Longitudinal susceptibility of uniaxial nematic

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A connection between the longitudinal susceptibility χ and the experimentally measured temperature dependence of the order parameter is obtained for uniaxial nematic liquid crystal in the scope of the theory of Landau and de Gennes. The absolute values of the susceptibility χ are obtained, with MBBA as the example, for the nematic phase from experimental data, and its critical behavior is investigated. It is shown that the Levanyuk–Ginzburg criterion is satisfied in the entire temperature range of the nematic phase, including the vicinity of the temperature T_c of the nematic–isotropic liquid phase transition. A connection is established between the nature of this transition and the width of the nematic-phase temperature interval connected with the position of the tricritical point on the nematic–smectic phase-transition line A.

1. INTRODUCTION

The order parameter of a uniaxial uniformly ordered nematic is the tensor[1]

$$S_{\alpha\beta}=S(n_\alpha n_\beta-{}^1/_3\delta_{\alpha\beta}), \qquad (1)$$

where $n_{\alpha,\beta}$ are the components of the director **n**, $S = \langle 3\cos^2\theta - 1\rangle/2$, θ is the angle between the longitudinal molecular axis and **n**, and the angle brackets $\langle ...\rangle$ denote averaging over the molecular ensemble. The response of a nematic to a field h thermodynamically conjugate to S is characterized by the longitudinal susceptibility[2]

$$\chi=(\partial S/\partial h)_{T;\, h\to 0}. \qquad (2)$$

Information on the value and the temperature dependence of χ in the nematic phase is necessary to select liquid-crystal materials with maximum response and to solve many physical problems. Let us note some of them.

1. Foremost are the description of the nematic–isotropic (N–I) liquid phase transition and the determination of the region of applicability of the mean-field Landau–De Gennes theory,[1,2] in the framework of which the thermodynamic-potential density $\Delta\Phi$ of the nematic is a power function of the invariants $\mathrm{Sp}\,\hat{S}^2 \propto S^2$ and $\mathrm{Sp}\,\hat{S}^3 \propto S^3$ and can be represented by the series

$$\Delta\Phi=\frac{1}{2}AS^2+\frac{1}{3}BS^3+\frac{1}{4}CS^4+\frac{1}{5}DS^5+\frac{1}{6}ES^6+\dots. \qquad (3)$$

Here $A = a\,(T - T^*)$, and the remaining coefficients are assumed to be independent of temperature in the relatively narrow interval of existence of the nematic phase. A distinctive feature of real nematics is that in the scope of the given theory the values and the temperature dependences of different physical parameters on both sides of the N–I transition turns out to be possible with account taken of a large number of terms of the series (3),[3,4] while the parameters $B<0$, $C<0$, $D>0$, and $E>0$ must be strongly correlated with one another, and all the terms of the series, starting with CS^4 turn out to be of the same order in the region of the N–I transition. Further increase of the number of terms in (3) makes such an approach purely empirical.

On the other hand, when describing one and the same set of experimental data the signs and the values of the coefficients of the series (3) depends substantially on the number the considered terms of this series.[3,5] This leaves indeterminate the expression of χ in terms of the coefficients of the series (3) when the following equation is used[2]

$$\chi=\left(\frac{\partial^2\Phi}{\partial S^2}\right)^{-1}_{S_p}, \qquad (4)$$

where the equilibrium value of S_p is the solution of the equation $\partial\Phi/\partial S = 0$. The latter must include a limited number of terms of the series (3), making indeterminate the estimate of the mean squared long-wave fluctuation $\langle(\partial S)^2\rangle v$ in the given volume V of the sample, a fluctuation obtained in the Gaussian approximation from the equation (Ref. 2)

$$\langle(\delta S)^2\rangle_V=kT\chi/V. \qquad (5)$$

The number of terms of the series (3) governs also the parameter

$$\varkappa=\langle(\delta S)^2\rangle_V/S^2, \qquad (6)$$

where $V = 4\pi r^3/3$ and r is the correlation radius of the spatial fluctuations of the modulus S. It is therefore impossible to establish through relations (3)–(6) the region in which the Levanyuk–Ginzburg criterion is satisfied[2]

$$\varkappa\ll 1, \qquad (7)$$

i.e., where the Landau–de Gennes theory is valid.

