although the order parameters  $S$  evaluated from above models are in good agreement. These order parameters have also been compared with that calculated by two other direct methods. All these results seem to be in good agreement. The variation of different parameters with the percentage concentration of sample has been reported and discussed giving probable reason.

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### KEYWORDS

Liquid crystals;  
Refractive index;  
Birefringence;  
Order parameter.

### INTRODUCTION

There are many liquid crystalline materials available for the device application. However the difficulty arises in identifying single liquid crystal that fulfils all the required conditions for a particular application. This has generated the interest of researchers to undertake the investigations in the area of mixing of different kind of liquid crystals. Liquid crystal mixtures are important in many other ways<sup>[1-3]</sup>. The complete miscibility study was the criterion originally used for the identification of dif-

ferent liquid crystalline phases. The liquid crystal mixtures are mainly used to widen the temperature range of phases, to investigate interaction between the molecules and subsequent change in order parameters in different phases, to investigate pre-transitional behavior and to identify unknown substances by mixing with substances with well known phase behavior. Hence a lot of work has been done on mixing by many other investigators<sup>[4-8]</sup>.

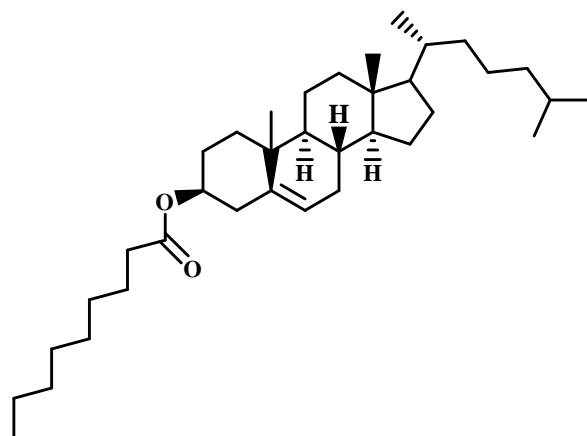
The importance for liquid crystals of their application in display devices and other scientific equipments

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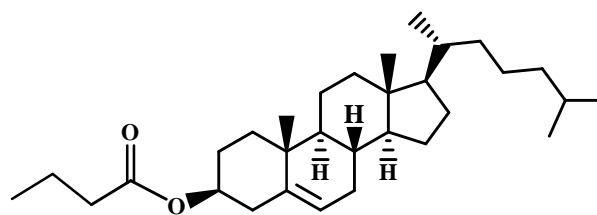
is now widely known<sup>[9]</sup>. The order parameter of a liquid crystal is an important parameter, which governs all its attributes and therefore its knowledge is essential for an effective use in material application in different areas. Various methods to calculate the order parameter are now being used. All anisotropic properties such as refractive indices, magnetic and electric susceptibilities and dielectric parameters etc. are related to the order parameter in some way or the other<sup>[10]</sup>. By studying the birefringence we can estimate the orientational ordering in liquid crystals at different temperatures. In the present paper we report principal polarizability and order parameter of two pure cholesteric liquid crystals and their three homogenous mixtures of different concentration. The effect of mixing has also been discussed on these properties.

Here three methods namely Neugebauer's anisotropic internal field model<sup>[11]</sup>, Vuks' isotropic internal field model<sup>[12]</sup> and Saupe and Maier's anisotropic model<sup>[13]</sup> with a single internal field parameter, have been used to calculate the effective polarizabilities and also the order parameter. Two other direct methods, as modified by Vuks and direct extrapolation method have also been used for evaluating the order parameter.

Experimental: Initial work on cholesterylakanoates are studied and reported in literature more than any other cholesteric materials. Two of the cholesterylakanoates chosen for our present study, have earlier been investigated by other investigators in pure form and related data are available. The Chemical structure of both the samples are presented as below-



Cholesteryl pelargonate



Cholesteryl butyrate

### Cholesteryl pelargonate

The cholesteric-isotropic transition temperature which are obtained by WHT (White light transmittance) technique with crossed polarizing microscope of two liquid crystals and their mixtures of different ratios are given below-

Cholesteryl Pelargonate (Ch.P.)      Ch  $\xleftarrow{92^{\circ}\text{C}}$  I

Ch. P.:Ch.B. :: 3:1                              Ch  $\xleftarrow{88^{\circ}\text{C}}$  I

