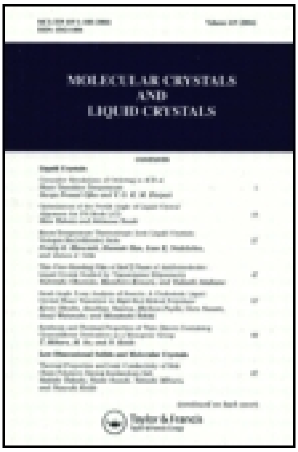

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Dielectric Properties of Nematic Liquid Crystals in Low Frequency Regime

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DIELECTRIC PROPERTIES OF NEMATIC LIQUID CRYSTALS IN LOW FREQUENCY REGIME

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Abstract Low-frequency dielectric properties of nematic liquid crystal (NLC) cells with and without polyimide alignment layers have been investigated with impedance spectroscopy. It is found that the dielectric behavior in the low frequency regime of NLC cells without polyimide alignment layers can be successfully explained in terms of the diffusive motion of impurity ions. The temperature dependences of the diffusion constant and the concentration of the impurity ions are determined from the dielectric behavior. NLC cells with polyimide alignment layers exhibit the interfacial polarization between the polyimide layers and NLC layer in the low frequency regime. The concentration of the impurity ions dissolved from polyimide layers to NLC layer is estimated.

INTRODUCTION

Dielectric properties of nematic liquid crystals (NLCs) have been extensively studied.[1] This is due to the fact that the dielectric anisotropy of NLCs is of fundamental importance in electro-optical application such as display devices and spatial light modulators. However, the dielectric properties in the low frequency regime have not been studied in detail. This is rather surprising, because charge transport of impurity ions in NLC and the complex impedance of NLC displays have been recently shown to affect strongly not only the electro-optical performance of actively addressed twisted NLC displays, but also the frequency dependences of the optical appearance of passively addressed twisted and supertwisted NLC displays.[2]

In this paper, we report the dielectric properties of NLC cells with and without polyimide alignment layers in the low frequency regime. The low-frequency dielectric properties contain much information concerning charge carrier transport as well as dielectric relaxation. From the dielectric properties we determine the diffusion constant and the concentration of the impurity ions. Influence of the polyimide layers on the concentration of the ions is also examined.

EXPERIMENT

The NLC used here was 4-cyano-4′-pentyl biphenyl (5CB). Polyimide layers were coated onto ITO glass substrates and were rubbed to align the NLC molecules homogeneously. The sample thickness was 6 μm. Samples without the polyimide layers were also prepared. The complex dielectric constants of the liquid crystal samples were obtained with a two-phase digital lockin amplifier interfaced with a computer for the data analysis and storage in the frequency range from 0.5 Hz to 200 kHz and in the temperature range from 298 K to 314 K. The measuring voltage was 20 mV_{pp}, which is well below the threshold voltage.

RESULTS AND DISCUSSION

NLC cells without polyimide layers

Figure 1 shows the frequency dependence of the real and imaginary parts of the complex dielectric constant ($\epsilon' - i\epsilon''$) of a 5CB cell without polyimide layer at 303 K. In the high frequency regime, a dielectric loss peak centered at about 10^5 Hz is observed. This loss peak exhibits the Debye-type relaxation and has been shown to be attributable to rotation around the molecular axis of NLC.[1] In the low frequency regime, ϵ' and ϵ'' vary as $f^{-1.5}$ and f^{-1}, respectively, where f is the frequency. The frequency dependence is due to the electrode polarization, and can be successfully explained in terms of the diffusive motion of impurity ions incorporated in the liquid crystal[3]:

$$\epsilon' = \frac{2nq^2D^{3/2}}{\epsilon_o\pi^{1/2}LkT}f^{-1/2} \tag{1}$$

and

$$\epsilon'' = \frac{2nq^2D}{\epsilon_o kT}f^{-1}, \tag{2}$$

where n is the ion concentration, q the electronic charge, D the diffusion constant of the ion, L the thickness of the cells, ϵ_o the free space permittivity, kT the thermal energy. We note that the diffusion constant and the concentration of the ion, which are important quantities for the characterization of the charge transport, can be determined by fitting these expressions to the experimental data in Fig. 1.