2. The presence of a cubic invariant in (3) upsets the universal connection between the temperature anomalies of the various physical quantities in the N–I transition region, and these anomalies are in the general case not described by simple laws. The universality, however, can be restored[3,6] by choosing as the "scale" not of the reduced temperatures $T_c - T$ but of the susceptibility χ or the relaxation time of the fluctuation of the modulus S

$$\tau=\chi\nu. \qquad (8)$$

The latter is valid if the kinetic coefficient ν in the nematic phase does not have a unique singularity similar to that in the isotropic phase.[7] Determination of $\chi(T)$ in the nematic phase from experimental data and comparison with an independently measured $\tau(T)$ would cast light on the universal properties of the N–I transition and of the $\nu(T)$ dependence.

The character of the temperature dependence of $\chi(T)$

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in the nematic phase is important also for ascertaining the causes of the large discrepancy between the observed width ($\Delta_T = 10^{-2} - 10^{-3}$, Ref. 3) and the nematic-phase and corresponding to the position of the tricritical point on the nematic–smectic A(N–A) phase-transition line. Here T_c and T_{NA} are the N–I and N–A transition temperature widths $\Delta = 1 - T_{\mathrm{NA}}/T_c$ predicted in the molecular-static theory ($\Delta_T = 0.13$, Ref. 8)

So far, however, there are no measured values of χ in the nematic phase. Direct determination of χ in a nematic from the change of the birefringence $\Delta n \sim S$ in a magnetic or electric field is impossible, since the change $\delta(\Delta n)$ in a field $\mathbf{H}\|\mathbf{n}$ by suppression of the transverse thermal fluctuations of the local direction of the director $\mathbf{n}(\mathbf{r})$ depends on H (or $|E|$) linearly,[1,9–11] and to determine χ we must know the change $\delta(\Delta n) \propto H^2$. In relatively weak fields the linear effect is stronger than the quadratic by two orders.[12]

We propose here a new approach to the determination of the longitudinal susceptibility χ of a nematic, in which the susceptibility can be expressed in terms of the temperature dependence of the modulus S. In Sec. 2 of the article we describe the formal aspect of the approach. Section 3 includes an analysis of the temperature dependence of χ in the nematic MBBA phase, a comparison with the measurements of other parameters having singularities near T_c, and the answers to the questions raised above.

2. LONGITUDINAL SUSCEPTIBILITY OF A NEMATIC AND THE TEMPERATURE DEPENDENCE OF THE ORDER PARAMETER

An important role will be played below by the character of the temperature dependence of the coefficient A in (3), a coefficient equal to the inverse susceptibility ($A = \chi_i$) of the isotropic phase. Its temperature dependence can be obtained from experiments on the critical behavior of various physical characteristics.[1,3] All the known data show that the relation $A \propto (T - T^*)$ holds for nematics having neither real nor virtual (manifested in mixtures) low-temperature smectic phases, in a wide temperature interval $T > T_c$, except for a narrow pretransition region of the order of fractions of a degree. We therefore write the density of thermodynamic potential of a uniaxial nematic in a field h conjugate to the modulus S in the form

$$\Delta\Phi = {}^1/_2 a(T - T^*)S^2 + \ldots - hS, \qquad (9)$$

where the number of the remaining terms of the expansion, with temperature-independent coefficients, is inessential for the sequel.

