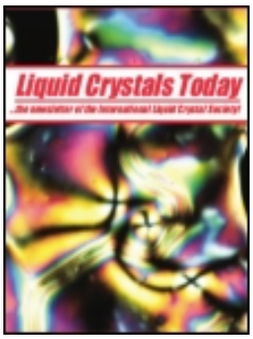

Liquid Crystals Today

ISSN: 1358-314X (Print) 1464-5181 (Online) Journal homepage: www.tandfonline.com/journals/tlcy20

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To cite this article: D. Madhavi Latha, V.G.K.M. Pisipati, P. Pardhasaradhi, P.V. Datta Prasad & D. Venkata Rao (2014) Estimation of order parameter from different models in symmetric dimeric liquid crystals, Liquid Crystals Today, 23:3, 54-60, DOI: 10.1080/1358314X.2014.917797

To link to this article: https://doi.org/10.1080/1358314X.2014.917797

Published online: 20 May 2014.

Liquid Crystals Today, 2014
Vol. 23, No. 3, 54–60, http://dx.doi.org/10.1080/1358314X.2014.917797

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Estimation of order parameter from different models in symmetric dimeric liquid crystals

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(*Received 17 March 2014; accepted 26 March 2014*)

The birefringence measurements with the temperature are carried out employing the wedge technique at the wavelength λ = 5893Å on symmetric dimeric liquid crystalline compounds of α,ω-bis(4-alkylanilinebenzylidene-4′-oxy) alkanes which are popularly known as m.OnO.m's with m = 3, 4 and 5 and spacer length n = 8, 9 and 10. The studied compounds are mono variant with long range of nematic phase. The birefringence data $\delta n = (n_e - n_o)$ along with the density results are employed to estimate the molecular polarisabilty anisotropy, $\delta\alpha = (\alpha_e - \alpha_o)$ assuming a particular local field (due to Vuks and Neugebauer) the nematic molecule experiences and the order parameter S is estimated from $\delta\alpha$ and $\Delta\alpha$, i.e., polarisabilty anisotropy which is estimated employing different methods. Further, S can also be obtained from Kuczynski model and also using the effective geometry parameter, α_g. Further, it is observed that from our analysis the S value obtained from Δn and α_g are identical. The results are analysed and compared with the data available in the literature.

Keywords: symmetric liquid crystal dimers; birefringence; orientational order parameter; molecular polarisabilities

1. Introduction

Liquid crystal dimers (DLCs) in which two mesogenic units are linked via a single flexible spacer which differ from conventional low molar mass mesogens attracted considerable research attention with recent studies including the role of the spacer in determining liquid crystal (LC) behaviour [1–6]. The DLCs may be broadly divided into two classes namely symmetric and non-symmetric. In a symmetric DLC, the two mesogenic units are identical whereas in a non-symmetric DLC they are different. These DLCs exhibit fascinating phase behaviour, quite different to that observed for conventional low molar mass LCs composed of molecules consisting of a single semi-rigid or mesogenic core attached to which are one or two terminal alkyl chains. Many workers have reported the synthesis of DLCs which include hydrogen-bonded dimers [7–9], their phase behaviour in carbonate-linked [10] and bent odd member dimers [11].

The orientational order parameter S is considered to be one of the most important material parameters of the nematic phase, which determines all of its anisotropic properties and the relations between macroscopic and microscopic properties. Different techniques are employed [4–7] for the determination of birefringence due to its importance. Recently, Alapati et al. [5] studied the order parameter variation with temperature in Schiff-based two symmetric DLCs employing the two field models viz., Vuks and Neugebauer.

This manuscript reports the estimation of order parameter and its variation with temperature in five Schiff base symmetric DLCs viz., α,ω-bis(4-alkylanilinebenzylidene-4′-oxy) alkanes which are popularly known as m.OnO.m's with m = 3, 4 and 5 and spacer length n = 8, 9 and 10 employing different methods.

2. Experimental

The refractive indices of a LC are measured with a wedge-shaped glass cell, similar to the one used to obtain birefringence by Haller et al. [12], using a 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 nematic 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.

