Conformational optical nonlinearity of the isotropic phase of a nematic liquid crystal

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The characteristics of the conformational optical nonlinearity of nematic liquid crystals near the nematic–isotropic phase transition is considered. A critical increase of the nonlinearity when the phase transition is approached is predicted and is observed by dynamic holography. It is shown that the effect is due to the increase of the correlations between the fluctuations of the photoconformer concentration and the polarizability of the liquid-crystal molecules near the phase transition.

One of the cubic optical nonlinearities of nematic liquid crystals (NLC) is the conformational linearity, which has gigantic values of the nonlinearity parameter $\varepsilon_2 = \; = 2n\Delta n/E^2$ (**E** is the lightwave field intensity and Δn is the change of the refractive index n).[1,2] The cause of this nonlinearity is the change of molecule conformation upon absorption of light.

Experimental investigations of the conformational nonlinearity, and its use to determine the conformer characteristics, were carried out mainly in the mesophase of nematics.[1–7] Data on conformational nonlinearity in the isotropic phase (IP) of NLC are much scantier.[3,4] Yet such investigations are of considerable interest, since the change of the intermolecular interaction in the IP compared with the mesophase should be reflected in the characteristics of the conformers. In addition, whereas far from the phase-transition (PT) point T_c the isotropic–nematic conformational nonlinearity should be the same as for ordinary nonmesogenic liquids, effects exhibited only by mesogenic liquids can appear near T_c, where the fluctuations of the orientational order of the molecules are substantial.

We report here theoretical and experimental investigations of the conformational optical nonlinearity in IP NLC. We show that the correlation between the conformer concentration fluctuations and the polarizability of NLC molecules, due respectively to anisotropy of the coefficient of light absorption by the NLC molecules and the anisotropy of the conformer polarizability in the NLC crystals, makes a nonzero contribution to the cubic-nonlinearity parameter, a contribution that is substantial near the IPC–NLC phase transition. In the vicinity of the PT, owing to the critical increase of the orientational correlations of the molecules as T_c is approached, this contribution leads to a critical behavior of the nonlinearity parameter. The theoreticaly predicted increase of ε_2 as the IP–NLC phase transition is approached was observed by dynamic holography.

THEORETICAL PART

We derive an expression for the cubic conformational nonlinearity parameter of an IP NLC. Since the phototransformed conformer molecules have a relatively long life (0.1–1 s),[1,3] we regard them as optically induced impurity molecules (OIM). Using the additivity of molecular refraction for mixtures, we can write for the change of the dielectric constant of IP NLC due to addition of a small OIM density N'.

$$\Delta\varepsilon_{\alpha\beta}=\overline{L_{\alpha\beta}N'\Delta\gamma_{\alpha\beta}},\quad \Delta\gamma_{\alpha\beta}=\gamma_{\alpha\beta}'-\gamma_{\alpha\beta}, \tag{1}$$

where $\gamma_{\alpha\beta}$ and $\gamma'_{\alpha\beta}$ are the polarizability tensors of the NLC molecules and of the OIM, and are assumed to be averaged over a large number of molecules in a physically small volume and to be macroscopic quantities referred to a single molecule, $L_{\alpha\beta} = t_{\alpha\alpha} t_{\beta\beta}$, where $t_{\alpha\alpha}$ is the diagonal tensor of the local field, and the overbar denotes thermal averaging.

The OIM density is determined from the kinetic equation

$$\partial N'/\partial t=D\Delta N'-N'/\tau+\sigma_{\mu\nu}kI_{\mu\nu}(N-N'). \tag{2}$$

Here D and τ are respectively the translational-diffusion coefficient and the OIM lifetime, $\sigma_{\mu\nu}$ is the molecular absorption coefficient tensor, k is the effective quantum yield and can be larger than unity if allowance is made of the reabsorption of the radiation and for the thermal activation of the conformers following local heating of the NLC near an excited molecule that relaxes without radiating,[8] $I_{\mu\nu} = E_\mu E_\nu/2$ is the exciting-radiation field-intensity tensor, and N is the NLC molecule density.