Ch. P.:Ch.B. :: 1:1                              Ch  $\xleftarrow{89^{\circ}\text{C}}$  I

Ch. P.:Ch.B. :: 1:3                              Ch  $\xleftarrow{94^{\circ}\text{C}}$  I

Cholesteryl butyrate (Ch. B.)              Ch  $\xleftarrow{106^{\circ}\text{C}}$  I

The extraordinary refractive index  $n_e$  and ordinary refractive index  $n_o$  in the cholesteric phase and refractive index  $n$  in the isotropic phase at different temperature have been measured by using the Abbe refractometer. The surface of glass prism in refractometer has been rubbed unidirectionally several times using soft cotton piece. The glass prism is first heated above isotropic temperature and then liquid crystal sample is placed on the lower prism surface. This procedure helps align the molecule of the liquid crystals along the prism face. The alignment was complete when the upper prism was clamped in the place and allows the system to cool slowly.

An analyzer has been introduced on the eyepiece of Abbe's refractometer, which blocks the ordinary or extraordinary ray at a time by rotating its analyzing axis. This clears the contrast of the boundary line at view of refractometer and makes it simple to record the observation. The constant temperature of the sample in the refractometer has been maintained by a Julabo F-25 refrigerated circulator.

The densities of the liquid crystal samples in cholesteric and isotropic phase were determined by introducing a weighted sample inside a capillary tube which

is placed in side the wall of a double walled glass jacket. The glass jacket having circulating oil by Julabo F-25 refrigerated circulator is maintained at constant temperature with an accuracy of  $\pm 0.1^\circ\text{C}$ . Changing meniscus level of the sample due to the temperature variation in the capillary has been recorded by a traveling microscope of least count 0.001 cm. The error in estimation of density was less than 1%.

## Theory

The effective polarizabilities  $\alpha_e$  and  $\alpha_o$  have been calculated by using the following three methods.

Neugebauer's method: Neugebauer's method of anisotropic internal field is given by

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

$$n_o^2 - 1 = 4\pi N\alpha_o(1 - N\alpha_o\gamma_o)^{-1} \quad (2)$$

Where N is the number of molecule per c.c.,  $n_e$  and  $n_o$  are refractive indices for ordinary and extraordinary rays and  $\gamma_e$  and  $\gamma_o$  are the internal field constants. The equation for calculating the polarizabilities  $\alpha_e$  and  $\alpha_o$  obtained from equation (1) and (2) and applying the condition  $\gamma_e + 2\gamma_o = 4\pi$ , gives

$$\frac{1}{\alpha_e} + \frac{2}{\alpha_o} = \frac{4\pi N}{3} \left[ \frac{n_e^2 + 2}{n_e^2 - 1} + \frac{2(n_o^2 + 2)}{n_o^2 - 1} \right] \quad (3)$$

Further, for isotropic phase

$$\gamma_e = 2\gamma_o = 4\pi/3, n_e = n_o = n, \alpha_e = \alpha_o = \bar{\alpha}$$

$$\alpha_e + 2\alpha_o = 3\bar{\alpha} = \frac{9}{4\pi N} \left[ \frac{n^2 - 1}{n^2 + 2} \right] \quad (4)$$

## Vuks' method

Vuks' method of isotropic local field, as modified from the Lorentz-Lorentz equation is as under-

$$\frac{n_e^2 - 1}{n^2 + 2} = \frac{4\pi N\alpha_e}{3} \quad (5)$$

$$\frac{n_o^2 - 1}{n^2 + 2} = \frac{4\pi N\alpha_o}{3} \quad (6)$$

The internal field constant 'a' was obtained by the relation-

$$\frac{n^2 - 1}{n^2 + 2} V_{\text{Iso}} = \frac{1}{3} \left[ \frac{n_e^2 - 1}{n_e^2 + 2 - 2a(n_e^2 - 1)} + \frac{2(n_o^2 - 1)}{n_o^2 + 2 + a(n_o^2 - 1)} \right] V_{\text{Ch}} \quad (7)$$

$\alpha_e$  and  $\alpha_o$  have been obtained by solving the equations (3) and (4); (5) and (6) and lastly (7) and (8)

presented above from three different approaches.

