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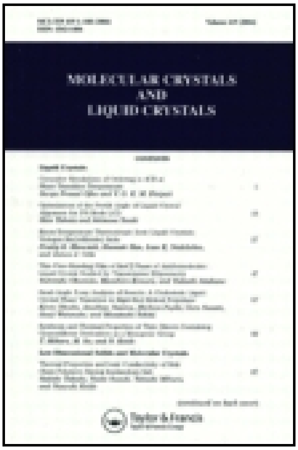

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NONLINEAR DIELECTRIC EFFECT IN ISOTROPIC POLAR NEMATOGENS

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Abstract The Nonlinear Dielectric Effect is measured in the isotropic phase of four nematogens. The NDE does not follow the classical $1/(T-T^*)$ behavior, due to noncritical contributions as well as to precritical slowing down. Novel non-mean-field type pretransitional contributions are observed in strongly polar nematogens.

INTRODUCTION

In isotropic nematogens the orientation of molecules is correlated on a mezoscale distance. Because of the coupling of the electric field and the dielectric anisotropy of the ordered subvolumes strong electrically induced effects arise in the medium, especially near the spinodal temperature T^*, where the correlation length rapidly increases[1] . The pretransitional birefringence[1,2] (the electrooptical Kerr effect - EKE) is well known, while the electric permittivity changes[3,4] (the nonlinear dielectric effect - NDE) are almost unknown. The basic difference between both these effects lies in the measuring frequency, which is of optical or kHz-MHz range, respectively. In consequence, the term related to the orientation of permanent dipoles dominates in the NDE[5] and quite strong pretransitional NDEs are expected in strongly polar nematogens. In this report we present the NDE study of such materials.

The NDE method is unique in that it provides the four-molecule orientational correlation factor g_4[5] and completes in this way information obtainable from the measurements of the electric permittivity, light

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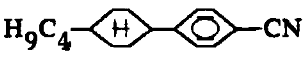
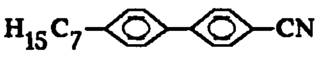
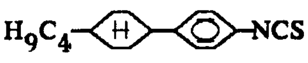
scattering and EKE in the paranematic phase.

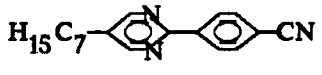
EXPERIMENTAL AND RESULTS

We studied nematogens with electric permittivity anisotropy ranging from ca. 10 (BCPIT) to 35 (HCPP). Their chemical formulas and acronims are:


(BCBN)


(HCBP)


(BCPIT)


(HCPP)

The short (1ms) high voltage pulses were applied to the samples to induce electric permittivity changes free of parasitic effects connected with flow of ionic impurities. Signals of the changes $\varepsilon_0\delta\varepsilon$ were registered by the oscilloscope and scaled with the precise reference capacitor connected in paralell to the thermostated capacitor filled with the liquid to be examined. The NDE constant, $D = \delta\varepsilon E^{-2}$, did not depend on the field intensity E up

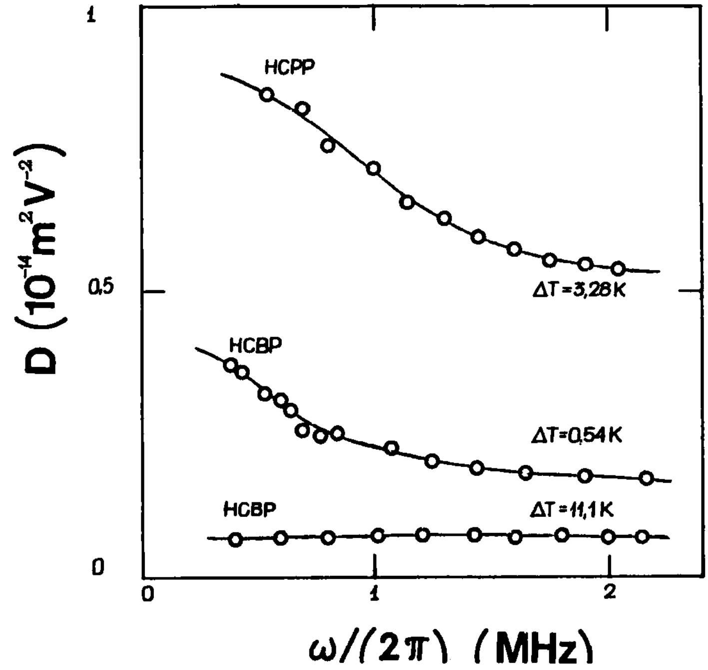


FIGURE 1 Frequency variation of the NDE constant.

to 30 kV/cm. The remarkable frequency dispersion in the pretransitional domain was observed, as it is shown in Fig.1.

The NDE vs. temperature, studied at the frequency of 1.5 MHz, where sensitivity of the experimental setup was maximal, is presented below. From Fig.2 it is seen that the NDE of the isotropic phase increases rapidly in the vicinity of the nematic region and pretransitional effects occur even 50K from the phase transition temperature.