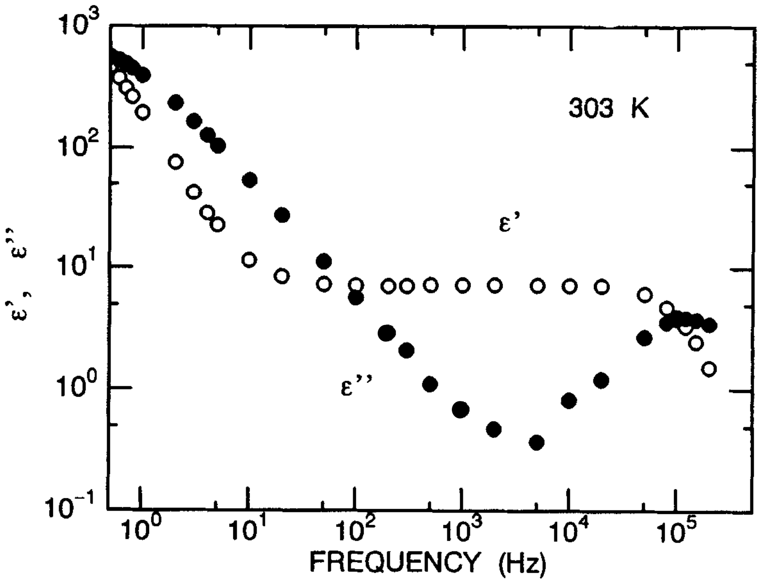

FIGURE 1 Frequency dependence of ϵ' and ϵ'' of a 5CB cell without polyimide layers at 303K.

Figure 2 shows the temperature dependence of the diffusion constant of the ion in 5CB. The discontinuity is seen at the clearing temperature. Such discontinuity has been observed in the temperature dependence of the drift mobility of a positive ion in 5CB, and furthermore the activation energies of the drift mobility are in agreement with those of the diffusion constant in Fig. 2 as expected from the Einstein relationship[4,5]: the activation energies of the diffusion constant are 0.37 eV in the nematic phase and 0.63 eV in the isotropic phase. Since we have found that the drift mobility of the positive ion in 5CB is much higher than that of a negative ion,[5] the diffusion constant and the concentration determined from Fig. 1 are of the positive ion. It is interesting to point out that the results in Fig. 2 are coincident with the temperature dependence of the diffusion constant of methyl red in 5CB measured with a forced Rayleigh scattering method.[6] This coincidence suggests that the ionic radius of the positive impurity ion in 5CB is almost equal to that of methyl red.

Figure 2 shows the temperature dependence of the concentration of the ion in 5CB as well. It should be noted that the ion concentration calculated from the low-frequency dielectric properties is equilibrium concentration. The ion concentration is almost constant

($\sim 4 \times 10^{13}$ cm^{-3}) in the nematic phase, and decreases with temperature in the isotropic phase. This behavior is not consistent with the prediction from the association-dissociation reaction; NLCs are regarded as weak electrolyte and hence the equilibrium ion concentration is expected to show thermally activated temperature dependence, $\exp(-U_o/kT)$, where U_o is the electrostatic binding energy of the ion pair.[7] We suggest that the adsorption of the ions onto the ITO electrode surfaces causes such temperature dependence of the ion concentration.

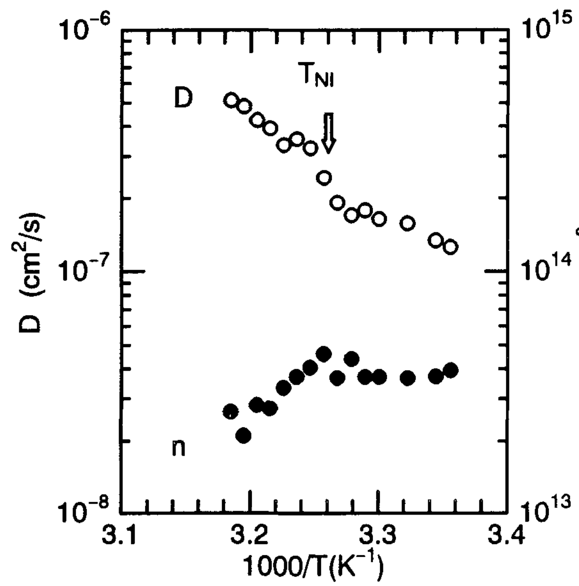

FIGURE 2 Temperature dependence of the diffusion constant, D, and the concentration, n, of the impurity ion in 5CB. T_{NI} is the clearing temperature.

