

**Original citation:**

Xu, H., Trushkevych, Oksana, Collings, N and Crossland, W. A.. (2009) Dielectric permittivity modulation of liquid crystals for microwave applications. *Molecular Crystals and Liquid Crystals*, 502 (1). pp. 235-244.

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<http://dx.doi.org/10.1080/15421400902817346>

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# **Dielectric Permittivity Modulation of Liquid Crystals for Microwave Applications**

H. Xu, O. Trushkevych, N. Collings, and W.A. Crossland

Electrical Engineering Division, Engineering Department, University of Cambridge,  
Cambridge, UK

**Keywords:** *dielectric anisotropy, birefringence, liquid crystals*

## **Introduction**

The intensive growth of microwave engineering industry, such as telecommunications, remote sensing and global navigation satellite systems etc., requires the development of materials with large tunability of dielectric properties in microwave region. In comparison to the existing candidates such as ferroelectric ceramic materials, liquid crystals (LCs) have drawn attention in recent years mainly because of low losses in microwave frequencies and comparatively high anisotropy [1-5]. For this reason, intensive research has focused on applications of LC in GHz. [6-12]. This work includes the assessment of selected nematic liquid crystals in terms of dielectric permittivity modulation in 0.01-6 GHz range. In particular, we discuss the correlation of the dielectric properties in GHz and optical frequencies, effects of alignment on the material tunability and relaxation properties of LCs.

## **Experimental**

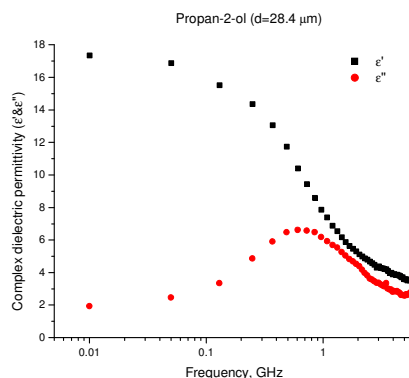
The RF measurement system consists of a network analyser (MS4623B, Anritsu) (frequency range 0.01-6 GHz), a RF test feature (BDS2200, Novocontrol), and a computer connected to the network analyser through GPIB interface for remote control. Solid (film-like) or liquid sample is sandwiched between a pair of parallel metal electrodes to form a capacitor at the end of the transmission line. The complex dielectric permittivity is calculated by measuring the complex reflection coefficient of both the filled and unfilled capacitor.

The liquid crystal samples are all commercial or research products, with birefringence values appropriate to cyano-biphenyls or higher. Planar alignment was achieved by coating the electrodes with polyimide alignment and rubbing. Electric field up to 40V AC (1 kHz) was applied to the sample to switch it into homeotropic geometry. The LC layer thickness was  $\sim 28\mu\text{m}$ . In this way, the parallel ( $\epsilon_{\parallel}$ ) and perpendicular ( $\epsilon_{\perp}$ ) components of dielectric permittivity can be measured.

## **Results and discussions**

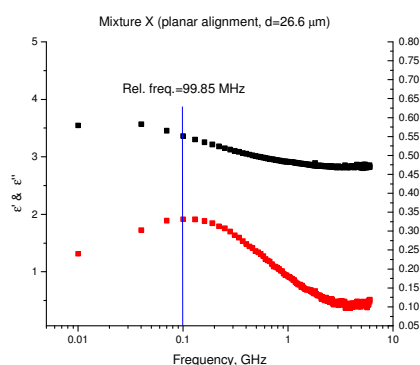
### *1. Molecular relaxation in GHz regime*

The dielectric spectroscopy in GHz regime can be applied to probe the molecular relaxations of polar materials, especially aqueous solutions and liquid crystals. With our RF system, we measured Propan-2-ol. Figure 1 shows a relaxation process of Propan-2-ol around 0.6 GHz. The relaxation time of the process is 237 ps, which is in good agreement with the published data [13].



