



Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

Publication details, including instructions for authors and
subscription information:

<http://www.tandfonline.com/loi/gmcl17>

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Version of record first published: 22 Sep 2006.

To cite this article: Sylwester J. Rzoska & Jan Chrapec (1990): Nonlinear Dielectric Effect for Studying Isotropic - Liquid Crystal Phase Transitions, *Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics*, 191:1, 333-337

To link to this article: <http://dx.doi.org/10.1080/00268949008038614>

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NONLINEAR DIELECTRIC EFFECT FOR STUDYING ISOTROPIC - LIQUID CRYSTAL PHASE TRANSITIONS

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Abstract The pretransitional effect in the isotropic phase in the vicinity of the nematic, smectic and blue phase has been studied by means of the Nonlinear Dielectric Effect. As a research method the NDE is complementary to the light scattering, Kerr and Cotton - Mouton effects.

INTRODUCTION

On approaching the point of the continuous phase transition strong, critical, fluctuations of the next phase appeared. It caused that many physical magnitudes may be described by a simple power relation with an universal critical exponent.¹ It also permits studying in the given phase properties of the next one. In some cases the critical fluctuations appeared although the phase transition is discontinuous. Such situation takes place for the isotropic - nematic transition.

In the isotropic phase it is possible to do measurements of such magnitudes as the light scattering (I), the Kerr (K) or Cotton - Mouton (CM) effects. They are very sensitive to critical fluctuations:²

$$I, K, CM \sim \frac{1}{T - T^*}, \quad T > T_c, \quad T^* = T_c - \Delta T, \quad (1)$$

where $K = \Delta n/E^2$, $CM = \Delta n/B^2$, E is the intensity of the electric field, B is the magnetic induction,

$\Delta n = n_{||} - n_{\perp}$ defines the anisotropy of the refractive index n for the completely ordered liquid crystal, T_c is the clearing temperature, T^* is the extrapolated temperature of the hypothetical continuous phase transition. The strong pretransitional anomaly, classically described^{1,2}, caused that these effects are particularly useful for determining the value of the phase transition's discontinuity ΔT .

The same type of behavior exhibits also, relatively less known, nonlinear dielectric effect (NDE). The method relied on measurements of the difference of the electric permittivity ($\Delta \epsilon^E = \epsilon_{||} - \epsilon$) in a strong ($\epsilon_{||}$) and in a weak (ϵ) electric field, respectively. The frequency of the weak measurement field is of the order of a few MHz (5 MHz in our studies). In almost all tested, up to now, cases $\Delta \epsilon^E \sim E^2$ was obtained, so that the measure of the NDE is the value of $\Delta \epsilon^E/E^2$.³ The same temperature character of the pretransitional effect for I , K , CM , NDE is not incidental because they are to a large extent complementary:^{2,4}

$$\begin{aligned} NDE &= C_{NDE} \frac{\Delta \epsilon_{\parallel} \Delta \epsilon_o}{a} t^{-\gamma}, & K &= C_K \frac{\Delta n \Delta \epsilon_o}{a} t^{-\gamma}, \\ CM &= C_{CM} \frac{\Delta n \Delta m}{a} t^{-\gamma} & I &= C_I \frac{\Delta \epsilon_{\lambda} \Delta \epsilon_{\lambda}}{a} t^{-\gamma}, \end{aligned} \quad (2)$$

where C denotes constants connected with the given method, $t = (T - T^*)/T^*$ is the dimensionless distance from T^* , 'a' is the coefficient of the quadratic term in the Landau free energy expansion^{1,2}, $\Delta \epsilon$ and Δm are dielectric and diamagnetic anisotropies, respectively. Indices by

$\Delta\epsilon$ specificate the frequency: the optical (λ), low frequency limit (0) and the NDE's measurement frequency (f).

RESULTS OF THE NDE MEASUREMENTS

The first measurements of the NDE near the nematic phase in MBBA⁵ and the next in C4NCS, CB5, ... confirm the above, classical, relations. The same temperature dependence was observed in the vicinity of the blue phase in cholesteryl oleate (CO) and cholesteryl oleyl carbonate (COC) ⁴ (Fig.1). It is noteworthy that for the isotropic - blue phase transition a numerical analysis did not exclude that the optimum exponent has a value little less than 1.