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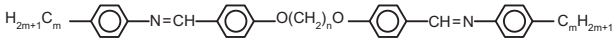
3. Results and discussion

The transition temperatures obtained from polarising microscope and DSC are in good agreement with the literature data [13,14] and the purity of the compounds is tested and obtained from the Perkin-Elmer DSC instrument [14]. The molecular structure of these symmetric DLC is given below.



3. O9O.3	m = 3	n = 9
3. O10O.3	m = 3	n = 10
4. O8O.4	m = 4	n = 8
4. O10O.4	m = 4	n = 10
5. O9O.5	m = 5	n = 9

The refractive index is measured for the five symmetric DLCs. It is observed that the value of refractive index (n_{iso}) is almost constant in the isotropic phase, and with the decrease of temperature from isotropic to nematic phase, at the IN phase transformation, the isotropic refractive index value splits into two, extraordinary refractive index ($n_e > n_{iso}$) and ordinary refractive index ($n_o < n_{iso}$), respectively. This splitting is clearly observed in the telescope of the modified spectrometer at the position of angle of minimum deviation at the IN transition. When the temperature is further decreased, the n_e increases while the n_o decreases slightly; with further decrease of temperature, in the deep nematic region, the refractive index values of both n_e and n_o saturate. The refractive indices variation with temperature is given in Figure 1 for the compound 5.O9O.5 as representative case.

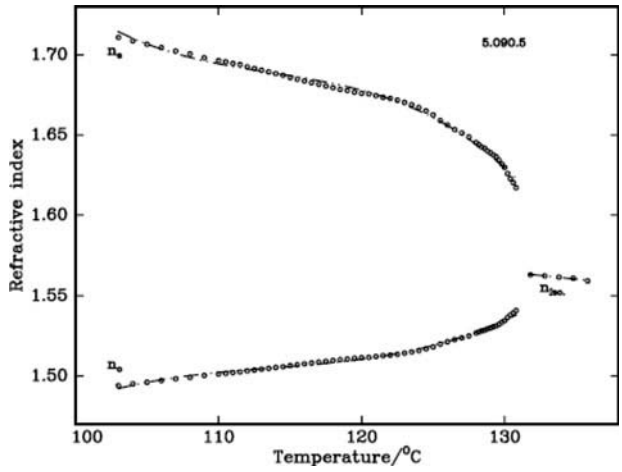


Figure 1. Variation of refractive indices with temperature for the compound 5.O9O.5.

3.1 *Estimation of order parameter*

The birefringence measurements using different techniques and the evaluation of molecular polarisabilities obtained using different internal field models mainly due to Vuks [15] and Neugebauer [16] are studied and applied in detail for the case of LCs by different workers [17–19] are used in estimating the orientational order parameter *S*.

The order parameter is calculated by different methods such as

- *S* calculated using birefringence δn.
- *S* from effective geometry parameter, α_g.
- *S* calculated using polarisability anisotropy calculated by Lippincott δ-function [20] method and the molecular polarisabilities obtained from Vuks model.
- *S* calculated using Vuks scaling factor.
- *S* calculated using polarisability anisotropy calculated using molecular vibration [21,22] method and the molecular polarisabilities obtained using Vuks model.
- *S* calculated using polarisability anisotropy calculated using Haller approximation method and the molecular polarisabilities obtained using Vuks model.
- *S* calculated using polarisability anisotropy calculated by Lippincott δ-function method and the molecular polarisabilities obtained from Neugebauer model [19].
- *S* calculated using Neugebauer f(B) parameter.
- *S* calculated using polarisability anisotropy calculated using molecular vibration method and the molecular polarisabilities obtained using Neugebauer model.
- *S* calculated using polarisability anisotropy calculated using Haller approximation method and the molecular polarisabilities obtained using Neugebauer model.

Further, the experimental results of refractive indices obtained using wedge technique and the density values are used to estimate the molecular polarisabilities α_e and α_o. Further, the expressions used for the estimation of molecular polarisabilities α_e and α_o assuming Vuks and Neugebauer internal field models and the estimation of order parameter *S* are presented in detail in the following sections.

