Critical behavior of dielectric permittivity in the isotropic phase of nematogens

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It has been shown that the temperature behavior of dielectric permittivity (ε) in the isotropic phase of nematogens can be described in the same way as in critical binary solutions. Hence, using a relation with the critical exponent $\phi=1-\alpha=0.5\pm0.03$ it was possible to portray the results of $\varepsilon(T)$ measurements in the isotropic phase of 5-heptyl-2-(4′-cyanobiphenyl)-pyrimidine and 4,4-n-octylcyanobiphenyl. The influence of the position of the permanent dipole moment on the results was tested by additional measurements in n-(p-methoxybenzylidene)-p'-butylaniline. It also has been shown that a fluidlike analogy can be applied to the nonlinear dielectric effect (NDE), which describes changes of dielectric permittivity induced by a strong electric field. Measurements were conducted for the lowest frequency used in NDE studies ($f=67$ kHz), so the condition $\tau f \ll 1$ (with τ the relaxation time) was always fulfilled. Values of discontinuities of the isotropic-nematic phase transition from both the analysis of $\varepsilon(T)$ measurements and NDE studies are in good agreement. [S1063-651X(96)06812-2]

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I. INTRODUCTION

The properties of the isotropic-nematic (I-N) phase transition has attracted considerable attention recently [1–14]. Some basic experimental facts were obtained from measurements of physical properties that are known for their particular sensitivity to fluctuations, conducted in the isotropic phase. For instance, the Kerr effect (KE), the light scattering (I), the Cotton-Mouton effect (CME) (see [5–8] and references therein), and the nonlinear dielectric effect (NDE) [9,10] were found to follow the same pretransitional behavior

$$\mathcal{E}_I,\mathcal{E}_{\mathrm{CME}},\mathcal{E}_{\mathrm{KE}},\mathcal{E}_{\mathrm{NDE}}=\frac{A}{(T-T^*)^{\gamma}}\quad \text{with}\quad \gamma=1,$$

$$T^*=T_{I\text{-}N}-\Delta T,\quad T>T_{I\text{-}N},\tag{1}$$

where $\mathcal{E}$ is a value of I, CME, KE, and NDE, respectively. T^* denotes the extrapolated temperature of a hypothetical, continuous phase transition, $T_{I\text{-}N}$ is the isotropic-nematic phase transition temperature (the clearing temperature), ΔT denotes the measure of the discontinuity of the phase transition, and A is the amplitude for the I, CME, KE, or NDE, respectively. Singularities of behavior of the electro-optic KE (EKE), NDE, CME, and I in the domain of phase transition may be described using the model proposed by de Gennes [the Landau–de Gennes (LdG) model] [5]. The experimental values of ΔT are usually in the range 0.5–2 K. These are much smaller than the ones theoretically predicted in the Maier-Saupe [3,11,12] or LdG mean-field [3,12,13] models. Recently, Mukherjee *et al.* [2–4] employed the equation of state near a coexistence curve where the I-N phase transition eventually falls, to obtain $\Delta T\approx3$ K.

It is believed that dielectric permittivity (ε) does not exhibit the pretransitional, "critical-like" behavior in close vicinity to the phase transition point I-N [6,7,15,16]. This seems unquestionable in nematogens with nonpolar molecules or with polar molecules where the permanent dipole moment is perpendicular to their long axis. In such cases $\varepsilon(T)$ is a linear function of temperature and $d\varepsilon/dT\leqslant0$ [6,16]. For nematogens with the dipole moment parallel to the long axis of the molecule, Bradshaw and Raynes [16] found, on approaching $T_{I\text{-}N}$, the opposite trend $d\varepsilon/dT>0$.