**Figure 1** Dielectric spectrum of Propan-2-ol.

Figure 2 exhibits a relaxation process at 99.85 MHz for a high birefringence nematic LC (Mixture X). Because the spectrum was taken in planar alignment geometry, the origin of the relaxation is assigned as the molecular re-orientation around long axis. In comparison with other nematic LCs, the relaxation frequency of this material is quite high. Figure 2 shows that the dielectric loss peak is located at  $\sim 0.1$  GHz and vanishes above 3GHz. The material possesses the highest dielectric modulation (22.5% at 3GHz) amongst the test LCs. However, in order to avoid large dielectric loss at  $\sim 1$ GHz and the tail up to 3GHz, for real dielectric modulation applications this LC mixture can only be used in frequencies above 3GHz.



**Figure 2** Dielectric spectrum of nematic liquid crystal (Mixture X)

## 2 Dielectric permittivity modulations of nematic LCs in GHz

We selected nine nematic LCs, with optical birefringence values ranging from 0.11 to 0.38, for the GHz dielectric measurements. The perpendicular ( $\epsilon_{\perp}$ ) and parallel ( $\epsilon_{\parallel}$ ) component of dielectric permittivity was measured under zero and applied electric field. The dielectric tunability, defined as below, was calculated from measured data.

$$T(\%) = \frac{(\epsilon_{\parallel} - \epsilon_{\perp})}{\epsilon_{\parallel}} \cdot 100\%$$

We compared some of the results with the data obtained from literature, in order to check validation of our measurement system.

Pentyl cyano-biphenyl (K15) is a well known nematic LC, which has been measured in the RF frequency range by various methods [1, 3, 14]. For comparison purposes, we intended to choose data measured similarly to our method (reflective coaxial method). Such a data found in the literature within our frequency range was measured using a parallel capacitor method (0.1-1 GHz) by Weil et al [1]. As can be found in Table I, the tunability obtained by us (3.5%) was smaller than the ref. data (4.3%). It can be also noticed that the  $\tan\delta$  loss measured by our RF system was 40-50% lower than the ref. data.

We have identified two reasons that could explain this disparity. Poor planar alignment may increase the value of  $\epsilon_{\perp}$ . It is difficult to check the alignment quality visually due to the use of metal electrodes in our system. The tuning voltage in our measurement system is limited to 40 Vp-p and the cell thickness is about 28 $\mu$ m. The resulting field strength of 1.43 V/ $\mu$ m may not be high enough to achieve maximum tunability. The other reason could be the possibility of affecting accuracies at the edge of measurement frequency ranges. The data at 1GHz by Weil et al. was measured at the edge of their measurement frequency range (0.1-1 GHz), whereas it was in the middle in our case (frequency range: 0.01-6 GHz).

**Table I** Data of K15 in comparison to the data in the literature [1]

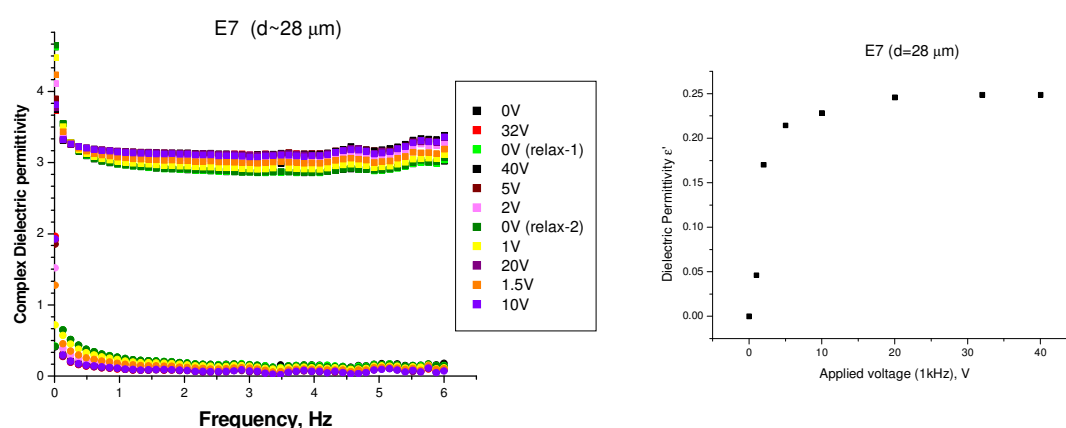
K15 (1GHz)	$\epsilon_{\parallel}$	$\epsilon_{\perp}$	$\Delta\epsilon$	$\tan\delta_{\parallel}$	$\tan\delta_{\perp}$	Tunability
Cambridge	3.18	3.07	0.11	0.055	0.095	3.5 %
Ref [1]	3.27	3.13	0.14	0.084	0.151	4.3 %

