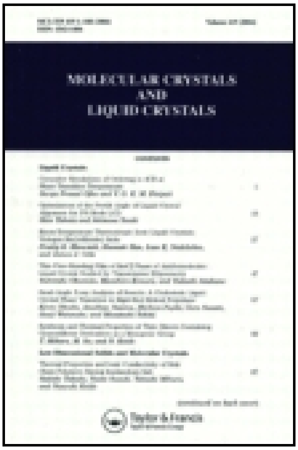

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Low Frequency Dielectric Relaxations in Surface Stabilized Ferroelectric Liquid Crystal

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Low Frequency Dielectric Relaxations in Surface Stabilized Ferroelectric Liquid Crystal

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Three relaxation modes are identified in the surface stabilized ferroelectric liquid crystal from the Tikhonov regularization analysis of the low frequency dielectric data. The field strength dependence of the relaxation frequency and mode strength for each relaxation mode was studied in detail so that we could help better clarifying for the characteristic relaxation mechanisms of the three low frequency modes recently found by various authors.

Keywords: low frequency modes; dielectric relaxation; ferroelectric liquid crystal; Tikhonov regularization

INTRODUCTION

In the ferroelectric liquid crystal[1] we have two low frequency modes, the soft mode and the Goldstone mode, where each represents the amplitude and phase fluctuation respectively of the mean molecular tilt from the layer normal[2]. In addition to the two bulk modes which couple linearly with the polarization in the ferroelectric liquid crystal several other modes can appear in the low frequency dynamics of the thin film samples of ferroelectric liquid crystal due to the finite size confinement effect[3].

In contrast to this case of bulk sample with helix structures very few dielectric studies are reported on the surface-stabilized ferroelctric liquid crystal samples where the helix structures are suppressed by the surface anchoring effect. In the thin film systems of surface-stabilized ferroelectric liquid crystal experimental observations are reported for the sample thickness dependent X-mode[4], the domain reversal mode[5, 6] and a layer deformation mode[7] in the low frequency region.

In the present work we want to analyze the broad dielectric spectrum overlapping with each other in the same low frequency region by use of the Tikhonov regularization method[8] to separate out each mode of characteristic relaxations. Each mode of the low frequency relaxations was then studied for the electric field dependence of the relaxation frequency and the mode strength, which should help to probe for the origin of each low frequency mode.

Tikhonov Regularization Method

Between the experimental data $\varepsilon(\omega)$ and a given kernel function $K(\omega,\tau)$ we can define the distribution function of relaxation time τ, $g(\tau)$, by an integral transform

$$\varepsilon(\omega) = \int_{-\infty}^{\infty} K(\omega,\tau) g(\tau) d\tau + \sum a_j h_j(\omega) \qquad (1)$$

where the 2nd term is a conductivity term well defined from the ionic ac current measurements.

This Fredholm integral equation of the first kind is an ill-posed problem, where the least square fitting solution of $g(\tau)$ is very unstable against a slightest change of $\varepsilon(\omega)$. Since the experimental values of $\varepsilon(\omega)$ include errors, we can not obtain a definitive solution but infinitely many solutions for g(τ) by use of the least square best fit algorithm.

A new algorithm for a definitive and stable solution of g(τ) in the ill-posed problem was introduced as Tikhonov regularization[7]. The Tikhonov regularization encounters the minimization of the function V(λ) as given by

$$V(\lambda)=\sum_{i=1}^{n}\frac{1}{\sigma_i^2}\left[\varepsilon(\omega_i)-\left(\int_{-\infty}^{\infty}K(\omega_i,\tau)g(\tau)d\tau+\sum_{j=1}^{m}a_j b_j(\omega_i)\right)\right]^2 \qquad (2)$$

$$+\lambda\int|g''(\tau)|^2\,d\tau$$

where σ_i represents the experimental error of $\varepsilon(\omega_i)$, and λ the regularization parameter to control the smoothness of the function g(τ) in competition with the first term of data tracking for the least square best fit. The calculation of g(τ) is then reduced to a programming to find the inversion of a determinantal matrix[9], where a commercial program is available[10].