3.1.1 *Estimation of order parameter 'S' from birefringence δn*

De Gennes [23] pointed out that the anisotropy of any physical quantity can be a measure of orientational

order. In the case of uniaxial LC, this parameter can be defined as

$$Q = \frac{\delta A}{\Delta A} \tag{1}$$

where δA is the anisotropy of any arbitrary physical quantity ΔA = ($A_{\|}$–$A_{\perp}$) and ΔA is the hypothetical anisotropy of A in the case of perfect order.

Kuczynski et al. [24,25] proposed a simple procedure for the determination of order parameter S from the birefringence measurements δn without considering the local field experienced by the molecule in a LC phase. The birefringence δn which is a function of temperature is fitted to the following equation,

$$\delta n = \Delta n \left(1 - \frac{T}{T^*}\right)^{\beta} \tag{2}$$

where T is the absolute temperature, T^* and β are constants. (T^* is about 1–4K 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

$$\log \delta n = \log \Delta n + \beta \log\left(\frac{T^* - T}{T^*}\right) \tag{3}$$

Thus, S is given by

$$S = \frac{\delta n}{\Delta n} \tag{4}$$

where Δn is the birefringence in perfect order or at 0K. The regression analysis is employed to get the best fit for the three parameters viz., T^*, Δn and β for the compounds studied. The best fit values are given in Table 1. The S values evaluated are shown in the Figure 2 for all the compounds. The advantage of this method is that the S value can be obtained in other liquid crystalline phases like smectics also as no internal field is considered in evaluating the Δn, the birefringence in perfect order.

Table 1. Parameters for the best fit through linear regression for the equation $\log \delta n = \log \Delta n + \beta . \log\left(\frac{T^* - T}{T^*}\right)$.

Compound	$T^* = (T_{NI} + \ldots)$	β	Log(Δn)	Δn	R
3.O9O.3	0.035	0.199	−0.499	0.316	0.971
3.O10O.3	0.011	0.170	−0.480	0.331	0.920
4.O8O.4	0.002	0.164	−0.493	0.321	0.920
4.O10O.4	0.040	0.176	−0.487	0.325	0.933
5.O9O.5	0.018	0.185	−0.492	0.322	0.960

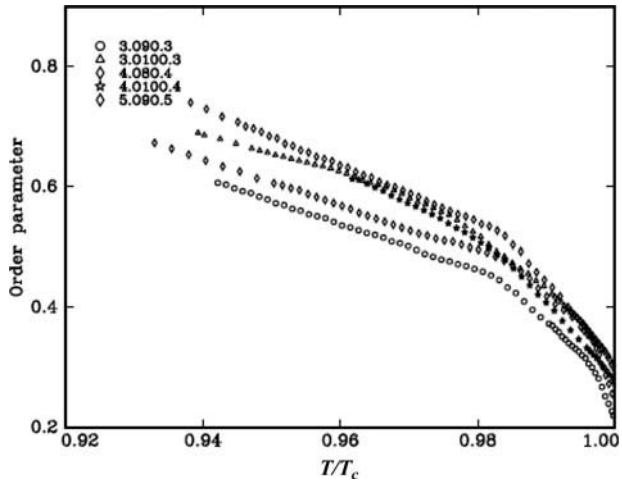


Figure 2. Variation of S values with reduced temperature (T/T_c) evaluated using Equation (4) for all the compounds.

3.1.2 *Estimation of order parameter 'S' from effective geometry parameter, α_g*

The equation for the determination of the order parameter S involving effective geometry parameter $\alpha_g = n_o/n_e$ [26] is given by

$$S = \frac{3\langle n \rangle (1 - \alpha_g)}{[2\alpha_g + 1](\Delta n)_0} \tag{5}$$

Where $<n^2>$ is the average refractive index obtained from the equation

$$\langle n^2 \rangle = 1/3\left(n_e{}^2 + 2n_o{}^2\right)$$

which decreases linearly with increase temperature as follows

$$\langle n \rangle = \mathrm{C} - \mathrm{DT} \tag{6}$$

The simplification of Equation (5) gives rise to Equation (4) and the variation of S with reduced temperature is identical with that one shown in Figure 2. The values C and D are obtained by plotting the temperature and average refractive index from above equation by linear regression. The values of C and D are given in Table 2.