Since the equation of state

$$a(T - T^*)S + \ldots - h = 0 \qquad (10)$$

is the functional connection between the variables $x = S$, $y = h$, and $z = T$, the use of the known identities[13]

$$\left(\frac{\partial x}{\partial y}\right)_z \left(\frac{\partial y}{\partial z}\right)_x \left(\frac{\partial z}{\partial x}\right)_y = -1, \qquad \left(\frac{\partial z}{\partial x}\right)_y = \left[\left(\frac{\partial x}{\partial z}\right)_y\right]^{-1}, \qquad (11)$$

yields

$$\left(\frac{\partial S}{\partial h}\right)_T = -\frac{1}{aS}\left(\frac{\partial S}{\partial T}\right)_h. \qquad (12)$$

In the limit as $h \to 0$ the value of S goes over here into the corresponding equilibrium values S_{eq} which satisfies Eq. (10) at $h = 0$ and describes the experimental $S(T)$ dependence when account is taken of a sufficient number of terms of the series (3). Indeed, for a large number of real nematics[4,14,15] the experimental $S(T)$ dependence can be described on the basis of the model (3) in the entire mesophase interval, and account must be taken of the corresponding number of terms for each object.

Changing to the variable $t = 1 - T/T_0$, where T_0 is the limiting superheating temperature of the nematic we obtain ultimately from (12) an expression for χ in terms of the measurable quantities:

$$\chi = \frac{1}{aT_0} \frac{\partial(\ln S)}{\partial t}, \qquad (13)$$

where the parameter a is obtained from the relation for the transition heat $\Delta H = aS_c^2 T_c/2$, $S_c = S(T_c)$. Since the derivation of (13) does not depend on the form and number of the remaining terms of the series (9), it can be used for a large group of systems whose thermodynamic potential in a field h conjugate to the order parameter S is of the form (9) and meets the requirements noted above. For theory variants that take into account a finite number terms of the series (3), substitution of the $S(t)$ dependences obtained from the equation $\partial\Phi/\partial S = 0$ in (13) leads to known results obtained for $\chi(t)$ (Refs. 1–3, 16) by using (4).

In particular, confining ourselves in (3) to terms of second, third, and fourth power in the modulus S, the $S(t)$ dependence takes the form

$$S = S_0 + S_0 \delta^{-1/2} t^{1/2}, \qquad (14)$$

where $S_0 = S(T_0)$, $\delta = 1 - T^*/T_0$. The effective susceptibility power-law exponential γ' defined approximately by $\chi \sim t^{-\gamma'}$ depends on the temperature and is obtained from (13) and (14) in the form

$$\gamma' = 1 - S_0/2S(t). \qquad (15)$$

For $T = T_0$ we obtain the know result $\gamma' = 1/2$ (Ref. 16). In addition, $\gamma' < 1$ for all $T \leqslant T_c$. By taking into account in (3) the gradient term $g(\nabla S)^2/2$ it is possible to obtain in the usual manner[2] an expression for the correlation radius $r = g(\chi)^{1/2}$ of the fluctuations of the modulus S in the nematic phase. The effective exponent $\nu' = \gamma'/2$ defined by the relation $r \sim t^{-\nu'}$ depends on temperature and varies in the interval $1/4 \leqslant \nu' < 1/2$.

Changing to some other temperature scale, i.e., introducing $\tilde{t} = 1 - T/T_1$, where $T_1 > T_0$, we can approximate quite accurately in a certain temperature interval $T < Tp_c$ the real $\chi(t)$ dependence by the formula $\chi(\tilde{t}) \sim \tilde{t}^{-1}$. This corresponds to Haller's known empirical procedure[15,17] of approximating the experimental $S(T)$ dependence in nematics and cholesterics by the formula

$$S = C(1 - T/T_1)^{\beta},$$

where $T_1 > T_c$ and explains the value $\nu' = 1/2$ obtained in Ref. 18 on NMR relaxation when $T_1 > T_0$ is chosen. Comparison of the results of various experiments on the critical change of the physical parameters in the nematic phase is possible only if identical values of the difference $T_0 - T_c$ are chosen.

