**Liquid Crystals**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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**To cite this Article** Kundu, Shyamal Kumar and Chaudhuri, B. K.(2005) 'Observation of scaling behaviour of dielectric relaxation in ferroelectric liquid crystals', Liquid Crystals, 32: 1, 27 — 30

**To link to this Article: DOI:** 10.1080/02678290412331320601

**URL:** http://dx.doi.org/10.1080/02678290412331320601

*Liquid Crystals*, Vol. 32, No. 1, January 2005, 27–30

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# Observation of scaling behaviour of dielectric relaxation in ferroelectric liquid crystals

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(*Received 17 May 2004; in final form 26 August 2004; accepted 29 August 2004*)

The temperature (25–120°C) and frequency (10 Hz–1 MHz) dependent dielectric relaxation spectra of different ferroelectric liquid crystals are found to show interesting scaling behaviour. Considering relaxation frequency ($\nu_{\max}$) as the scaling frequency, the imaginary part of the dielectric constants ($\varepsilon''$) of the ferroelectric liquid crystals are found to follow the universal scaling formula $\frac{\varepsilon''(f)}{\varepsilon''_{\max}} \alpha \left(\frac{f}{\nu_{\max}}\right)^m$, i.e. when ($\varepsilon''/\varepsilon''_{\max}$) is plotted as a function of ($f/\nu_{\max}$), the scaling behaviour of a ferroelectric liquid crystal can be represented by a single master curve irrespective of film thickness, bias field and temperature; $\varepsilon''_{\max}$ is the maximum value of $\varepsilon''$ at the frequency $\nu_{\max}$ and $m$ is a parameter $\sim 1$.

## 1. Introduction

Some time ago Nagi reported ‘universality’ behaviour of low frequency fluctuation, dissipation and relaxation properties in condensed matter [1]. Such behaviour, however, has not been well demonstrated in ferroelectric liquid crystal (FLC) systems, but is very important for data evaluation programs. The scaling of different data sets to collapse all to one common curve indicates that the process can be separated into a common physical mechanism modified by the thermodynamic scales [2]. The usefulness of this scaling concept may be shown in the gas–liquid coexistence of many molecular fluids. When the coexistence curves of different fluids are scaled by the density and temperature at the critical point, they collapse to a single universal curve [3, 4]. Recently, renewed interest has focused on the scaling observed in the frequency dependence of the ionic conductivity of ion-containing glasses [5–7].

We consider one of the fundamental problems, associated with the ‘relaxation properties’ of FLCs, is to find a universal relationship describing their complicated dielectric relaxation behaviour. Many empirical model equations have so far been proposed to describe the relaxation behaviour of semiconducting glassy oxides and liquid crystalline materials [8–10]. Among these models, the Cole–Cole relationship [8] represented by the equation

$$\varepsilon^*(\nu) - \varepsilon_\infty \alpha \frac{1}{1 + (\mathrm{i}2\pi\nu\tau)^{1-\alpha}} \tag{1}$$

has been widely used to describe the dielectric spectra of different systems. Devidson–Cole [9] also proposed another form of this equation,

$$\varepsilon^*(\nu) - \varepsilon_\infty \alpha \frac{1}{[1 + \mathrm{i}2\pi\nu\tau]^{\delta}}. \tag{2}$$

The Williams–Watts [10] formalism for fitting the dielectric spectra is given by

$$\varepsilon^*(\nu) \alpha \left\{ \textit{the Fourier transform of} \left(\frac{d}{dt}\right) \left[ - \exp - (t/\tau)^{\beta} \right] \right\} \tag{3}$$

None of these relationships can, however, be uniquely applied to represent the dielectric spectra of different FLC systems. Many researchers [2, 11–13] have used various forms of the scaling frequency and attempted to show the universality character of the dielectric spectra of different solid systems. For example, Sidebottom [11] used ($\sigma_{\mathrm{d.c.}}/\varepsilon_0\Delta\varepsilon$) as the scaling frequency, where $\varepsilon_0$ is the free space permittivity and $\Delta\varepsilon[=\varepsilon_0 - \varepsilon_\infty]$ is the permittivity change from the unrelaxed base line $\varepsilon_\infty$ to the fully relaxed level $\varepsilon_0$. According to the model used by Dyre [14], the scaling frequency is equivalent to some numerical factor multiplied by the crossover of the hopping frequency. Recently, several attempts have been made to scale the conductivity spectra of several semiconducting glass compositions [2, 11, 13, 15, 16] in terms of temperature and composition. The dielectric

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*Liquid Crystals*
ISSN 0267-8292 print/ISSN 1366-5855 online
http://www.tandf.co.uk/journals
DOI: 10.1080/02678290412331320601

spectra of some supercooled organic liquids [16, 17] have also been scaled in terms of the width and the peak position and critical mixture [18].