Neglecting small terms of order $N'/N \ll 1$, we can write the solution of Eq. (2) with zero initial and boundary conditions in the form

$$N'(\mathbf{r},t)=Nk\iint d\mathbf{r}'\,dt'\,G(\mathbf{r}-\mathbf{r}',t-t')\sigma_{\mu\nu}I_{\mu\nu}(\mathbf{r}',t'), \tag{3}$$

where $G(\mathbf{r}-\mathbf{r}', t-t')$ is the Green's function for Eq. (2). The coefficients D, τ, and $\sigma_{\mu\nu}$ in (2) and (3) are, as usual, averaged over a sufficiently large number of molecules occupying a physically small volume. Owing to the molecular motions, these coefficients fluctuate and bring about corresponding fluctuations $\delta N'$ of the OIM density. Obviously, $\Delta\gamma_{\alpha\beta}$ and $L_{\alpha\beta}$ also fluctuate. We are interested in the fluctuations due to orientational motion of the molecules, for it is these very fluctuations which are large near the point of the PT from an orientationally disordered liquid to an orientationally ordered one, i.e., in a nematic.

For the orientational fluctuations of the tensors $\sigma_{\mu\nu}$ and $\Delta\gamma_{\alpha\beta}$ we can write the expressions:

$$\delta\sigma_{\mu\nu}=BS_{\mu\nu}(\mathbf{r},t),\quad \delta(\Delta\gamma_{\alpha\beta})=MS_{\alpha\beta}(\mathbf{r},t), \tag{4}$$

where B is of the order of the anisotropy of the molecular absorption coefficient, M is of the order of the anisotropy γ_a of the NLC molecule polarizability, and $S_{\alpha\beta}(\mathbf{r},t)$ is the tensor order parameter.[9]

If fluctuations of the local-field factor are neglected, Eq. (1) takes the form

$$\Delta\varepsilon_{\alpha\beta}=\bar{L}[\overline{N'}\,\overline{\Delta\gamma}\delta_{\alpha\beta}+\overline{\delta N'\delta(\Delta\gamma_{\alpha\beta})}]. \quad (5)$$

The first term describes here the conformational nonlinearity of an ordinary nonmesogenic liquid, while the second takes into account the contribution made to the nonlinearity of the orientational fluctuations that are substantial in IP NLC at temperatures close to T_c.

We carry out the calculation for a specific experimental situation encountered when the method of dynamic holography is used.[1] In this case two coherent light beams of like polarization and having intensities I_1 and I_2 intersect in the cell with the NLC, and the intensity distribution produced in the plane of the cell is

$$I(x)=I_0[1+m\cos(qx)],\quad m=2(I_1I_2)^{1/2}/I_0,\quad I_0=I_1+I_2. \quad (6)$$

If the NLC cell thickness is much less than the diameter of the light beam and the cell is regarded as thin, the OIM density depends only on the coordinate x. Recognizing next that $\sigma_{\mu\nu}I_{\mu\nu}=\delta_{\mu\nu}\sigma_{\mu\nu}I(x)$ in Eq. (3) and substituting the value of the Green's function,[10] we obtain for $\overline{N'}\;\overline{\Delta\gamma}$ in the stationary case the expression

$$\overline{N'}\,\overline{\Delta\gamma}=I_0\bar{L}N\bar{\sigma}k\overline{\Delta\gamma}[m\cos(qx)/(\bar{D}q^2+\bar{\tau}^{-1})+2\bar{\tau}], \quad (7)$$

where $\bar{\sigma}$ is the average molecular absorption of light with a given polarization ν.

To calculate $\overline{\delta N'\delta(\Delta\gamma_{\alpha\beta})}$ we confine ourselves to the case when the fluctuations $\delta N'$ are due mainly to orientational fluctuations of the molecular absorption coefficient $\sigma_{\mu\nu}$. It follows then from (3) and (4) that $\overline{\delta N'\delta(\Delta\gamma_{\alpha\beta})}$ is expressed in terms of an integral of the correlation function $K_{\alpha\beta\mu\nu}=\overline{S_{\alpha\beta}(\mathbf{r},t)S_{\mu\nu}(\mathbf{r}',t')}$. Since $N'\ll N$, we can approximately replace $K_{\alpha\beta\mu\nu}$ by the correlation function $K^0_{\alpha\beta\mu\nu}$ corresponding to an IP NLC containing no OIM. Using the explicit form of $K^0_{\alpha\beta\mu\nu}$ obtained in Ref. 11, we have the following expressions for the second term of (5):

$$\overline{\delta N'\delta(\Delta\gamma_{\alpha\beta})}=I_0[A_1+A_2m\cos(qx)]\delta_{\alpha\beta}(\delta_{\nu\alpha}-{}^1/_3),$$
$$A_2=4kNk_BTMBt_c^2\varphi/\eta r_c^3, \quad (8)$$

where

$$\varphi=\iint_0^\infty dr\,dt\frac{\exp(-\tau_D'r^2/4t-t/\tau')}{2(\pi t/\tau_D')^{1/2}}f(r,t)\cos(qr_cr). \quad (9)$$

