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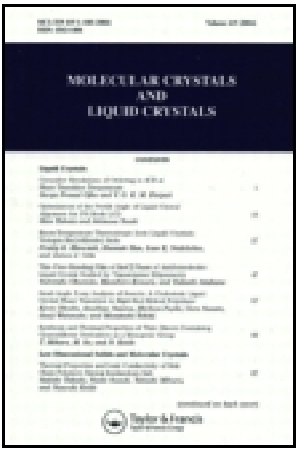

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Dielectric Anomaly in Ferroelectric Liquid Crystals with Polarization Inversion

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The dielectric anomaly in ferroelectric liquid crystals, showing polarization inversion, is observed as functions of temperature and external field. The dielectric constant shows an anomalous nonlinear behavior as a function of the electric. In describing the dielectric anomaly, the effective dipole moment, deduced from the field-dependence of the dielectric constant, is introduced into a model of a dynamically fluctuating mixture of at least two interconvertible conformers whose relative densities change with temperature and an external electric field.

Keywords: ferroelectric LC; dielectric anomaly; polarization inversion

INTRODUCTION

Since the discovery of ferroelectricity in tilted chiral smectic phase[1], ferroelectric liquid crystals (FLCs) has been studied extensively for the scientific understanding as well as device application. In most FLCs, due to the contribution of Goldstone mode corresponding to the collective director reorientations around the smectic cone, the dielectric constants are increasing through smectic A-smectic C* (S_mA-S_mC*) phase transition and have a broad maximum in a few degrees below S_mA-S_mC* phase transition temperature, T_c[2]. In this temperature regime, there is a peak of dielectric constant near zero field and the dielectric constant is reduced to a constant value over a certain critical field due to Goldstone mode suppressed.

Depending on the subtle change in the chemical structure, however, some chiral compounds have shown an anomalous behavior in a dielectric constant with polarization inversion in S_mC* phase. In such compounds, the dielectric constant has a local broad minimum value at a temperature T_i ($< T_c$), at which the spontaneous polarization vanishes, in the case of low dc bias field. Near T_i the dielectric constant increases with increasing an applied electric filed after suppressing the Goldstone mode by the electric field.

In this work, to describe this anomalous dielectric behavior we introduce the effective dipole moment obtained from the field dependence of the dielectric constant into a model of a dynamically fluctuating mixture of at least two interconvertible conformers whose relative densities change with temperature and external field.

EXPERIMENTALS

The chiral compound used in this work was s-2-methylbuthyl 4-n-nonanoyl-oxybiphenyl-4'-carboxylates (C9) synthesized by one of authors. It has the following phase sequence : isotropic→S_m A→(43.0 °C)→S_m C^*→crystalline. The sample cell was made of conductive indium-tin-oxide coated glasses with buffed polymer layers on the inner surfaces of the cell to promote homogeneous alignment. The cell thickness was maintained by using glass spacers of 3 μm. The dielectric constant was measured by impedance analyzer (HP 4192A). The cell mounted in a microfurnace (FP90) for temperature control, and temperature fluctuations were approximately 0.1 °C.

RESULTS AND DISCUSSION

Fig. 1 shows that the temperature dependence of dielectric constant ε for various dc bias fields at 500 Hz. The ε increases through S_m A-S_m C^* phase transition and reach the maximum value near T_c = 43 °C as usual FLCs. However, it gradually decreases with further decreasing temperature and has the local broad minimum value in case of low dc bias field (≤ 3 V/μm). Such anomaly has not been observed in other FLCs like SCE12 and DOBAMBC.

Using a model of a dynamically fluctuating mixture of two inter-convertible conformers, the dielectric constant ε was represented as $\varepsilon = \varepsilon_G + \varepsilon_I$, where ε_G and ε_I are the dielectric constant of the Goldstone mode and the induced dielectric constant due to the changing of molecular densities by the applied electric field, respectively.