To the best of our knowledge there is no analytical portrayal of this anomalous, pretransitional $\varepsilon(T)$ behavior. Such an attempt is presented in this paper. It is based on experimental studies of linear and nonlinear components of the dielectric permittivity ($\varepsilon,\mathcal{E}_{\mathrm{NDE}}$) in the isotropic phase of three nematogens: octylcyanobiphenyl (8CB) [5–8,16,17], 5-heptyl-2-(4′-cyanobiphenyl)-pyrimidine (HCPP) [14,18] (with the parallel dipole moment), and p-methoxy-benzlidene-p'-n-butylaniline (MBBA) [5–8,19] (with the perpendicular dipole moment). A considerable value of the anisotropy of dielectric permittivity in HCPP ($\Delta\varepsilon^0\approx35$) [14,18] allows one to expect a spectacular pretransitional behavior. The investigations presented in this paper make use of the idea mentioned above of treating T^* as a fluidlike, near-critical point [2–4]. To minimize the possible detrimental influence of relaxation processes, investigations of the nonlinear dielectric effect [$\mathcal{E}_{\mathrm{NDE}}=(\varepsilon^E-\varepsilon)/E^2$, where ε and ε^E denote dielectric permittivities in a weak and a strong electric field E, respectively] [20] were conducted with the lowest (as far as we know) measurement frequencies ever applied [21,22].

II. EXPERIMENT

Tests were performed in the isotropic phase of two nematogens with the dipole moments parallel to the long axis of the molecule HCPP ($\Delta\varepsilon^0\approx35$, $T_{I\text{-}N}=51.3$ °C) [14,18], prepared by Wiesław Pyżuk from Warsaw University, and 8CB ($\Delta\varepsilon^0\approx10$, $T_{I\text{-}N}=39.6$ °C) [5–7,17,23–25], purchased from BDH-Merck. They were supplemented by measurements in MBBA ($\Delta\varepsilon^0\approx-0.5$, $T_{I\text{-}N}=43.6$ °C), from Aldrich Chemicals [5–7,9,10,19,27], for which the dipole moment is approximately perpendicular to the long axis of the molecule.

All samples were degassed immediately prior to each measurement.

The nonlinear dielectric effect [20] measurements were performed using an apparatus based on the frequency modulation of an LC generator, described in detail in our previous paper [21]. In the research presented we applied frequencies of a weak measurement field much lower than those ever applied in NDE studies [21,22]: $f=67$ kHz (HCPP and 8CB) and $f=250$ kHz (MBBA). This made it possible to avoid the influence of the coincidence between the measurement radio frequency (f) and the relaxation time (τ) of pretransitional relaxation processes near $T_{I\text{-}N}$ [21,22]. Thus the condition

$$\frac{f^{-1}}{\tau} \gg 1 \tag{2}$$

was fulfilled up to the clearing point.

To minimize the influence of heating or hydrodynamic motions [5] the strong electric field was applied in the form of rectangular pulses of duration $\tau_D \approx 4$ ms. The voltage of the weak measurement field was 5 V and for the strong, steady electric field (E) it ranges as follows: $U_E=50$–900 V in HCPP, 300–1000 V in 8CB, and 500–1100 V in MBBA. The values decreased on approaching $T_{I\text{-}N}$ so that the registered changes of the capacitance $\Delta C(E)$ were always in the range 5–30 fF. The samples were placed in a flat-parallel capacitor made of Invar [28] with gaps $d=1$ mm ($C_0 \approx 4$ pF) for HCPP and 8CB and 0.3 mm ($C_0 \approx 9.3$ pF) for MBBA. At each temperature distance from $T_{I\text{-}N}$ the condition $(\varepsilon^E - \varepsilon) \propto E^2$ was always satisfied with an error less than 1%.

The dielectric constant was measured using the same capacitor and SOLARTRON 1260A impedance analyzer, with five-digit resolution. Tests conducted for $f=10$ kHz, 100 kHz, and 1 MHz did not show any influence of this frequency shift on results. This agrees with previous studies in the isotropic phase in 8CB [6,15] and MBBA [6], which clearly showed that these frequencies are beyond the dispersion region. Results presented below are for $f=100$ kHz. The temperature of the capacitor, measured by a platinum resistor (A1 class, DIN 43 760) and Keithley 195 multimeter with a resolution of 0.005 K, was stabilized by means of a double-stage water thermostat system with an accuracy 0.01 K/h. Fits were done by means of the ORIGIN 3.5 software (Microcal Inc.). All errors are given as three standard deviations.