### 3 The effect of alignment on the dielectric permittivity modulations of nematic LCs

In the voltage controlled tunability measurements, we paid attention to the effect of the surface alignment. In order to investigate the effect of surface alignment on the measurement reproducibility, we carried out measurements on cells with/without surface alignment treatments. We found that, for a test cell without planar alignment, the measured  $\epsilon_{\perp}$  value could not be reproduced. After external field was applied, the value of  $\epsilon_{\perp}$  (dielectric permittivity at planar state under zero voltage) became higher than the initial value. However, for the test cell coated with a polyimide layer and rubbed uniaxially, the value of  $\epsilon_{\perp}$  at planar state (0V) remained the same when the voltage was returned to zero. Figure 3(a-b) shows the dielectric permittivity against applied voltages for E7 in a test cell treated by planar alignment. The dielectric spectra illustrate that, with increasing voltage, the dielectric permittivity increases initially at about 1V (the Frederick's transition threshold voltage) and tends to saturate at 40V (molecules aligning with the field direction). As shown in Figure 3a, the curves representing the initial zero

voltage state (0V), the voltage returned to zero for the first time state (0V relax-1), and the voltage returned to zero for the second time state (0V relax-2) are almost fold-over.

In conclusion, the aligned test cell could be switched multiple times without changes in the measured  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$ . Because the measured dielectric permittivity is very sensitive to the alignment of the molecules, a surface alignment is essential in order to provide strong enough surface anchoring energy for maintaining the initial planar alignment state.



**Figure 3a** (left) Dielectric spectra of E7 under variable applied voltages.  
**3b** (right) Dielectric permittivity vs applied voltage for E7.

### 3 Correlation between dielectric anisotropy in microwave (MMW) regime and optical anisotropy in optical frequency

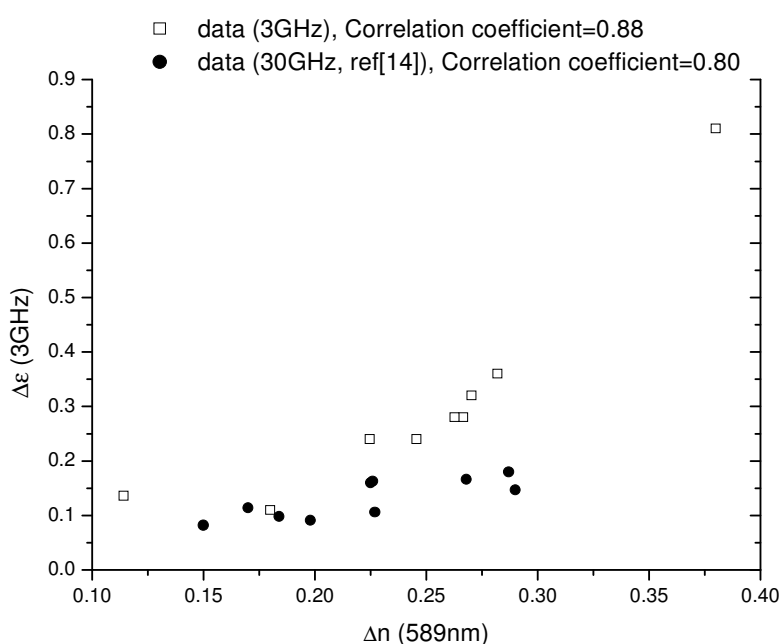
**Table II** lists the real and imaginary parts of the dielectric permittivity at 3GHz and the refractive index values at 589nm for the tested nematic liquid crystals. The studied materials exhibit a dielectric anisotropy varying between 0.25 and 0.83 with low associated losses (typically  $\tan\delta_{\perp} \sim 0.05$  and  $\tan\delta_{\parallel} \sim \tan\delta_{\perp}/2$ ). Figure 4 shows dielectric anisotropy  $\Delta\epsilon$  (in microwave regime) vs birefringence  $\Delta n$  (589nm) for two sets of data. One set of data was measured by us at 3GHz for the 9 nematic LCs, as listed in Table II. The other set of data was obtained from the work done by Lim K.C. et al at 30GHz [14].