Table 2. The intercept and slope values C and D, respectively, of all the compounds.

Compound	C value	D value
3.O9O.3	1.5011	−0.000149
3.O10O.3	1.5982	−0.000119
4.O8O.4	1.6175	−0.000124
4.O10O.4	1.6284	−0.000145
5.O9O.5	1.6061	−0.000100

3.1.3 *Estimation of order parameter 'S' from polarisabilities*

For the estimation of the order parameter S from the molecular polarisabilities α_e and α_o the equation used is

$$S = (\alpha_e - \alpha_o)/(\alpha_{||} - \alpha_{\perp}) \tag{7}$$

where $\alpha_{||}$ and $\alpha\perp$ are the principal polarisabilities and they can be estimated using different methods including the semi empirical methods due to Lippincott and molecular vibration techniques. Further, the molecular anisotropy can be obtained from the Haller extrapolation technique using the experimentally evaluated molecular polarisabilities from refractive index and density data. The molecular polarisabilities α_e and α_o are evaluated assuming a local field that the nematic molecule experiences. Vuks and Neugebauer proposed two different types of local fields. According to Vuks, the nematic molecule experiences an isotropic field while Neugebauer proposed an anisotropic field to the nematic molecule. The methods used, the expressions employed and the procedures adopted are described below in each case for a ready reference.

3.1.4 *Estimation of molecular polarisabilities from refractive indices and density using Vuks and Neugebauer methods*

For the estimation of the molecular polarisabilities of LC molecules, the authors have considered both the models, one due to Vuks and the other due to Neugebauer. The relevant equations of the two models for the calculation of molecular polarisabilities are given below.

3.1.4.1 Vuks method. This model was first applied to LC molecules by Chandrasekhar et al. [19] assuming the internal field as isotropic even in anisotropic crystal. The assumptions lead to the following equations.

$$\alpha_e = \left[\frac{3}{4\pi N}\right]\left[\frac{n_e^2-1}{\bar{n}^2+2}\right]$$
$$\alpha_o = \left[\frac{3}{4\pi N}\right]\left[\frac{n_o^2-1}{\bar{n}^2+2}\right] \tag{8}$$

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.

$$\bar{n}^2 = \left[\frac{n_e^2 + 2n_o^2}{3}\right]$$

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

3.1.4.2 Neugebauer method. Subramanyam et al. [22] applied this method to LC molecule. According to this method, the molecular polarisabilities are

$$\alpha_e = \left(AB - 3 \pm \sqrt{(AB-3)^2 - 4AB}\right)/2A \tag{9}$$

$$\alpha_o = \left(AB + 3 \pm \sqrt{(AB+3)^2 - 16AB}\right)/4A \tag{10}$$

where

$$A = \frac{1}{\alpha_e} + \frac{2}{\alpha_o} = \frac{4\pi N}{3}\left[\frac{n_e^2+2}{n_e^2-1}\right] + \left[\frac{2(n_o^2+2)}{n_o^2-1}\right] \tag{11}$$

$$\begin{aligned} B &= \left(\alpha_{||} + 2\alpha_{\perp}\right) = (\alpha_e + 2\alpha_o) \\ &= 3\alpha = 9\left(\bar{n}^2-1\right)/\left[(4\pi N_i)\left(\bar{n}^2+2\right)\right] \end{aligned} \tag{12}$$

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

Using the two models the molecular polarisabilities, α_e and α_o of all the five DLCs are evaluated from the refractive indices and density data.

3.1.5 *Estimation of molecular polarisability anisotropy from Lippincott δ-function model and the molecular vibrational methods*

The principal molecular polarisability anisotropy and the mean polarisability are evaluated for all the above said five number of compounds using the Lippincott δ-function model [20] and the molecular vibration method [21,22]. Table 3 represents the polarisability values of these compounds.

Table 3. Parallel, perpendicular components and mean Polarisabilities of (10^{-24} cm^3) m.OnO.m compounds.