3. LONGITUDINAL SUSCEPTIBILITY OF THE NEMATIC PHASE OF MBBA

Comparison of (5) with (13) establishes a connection between the equilibrium fluctuations of the modulus S and its temperature dependence. If experimental values of S are used in (13), the obtained value of χ contains a contribution from transverse thermal fluctuations of the local direction of the director $\mathbf{n}(\mathbf{r})$ by virtue of the principle of the conservation of the modulus of a nematic in a degenerate system.[19,20] Since the relative change $\delta(\Delta n)/\Delta n$ due to the suppression of the director fluctuations[1,9-12] is negligibly small, the main contribution to χ is made by the temperature dependence of the local orientational ordering of the molecules.

When χ is calculated by Eq. (13) it is important to choose the physical quantity from whose value one can determine the modulus S. The real retardation of the molecule rotation about their longitudinal axes and the biaxial character of the second-rank molecular tensors $\tilde{\gamma}$ causes the macroscopic anisotropy

$$\Delta M \infty S\hat{\Delta}\gamma + {}^1/_2 G\hat{\Delta}\gamma' \qquad (16)$$

of the corresponding physical quantity to be determined not only by the component S_{zz}, but also by the biaxiality $G = S_{xx} - S_{yy}$ of the microscopic tensor of the orientational ordering of the molecules. Here

$$S_{ii} = \langle 3\cos^2\theta_{in} - 1\rangle/2 \quad (i = x,\ y,\ z), \qquad (17)$$

where θ_{in} is the angle between the ith axis of the molecular system of coordinates and the director $\mathbf{n}$. We use in (16) the notation

$$\Delta\hat{\gamma} = \hat{\gamma}_{zz} - {}^1/_2(\hat{\gamma}_{xx} + \hat{\gamma}_{yy}),\ \ \Delta\hat{\gamma}' = \hat{\gamma}_{xx} - \hat{\gamma}_{yy}. \qquad (18)$$

The temperature dependence of G is the reciprocal of the temperature dependence of S: as $T \to T_c$ the parameter G increases, reaching a maximum value $G \approx 0.15S$ at $S \approx 0.4$, and decreases with further increase of T.[21] Therefore at a ratio $\Delta\hat{\gamma}'/\Delta\hat{\gamma}$ that is not small as in the case of diamagnetic susceptibility,[21] the temperature dependences of ΔM and S near T_S can differ noticeably.[22] The determination of S from the anistropy of the diamagnetic susceptibility ΔM (Ref. 1) can therefore yield a distorted temperature dependence of χ calculated from (13).

For the determination of S it is preferable in this respect to use in place of ΔM the anisotropy $\Delta\varepsilon$ of the dielectric function in the optical band [formally defined by the same equation (16)], since the biaxiality of the molecular tensors $\hat{S}$ (17) of the polarizability $\hat{\gamma}$ (8) is manifested here to a much lesser degree.[22,23] Allowance for the anisotropy of the local field of the light wave is of no principal importance in this case (cf. Refs. 1 and 3), since it leads to multiplication of the right-hand side of (16) by the quantity $(1 + \sigma_0 + \sigma_1)^{-1}$, where σ_0 is independent of temperature and $\sigma_1 \sim S$, but even for nematics with large local-field anisotropy we have $\sigma_0 \approx 0.2$ and 2 and $\sigma_1/\sigma_0 \approx 0.1$ (Ref. 23). By choosing the refractive indices of the nematic in the proper spectral region one can reduce the sum $\sigma_0 + \sigma_1$ to zero and simultaneously lower the ratio $\Delta\hat{\gamma}'/\Delta\hat{\gamma}$ (Ref. 23). To take into account the temperature dependence of the nematic it is necessary to use in place of the anisotropy $\Delta\varepsilon = n_\parallel^2 - n_\perp^2$ the quantity

$$\Delta\varepsilon/(\bar{\varepsilon} - 1) = \mathrm{const}\cdot S, \qquad (19)$$