**Table II.** Dielectric permittivity (3GHz) and refractive index (589nm) of the tested NLCs

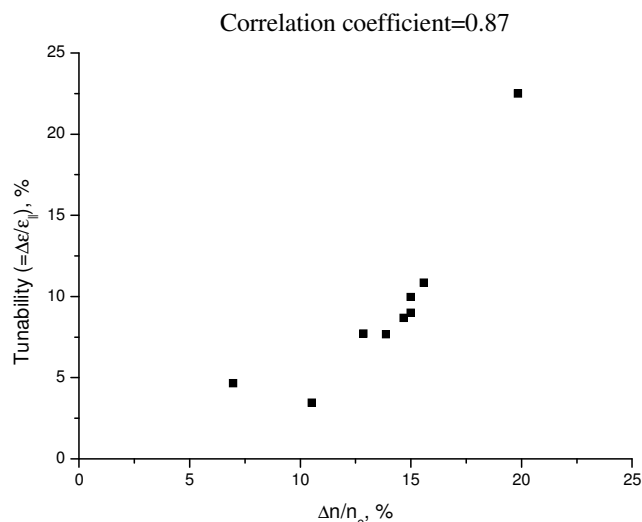
Nematic LC	$\epsilon_{\perp}$	$\epsilon_{\parallel}$	$\Delta\epsilon$	$\tan\delta_{\perp}$	$\tan\delta_{\parallel}$	Tunability	$n_e$	$n_o$	$\Delta n$
	3GHz						589nm		
L048	2.95	3.23	0.28	0.04	0.02	8.7 %	1.7904	1.5277	0.2627
BL037	2.96	3.32	0.36	0.06	0.03	10.8%	1.808	1.526	0.2820
BL111	2.89	3.21	0.32	0.06	0.03	10.0%	1.8025	1.5321	0.2704
BL093	2.89	3.13	0.24	0.06	0.02	7.7%	1.7697	1.5241	0.2456
Mixture X	2.79	3.6	0.81	0.05	0.035	22.5%	1.915	1.535	0.38
E7	2.87	3.11	0.24	0.05	0.025	7.7%	1.7462	1.5216	0.2246
MDA1602	2.83	3.11	0.28	0.06	0.04	9.0%	1.7779	1.5113	0.2666