III. RESULTS AND DISCUSSION

Figure 1 shows reciprocals of measured low-frequency $\mathcal{E}_{\text{NDE}}$ values for HCPP, 8CB, and MBBA in a normalized scale, relative to the amplitude A_{NDE} [relation (1)], corresponding to the given liquid-crystalline material. It is clearly seen that for each case the same type of pretransitional behavior occurs. It is noteworthy that the range of validity of relation (1) (about 50 K) is much larger than the one found previously in NDE, KE, CME, or I studies (5–15 K) [5–10,14,17,19,22–27]. A very sensitive apparent scale analysis, presented in the inset, additionally confirms the validity of relation (1), with no distortions near $T_{I\text{-}N}$. It also makes it easier to compare values A_{NDE} and ΔT obtained for tested nematogens. The values of the discontinuity of the phase transition, 0.7 K for HCPP and 1.6 K for 8CB, are, within the limit of experimental error, in agreement with the ones previously obtained [14,17,23–25]. For MBBA the value of ΔT (≈ 2 K) is greater than the one found from I, KE, or CME studies (0.7–1 K) [9,10,19,26,27]. It is noteworthy that NDE investigations of MBBA performed for $f=6$ MHz also gave $\Delta T \approx 0.7$ K [9,10], but with a significant disagreement with the amplitude A_{NDE} derived from the LdG model [10]:

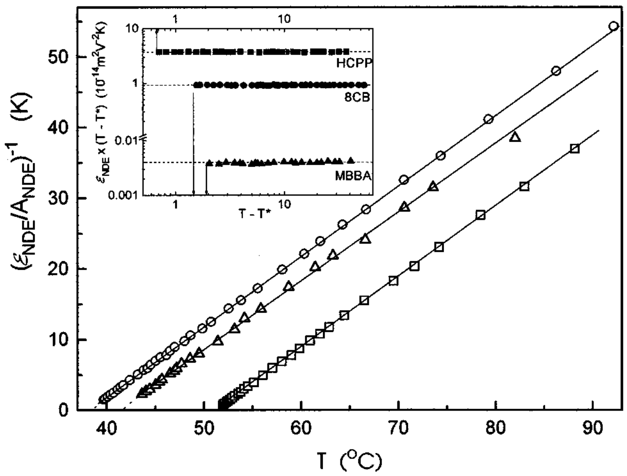

FIG. 1. Reciprocals of the measured low-frequency NDEs in the isotropic phase of HCPP (□), 8CB (○), and MBBA (△). Data are normalized by the amplitude A_{NDE} appropriate for the given liquid-crystalline material. The inset enables a comparison of values of A_{NDE} and ΔT and shows the lack of distortions from the classical behavior [relation (1)].

$$\mathcal{E}_{\text{NDE}} = \frac{A_{\text{NDE}}}{T-T^*} = \frac{2}{3a}\, \varepsilon_0\, \frac{\Delta\varepsilon^0 \Delta\varepsilon^f}{T-T^*}, \tag{3}$$

where $\Delta\varepsilon^f, \Delta\varepsilon^0$ are anisotropies of dielectric permittivity for the measurement frequency and in the zero-frequency limit, respectively. The coefficient a denotes the constant amplitude of the second-rank term in the LdG series [6]. In the presented studies it was assumed that $\Delta\varepsilon^f \approx \Delta\varepsilon^0$. Based on the results presented in Table I and on values of $\Delta\varepsilon^0$ mentioned above, the value of an important phenomenological coefficient a was found to be $a \approx 0.089$ J cm^{-3} K^{-1} (0.09 J cm^{-3} K^{-1} in Ref. [23]) for 8CB and $a \approx 0.055$ J cm^{-3} K^{-1} (0.056 J cm^{-3} K^{-1} in Ref. [27]) for MBBA. The agreement between obtained values and reference data from KE measurements confirms the quantitative validity of relation (3) for the low-frequency NDE. For yet untested HCPP one can estimate $a \approx 0.19$ J cm^{-3} K^{-1}.