Since the order parameter is given by  $S = (\alpha_e - \alpha_o) / (\alpha_e + \alpha_o)$ , where we have only  $\alpha_e$  and  $\alpha_o$  values but not the principal polarizabilities  $\alpha_{\parallel}$  and  $\alpha_{\perp}$ , the parallel and perpendicular components respectively to the long axis of the molecule, we use an extrapolation method for calculating the  $\alpha_{\parallel}$ - $\alpha_{\perp}$  values. The extrapolation has been done for the value of  $(\alpha_e - \alpha_o)$  on the plot  $(\alpha_e - \alpha_o)$  against  $\ln(1 - T/T_c)$  for  $T=0$ , i.e. for absolute temperature.

## Direct extrapolation method

The other two direct methods for calculating the order parameter is based on the variation of ordinary and extraordinary refractive indices which do not need the molecular weight and density values. The relationship between refractive index parallel and perpendicular to the direction of molecular axis and order parameter can be obtain by modifying the appropriate equation for  $n_{\parallel}$  and  $n_{\perp}$  as follows.

$$n_{\parallel} = \bar{n} + \frac{2}{3} S \cdot \Delta n \quad (10)$$

$$n_{\perp} = \bar{n} - \frac{1}{3} S \cdot \Delta n \quad (11)$$

Where,  $\bar{n}$  is the average refractive index and  $\Delta n$  is the birefringence corresponding to complete alignment for the material. From equations (10) and (11), we get order parameter,

$$S = \frac{n_{\parallel} - n_{\perp}}{\Delta n} = \frac{\delta n}{\Delta n}$$

The value of order parameter equal to 1 represents a complete order. At absolute zero,  $\delta n = \Delta n$ . So the order parameter may be obtained by extrapolating  $\delta n$  for  $T=0\text{K}$ . This extrapolation is done on the linear portion of birefringence ( $\delta n$ ) curve plotted against  $\ln(1 - T/T_c)$  for  $T = 0\text{K}$ .

In this second direct method, in order to find out the order parameter (S), the refractive indices  $n_e$  and  $n_o$  have been analyzed by following method of Haller<sup>[14]</sup> and Horn<sup>[15]</sup>. This method uses Vuks relation:

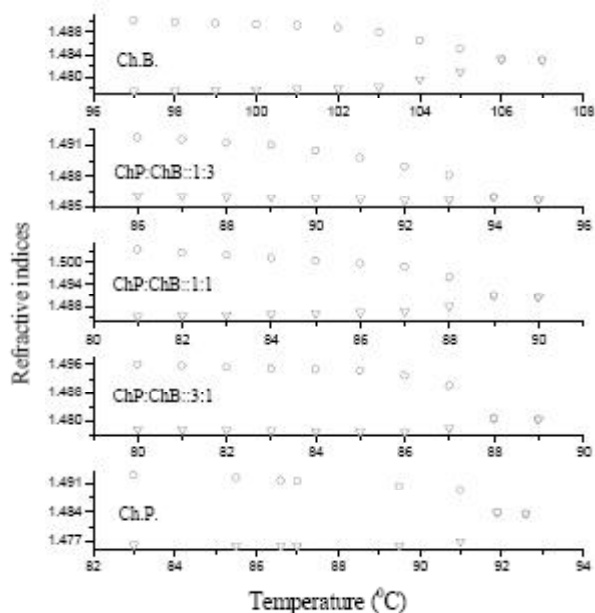
$$S \left( \frac{\delta\alpha}{\alpha} \right) = \frac{3(n_e^2 - n_o^2)}{(n_e^2 + 2n_o^2 - 3)} \quad (12)$$

Where  $\delta\alpha$  is the anisotropy of polarizability and  $\alpha$  the molecu-

lar polarizability. To determine S we plot  $\left\{ \frac{3(n_e^2 - n_o^2)}{(n_e^2 + 2n_o^2 - 3)} \right\}$

against  $\ln \left( 1 - \frac{T}{T_c} \right)$  and the straight line so obtained is extrapo-

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**Figure 1:** Plot of extraordinary (o) and ordinary (∇) refractive indices with the variation of temperature for cholesteryl butyrate, cholesteryl pelargonate and their mixture in different ratio