The function $f(r,t)$ is expressed here in terms of the error probability integral and is given in Ref. 11, $\tau_D'=r_c^2/\bar{D}t_c$, $\tau'=\tau/t_c$, t_c' and r_c are the correlation time and radius of the orientational fluctuations, and η is a constant having the meaning of a viscosity coefficient.[11] The constant A_1 is determined in the same manner as A_2, except that $\cos(qr_cr)$ under the integral sign in (9) is replaced by unity.

Substituting (7) and (8) in (5), we obtain for $\Delta\varepsilon_{\alpha\beta}$ an expression in which the term describing the phase diffraction grating in the stationary case takes the form

$$\Delta\varepsilon_{\alpha\beta}'=\varepsilon_{2\alpha\beta}I_0m\cos(qx),$$

where the cubic-nonlinearity parameter is

$$\varepsilon_{2\alpha\beta}=\bar{L}\left[\frac{\overline{Nk\bar{\sigma}\,\Delta\gamma}}{\bar{D}q^2+\bar{\tau}^{-1}}+A_2\left(\delta_{\alpha\nu}-\frac{1}{3}\right)\right]\delta_{\alpha\beta}. \quad (10)$$

To estimate the contribution of the second term in (10), we use the fact that $\tau'\gg1$ (Ref. 11) and that τ_D' does not depend on temperature and is usually of order unity. In this case the region of substantial values of the integrand of (9) is defined by $t<1$ and $r^2\lesssim 4t$, and the function $f(r,t)$ can be approximated by expanding it in powers of r and retaining the first terms of the expansion. Evaluation of the integral (9) yields $\varphi\approx(-64\pi\tau_D')^{-1}$ and after substituting A_2 in (10) we have for the case of self-diffusion the estimate

$$\varepsilon_{2\nu\nu}\equiv\varepsilon^r_{2\nu\nu}+\varepsilon^f_{2\nu\nu}\approx\frac{\overline{N}\bar{L}k\bar{\sigma}\,\overline{\Delta\gamma}}{\bar{D}q^2+\bar{\tau}^{-1}}-\frac{\overline{N}\bar{L}k\bar{\sigma}\gamma_ak_BT\bar{D}}{24\pi\eta}\frac{t_c^3}{r_c^5}, \quad (11)$$

where $\varepsilon^r_{2\nu\nu}$ and $\varepsilon^f_{2\nu\nu}$ are the regular and fluctuating parts of the nonlinearity parameter, $t_c^3/r_c^5=a/(T-T^*)^{1/2}$, T^* is the IP stability-loss temperature, and the coefficient a is independent of temperature. We have put in (11) $B\approx\bar{\sigma}$ and $M\approx\gamma_a$.

Expression (11) shows that the fluctuations of the average molecule orientation in IP NLC lead to the appearance of an additional, compared with an ordinary nonmesogenic liquid, term $\varepsilon^f_{2\nu\nu}$, that increases critically near T^*. It is difficult to predict the exact temperature dependence of the nonlinearity parameter $\varepsilon_{2\nu\nu}$ of an IP NLC, since the parameters $\bar{D}$, $\bar{\tau}$, $\bar{L}$, η, and N are also temperature dependent, and $\overline{\Delta\gamma}$ and γ_a are known only in order of magnitude. Furthermore, no account was taken in the derivation of (11) of the contribution made to $\delta N'$ by the possible fluctuations of $\bar{\tau}$ and $\bar{D}$, and also of the fluctuations of the local-field tensor. Nonetheless, if we use the MBBA parameters $\bar{D}=10^{-6}\ \mathrm{cm^2\cdot s^{-1}}$, $\eta=0.1$ P (Ref. 9), $q=10^4\ \mathrm{cm^{-1}}$, $\tau=1$ s (from Ref. 1), $a=10^{12}\ \mathrm{s^3\cdot cm^{-5}\cdot deg^{1/2}}$ (Ref. 9), $|\overline{\Delta\gamma}/\gamma_a|\approx10^{-3}$ (Ref. 5), and it is assumed that a contribution of the same order as (7) is made to $\varepsilon_{2\nu\nu}$ by the fluctuations of $\bar{L}$, $\bar{D}$, and $\bar{\tau}$, the second term of (11) becomes comparable with the first at $T-T^*\approx\approx0{,}1^\circ$. This gives grounds for assuming that the entire fluctuation-induced contribution to the nonlinerity can be determined in experiment.