Figure 2 presents results of measurements of dielectric permittivity in the isotropic phase of HCPP. It exhibits the pretransitional behavior on approaching the clearing point as mentioned in the Introduction [16]. It appears that in the temperature range $T_{I\text{-}N}$–$T_{I\text{-}N}+50$ K dielectric permittivity can be described by the relation

$$\varepsilon(T-T^*) = \varepsilon^* + a^*(T-T^*) + A^*(T-T^*)^{\phi}$$
$$\text{with } \phi = 0.5 \pm 0.02, \quad T > T_{I\text{-}N}, \tag{4}$$

TABLE I. Values of parameters describing the pretransitional behavior of dielectric permittivity [relation (4)] and the nonlinear dielectric effect [relation (3)] in the isotropic phase of tested liquid-crystalline materials.

	$\varepsilon_{\mathrm{NDE}}=\dfrac{A_{\mathrm{NDE}}}{T-T^*}$		$\varepsilon=\varepsilon^*+a^*(T-T^*)+A^*(T-T^*)^{\phi}$				
Liquid-crystalline materials	A_{NDE} (10^{-14} $\mathrm{m^2\,V^{-2}}$) (χ_ν^2)	T^* (°C) ΔT	ε^* (χ_ν^2)	a^* ($\mathrm{K^{-1}}$)	A^* ($\mathrm{K^{-1}}$)	ϕ	T^* (°C) ΔT
HCPP	$3.72_{\pm 0.02}$ (1.1)	$51.09_{\pm 0.02}$ $0.68_{\pm 0.03}$	$18.04_{\pm 0.02}$ (1.2)	$-0.043_{\pm 0.005}$	$0.265_{\pm 0.01}$	$0.503_{\pm 0.02}$	$51.07_{\pm 0.04}$ $0.7_{\pm 0.05}$
8CB	$0.93_{\pm 0.01}$ (1.3)	$38.15_{\pm 0.02}$ $1.55_{\pm 0.03}$	$10.29_{\pm 0.02}$ (1.3)	$-0.37_{\pm 0.005}$	$0.260_{\pm 0.01}$	$0.498_{\pm 0.02}$	$38.65_{\pm 0.04}$ $1.6_{\pm 0.05}$
MBBA	$3.9\times 10^{-3}_{\pm 0.02}$ (1.5)	$41.6_{\pm 0.06}$ $1.9_{\pm 0.1}$					

where ε^* is the value of ε at $T=T^*$ and a^* and A^* are amplitudes. Values of the parameters are given in Table I. The form of this relation is analogous to that applied in the homogeneous phase of critical, binary solutions [29,30]:

$$\varepsilon(T-T_{\mathrm{crit}})=\varepsilon_{\mathrm{crit}}+a(T-T_{\mathrm{crit}})+A(T-T_{\mathrm{crit}})^{1-\alpha} \quad \text{with } 1-\alpha\approx 0.88, \tag{5}$$

where $\alpha\approx 0.12$ is the critical exponent of the specific heat for the three-dimensional Ising ($d=3$, $n=1$) universality class [8] and $\varepsilon_{\mathrm{crit}}$ is the value of dielectric permittivity at the critical consolute temperature T_{crit}.

Results of studies in 8CB presented in Fig. 3 and in Table I further confirm the validity of relation (4). For both HCPP and 8CB the same value of the exponent $\phi\approx 0.5$ was found. The values of ΔT obtained from $\varepsilon(T)$ fits using relation (4) and from NDE measurements presented above are in a good agreement. Indeed, the deviation of the experimental data from the fitted function does not exceed $\pm 2.8\times 10^{-4}$ up to $T_{I\text{-}N}+50$ K.