NLC cells with polyimide layers

The frequency dependence of the real and imaginary parts of the complex dielectric constant of a 5CB cell with polyimide layer is shown in Fig. 3. Dielectric loss peaks at 10 Hz and 10^5 Hz are observed. The origin of the high-frequency peak is the same as that in Fig. 1. The Cole-Cole diagram shown in Fig. 4 reveals that the low-frequency loss peak is due to the Debye-type relaxation. The solid curve in the figure is calculated by assuming the interfacial polarization between 5CB and polyimide layers.[8] In the calculation, $\epsilon_{LC\perp} = 7.1$, $\sigma_{LC} = 9.2 \times 10^{-10}$ S/cm, and $\epsilon_{PI} = 3.4$ were used, where $\epsilon_{LC\perp}$ and σ_{LC} are the dielectric constant in the direction perpendicular to the long molecular axis and the conductivity of 5CB, respectively, and ϵ_{PI} is the dielectric constant of polyimide. The solid semicircle is in excellent agreement with the experimental data, demonstrating that the low-frequency dielectric relaxation is attributable to the interfacial polarization.

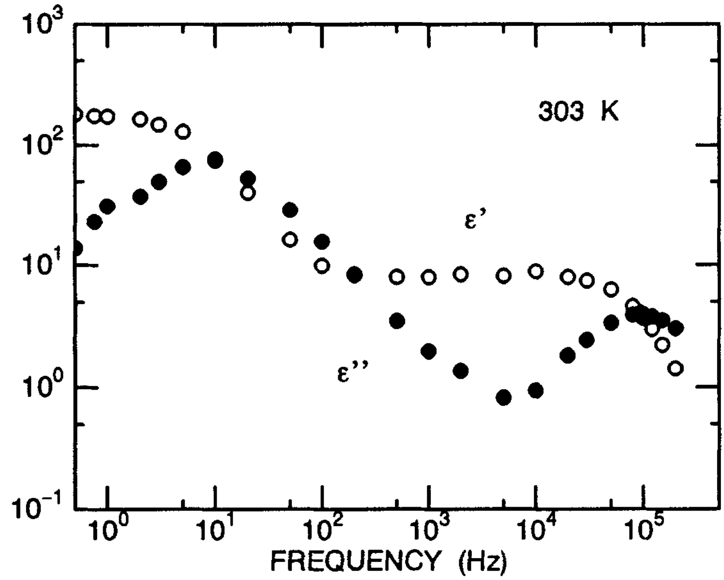

FIGURE 3 Frequency dependence of ϵ' and ϵ'' of a 5CB cell with polyimide layers at 303K.

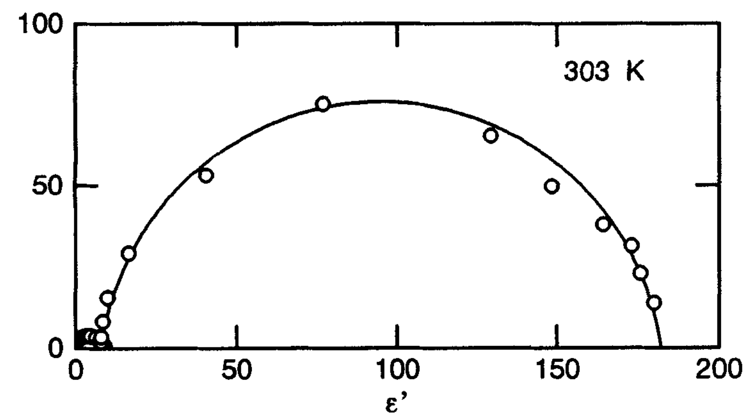

FIGURE 4 Cole-Cole plot for the relaxation at low frequency. The solid semicircle is calculated by assuming the interfacial polarization between 5CB and polyimide layers.