Using relaxation frequency, obtained from equation (1), as the scaling frequency, we show in this paper that the dielectric relaxation data of FLCs such as Felix-15/100 and CS-1027, exhibit interesting scaling behaviour. Furthermore, this scaling behaviour is found to be independent of temperature, cell thickness and also of the applied bias field. The dielectric constant data of several other FLC samples having very different structure and dipole moments, including, CS-1022 [19], FLC-250 [20] and a ferroelectric copolysiloxane [21], have also been tested to show the validity of the scaling law reported here. Interestingly, all the different samples studied are found to show scaling behaviour.

## 2. Experimental

A shielded parallel plate capacitor was used to measure the complex dielectric permittivity ($\varepsilon^*$) of the FLC samples for different thicknesses of the cell (10–80 µm). Indium tin oxide (ITO)- coated transparent glass plates were used for electrodes. The Felix-15/100 and CS-1027 type FLC mixtures, were introduced into the cell by means of the capillary action technique in their isotropic phases. The CS-1027 sample used undergoes a ferroelectric SmC* to paraelectric SmA phase transition at 62.0°C; Felix-15/100 undergoes a similar transition from the ferroelectric SmC* to the paraelectric SmA phase at 72.0°C. For homogeneous alignment (unidirectional alignment), we applied, as in earlier work [22], a low frequency a.c. field ($\cong 0.2\,\mathrm{V\,\mu m^{-1}}$) for about 60 min. The good quality of the alignment was verified by polarizing microscopy. The sample temperature (25.0–120°C) was controlled by a Eurotherm temperature controller to within $\pm 0.1$°C. The frequency dispersion of the complex dielectric permittivity $\varepsilon^*(\omega)=(\varepsilon', \varepsilon'')$ was measured in the frequency range 10 Hz–1 MHz, under different bias fields varying from 0 to 4 V, using a computer-controlled HP 4192A impedance analyser.

## 3. Results and discussion

The variations of the frequency dependent $\varepsilon''(\omega)$ for two typical FLC samples, Felix-15/100 and CS-1027, for different fixed temperatures, are shown in figures 1 (*a*) and 1 (*b*), respectively, for the G mode in the SmC* phase. It can be seen that the dielectric spectra merge into a single common curve when $\varepsilon''$ (along the $y$-axis) is scaled with respect to $\varepsilon''_{\max}$ relaxation and the frequency (along the $x$-axis) is scaled with respect to relaxation frequency ($\nu_{\max}$). The relaxation frequency and $\varepsilon''_{\max}$ were first obtained from fitting the experimental dielectric constant data with equation (1), and then each value of $\varepsilon''_{\max}$ for the FLC was scaled. A perfectly superposed master curve for the dielectric relaxation spectra was obtained for the material Felix-15/100, as shown in figure 2 (*a*). It can be seen that all the curves, irrespective of frequency and temperature, follow the unique relationship

$$\frac{\varepsilon''(f)}{\varepsilon''_{\max}} \alpha \left(\frac{f}{\nu_{\max}}\right)^m \text{ or } \frac{\varepsilon''(f)}{\varepsilon''_{\max}} = F\left(\frac{f}{\nu_{\max}}\right)^m \tag{4}$$

where $F$ is a ‘universal’ master function; here $\varepsilon''/\varepsilon''_{\max}=(f/\nu_{\max})^m$ for $f \gg \nu_{\max}$, where $\varepsilon''_{\max}$ is the maximum value of $\varepsilon''(f)$ at the relaxation frequency $\nu_{\max}$ and $m \sim 1$ [17, 23]. The other sample used in our present investigation, CS-1027, follows the same scaling behaviour, see figure 2 (*b*). Similar power dependence behaviour, equation (4), was exhibited by some supercooled liquids [16, 17] and also by some amorphous materials where almost Debye-type behaviour of the dielectric constant was found. Such a scaling relationship indicates that the relaxation mechanism in ferroelectric liquid crystals is temperature independent.