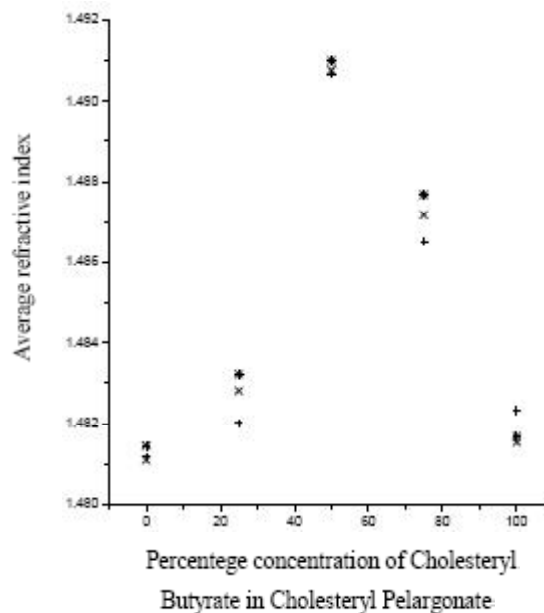
**TABLE 1:** Effective polarizabilities  $\alpha_c$  and  $\alpha_o$  in the unit of  $10^{24} \text{ cm}^3$  for cholesteryl pelargonate (ChP)

Temperature (°C)	Neugebauer		Vuks		Saupe and Maier	
	$\alpha_o$	$\alpha_c$	$\alpha_o$	$\alpha_c$	$\alpha_o$	$\alpha_c$
83	62.9441	65.2291	63.5550	66.2923	62.1815	64.0311
85.5	63.1426	65.3687	63.7382	66.4037	62.3295	64.1298
86.6	63.2347	65.4043	63.8155	66.4123	62.3879	64.1418
87	63.2896	65.4138	63.8585	66.4002	62.4207	64.1374
89.5	63.5869	65.5175	64.1049	66.4121	62.6141	64.1721
91	63.9139	65.6064	64.3689	66.3890	62.8278	64.1916
	$(\alpha_o \alpha_c) = 33.344$		$(\alpha_o \alpha_c) = 40.107$		$(\alpha_o \alpha_c) = 28.764$	

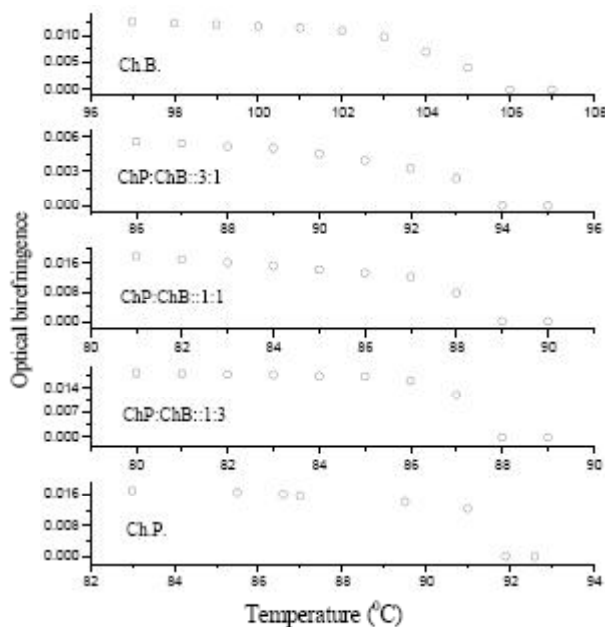
lated to  $T=0\text{K}$ . The intercept at  $T=0\text{K}$ , where the complete order occurs give the value of  $(\delta\alpha/\alpha)$ . Inserting the value of  $(\delta\alpha/\alpha)$  in equation (5) gives the value of order parameter for different temperatures.

## RESULTS

Figure 1 shows the variation of ordinary and extraordinary refractive indices with the temperature for Cholesteryl Pelargonate, Cholesteryl Butyrate and their mixtures of ratio 3:1, 1:1 and 3:1. Figure 2 represents the Variation of average refractive index with percentage concentration of Cholesteryl Butyrate in Cholesteryl Pelargonate. Variation of birefringence ( $\delta n = n_e - n_o$ ) with temperature for all the samples are given in figure 3. Figures 4a to 4e show the change in the values of order parameter evaluated by the different methods with tem-



**Figure 2:** Variation of optical anisotropy with percentage concentration of cholesteryl butyrate in cholesteryl pelargonate at  $T_c-1(+)$ ,  $T_c-3(\Delta)$  and  $T_c-5(*)$