From the frequency at the dielectric loss peak, f_{peak}, the conductivity of the 5CB layer is determined from

$$f_{peak} = \frac{\sigma_{LC}}{2\pi(\epsilon_{LC\perp} + \epsilon_{PI} d_{LC}/d_{PI})}, \tag{3}$$

where d_{LC} and d_{PI} are the thickness of the 5CB layer and the polyimide layer, respectively. The measurement of σ_{LC} enables us to estimate the ion concentration in the 5CB layer using the drift-mobility data.[5] It is thus possible that by measuring f_{peak} of a 5CB cell with polyimide layer after the cell preparation, we estimate the temporal variation of the concentration of ions dissolved from the polyimide layers. The results are shown in Fig. 5. The ion concentration increases with time after the cell preparation and is much higher than that of the 5CB cells without the polyimide layers. The time constant for the increase in the ion concentration is 4000 h. Since the ion concentration of the 5CB cell without the polyimide layers is almost constant or slightly increases in the time range similar to Fig. 5, we conclude that considerable numbers of impurity ions ($\sim 3 \times 10^{16}$ cm^{-3}) are dissolved from the polyimide layers. The saturated concentration of the ion is independent of temperature in the nematic phase, which is similar to the results in Fig. 2.

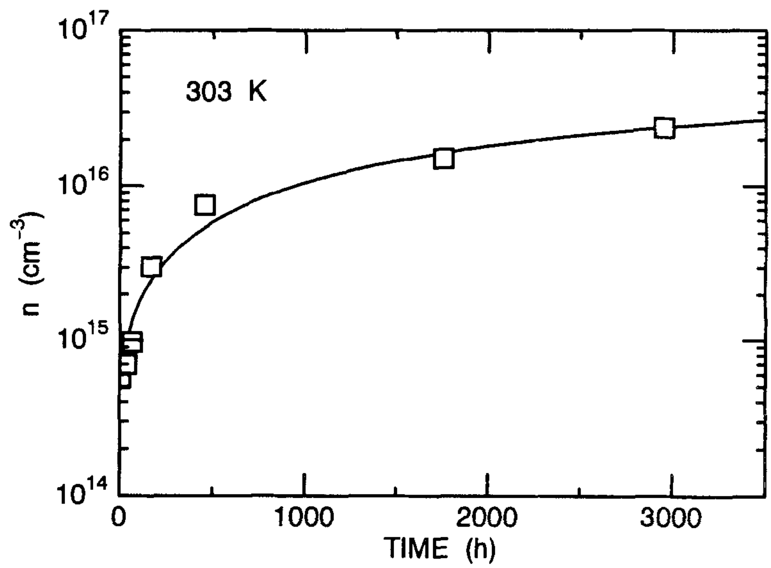

FIGURE 5 Temporal variation of the concentration of the impurity ion in a 5CB cell with polyimide layers after the cell preparation.

CONCLUSIONS

We have studied the low-frequency dielectric properties of 5CB cells with and without polyimide alignment layers. We find that the dielectric behavior in the low frequency regime of 5CB cells without polyimide alignment layers is due to the electrode polarization and can be successfully explained in terms of the diffusive motion of impurity ions. From the dielectric behavior, the temperature dependences of the equilibrium concentration and the diffusion constant of the positive ions are determined; the ion concentration is almost constant in the nematic phase and decreases with temperature in the isotropic phase, while the diffusion constant exhibits the thermally activated temperature dependence with the discontinuity at the clearing point. It is also found that the 5CB cells with polyimide alignment layers show the interfacial polarization between the polyimide and the 5CB layers in the low frequency regime. We estimate the temporal variation of the concentration of the impurity ions dissolved from the polyimide layers to the 5CB layer after the cell preparation. The ion concentration increases with the time constant of 4000 h, and the 5CB cells with polyimide layers have about 1000 times higher saturated concentration than the 5CB cells without polyimide layers.

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