The dielectric permittivity anomalously decreases both on approaching the clearing point ($A^*>0$) and the critical consolute point ($A>0$) (see [30] and references therein). However, the manifestation of the pretransitional behavior is definitely much weaker on approaching the critical consolute point than for the I-N transition. It is also noteworthy that in critical solutions, apart from the immediate vicinity of T_{crit}, additional correction-to-scaling terms should be taken into account [29,30].

The inset in Fig. 3 shows the behavior of $\varepsilon(T)$ in the isotropic phase of MBBA, a nematogen with the dipole moment perpendicular to the main axis of the molecule. Within the limit of experimental error, the dielectric constant remains a linear function of temperature up to $T_{I\text{-}N}$. This is in agreement with the results of previous experimental studies in liquid-crystalline materials of the same type [6,16].

IV. CONCLUSION

Results presented above suggest that dielectric permittivity on approaching the I-N phase transition may follow the same pattern as in binary solutions on approaching the critical consolute point. A comparison of relations (4) and (5) gives the value of the exponent $\phi=1-\alpha$ for the isotropic-

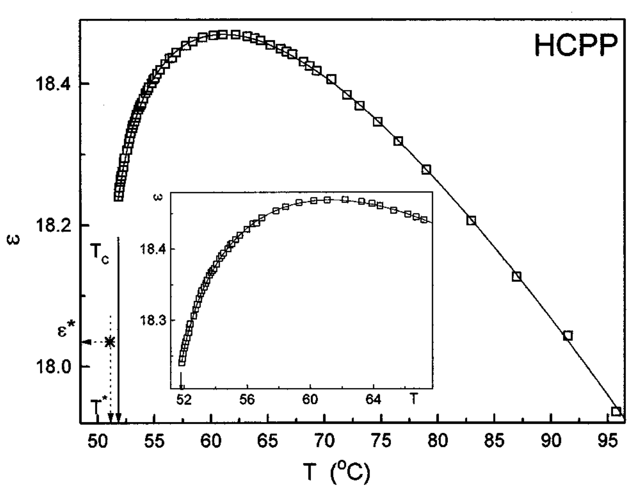

FIG. 2. Behavior of dielectric permittivity ($f=100$ kHz) in the isotropic phase of HCPP. The positions of the clearing point (T_{I-N}) and the fitted point (T^*,ε^*) of the hypothetical continuous phase transition are also shown.

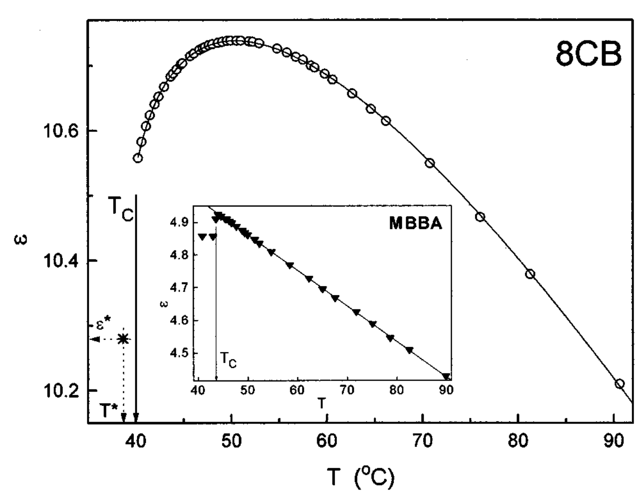

FIG. 3. Behavior of dielectric permittivity ($f=100$ kHz) in the isotropic phase of 8CB. The positions of the clearing point ($T_{I\text{-}N}$) and the fitted point (T^*,ε^*) are also shown. The inset shows the behavior of $\varepsilon(T)$ in the isotropic phase of MBBA.

nematic phase transition and consequently $\alpha=0.5\pm0.03$. This value is beyond the mean-field approximation and can be obtained from the Ornstein-Zernike approximation [8] or from the Gaussian model [31] that assumes the existence of weakly interacting or non interacting fluctuations. The appearance of precritical anomalies associated with exponent α was shown previously in the specific-heat studies in 8CB [8,32] and MBBA [8,33]. The weakness of the anomalies obtained and the limited region of their appearance ($T-T_{I\text{-}N}<2$ K) made the analysis very difficult; nevertheless, the value of $\alpha\approx0.5$ was considered as the most reliable. In Ref. [33] it was also pointed out that $\alpha\approx0.5$ leads to the fulfillment of the Josephson's scaling law with $d=3$.