**Figure 3:** Variation of optical birefringence ( $\delta n$ ) with temperature in °C for cholesteryl pelargonate, cholesteryl butyrate and their mixtures in different ratio

perature for pure liquid crystals and their mixtures. Variation of average polarizability and order parameter with percentage concentration of cholesteryl butyrate in cholesteryl pelargonate is shown in figure 5. TABLES

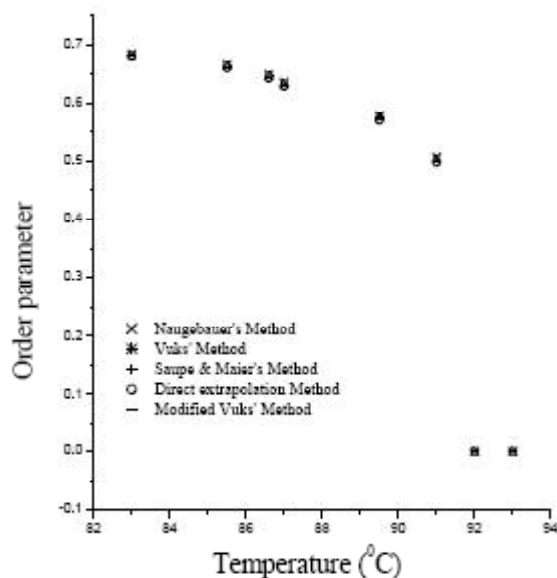


Figure 4a : Plot of order parameter with temperature in °C for cholesteryl pelargonate

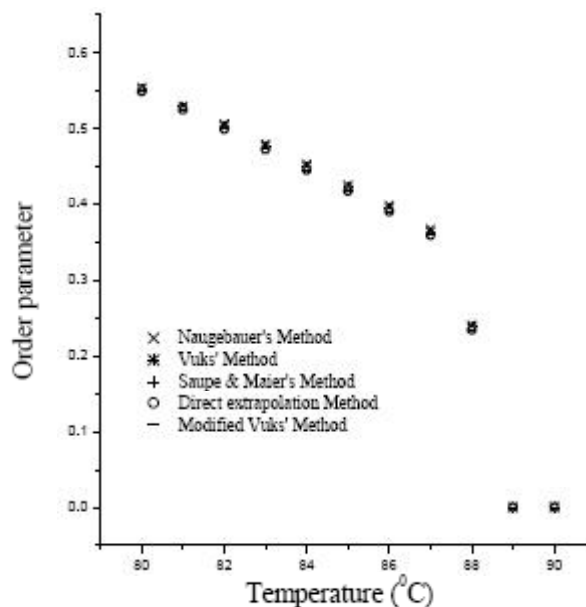


Figure 4c : Plot of order parameter with temperature in °C for and cholesteryl butyrate in ratio 1:1

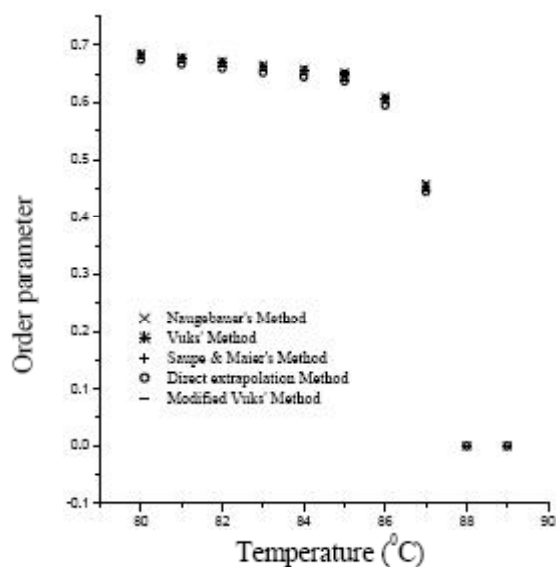


Figure 4b : Plot of order parameter with temperature in °C for mixture of cholesteryl pelargonate and cholesteryl butyrate in ratio 3:1