It should be noted that the estimate above was obtained assuming a relatively large anisotropy of the NLC molecules and of the conformers ($\beta\sim\bar{\sigma}$, $|\gamma_a|\approx10^3|\overline{\Delta\gamma}|$). This assumption is apparently justified for conformers in MBBA (see Ref. 5). For weaker anisotropy, however, the contribution of the term $\varepsilon^f_{2\nu\nu}$ to the nonlinearity parameter may turn out to be less significant.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

The investigations were carried out by the dynamic-holography method (Fig. 1). The beam of an He–Cd laser (divergence 1 mrad, power 20 mW, $\lambda=0.44\ \mu$m) operating in the lowest transverse mode was split into two parallel coherent beams of equal intensity. The beams crossed in the focal plane of a large-aperture objective O, where a cell with the liquid crystal MBBA was placed. The period $\Lambda=2\pi/q$ of the interference pattern in the NLC plane was determined by

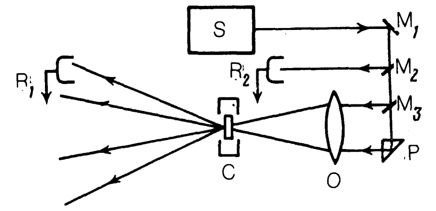

FIG. 1. Setup for recording dynamic holograms: S—laser, M_1–M_3—mirrors, P—prism, C—cell with NLC in a thermostat, R_1, and R_2—photoreceivers, O—objective.

the distance between the beams ahead of the objective and could be varied in the range 5–70 μm by parallel displacement of the prism P. Both flat-polished and unfinished glass substrates were used in the cell. The experimental results were independent of the substrate finish within the limits of error. The NLC thickness was 50 μm, its absorption coefficient $\alpha = 11\ \text{cm}^{-1}$ ($\lambda = 0.44\ \mu$m), and the bleaching temperature $T_c = 43.3\ ^\circ$C. The cell was placed in a thermostat whose temperature was set accurate to 0.025° and maintained accurate to 0.01°.

A thin sinusoidal holographic phase grating was recorded in the region of the light beam intersection and was manifested by the appearance of the first orders of self-diffraction. The self-diffraction intensity was recorded by photodiode PR_1 with a digital voltmeter. The power was monitored with a PR_2 photodiode.

We measured the temperature dependences of the first-order self-diffraction intensity I_d (Fig. 2a) at different periods Λ. Since the Raman–Nath self-diffraction regime set in under the experimental conditions, the self-diffraction intensity was determined from the expression[1]

$$I_d \approx (2\pi z/\lambda)^2 \varepsilon_2^2 I_0^3, \qquad (12)$$

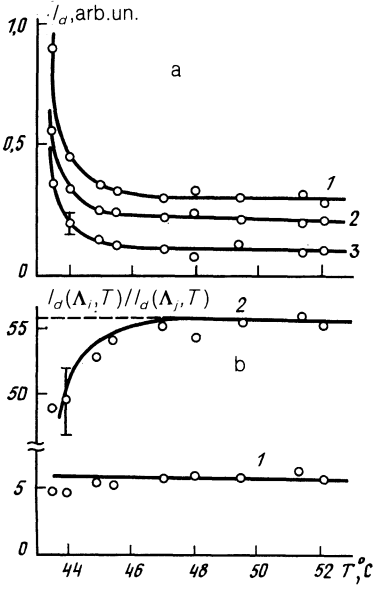

FIG. 2. a) Plots of $I_d(T)$ for different values of Λ: $1 - I_d(\Lambda_1 = 25\ \mu\text{m},\ T)\cdot 200$, $2 - I_d(\Lambda_2 = 16\ \mu\text{m},\ T)\cdot 5$, $3 - I_d(\Lambda_3 = 9\ \mu\text{m},\ T)$; b) plots of $I_d(\Lambda_i,\ T)/I_d(\Lambda_j,\ T)$ for $\Lambda_i = 25\ \mu$m: $1 - \Lambda_j = 16\ \mu$m, $2 - \Lambda_j = 9\ \mu$m.

where I_0 is the intensity of the writing beams.