It is noteworthy that the same dimensionality can be obtained from the Gaussian model [31], where $\alpha=(4-d)/2$ and consequently $\alpha=0.5$ for $d=3$. Additionally, the approximation adopted in the derivation of relations (1) and (3) makes it valid for this simple model.

The decrease of dielectric permittivity in 8CB and HCPP ($A^*>0$) near $T_{I\text{-}N}$ reflects the cancellation of the contribution to ε coming from the antiparallel ordered permanent dipole moments of molecules contained in pretransitional fluctuations. This mechanism is absent in MBBA, where the dipole moment is perpendicular to the long axis of the molecule (Fig. 3) [6,16,34].

It seems that the analogy with critical solutions can be extended also to the nonlinear dielectric effect or (EKE). In Refs. [21,35,36] it was shown that on approaching the critical consolute point

$$\mathcal{E}_{\mathrm{NDE}},\mathcal{E}_{\mathrm{EKE}}\propto\langle\Delta S^2\rangle_V\chi, \tag{6}$$

where $\langle\Delta S^2\rangle_V$ is the mean square of fluctuations of the order parameter S, and χ denotes the generalized susceptibility.

In a critical solution the strong electric field induces a uniaxial, quasinematic ordering. In such conditions the correlation length has the form $\xi=\xi(\xi_{\parallel},\xi_{\perp},\xi_{\perp})$. The component $\xi_{\parallel}$ obeys the nonclassical behavior, while components $\xi_{\perp}$ cross over the Ginzburg criterion and become classical. Hence the susceptibility $\chi\propto(T-T_{\mathrm{crit}})^{-\gamma}$ becomes classical ($\gamma=1$) [21,35]. This made it possible to elucidate the puzzling critical behavior of the NDE and EKE in critical, binary solutions [37]. Prenematic fluctuations are ''naturally'' anisotropic and stiff. Thus, these features, together with the discontinuity of the I-N phase transition, may account for the approximate lack of interactions between fluctuations:

$$\langle\Delta S^2\rangle_V\propto\langle|\Delta S|\rangle_V^f\langle|\Delta S|\rangle_V^0\propto\Delta\varepsilon^0\Delta\varepsilon^f=\mathrm{const.} \tag{7}$$

Substituting T^* for T_{crit} in relation (6), one can obtain, in an agreement with relation (3),

$$\mathcal{E}_{\mathrm{NDE}},\mathcal{E}_{\mathrm{EKE}}\propto\mathrm{const}\chi_0^*\,\frac{\Delta\varepsilon^f\Delta\varepsilon^0}{T-T^*}, \tag{8}$$

where χ_0^* denotes the amplitude of the susceptibility [coefficient a^{-1} in relation (3)].

It seems that the analysis presented may offer an adequate description of the pretransitional behavior of ε and the NDE and EKE both in the isotropic phase of nematogens and in critical solutions [37]. The fluidlike analogy has been successfully investigated theoretically by Mukherjee *et al.* [2–4]. They also pointed out the possibility of critical behavior with $d=3$ for the I-N phase transition.

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[36] S. J. Rzoska, V. Degiorgio, and M. Giardini, Phys. Rev. E **49**, 5234 (1994).
[37] In critical, binary solutions $\mathcal{E}_{\mathrm{NDE}} \propto (T-T_{\mathrm{crit}})^{-\psi}$, with $\psi=0.59$ or 0.4 [21,35]; $\mathcal{E}_{\mathrm{EKE}} \propto (T-T_{\mathrm{crit}})^{-\varphi}$, with $\varphi=0.59$–0.85 [35,36]; and $\mathcal{E}_{I} \propto (T-T_{\mathrm{crit}})^{-\gamma}$, with $\gamma \approx 1.23$ [8].