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THE NONLINEAR DIELECTRIC EFFECT IN THE ISOTROPIC PHASE OF NEMATOGENS

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ABSTRACT Results are presented of studies of the nonlinear dielectric effect (NDE) for two frequencies (300 kHz and 6.1 MHz) of the weak measuring field in the isotropic phase of 4 heptyl 4' cyanobiphenyl (7CB). For the smaller frequency the NDE increases markedly towards positive values on approaching the clearing point. For the higher frequency the increase is negative. Despite this difference both pretransitional effects exhibit similar, mean - field temperature character. The observed behaviour coincide with relations between the relaxation time of prenematic fluctuation and measurement frequencies.

INTRODUCTION

Large number of experiments on the Kerr effect (KE), the Cotton - Mouton effect (CME) and the light scattering (I) confirmed the correctness of the Landau - de Gennes model in the isotropic phase of nematogens^{1,2}.

$$I, KE, CME = \frac{A^{I, KE, CME}}{T - T^*}, \quad T^* = T_c - \Delta T, \quad T > T_c \quad (1)$$

where: T^* is the temperature of the hypothetical, continuous phase transition, T_c and ΔT are the clearing temperature and the discontinuity of the isotropic - nematic transition, respectively. A denotes the amplitude for the I, KE and CME, respectively.

This result enabled the determination of certain properties of liquid crystalline phases still in the isotropic liquid. A deviation from the above, mean - field, description are observed in the immediate vicinity of the clearing point. Unfortunately, its relatively small value make a definitive explanation of its origins very difficult.^{2,3}

From recent observations on the nonlinear dielectric effect (NDE):⁴

$$NDE = \frac{\Delta \epsilon^E}{E^2} = \frac{\epsilon^E - \epsilon}{E^2} \quad (2)$$

where: $\varepsilon^E, \varepsilon$ denotes electric permittivity in the strong and weak electric field E , respectively.

interesting new aspects in this "old" field of research^{4,5,6,7} seems to be opened up. New possibilities introduced by the NDE are associated with the fact that the radio - frequency of the weak, measuring electric field coincide with relaxation times of pre-nematic fluctuations.

This conclusion is supported by results presented in this paper of measurements in 4 heptyl 4' cyanobiphenyl (7CB) in an as yet untested span of frequencies: 300 kHz - 6.1 MHz.

EXPERIMENTAL

The NDE measurement apparatus for a frequency of about 6 MHz has been described in paper⁸. The apparatus for $f_m^{(1)} = 300$ KHz is based essentially on the same idea but certain important new problems necessitated the application of strong, steady measurement field in the form of rectangular pulses having time of duration not appreciably exceeding 10^{-2} s (details will be published).

Such short pulses make it possible to avoid problems with hydrodynamic motions in the sample.⁷ In our studies the duration of the pulse of E was 2 ms for $f_m^{(2)} = 6.1$ MHz and 8 ms for $f_m^{(1)} = 300$ kHz. The sample was placed in a flat parallel capacitor with gap 0.33 mm. The steady electric field voltage applied was in the range 300 V - 1000 V. Changes of the electric permittivity $\Delta\varepsilon^E$ induced by the strong electric field was always proportional to E^2 , with error less than 2 %. The temperature was stabilised with an accuracy of ± 0.01 K.

The sample of 7 CB was obtained from the BDH-Merck Company and had $T_C = 42.8^\circ\text{C}$ and a very small conductivity hence it could be used without any additional purification.

RESULTS AND DISCUSSION

On Figure 1 results of the NDE measurements in the isotropic phase of 7 CB are presented. For the measurement frequency $f_m^{(1)} = 300$ kHz the effect is always positive and increases markedly on approaching T_C . The sign of the NDE is in agreement with well confirmed experimental predictions that fluctuations associated with nonhomogeneities of dielectric properties of the medium should always give a positive contribution to the NDE^{4,9}

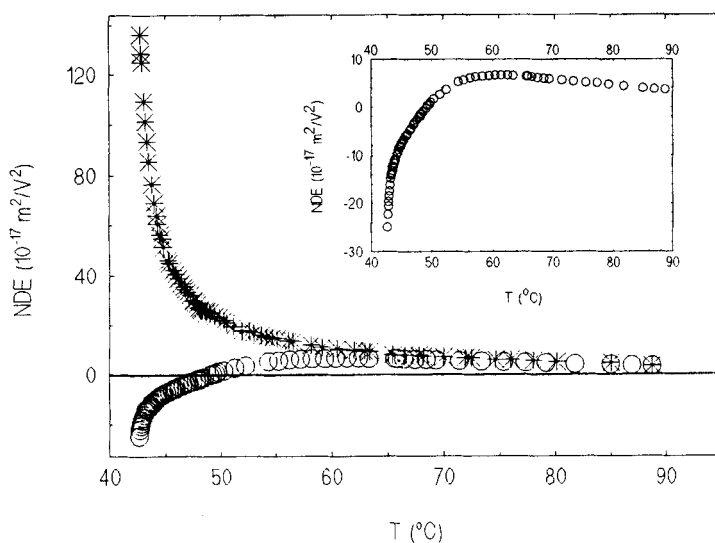


Figure 1 Results of NDE measurements for two frequencies of the weak electric field: $f_m^{(1)} = 300\text{kHz}$ (crosses) and $f_m^{(2)} = 6.1\text{MHz}$ (circles).