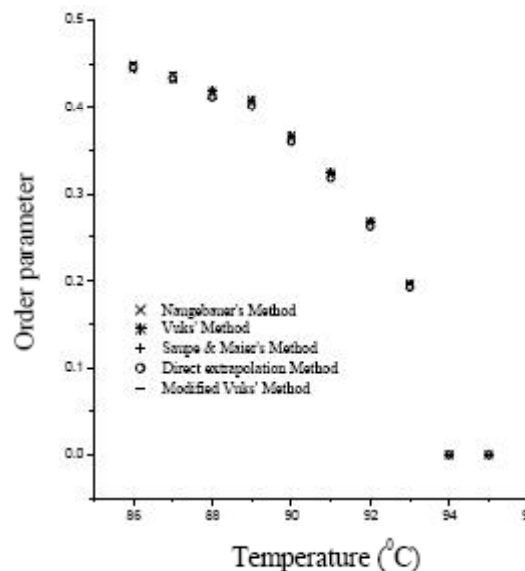


Figure 4d : Plot of order parameter with temperature in °C for mixture of cholesteryl Pelargonate and cholesteryl butyrate in ratio 1:3

1-4 represent the effective polarizabilities  $\alpha_c$  and  $\alpha_o$  calculated by Neugebauer's, Vuks' and Saupe and Maier's method for all the systems.

## DISCUSSION

As can be seen from figure 1, in the case of Cholesteryl Pelargonate ordinary refractive index decreases smoothly with the temperature from its value of 1.492 at 83°C and it shows a sudden drop after 91°C

and thereafter the refractive index becomes 1.484 at 92°C indicating cholesteric to isotropic transition at this temperature. On further increase in temperature refractive index remain almost constant like any other normal organic liquid<sup>[16]</sup>. At the same time the extraordinary refractive index increases with increase in temperature and at Ch-I transition point, it is equal to the average refractive index.

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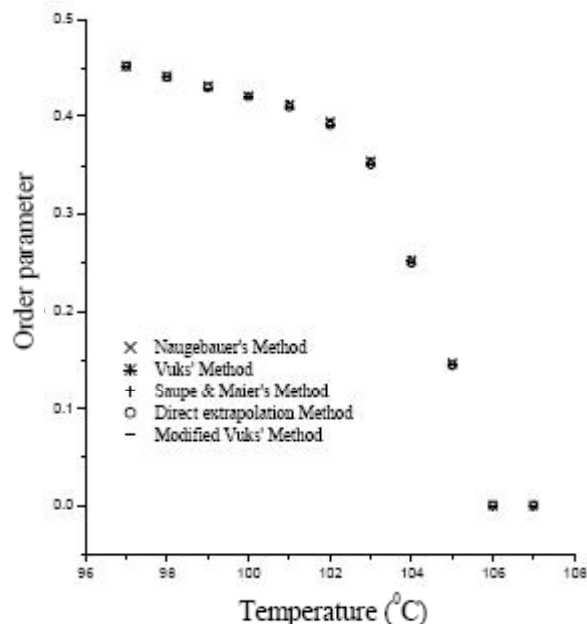


Figure 4e : Plot of order parameter with temperature in °C for cholesteryl butyrate

Similar behaviour has been shown by Cholesteryl Butyrate with slightly different value of refractive index and phase transition temperature point of 106°C. Three mixtures of various concentrations also show same type of behaviour with one difference where the value of transition temperature shifts towards the higher side with increase of concentration of sample with higher transition temperature.

Figure 2 shows the plot of average refractive indices vs percentage concentration of cholesteryl butyrate in Cholesteryl Pelargonate for three different temperatures. With increase in concentration of cholesteryl butyrate in cholesteryl pelargonate the refractive index increases first from 1.49, reaches a peak and then falls to a value of 1.481 at 100% butyrate in mixture. This nature remains same for all the three temperatures shown here by representative graphs at temperature ( $T_c - 1$ ), ( $T_c - 3$ ) and ( $T_c - 5$ ) where  $T_c$  is the Ch-I transition temperature for corresponding samples. The reason for this anomalous increase is not very clear. It may be possible that the molecular arrangement for 50% concentration in the mixture is such that it gives rise a maximum value of refractive index in comparison to other system. Similar behaviour has been reported by Sandhya et al.<sup>[17]</sup> for the photocurrent anisotropy at particular concentration of compounds 2-(4'-octylphenyl)-6-

TABLE 2: Effective polarizabilities  $\alpha_e$  and  $\alpha_o$  in the unit of  $10^{-24}\text{cm}^3$  for cholesteryl butyrate (ChB)