Our concerning integral equations for g(τ) are

$$\varepsilon''(\omega)=\frac{\sigma_0}{\varepsilon_0}\omega^{-1}+\Delta\varepsilon\int\frac{g(\tau)\omega\tau}{1+\omega^2\tau^2}d(\ln\tau) \qquad (3\text{-}1)$$

$$\varepsilon'(\omega)=\varepsilon_\infty+\Delta\varepsilon\int\frac{g(\tau)}{1+\omega^2\tau^2}d(\ln\tau) \qquad (3\text{-}2)$$

where d(lnτ) represents the logarithmic scale of τ axis.

DIELECTRIC CONSTANT MEASUREMENTS

We have employed the lock-in amplifier system (EG&G, DSP 7260) to measure the low frequency dielectric constants as depicted in Fig.1. The standard capacitor C_s was chosen to be 20μ F while the sample has a room temperature value of capacitance C_d of about 20 nF. Since we have $C_s \gg C_d$ we obtain for various voltage signals of Fig.1

We can thus find the capacitance of the sample C_d by measuring V_{out}, the voltage across the standard capacitor C_s by use of the lock-in amplifier.

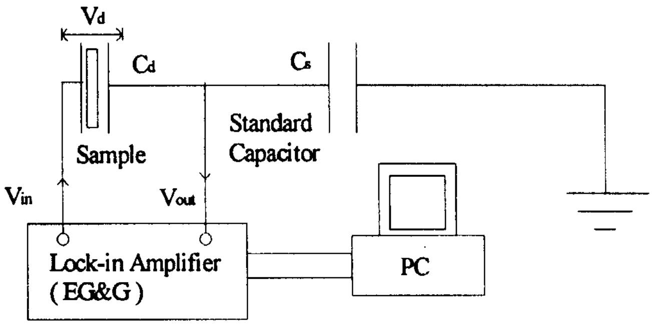

FIGURE 1 Experimental set up

The present sample (Chisso, CS-1025) is known to have the SmC* phase in the broad range of temperature from –3 °C to 62 °C with a long helical pitch of 10μ m. The sample thickness of the cell was 6μ m and the sample cell was made up of conductive indium-tin-oxide coated glass. Our dielectric measurements in the frequency range of 1 Hz to 100 KHz were performed at room temperature with applied voltages of 0.01 Vrms to 3.0 Vrms and the phase delay in the lock-in amplifier was compensated by use of the empty cell.

EXPERIMENTAL RESULTS AND DISCUSSIONS

In Fig.2 we have shown the calibrated data of imaginary dielectric constants $\varepsilon''(\omega)$ measured at various probe voltages.

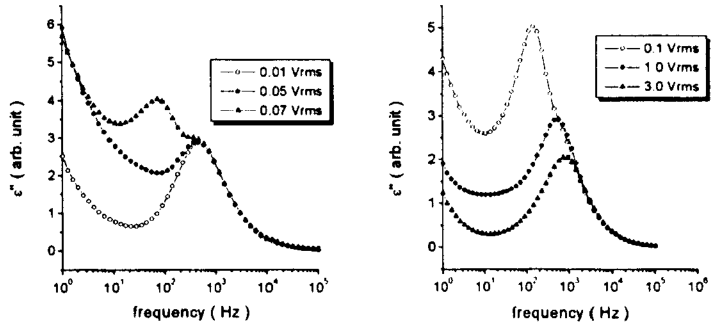

FIGURE 2 Compensated data of ε"(ω) measured at various probe voltage from 0.01 Vrms to 3.0 Vrms

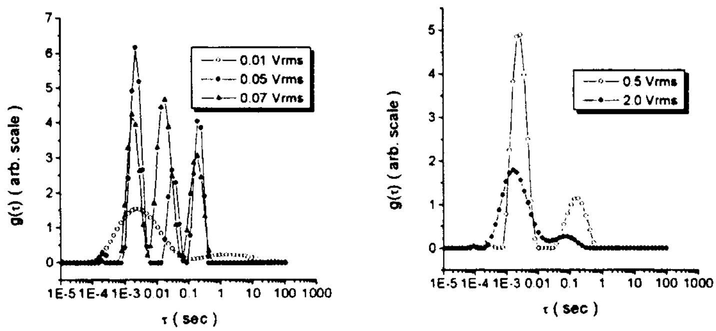

FIGURE 3 Relaxation time distribution g(τ) obtained from Tikhonov regularization analysis of ε"(ω) shown in Fig.2

These dielectric data of measurements were analyzed by use of the Tikhonov regularization program to obtain the relaxation time distributions g(τ) as depicted in Fig.3, where discrete peaks implicate discrete sets of relaxation times.