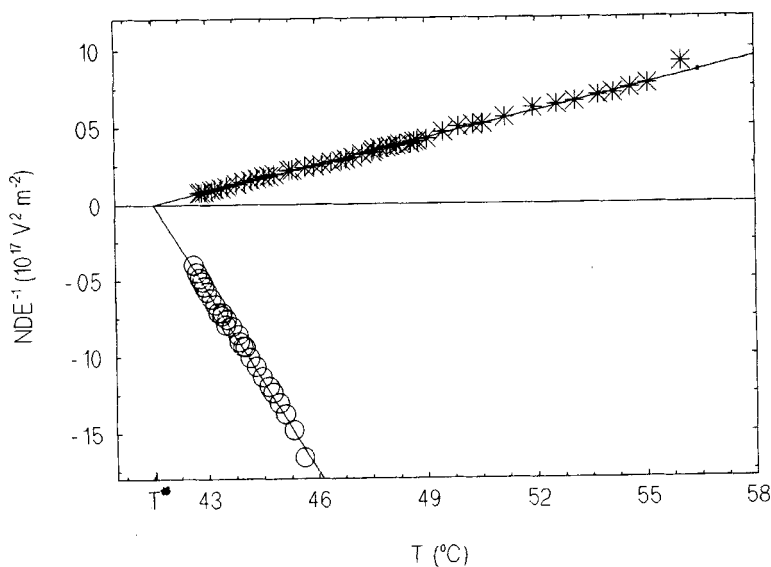


Figure 2 The reciprocal of the NDE for $f_m^{(1)}$ and $f_m^{(2)}$. The extrapolation beyond T_c determine $T^* = 41.5_4 \pm 0.1^\circ\text{C}$ and $\Delta T \approx 1.3^\circ\text{C}$.

For the frequency $f_m^{(2)} = 6.1 \text{ MHz}$, on approaching T_c the NDE changes its sign to the negative one and increases markedly towards negative values.

For $T - T_c > 30 \text{ K}$ the NDE for both measurement frequencies coincide.

Simple analysis as presented on Figure 2 shows that for both frequencies pretransitional increases can be described by the same mean - field relation:

$$NDE \propto \frac{1}{T - T^*}, \quad (3)$$

Within the limit of experimental error values of T^* and ΔT are the same for both frequencies and in agreement with previous results obtained in studies on the Kerr effect^{2,3}.

Comparing the evaluation of relaxation times of prenematic fluctuations (τ_r) on approaching the clearing point¹⁰ with angular frequencies of the applied, weak measuring fields it may be concluded that :

- far from T_c , where the NDE for both frequencies coincide the condition:

$$\omega_m^{(1,2)} \tau_r \ll 1 \quad \text{is fulfilled}$$

- for the smaller measurement frequency the condition: $\omega_m^{(1)} \tau_r < 1$ is valid up to T_c

- for the greater measurement frequency, in the region where the LdG - like description is valid: $\omega_m^{(1,2)} \tau_r \gg 1$

CONCLUSIONS

Results presented show that the relaxation of critical fluctuations have a crucial influence on the behaviour of the nonlinear dielectric effect in the isotropic phase of nematogens.

These results raise questions on the different signs of contributions from pretransitional, prenematic, fluctuations for $\omega_m \ll \tau_r^{-1}$ and $\omega_m \gg \tau_r^{-1}$. This difference appears to be related only to the signs and values of amplitudes in the expression of the type proposed by the Landau - de Gennes model (Figure 2).

Probably yet another factor is of importance in understanding the properties of by the NDE in the paranematic region. In MBBA and EBBA, where the dipole moment is almost perpendicular to the main axis of the rod - like molecule for, $f_m^{(2)} \approx 6.1 \text{ MHz}$ the NDE is always positive and increases markedly towards positive values on approaching T_c . In paper¹¹ was been shown that this effect can be qualitatively described after substituting the molecular polarisability instead the anisotropy of the electric permittivity in the expression derived for the NDE from the Landau - de Gennes model.

For 7CB tested in this paper the dipole moment is parallel to the main axis of the rod-like molecule.

In the opinion of the present authors an important role in understanding the NDE in the isotropic phase of nematogens could be the model proposed by Kielich, which take into account all factors of importance^{12,13}:

$$\frac{\Delta\epsilon}{E^2}(\omega, \tau) = f(R_k, m^2 / \gamma, T)$$

where: $R_k = (1 + j\omega\tau_k)^{-1}$, m is the dipole moment, γ is anisotropy of polarisability.

This relation includes lead to the change of the sign of the NDE in the dispersion region as well as its dependence from factor which seems to be important here.

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