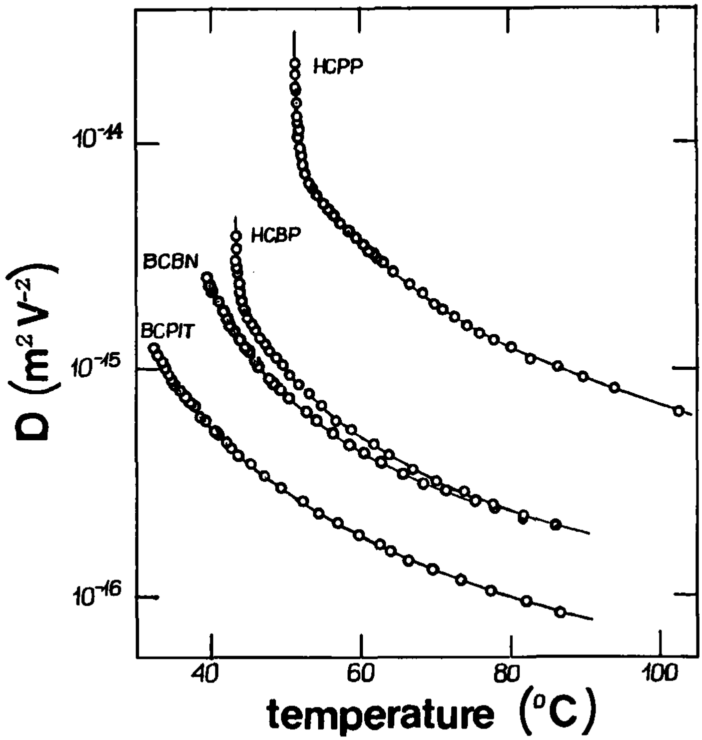


FIGURE 2 Temperature variation of the NDE constant.

DISCUSSION

The 1/(T-T*) type behavior of the NDE constant D in the paranematic phase, following from the Landau-De Gennes theory[1], is not confirmed by the standard D^{-1} vs. T plot which is curvilinear for all examined compounds. Neither is the classical behavior confirmed by the more informative plot of the apparent critical amplitude Dt vs. t ($t=T/T^*-1$). The amplitude should be constant for the phenomenological De Gennes model or vary slightly in

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the molecular Maier-Meier approach. In fact, a distinct and complex variation of Dt is observed (see Fig.3).

In order to interpret Fig.3 we assume that two factors are responsible for the deviations from the mean-field type behavior. One, that slightly decreases the apparent amplitude with increasing temperature far from the N-I transition point, is the noncritical contribution probably related to the nonlinearity of the mean electric permittivity. The second one, that decreases the effect near the transition to the nematic phase (up to 10 K), is the result of the critical slowing down and NDE dispersion in the pretransitional domain, as our tentative frequency measurements seem to indicate.

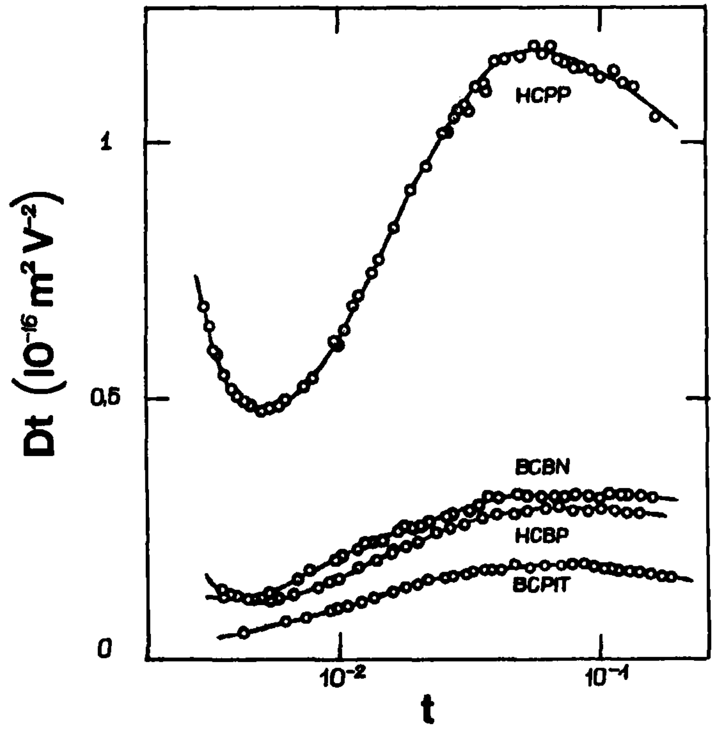

FIGURE 3 Dt vs. reduced distance from T^*.

In Fig.3 additional non-mean-field increments are seen in the close vicinity (below 1K) of the I→N phase transition. Although their origin is not clear as yet, they are related to the dielectric anisotropy, and so the non-mean-field increments are the greatest for HCPP, much smaller for HCBP or even unobservable for BCBN and BCPIT, this being the sequence of decreasing anisotropy of electric permittivity.

The non-mean-field type increments, which are of interest from the theoretical point of view, have been only barely observed previously by the Kerr and Cotton-Mouton techniques[6-8]. In contrast, the pretransitional NDE contributions in HCPP are sufficiently high to be analysed numerically. By fitting the data to the empirical equation:

$$D = A_1\, t^{-1} + A_2\, t^{-\Lambda} + A_3\,, \qquad (1)$$

where the form of the amplitude of the mean-field term:

$$A_1 = A_0\, (1 + \omega^2\tau^2)^{-1}\,, \qquad \tau = \tau_0 t^{-1} \qquad (2)$$

reflects the dispersion of the electric permittivity and the critical slowing down, we found the apparent exponent $\Lambda = 2.5 \pm 0.5$ from the attainable temperature range $t > 3\times10^{-3}$. From the fitted amplitude $A_0 = 1.3\times10^{-16}\ m^2V^{-2}$ the free energy density and the viscosity coefficients are found: $a = a_0/T^* = 0.17\times10^6\ Jm^{-3}K^{-1}$ and $\eta = \tau_0 a_0 = 6.7\times10^{-2}\ kgm^{-1}s^{-1}$. These values are almost the same as those determined from the birefringence studies for PCB [9] which has a molecular structure similar to HCPP.

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