Temperature (°C)	Neugebauer		Vuks		Saupe and Maier	
	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$
99	52.6254	54.0175	52.9995	54.6613	53.2609	54.4505
100	52.7104	54.0709	53.0762	54.7000	53.3225	54.4844
101	52.7956	54.1245	53.1530	54.7387	53.3842	54.5185
102	52.8811	54.1783	53.2300	54.7776	53.4461	54.5527
103	52.9667	54.2321	53.3072	54.8165	53.5082	54.5870
104	53.0610	54.2723	53.3871	54.8313	53.5705	54.6025
105	53.2001	54.2901	53.4939	54.7924	53.6521	54.5799
	$(\alpha_o\alpha_e) = 30.691$		$(\alpha_o\alpha_e) = 6.701$		$(\alpha_o\alpha_e) = 26.274$	

TABLE 3 : Effective polarizabilities  $\alpha_e$  and  $\alpha_o$  in the unit of  $10^{-24}\text{cm}^3$  for mixture ChP:ChB:: 3:1

Temperature (°C)	Neugebauer		Vuks		Saupe and Maier	
	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$
80	60.8263	63.1678	61.4509	64.2602	60.4459	62.3605
81	60.8948	63.2139	61.5136	64.2955	60.4952	62.3909
82	60.9638	63.2606	61.5769	64.3313	60.5451	62.4218
83	61.0336	63.3078	61.6408	64.3676	60.5955	62.4533
84	61.1039	63.3556	61.7053	64.4045	60.6466	62.4853
85	61.1749	63.4039	61.7704	64.4418	60.6982	62.5178
86	61.2912	63.3786	61.8497	64.3488	60.7500	62.4528
87	61.6859	63.2506	62.1065	63.9740	60.9308	62.2043
	$(\alpha_o\alpha_e) = 34.169$		$(\alpha_o\alpha_e) = 41.175$		$(\alpha_o\alpha_e) = 28.766$	

TABLE 4 : Effective polarizabilities  $\alpha_e$  and  $\alpha_o$  in the unit of  $10^{-24}\text{cm}^3$  for mixture ChP: ChB:: 1:1

Temperature (°C)	Neugebauer		Vuks		Saupe and Maier	
	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$
81	56.8794	59.0003	57.4432	59.9935	57.1399	58.9091
82	57.0063	59.0267	57.5440	59.9718	57.2174	58.9015
83	57.1344	59.0517	57.6452	59.9475	57.2951	58.8919
84	57.2638	59.0753	57.7469	59.9206	57.3729	58.8804
85	57.3946	59.0974	57.8492	59.8910	57.4509	58.8669
86	57.5266	59.1181	57.9520	59.8588	57.5291	58.8514
87	57.6725	59.1396	58.0651	59.8216	57.6160	58.8339
88	58.0778	59.0388	58.3361	59.4831	57.8146	58.6104
	$(\alpha_o\alpha_e) = 39.987$		$(\alpha_o\alpha_e) = 48.312$		$(\alpha_o\alpha_e) = 33.531$	

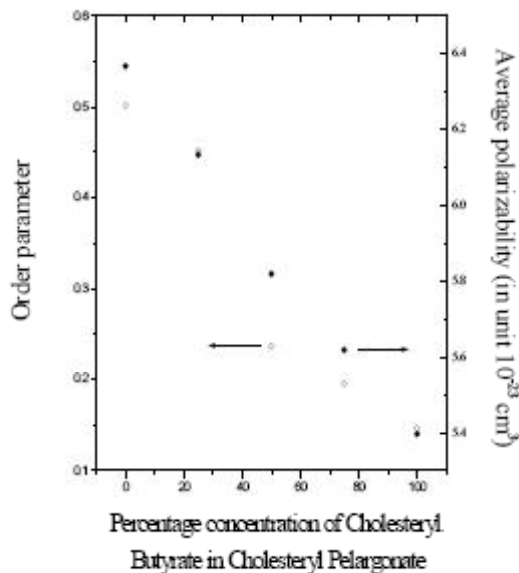
dodecyloxynaphthalene and 2-(4'-pentylphenyl)-6-ethynaphthalene.

In figure 3 optical anisotropy has been plotted with temperature for the pure samples and their mixtures. For Cholesteryl Pelargonate the value of optical anisotropy decreases gradually with temperature but sharply near the cholesteric-isotropic transition and become zero after transition temperature. Cholesteryl Butyrate and other mixtures show similar variation giving different transition temperature as expected. Similar behavior has also been reported for other liquid crystal samples and mixture by several other workers<sup>[18-21]</sup>.