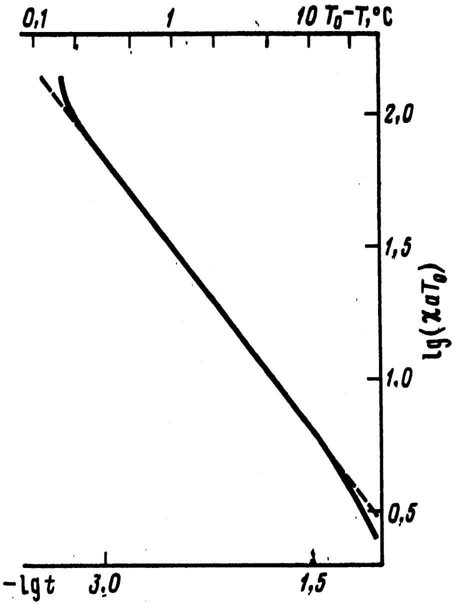

FIG. 1. Temperature dependence of the product $\chi a T_0$ (13) for the nematic phase of MBBA, calculated from Eqs. 19 and (20) at $t = 1 - T/T_0$, $T_0 = T_c + 0.1°$ (Ref. 29), and $T_c = 318.46$ K (Refs. 17 and 24).

where $\bar{\varepsilon} = (n_\parallel^2 + 2n_\perp^2)/3$, $n_{\parallel,\perp}$ are the refractive indices of the nematic.

In the present study we investigated an MBBA nematic independently researched by independent methods. Precision measurements have yielded for it the refractive indices $n_{\parallel,\perp}$ ($\lambda = 589$ nm),[24] which are well approximated in the temperature interval $0.06° \leqslant T_c - T \lesssim 30°$ by the equations[24]

$$n_\perp = 1{,}5765 - 0{,}174\tau^{1/2} + 0{,}253\tau,$$
$$n_\parallel = 1{,}6935 + 0{,}282\tau^{1/2} + 0{,}243\tau, \qquad (20)$$

where $\tau = 1 - T/T_c$, $T_c = 318.46$ K.[17,24] The anisotropy of the local field in the MBBA is small[25] and introduces no noticeable change whatever in the temperature dependence of S (19). This explains the equality of the normalized $S(T)$ temperature dependences obtained by independent methods.[26,27] It is known from precision measurements of the heat capacities of MBBA[28,29] that T_0 exceeds by 0.1° the lower limit $T_c^{(-)}$ of the real two-phase region assumed for T_c in refractometric experiments for single-domain nematic samples.

The figure shows the temperature dependence of the product aT_0 calculated from (13), (19), and (20) using $T_0 = T_c + 0.1°$. In the entire temperature interval $0.1° \leqslant T_c - T \lesssim 20°$ the variation of χ is described by the relation $\chi \sim t^{-\gamma'}$ with a constant value $\gamma' = 0.68$. On the other hand, in the same temperature interval, according to acoustic measurements in MBBA[3,6] the ratio $Y = \alpha/z = 1/2$ of the effective heat-capacity exponents (α) and of the relaxation times (z) of the fluctuations of the modulus S remain constant. Using the effective values $\alpha = 0.32$–0.35[28,29] which are constant in this temperature interval, we obtain $z = 0.64$–0.70. This agrees with the value given above for and shows that for the nematic phase of MBBA the kinetic coefficient γ' does not have in relation (8) an independent singularity near T_c. Note that in the isotropic phase of MBBA the anomalous change of ν, which differs from the

$\nu = \nu_0 \exp(\theta/T)$ dependence,[6] is much less pronounced near T_c than for the nematic BMOAB.[7] Thus, the results of the independent investigations of the $\chi(t)$ and $\tau(t)$ dependences in the MBBA nematic phase agree well within the framework relation (8).