The $I_d(t)$ dependence was obtained in the course of the measurements starting with the larger values of T. Once a certain temperature set in, the readings were made after a time sufficient for the results to be reproducible. This was about five minutes at $T - T_c \approx 10°$ and reached several hours near T_c.

The obtained experimental dependences agree qualitatively with expression (12) with ε_2 estimated from (11), if the polarization change during the phototransformation is $\overline{\Delta\gamma} < 0$. The condition $\overline{\Delta\gamma} < 0$ is met in the mesophase,[5,12] but in the IP the characteristics of the conformers can be different. We have therefore performed additional experiments to determine the sign of $\overline{\Delta\gamma}$ in the IP MBBA.

If $T \gg T_c$, the sign of $\overline{\Delta\gamma}$ is the same as that of the change Δ_n of the refractive index [see (11)], which can be determined by the method of induced nonlinear lens. The experimental setup is shown in Fig. 3. The beam from an He–Cd laser was focused into a cell with MBBA ($T = 60\ ^\circ$C) and the light intensity $I(l,\ d_0)$ at the center of the beam passing through the cell was recorded as a function of the distance l between the converging lens and the cell. Since the light intensity at the beam center is uniquely related to the half-width of a Gaussian distribution (the diameter of the recording region was much smaller than the beam width), the variation of the intensity $I(d_0)$ with l attests to the formation of a Gaussian nonlinear lens in the MBBA. The intensity $I(l)$ is then qualitatively different for converging ($\Delta n > 0$) and diverging ($\Delta n < 0$) lenses (Fig. 4).[12] The experimental results in this figure show that the sign of Δn, meaning also $\overline{\Delta\gamma}$, is negative in our case, just as in the mesophase.

Let us analyze the contribution made to the $I_d(T)$ temperature dependence near the PT point T_c by the first term in (11), which is not connected with fluctuations. Since the refractive index $\bar{n} = \overline{L\gamma N}$ of the IP NLC reveals no critical behavior near T_c and depends little on the temperature,[13] it is reasonable to assume that the products $\overline{L\Delta\gamma N}$ and $\overline{L\gamma_a N}$ in (11) depend little on T. In addition, our measurements have shown that the absorption coefficient $\bar{\alpha} = \overline{L\sigma N}$ of the IP NLC is independent of temperature practically in the entire range of T. Contributions to $I_d(T)$ in the first term of (11) are therefore made only by the $\overline{D}(T)$ and $\bar{\tau}(T)$ dependences, which can be quite strong.[3,6]

The dependences of the lifetime $\bar{\tau}_g = (\overline{D}q^2 + \bar{\tau}^{-1})^{-1}$ of the holographic gratings on their period Λ (Refs. 1 and 6), obtained by us far from and close to T_C, have shown that at the values employed $q\tau_g^{-1}$ is proportional to q^2, i.e., the

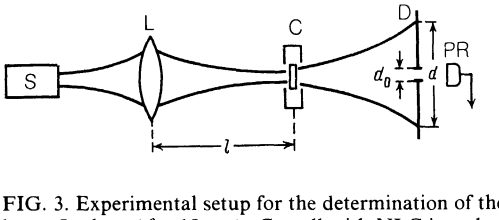

FIG. 3. Experimental setup for the determination of the sign of Δn: S—laser, L—lens ($f = 10$ cm), C—cell with NLC in a thermostat, D—diaphragm, PR—photoreceiver.