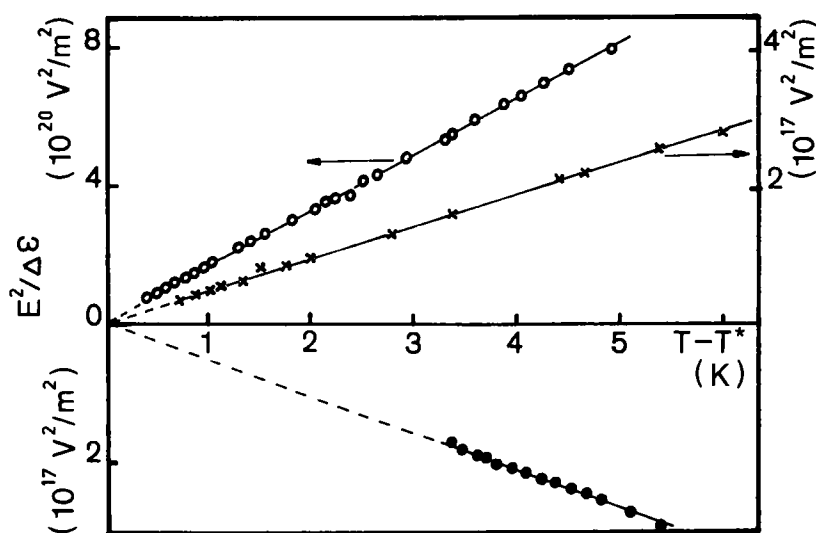


Figure 1. Inverse of the NDE versus $T - T^*$ for the transition to the nematic (MBBA, crosses), blue phase (CO, open circles) and to the smectic in DOBAMBC (full circles).

A special case is the transition to the smectic phase. For 6 and 7 DBT⁶ the classical behavior was observed only for $T > T_c + 4$ K. Elucidation of this behavior is not unequivocal. For instance it may be connected with the value of the measurement frequency.

In all previous research on liquids (i.e. also in critical solutions of ordinary liquids) fluctuations always caused positive changes in the electric permittivity ($\Delta\epsilon^E > 0$). For DOBAMBC, on approaching the smectic A phase, the sign of these changes is negative ($\Delta\epsilon^E < 0$) (fig.1). It may be due to the influence of the ferroelectric smectic* C fluctuations or the influence of the frequency of the measurement field. Maybe the dominating mechanism responsible for the critical effect is of the orientational nature like in the solid state.⁷

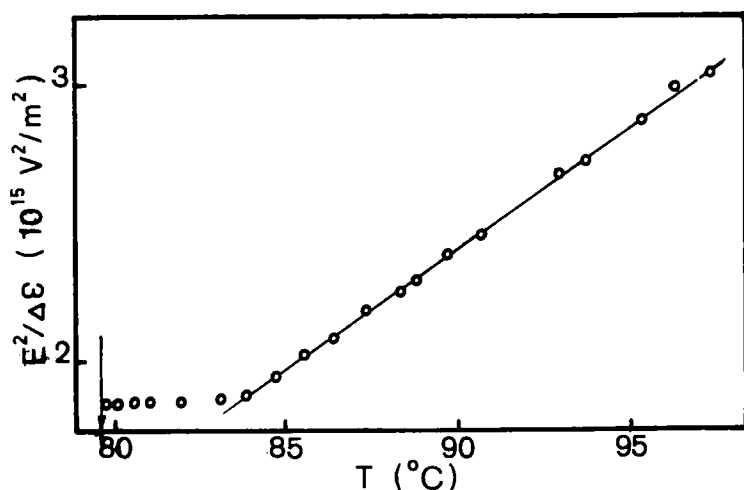


Figure 2. The temperature dependence of the $(NDE)^{-1}$ for 6-DBT.

Presented above results concerned pure systems. One of the most common impurities is, in practice, a

nonmesogenic dopant. It may cause decreasing of T_c and the existence of the two-phase region between the nematic and isotropic phase⁴. It takes place for example in the MBBA - benzene mixture. Measurements of the NDE showed that also in this case the critical effect preserves the classical temperature character (eq.1,2). The addition of benzene changes the value of ΔT and the amplitude of the critical effect.⁸

In our opinion the above results prove that the NDE may be a very usefull instrument for studying properties of the isotropic - liquid crystal transitions.

ACKNOWLEDGEMENTS

The disscused in this paper studieswherecarried out under the Polish Central Project for Fundamental Research CPBP 01 06.

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