TABLES 1 to 5 represent the values of effective polarizabilities ( $\alpha_e$ ,  $\alpha_o$ ) obtained by Neugebauer's, Vuks' and Saupe & Maier's methods for liquid crystals in pure and mixture form. The values of principal

**TABLE 5 : Effective polarizabilities  $\alpha_e$  and  $\alpha_o$  in the unit of  $10^{-24} \text{ cm}^3$  for mixture ChP: ChB: 1:3**

Temperature (°C)	Neugebauer		Vuks		Saupe and Maier	
	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$	$\alpha_o$	$\alpha_e$
86	55.7096	56.3552	55.8976	56.6522	55.7174	56.2589
87	55.7785	56.4064	55.9618	56.6952	55.7680	56.2945
88	55.8524	56.4498	56.0138	56.7245	55.8187	56.3194
89	55.9203	56.5034	56.0778	56.7715	55.8696	56.3581
90	56.0057	56.5292	56.1472	56.7697	55.9206	56.3589
91	56.0853	56.5492	56.2108	56.7620	55.9658	56.3540
92	56.1687	56.5513	56.2723	56.7268	56.0066	56.3266
93	56.2770	56.5589	56.3535	56.6880	56.0646	56.3001
	$(\alpha_{  }\alpha_{\perp}) = 14.364$		$(\alpha_{  }\alpha_{\perp}) = 16.951$		$(\alpha_{  }\alpha_{\perp}) = 12.184$	

**Figure 5 : Variation of order parameter and average polarizability for mixture of cholesteryl butyrate in cholesteryl pelargonate at corresponding temperature ( $T_c-1$ )°C**

polarizability anisotropy are also given for each model. A Plot of order parameter with varying temperature for all the samples are given in figure 4a to 4e. It may be seen that for all the samples, the order parameter obtained by Vuks', modified Vuks' and direct extrapolation method remains almost same, probably because these are achieved from a similar approach. The anisotropy of the effective polarizability ( $\alpha = \alpha_e - \alpha_o$ ) obtained for Cholesteryl Pelargonate have been found to be 22.8, 27.3 and  $18.4 \times 10^{-24} \text{ cm}^3$  for temperatures ( $T_c-1$ ), ( $T_c-3$ ) and ( $T_c-5$ ), using Neugebauer's, Vuks' and Saupe and Maier's model respectively. The value of order parameters obtained from the three models have been found to be 0.6961, 0.6941 and 0.6939. Since the order parameter is given by  $S = (\alpha_e - \alpha_o) / (\alpha_{||}\alpha_{\perp})$  where anisotropy of principal polarizability  $\alpha_{||}\alpha_{\perp}$  is obtained from extrapolation of  $\alpha_e - \alpha_o$  to absolute zero, the

ratio  $(\alpha_e - \alpha_o) / (\alpha_{||}\alpha_{\perp})$  remains almost constant for a given temperature. This shows that the order parameter for a given temperature is independent of the model used to evaluate.

Corresponding variation in average polarizability and order parameter with percentage concentration of cholesteryl butyrate in cholesteryl pelargonate at temperature ( $T_c-1$ ) is represented in figure 5. Both the values that are average polarizability and order parameter have maximum value for Cholesteryl Pelargonate and minimum for cholesteryl butyrate varies almost linearly decreasing concentration of Cholesteryl Butyrate. This fact can be utilized to prepare mixture of suitable order parameter or average polarizability for systems having components which are member of same homologous series.

## CONCLUSION

It may be seen that that order parameter computed from optical anisotropy data using the isotropic field model (Vuks approach), the anisotropic field model (Neugebauer's approach), Saupe and Maier's method agrees well and the average deviation is found to be 0.8 % for both liquid crystals and their mixtures in all the phases. The agreement in the values of the order parameter calculated from different approaches may appear strange but it is due to the fact that the order parameter is the ratio of  $(\alpha_e - \alpha_o)$  to  $(\alpha_{||}\alpha_{\perp})$ . So, although  $\alpha_o$  and  $\alpha_e$  are sensitive to the nature of local field, this ratio is independent of the model. A slight variation in the solid phase is due to the fact that the anisotropy is considerable in this phase. It appears from the results that Neugebauer's approach is more realistic than Vuks approach as it includes the fact that local field is anisotropic in nature.

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