ZLI2222/000	2.77	2.91	0.14	0.04	0.025	4.6%	1.6134	1.5010	0.1124
K15	3.07	3.18	0.11	0.095	0.055	3.4%	1.71	1.53	0.18

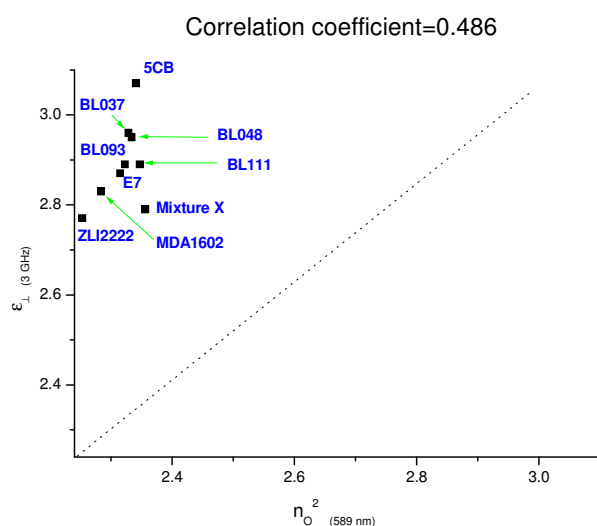
As found in Figure 4, for both the data measured at 3GHz and at 30GHz, the dielectric anisotropy  $\Delta\epsilon$  (GHz) was larger in the material with higher birefringence (589nm). For our measured data, a fairly high correlation coefficient of 0.88 between  $\Delta\epsilon$  (3 GHz) and  $\Delta n$  (589nm) was obtained. Similar correlation coefficient (0.8) between  $\Delta\epsilon$  (30 GHz) and  $\Delta n$  (589nm) was found for the data measured by Lim et al [14]. For the purpose of normalisation, we plot the dielectric tunability (T%, 3GHz) against  $\Delta n/n_e$  (% , 589nm) (Figure 5). The correlation coefficient is 0.87, which is similar to that between  $\Delta\epsilon$  (3 GHz) and  $\Delta n$  (589nm).



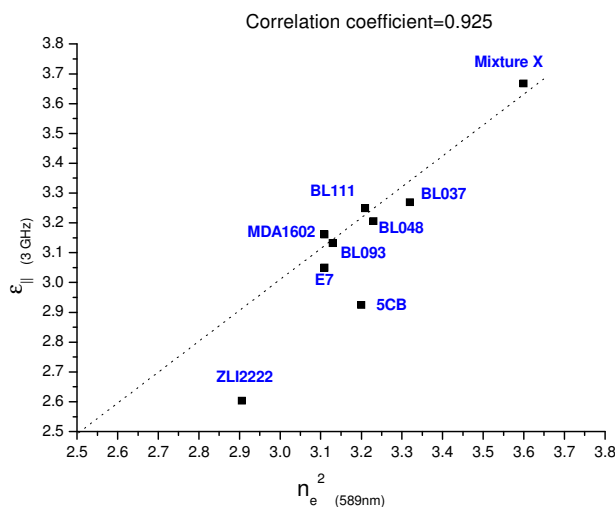
**Figure 4** Dielectric anisotropy  $\Delta\epsilon$  (in microwave regime) vs birefringence  $\Delta n$  (589nm), open square for data of the LCs tested by us at 3GHz; filled circles for data obtained from the literature [14] at 30 GHz.



**Figure 5** Dielectric tunability T (% , 3GHz) vs  $\Delta n/n_e$  (% , 589nm) for tested LCs. The fairly high correlation coefficients (for 3GHz and 30GHz) indicated in Figure 4 mean that, for variety of LCs, the dielectric anisotropy at microwave regime is correlated quite well with the birefringence in visible (589nm). This finding is indirectly supported by another research work in the MMW regime (Nose et al [4]). They discovered that there was strong correlation between the refractive indices for MMW and for the visible wavelengths. Both the results of dielectric anisotropy (by us) and birefringence (by Nose et al [4]) confirm there is a correlation between the dielectric /optical properties in MMW and visible regions. This behavior is not only interesting but also useful. It means that, to obtain a certain dielectric modulation  $\Delta\epsilon$  in MMW regime, we can select a LC material according to its birefringence value at optical frequencies.