The pre-transition increase of γ' at $T_c - T < 0.1°$ can be due both to natural violation of the dependences (20) near T_c and to the growth of the fluctuations of the modulus S in this region. The latter can be ascertained by estimating the parameter $\varkappa$ [see (6)] using the values, shown in the figure of χ, of the temperature dependence of the correlation radius $r = r_0 t^{-0.34}$, and also the values of the parameter $r_0 = 6$ Å (Ref. 3) and the value $a = 0.093$ J/cm$^3\cdot$K obtained from independent data on induced electric and magnetic birefringence in the isotropic phase.[30] As a result we obtain for $T = T_c - 0.05°$ and $S = 0.32$ (Refs. 26 and 30) in the correlation volume of the sample $\varkappa = 0.056$, i.e., the Levanyuk–Ginzburg criterion (7), which is well satisfied at the N–I transition point itself. This makes the Landau–de Gennes theory applicable to the description of the N–I transition in the given specific compound. This probably holds also for a larger group of objects, if it is recognized that the parameters S, r_0, ν', and a used here are typical also of other known compounds.[3]

As seen from the figure, at the value $T_0 - T_c \approx 0.1°$ typical of nematics the value of χ decreases compared with $\chi(T \approx T_c$ by one order already at $T_c - T \approx 3$–$4°$, and b two orders only at $\Delta T = 40$–$50°$. This is due to the nature of the N–I transition and to the inequality $\gamma' < 1$. As a result, the principal change takes place in a narrow temperature region $t \lesssim T_c$. This explains the narrowness of the interval of the nematic phase $\Delta_T = 1 - T_{\rm NA}/T_c$, which corresponds to the position of the tricritical point on the line of the N–A phase transitions. In fact, a tricritical point is the result of the vanishing of the coefficient[1]

$$\beta = \beta_0 - {}^1/_2 \lambda^2 \chi \qquad (21)$$

in the invariant $|\psi|^4$ in the expansion of the density of the thermodynamic potential of the smectic A in the modulus of the smectic order parameter. The renormalization resulting from the interaction of the parameters of the nematic and smectic orders is determined by the longitudinal susceptibility χ, which is appreciable in real nematics only in a narrow vicinity of T_c. It is this which leads to the smallness of Δ_T.

On the other hand, within the framework of the Maier-Saupe theory, on which the MacMillan theory is based,[8] T_0 values ≈ 300 K correspond to the interval $T_0 - T_c \approx 3.5°$ and a decrease of χ by an order compared with $\chi(T \approx T_c)$ takes place at $\Delta T \approx 30$–$40°$, corresponding exactly to the value $\Delta_T = 0.13$ (Ref. 8). Thus, the smallness of the real values $\Delta_T = 10^{-2}$–10^{-3} (Ref. 3) is connected with the very nature of the N–I phase transition and the aforementioned singularities of the behavior of $\chi(t)$. An additional factor that decreases the interaction constant λ in (21) and correspondingly the value of Δ_T is the orientational melting of the end fragments of the flexible molecule chains in the N–A transition.[32]

We note in conclusion that it becomes possible in practice to obtain quantitative information on the susceptibility χ. It is an additional property of liquid-crystal materials along with the usually considered quantities such anisotropy of the static dielectric constant $\Delta\varepsilon$, the birefringence Δn, the ratio of the elastic moduli K_{33}/K_{11}, and the order parameter S. The response $\chi_q = K_q \chi$ of a nematic to any physical action that leads to a change of the modulus S and to which one can set in correspondence in the thermodynamic potential (9) the term $K_q hS$ can be expressed in terms of χ (13). The role of the external perturbation can be played also by resonant actions on the intramolecular degrees of freedom which leads to a change of the internal state of the molecules, of the intermolecular interaction, and of the modulus S in the sample.

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