	Polarisabilities by Lippincott δ – function model			Vibrational method
Compound	$\alpha_{\|\|}$* 10^{-24} cm^3	$2\alpha_{\perp}$ 10^{-24} cm^3	α_M 10^{-24} cm^3	α_M 10^{-24} cm^3
3.O9O.3	148.14	92.16	80.10	75.61
3.O10O.3	151.79	94.40	82.06	77.81
4.O8O.4	151.79	94.40	82.06	77.87
4.O10O.4	159.08	98.89	85.99	82.32
5.O9O.5	162.76	101.13	87.96	84.49

Note: *$\alpha_{\|n}$ value (1.3844) included in parallel component of polarisability.

3.1.6 *Estimation of order parameter, S from Haller's extrapolation, scaling factor (Vuks) and f(B) parameter (Neugebauer)*

The corresponding expressions are given below

3.1.6.1 Vuks method. The order parameter is given by [20,21]

$$S = \left[\frac{\alpha}{\alpha_{||} - \alpha\perp}\right]\left[\frac{n_e^2 - n_o^2}{\bar{n}^2 - 1}\right] \tag{13}$$

where

$$\bar{n}^2 = \left(\frac{n_e^2 + 2n_o^2}{3}\right)$$

3.1.6.2 Neugebauer method. In the Neugebauer method, the order parameter S is given [20,22]

$$S = \left[\frac{\alpha}{(\alpha_{||} - \alpha\perp)}\right] f(B) \tag{14}$$

where

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

and

$$B = \frac{n^2 - 1}{n^2 + 1}\left(\frac{n_e^2 + 2}{n_e^2 - 1} + 2\frac{n_o^2 + 2}{n_o^2 - 1}\right)$$

The scaling factors for the determination of order parameter are obtained in both the cases by plotting log–log plots between $\left[\frac{n_e^2 - n_0^2}{\bar{n}^2 - 1}\right]$ and $f(B)$ in Vuks and Neugebauer cases respectively against $(T_C - T)/(T_C - T_{NC/NK})$, i.e., the reduced temperature.

The order parameter S from the molecular polarisabilities is estimated by assuming Vuks and Neugebauer internal field models from the Lippincott δ-function, molecular vibration, Haller's extrapolation and scaling factor compared with obtained from effective geometry parameter are depicted in Figures 3 and 4 for the case of compound 4.O10O.4. The denominators, i.e., the

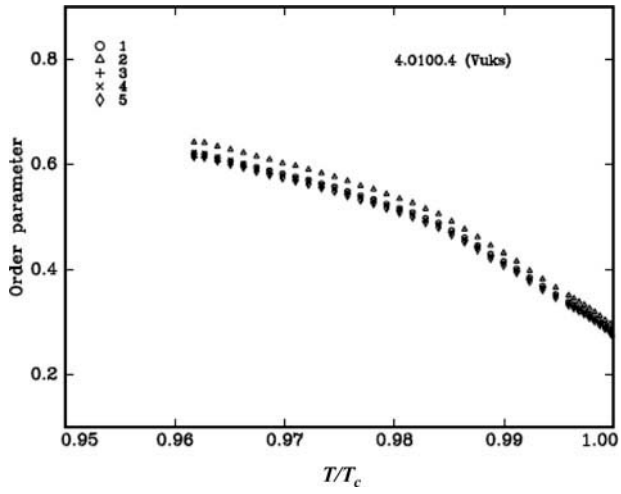


Figure 3. Variation of S with the reduced temperature (T/T_c) obtained in the case of Vuks obtained from 2. Lippincott 3. Vibration method 4. Haller's extrapolation. 5. Scaling factor in 4.O10O.4 compound and compared with 1. The S values obtained independently with the effective geometry parameter, α_g.