The dielectric constant data of Fig.2 displays a central low frequency continuum of the ω^{-1} conductivity contribution extended to very broad relaxation peaks with peak positions and relative strengths varying with the amplitudes of the probe fields.

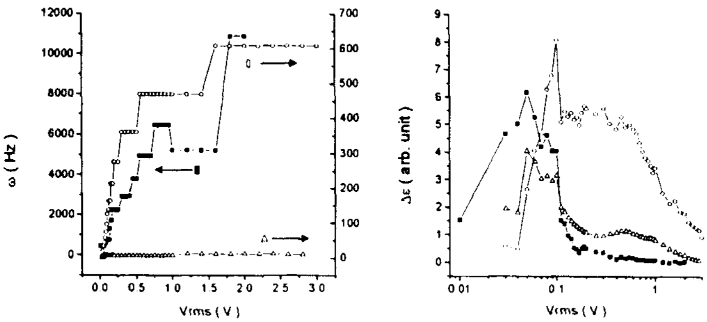

FIGURE 4 Dielectric strength (Δε) and relaxation frequency (ω) dependence on probe voltage (Vrms) for low frequency modes of g(τ): -o - Domain mode, -■ - X-mode, and -△- L-mode

These broad continuum spectra ε"(ω) of overlapping bands can be seen to be transformed into a set of discrete peaks in g(τ) of Fig.3. The discrete peaks of g(τ) correspond to the Debye relaxation peaks of the characteristic mode frequencies. The dielectric strength Δε and relaxation frequencies ω of the three low frequency modes are obtained as a function of the field amplitude from the data of g(τ) and shown in Fig.4.

The so-called X-mode, corresponding to the director reorientational mode along the sample thickness X-direction, is observed to increase in both relaxation frequency and dielectric strength with increasing voltage of the probe field from 0.01 Vrms to 0.05 Vrms. This X-mode, however, was decreasing in its dielectric strength with increasing applied voltage above 0.05 Vrms and tending to vanish at higher applied voltages above 0.1 Vrms.

The domain mode as due to the domain reversal processes started to appear at 0.03 Vrms and increase in its dielectric strength in the input voltage range from 0.03 Vrms to 0.1 Vrms where the X-mode was observed to decrease with increasing input voltage.

Another low frequency relaxation peak was appearing at applied voltages from above 0.03 Vrms. This relaxation peak, attributed to a layer distortion of unknown origin and denoted as L-mode[7], was found to be most weakly dependent on the applied field in both its frequency and dielectric strength.

CONCLUSION AND SUMMARY

Three distinctive relaxation modes were identified from the relaxation time distribution function $g(\tau)$ obtained by the Tikhonov regularization analysis in good agreements with previous works[4-7] : the X-mode corresponding to the director relaxation along the thickness direction of the cell, the domain reversal mode, and the layer deformation mode. The X-mode was found to increase in the relaxation frequency but decrease in the dielectric strength with increasing strength of the applied probe field. The domain mode appeared only above a threshhold field and was increased in both frequency and dielectric strength with increasing field strength above the threshhold. The layer deformation mode was found to be weakly dependent on the probe field strength in both relaxation frequency and mode strength. In conclusion, the Tikhonov regularization analysis can be well applied to the low frequency dielectric relaxation spectra of the surface stabilized ferroelectric liquid crystal to determine the discrete mode structures in the time (τ) domain from the observed broad continuum spectra in the frequency (ω) domain.

Acknowledgments

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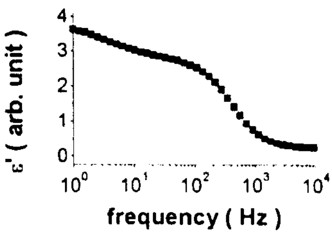
Note Added in Referee Response



A representative data set of ε'(ω) which were used in the simultaneous fit of ε'(ω) and ε"(ω) by the Tikhonov regularization to determine g(τ)

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