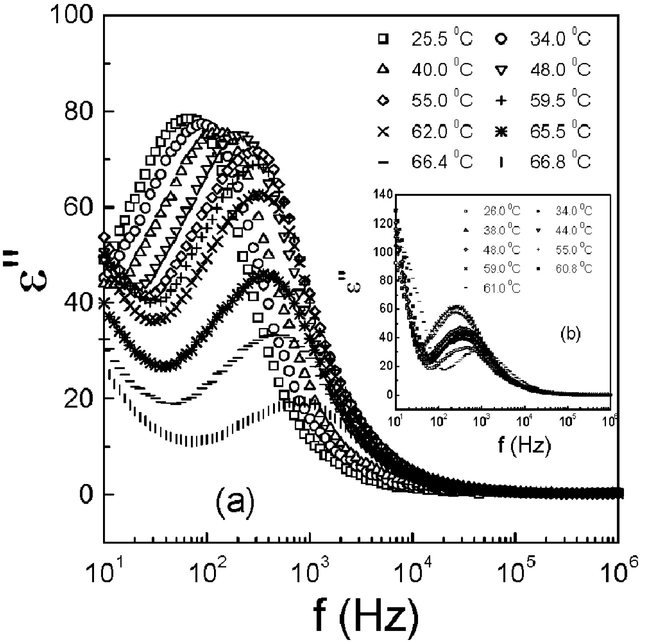

Figure 1. Variation of the imaginary part of the dielectric constant ($\varepsilon''$) in the SmC* phase as a function of frequency at different fixed temperatures: (*a*) 11 µm thick cell of Felix-15/100 and (*b*) 18 µm thick cell of CS-1027.

We now consider whether similar scaling behaviour is also observed in the G mode in the SmC* phase for different fixed bias voltages. For this purpose, the relaxation frequency $\nu_{\max}$ and $\varepsilon''_{\max}$ were again obtained by fitting the experimental dielectric constant data,

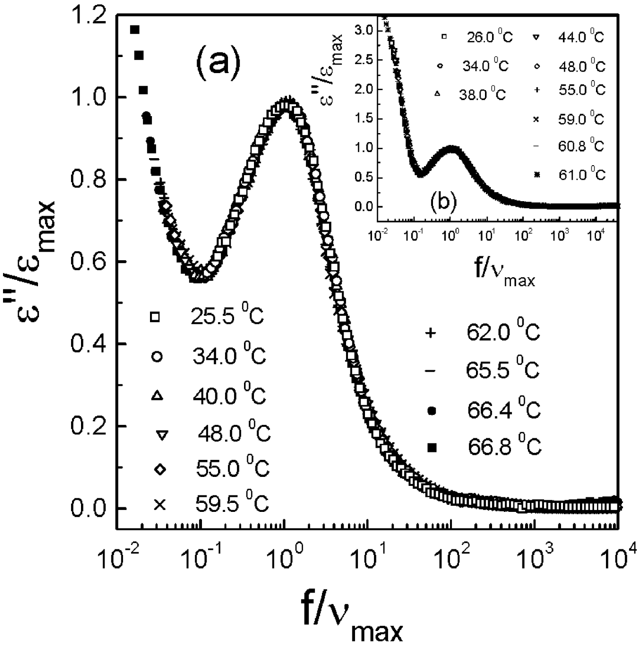

Figure 2. The frequency spectra of the imaginary part of the dielectric constant for (*a*) Felix-15/100 (11 μm cell thickness) and (*b*) CS-1027 (18 μm cell thickness) for different temperatures. $\varepsilon''$ and frequency axes are scaled, respectively, by $\varepsilon''_{\max}$ and relaxation frequency $\nu_{\max}$ obtained from the best fit of dielectric constant data with equation (1).

measured under different bias fields, with equation (1), and then each $\varepsilon''_{\max}$ for the FLC material (Felix-15/100) was scaled as before. A very good superposed master curve is again obtained for the dielectric relaxation spectra of the sample, as shown in figure 3, at an arbitrarily fixed temperature (55.0°C). This indicates that the relaxation mechanism is also independent of bias field.

Finally, to show that the scaling behaviour described by equation (4) is independent of sample thickness, the dielectric spectra were analysed for different arbitrary cell thicknesses at any arbitrary temperature (28.2°C), see figure (4). Interestingly, it is observed that except for the very low frequency region, the spectra for all the different thicknesses of sample superpose on a single master curve. The small deviation seen only in the low frequency region arises due to the different ionic contribution of FLC cells with different thicknesses.

From figures 2, 3 and 4, one can see the universality of the dielectric spectra of the FLC systems. To the best of our knowledge, such behaviour has not been reported to date. We have also examined the scaling behaviour of the dielectric relaxation data of several other samples including CS-1022 [19], FLC-250 [20], and a ferroelectric copolysiloxane [21], measured earlier by different research groups. Interestingly, all these samples are found to follow the scaling behaviour described by equation (4), which may, therefore, be regarded as a universal scaling law for ferroelectric liquid crystal systems.