The Goldstone mode is described in terms of spontaneous polarization P, tilt angle θ, and wavevector q as $\varepsilon_G = (1/K_3)\,(P/q\theta)^2$, where K_3 is twist elastic constant[2]. In our compound unlike normal FLCs, the polarization near T_i can be simplified as $P \propto (1/T - 1/T_i)$ introducing the model of dynamical fluctuation with two different molecular species[3] such as

$$P = \left[\frac{P_a}{1+\exp(-U/k_B T)} + \frac{P_b}{1+\exp(U/k_B T)}\right](T_c - T)^\alpha, \tag{1}$$

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where U, α, and k_B are an activation energy between two species, a critical exponent, and the Boltzmann constant, respectively. Here P_a and P_b denote the spontaneous polarizations of each species. Therefore, the Goldstone mode is directly expressed as $\varepsilon_G \propto (1/K_3)\{(PU/k_B q\theta)(1/T-1/T_i)\}^2$, which has the minimum value at T_i. From the experimental results for temperature dependences of the dielectric constant and the spontaneous polarization with polarization inversion, we obtained the following parameters using the least square fitting processes: $U = 0.020$ eV, $\alpha = 0.37$, and $T_i = (30.94 \pm 0.01)$ °C.

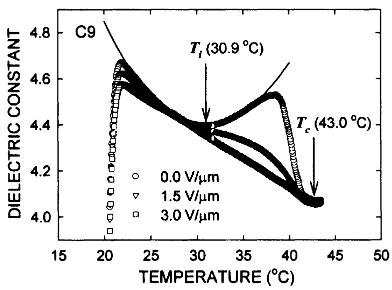

FIGURE 1 : The temperature dependences of dielectric constant ε for C9, measured for various dc bias fields at 500 Hz. The circular, rectangular, and triangular symbols represent E = 0 V/μm, 1.5 V/μm, and 3 V/μm, respectively.

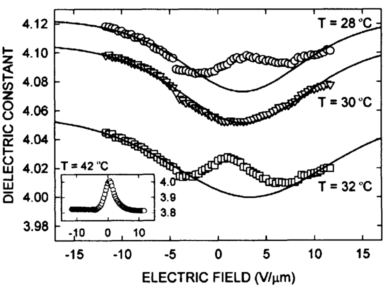

FIGURE 2 : The dielectric constant as a function of dc bias field at 500 Hz near T_i (31.0 °C). The solid lines are the results of the least square fits for each temperature. The graph in small box shows the normal behavior of dielectric constant near T_c (43.0 °C).

Fig. 2 shows the dielectric constant as a function of dc bias field near T_i, at which the spontaneous polarization vanishes. As shown in small box in the figure, the dielectric constant has the peak near zero field and is suppressed with a certain constant value in high temperature regime (near T_c). However, at T_i the peak disappears and near T_i the dielectric constant increases with increasing an applied electric filed after suppressing the Goldstone mode by the electric field.

The electric field applied to FLC molecules changes the density of the conformer as well as suppresses the Goldstone mode over a critical field. The change of the molecular density by an electric field produces the induced dielectric susceptibility and the interaction energy associated with the effective dipole moment μ_{eff} is described by $\Delta E = \mu_{eff}E$. Considering the

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effective dipole interaction energy ΔE as well as the activation energy U, the spontaneous polarization in FLC with two different molecular species is expressed by replacing U with $U+\Delta E$ in Eq. (1). When the electric field was applied over the critical field, the component of the Goldstone mode was suppressed and thus became the constant value with increasing field. Therefore, the induced dielectric constant is dominant with increasing field and is expressed as

$$\varepsilon_I = \frac{\mu_{eff}}{\varepsilon_0 k_B T}(T_c - T)^{\alpha} \frac{P_a - P_b}{\{\exp(-U'/2k_B T) + \exp(U'/2k_B T)\}^2} + \varepsilon', \tag{2}$$

where $U' = U+\Delta E$ and ε' is a constant.

Using the least square fitting processes for Eq. (2), we obtained the solid lines in Fig. 2 for each temperature. At inversion temperature T_i, the fitted result well explains the anomalous behavior. In the fitting procedure, we used the parameters achieved from the fitting results for the temperature dependence of the polarization. From Fig. 2, it was found that the effective dipole moment, μ_{eff}, was described by the simple power law as a function of the temperature.

CONCLUDING REMARKS

We have introduced the effective dipole moment into a model of a dynamically fluctuating mixture of at least two interconvertible conformers whose relative densities change with temperature and external field in FLCs with the polarization inversion. Within the framework of our model, we explained the anomalous behavior in dielectric permittivity in $S_m C^*$ phase with polarization inversion and found that the effective dipole moment can be described by the simple power law as a function for temperature.

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