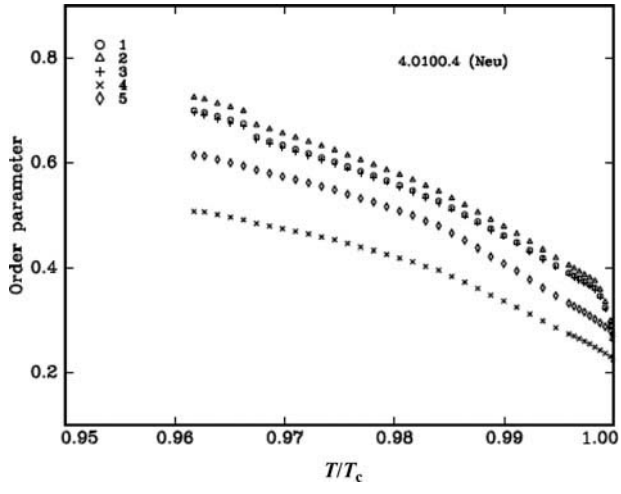


Figure 4. Variation of S with the reduced temperature (T/T_c) obtained in the case of Neugebauer obtained from 2. Lippincott 3. Vibration method 4. Haller's extrapolation. 5. Scaling factor in 4.O10O.4 compound and compared with 1. The S values obtained independently with the effective geometry parameter, α_g.

Table 4. Values of $(\alpha_{\|}-\alpha\perp) \times 10^{-24}$ cm^3 used for the evaluation of order parameter S by Lippincott, Molecular vibration methods and from log–log plots extrapolated to absolute zero (Haller and Scaling factor).

	Lippincott	Molecular vibration	Haller		Scaling factors	
			$(\alpha_{\|}-\alpha\perp)$		Sc	fB
Compound	$(\alpha_{\|}-\alpha\perp)$	$(\alpha_{\|}-\alpha\perp)$	Vuks	Neug.	Vuks	Neug.
3.O9O.3	55.98	54.40	56.49	56.75	1.4434	1.4340
3.O10O.3	57.39	55.63	57.41	57.94	1.4080	1.3870
4.O8O.4	57.39	55.51	57.54	57.80	1.4598	1.4500
4.O10O.4	60.19	58.20	60.81	60.67	1.4370	1.4390
5.O9O.5	61.63	59.20	61.37	59.18	1.4533	1.4418

principal polarisabilities, are obtained by four different methods as stated above. For the sake of ready reference, the values obtained for all the five m.OnO.m dimeric compounds from the four different methods viz., the Lippincott δ-function, molecular vibration, Haller's extrapolation and scaling factor are given in the Table 4.

4. Conclusions

For the sake of comparison of S from different methods for the two field models, with S from Δn, the percentage deviation of S value from all the methods to that calculated from Δn, the birefringence in perfect order (the error in the value of S from Δn is about 10%). If the deviation is ≤10%, the values can be considered to be in agreement with S from Δn with in the experimental error. From the figures, the tables, and the from the results on the other compounds which are not shown in figures the salient features envisaged are

- the S value obtained from Lippincott δ-function, vibrational method, Haller extrapolation method, scaling factor in the case of Vuks model agrees in all compounds.
- In the case of Neugebauer, the results are in good agreement for the following compounds: 3.O10O.3, 4.O8O.4, 4.O10O.4 in Lippincott δ-function, where as in the vibration method agreement is in the case of 4.O8O.4 only. In Haller method, the following compounds agree: 4.O8O.4, 3.O10O.3 with the S value obtained from birefringence.
- S calculated using $f(B)$ parameter from Neugebauer shows lower values to that of S from Δn in all the compounds.

Finally, from the data, it may be concluded that the compounds favour Vuks model over that of Neugebauer model. Further, one common thing that has been observed is that the S values obtained from $f(B)$ parameter always low whereas S values obtained from Haller extrapolation method are higher. The same trend is obtained in the case of monomers and dimmers studied previously [27,28].

Acknowledgements

The authors P. Pardhasaradhi and P.V. Datta Prasad thank the Management, and the Principal of The Hindu College, Machilipatnam, for providing facilities to carry out the measurements. The author Dr V.G.K.M. Pisipati express his thanks to The Head, ECE Department and the Management of K.L. University, Vaddeswaram 522 502, India for providing facilities.

Funding

This work is supported by the Department of Science and Technology [grant number SR/S2/CMP-0071/2008]. D. Madhavi Latha acknowledges the financial support of DST-WOS-A through [grant number SR/WOS-A/PS-05/2010].

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