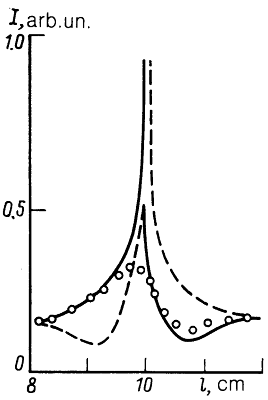

FIG. 4. Calculated $I(l)$ curves for $n>0$ (dashed) and $n<0$ (solid line), and the experimental points.

contribution of the conformer lifetimes to the values of τ_g and I_d can be neglected.

To determine the temperature dependence of the diffusion coefficient we use the fact that the temperature dependence of the regular term $\varepsilon^r_{2\nu\nu}$ in expression (11) for $\varepsilon_{2\nu\nu}$ is determined only by the $D(T)$ dependence, and the contribution of the fluctuating term $\varepsilon^f_{2\nu\nu}$ to the value of I_d becomes negligibly small at sufficiently large periods Λ, as can be seen from (11). The physical reason is that the regular term of $\varepsilon_{2\nu\nu}$ is determined by the amplitude of the sinusoidal distribution of the average density of the conformers, and the diffuse smearing of this distribution decreases this density when the period Λ is decreased (as Λ^2 for $Dq^2 \gg 1/\tau$). The fluctuating term, on the other hand, depends on the degree of correlation of the light-absorption-coefficient fluctuations with the polarizability of the NLC molecules, and is independent of the lattice period. The $\overline{D}(T)$ dependence can therefore be determined from experimental data obtained for $I_d(T)$ under conditions when the fluctuation term in I_d is small. An experimental criterion of satisfaction of this condition can be absence of a temperature dependence of the ratio of the self-diffraction intensities measured at different periods Λ. In fact, it follows from (11) and (12) that this ratio is

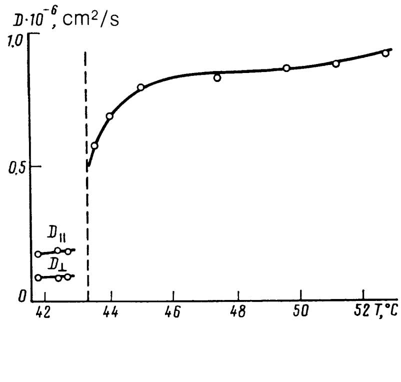

FIG. 5. $\overline{D}(T)$ dependence. The data for the mesophase are taken from Ref. 6.

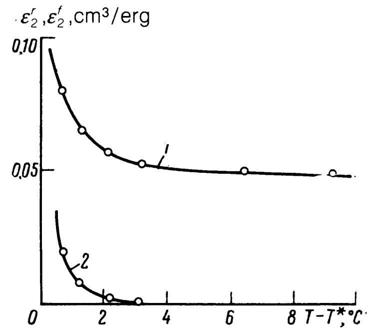

FIG. 6. Plots of $\varepsilon^r_2(T)$ (1) and $\varepsilon^f_2(T)$ (2) for $\Lambda = 9\ \mu$m.

$$\frac{I_d(\Lambda_i)}{I_d(\Lambda_j)} \approx \left(\frac{\Lambda_i}{\Lambda_j}\right)^4 \left[\frac{1+4\pi\overline{D}^2(T)\,bT/\Lambda_i^2(T-T^*)^{1/2}}{1+4\pi\overline{D}^2(T)\,bT/\Lambda_j^2(T-T^*)^{1/2}}\right]^2 \qquad (13)$$

and is equal to $(\Lambda_i/\Lambda_j)^4$ if the fluctuation contribution is small.

Such a situation is realized in experiment for lattice periods $\Lambda \gtrsim 16\ \mu$m (see curve 1 of Fig. 2b). It can therefore be assumed that for these periods we have $\varepsilon_{2\nu\nu} = \varepsilon^r_{2\nu\nu}$ and

$$I_d \approx \left(\frac{2\pi z}{\lambda}\frac{\overline{N}\overline{L}k\bar{\sigma}\,\overline{\Delta\gamma}}{\overline{D}q^2}\right)^2 I_0^3. \qquad (14)$$

The temperature dependence $\overline{D}(T)$ obtained from the experimental data for $I_d(\Lambda = 25\ \mu\text{m}, T)$ using Eq. (14) is shown in Fig. 5. The numerical values of D were obtained by mesuring the $\tau_g(\Lambda)$ dependence at $T - T_c = 10°$ by the procedure described in Refs. 1 and 6.