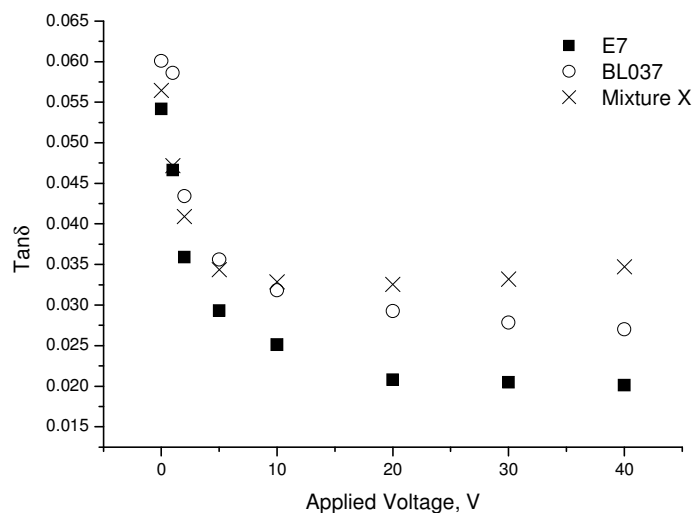
**Figure 6**  $\epsilon_{\perp}$  (3GHz) vs  $n_o^2$  (589nm) for tested LCs.



**Figure 7**  $\epsilon_{||}$  (3GHz) vs  $n_e^2$  (589nm) for tested LCs.

We also evaluate the data in terms of the correlation between the components of the anisotropy,  $\epsilon_{||}$  or  $\epsilon_{\perp}$  at 3GHz, with  $n_e^2$  or  $n_o^2$  ( $\lambda=589\text{nm}$ ), as shown in Figures 6-7.

An interesting feature found in Figures 6-7 is that the parallel and perpendicular dielectric and optical components showed different correlation coefficients. The correlation coefficient for parallel components was 0.925. For the perpendicular components, a lower correlation coefficient 0.486 was obtained. To understand the physical origin of this behavior is currently the subject of further work.



**Figure 8** Loss  $\text{tan}\delta$  vs applied voltage for three LCs.

Another experimental finding is that, in general the measured dielectric loss  $\text{tan}\delta_{||}$  was almost half of  $\text{tan}\delta_{\perp}$  (see Table II). Figure 8 shows, for three Lbs., how the loss



$\tan\delta_{\perp}$  ( $V=0$ ) decreases towards  $\tan\delta_{\parallel}$  ( $V=40V$ ), as the LC directors are re-oriented to approach the homeotropic state by increasing the voltage. Similar behavior was also found by other researchers [15]. This may be attributed to different molecular relaxations in the planar or homeotropic states [16]. In the initial planar state (with no electric field), the dielectric dispersion at high MHz frequencies is caused by the LC molecular orientation around long axis with a characteristic relaxation frequency  $f_{\perp}$ . In the homeotropic state (with an external field), the dielectric absorption at MHz regime is dominated by reorientation about the molecular short axis with a relaxation frequency ( $f_{\parallel}$ ). When measuring the dielectric loss at 3GHz, the measurement field senses the tails of the two different relaxations located at different frequencies. Due to the different rotational viscosities for the two relaxation processes, generally  $f_{\perp} > f_{\parallel}$  [16]. Therefore, we expect that the  $\tan\delta_{\perp}$  loss is the more significant. This is still an open question for further investigation.

## Conclusions

In summary, we have measured dielectric properties, in the region from 0.01GHz up to 6GHz, of nematic liquid crystals with a range of birefringence values. At 3GHz, the maximum dielectric modulation was found to be 22.5% for a high birefringence LC (Mixture X). The modulation is accompanied by low dielectric loss. We also investigated the effect of alignment on the reproducibility of dielectric modulation. It was found that a good surface alignment is essential to achieve reproducibility of the dielectric modulation. Finally, we experimentally confirmed that there is a strong correlation between the dielectric anisotropy in the microwave region and the birefringence at visible frequencies.

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### **Acknowledgments**

The authors acknowledge funding from the Centre for Advanced Photonics and Electronics (CAPE). Current CAPE partners are Dow Corning Ltd. and Alps Electric Co., Ltd.