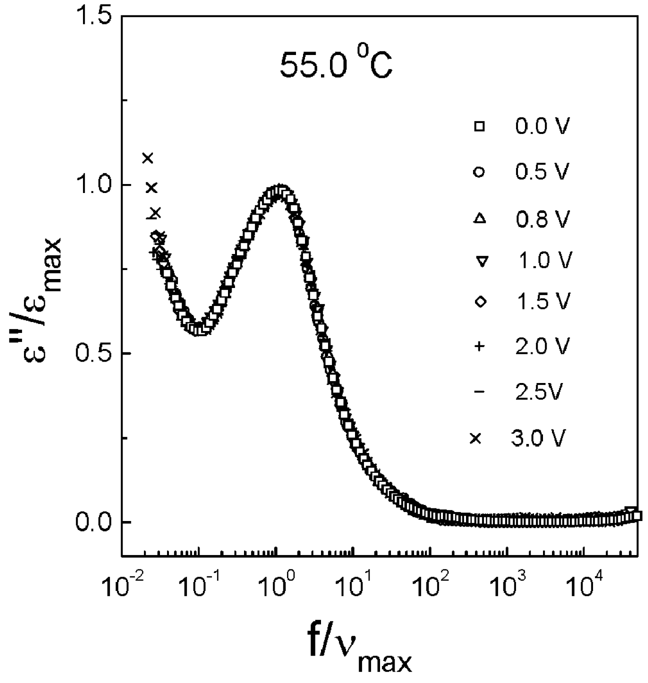

Figure 3. The frequency spectra of the imaginary part of the dielectric constant of the FLC mixture Felix-15/100 with different bias voltages at an arbitrarily fixed temperature (55.0°C). $\varepsilon''$ and frequency axes are scaled, respectively, by $\varepsilon''_{\max}$ and relaxation frequency $\nu_{\max}$ obtained from the best fit of dielectric constant data with equation (1) for 11 μm thick cell.

## 4. Conclusion

In conclusion, scaling is an important feature of the dielectric constant in FLCs, which resembles the behaviour of many other systems. The dielectric relaxation data of a FLC, over a wide range of temperature and frequency, can be scaled in such a way as to lie on a single master curve. The dielectric relaxation data for different bias fields and for different thicknesses of the FLC samples can also be scaled in such a way that they fit to the same master curve. The dielectric relaxation frequency obtained from the Cole–Cole fitting of the experimental data is considered as an appropriate parameter for the scaling of the dielectric relaxation spectra for the ferroelectric liquid crystals. We notice that similar scaling behaviour is exhibited by the dielectric relaxation spectra of the SmC* and SmA phases in the soft mode as well as in the nematic phase. It has been shown that in the case of supercooled organic liquids, the scaling behaviour is not appropriate when a second relaxation process appears [17]. However, it can be seen that for the FLC system, the

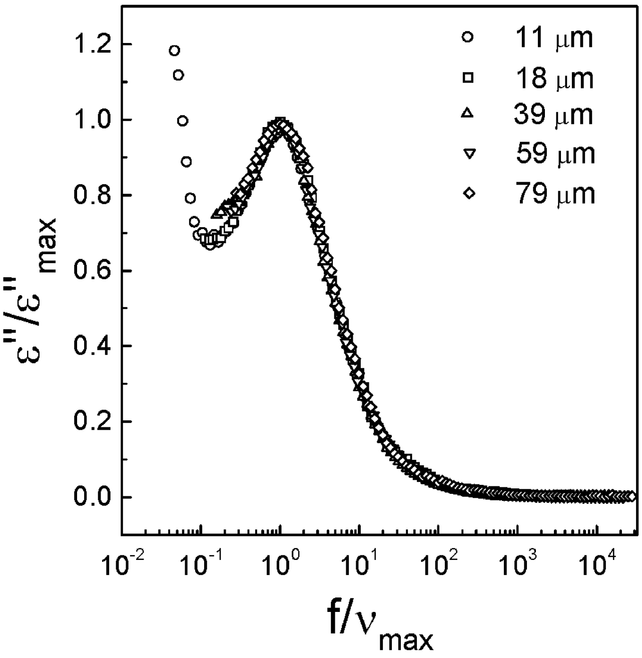

Figure 4. The frequency spectra of the imaginary parts of the dielectric constant of the FLC mixture Felix-15/100 with different sample thickness (11, 18, 39, 59 and 79 $\mu$m) at an arbitrarily fixed temperature (28.2°C). $\varepsilon''$ and frequency axes are scaled, respectively, by $\varepsilon''_{\max}$ and relaxation frequency $\nu_{\max}$ obtained from the best fit of dielectric constant data with equation (1).

scaling behaviour can be observed in a system even with a double relaxation, if the average of the two-relaxation frequencies is taken as the scaling frequency. This scaling observation in FLCs is interesting and may be extended to other similar systems in any data evaluation program, in the verification of the experimental data and above all to obtain a unified picture of the scaling behaviour in many different systems.

## Acknowledgements

The authors are grateful to the CSIR Government of India for a research grant and fellowship to carry out this project. The authors are also grateful to the AvH Foundation, Germany, and Prof. W. Haase for providing the instruments for measuring dielectric relaxation of the liquid crystals.

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