For periods $\Lambda \lesssim 9\ \mu$m, the fluctuating term $\varepsilon^f_{2\nu\nu}$ is already appreciable near the PT, since the ratio $I_d(\Lambda = 25\ \mu\text{m})/I_d(\Lambda = 9\ \mu\text{m})$ decreases as T_c is approached (curve 2 of Fig. 2b). The data obtained on the temperature dependences of $I_d(\Lambda = 9\ \mu\text{m})$ and $I_d(\Lambda = 25\ \mu\text{m})$ make it possible to separate the temperature dependence of the fluctuation term $\varepsilon^f_{2\nu\nu}$ $(\Lambda = 9\ \mu\text{m})$ from the $\varepsilon_{2\nu\nu}(\Lambda = 9\ \mu\text{m}, T)$ dependence. In fact, the total value $\varepsilon_{2\nu\nu}(\Lambda = 9\ \mu\text{m}, T)$ can be obtained directly from the experimental temperature dependence $I_d(\Lambda = 9\ \mu\text{m}, T)$ [see Eq. (12)]. The regular term for this period is calculated from the values $\varepsilon_{2\nu\nu}(\Lambda = 25\ \mu\text{m}, T) \approx \varepsilon^r_{2\nu\nu}$ $(\Lambda = 25\ \mu\text{m}, T)$ with allowance for the decrease of the lattice period:

$$\varepsilon^r_{2\nu\nu}(\Lambda_i/\varepsilon^{\mu\text{m}}_{2\nu\nu}(\Lambda_j) = (\Lambda_i/\Lambda_j)^2.$$

We next obtain the fluctuation term $\varepsilon^f_{2\nu\nu}$ $(\Lambda = 9\ \mu\text{m}, T)$ by subtracting $\varepsilon^r_{2\nu\nu}$ $(\Lambda = 9\ \mu\text{m})$ from the total value $\varepsilon_{2\nu\nu}(\Lambda = 9\ \mu\text{m})$. The dependence $\varepsilon^f_{2\nu\nu}$ $(\Lambda = 9\ \mu\text{m}, T)$ obtained in this manner is shown in Fig. 6. It can be seen that $\varepsilon^f_{2\nu\nu}$ (Λ, T) increases critically as the PT is approached. Since estimates that follow from Ref. 9 give $T_c - T^* \approx 0{,}7°$, the experimental points closest to the temperature T_c correspond to a value $T - T^* \approx 0.8°$. Estimates made for this temperature at the parameter values indicated at the end of the preceding section yield for the $\varepsilon^f_2/\varepsilon^r_2$ ratio a value 10^{-1}, in good enough agreement with the experimental $\varepsilon^f_2/\varepsilon^r_2 \approx 0.25$.

CONCLUSION

The experimental data offer thus evidence that near the PT point of an IP NLC a substantial contribution to the conformational optical nonlinearity and to its temperature dependence is made by orientational correlations between the OIM concentration and the polarizability of the NLC molecules.

It must be noted, however, that the experimental conditions chosen by us turned out to be not optimal for the study of the influence of fluctuations on optical nonlinearity, since the regular part ε_2^r of the nonlinearity parameter turned out to be predominant. To determine the contribution of the fluctuations it is more convenient to use the smallest possible holographic-grating periods, i.e., operate with counter-propagating beams (in this case $\Lambda \approx \lambda/2$). It is also desirable to work with NLC having a weak $D(T)$ dependence near T_c, since this dependence distorts the fluctuation-induced temperature dependence of ε_2. It seems to us that if these conditions are met an investigation of the characteristics of the conformational nonlinearity near T_c can serve as a basis for a new effective method of studying critical phenomena in NLC by dynamic holography. Among its undisputed advantages over the light-scattering method are high sensitivity and the possibility of using thin NLC layers, i.e., of investigating the influence of the surface on the critical phenomena, and also the fact that multiple scattering need not be taken into account.

We note in conclusion that the fluctuation contribution, investigated by us, to the conformational nonlinearity is apparently the first observed effect in which are manifested heretofore unknown correlations of fluctuations of the polarizability of molecules with the photoconformer density. One can also expect the correlation of the indicated quantities can lead, near the IP–NLC PT, to singularities of the temperature dependences of the intensity of Raman and Rayleigh scattering on the OIM.

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