

# Manifestations of the Higher Moments of the Orientation Distribution Function of Molecules in the Spectral Properties of an Impurity Nematic

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**Abstract**—The polarized electronic absorption spectra, orientation ordering, and the special local field features were studied for push-pull linear dye molecules with strong donor–acceptor electronic conjugation of terminal fragments in the matrix of a nematic liquid crystal. The temperature-induced inversion of the sign of the splitting of polarized impurity absorption bands was observed. This effect was shown to be caused by the statistical character of orientation ordering of impurity molecules and manifestation of the higher moments of the orientation distribution function. The dependence of local field parameters (Lorentz tensor components) of impurity molecules on their orientation ordering was established. This dependence was used to reproduce the temperature dependence of the orientation order parameter of the matrix. © 2004 MAIK “Nauka/Interperiodica”.

## 1. INTRODUCTION

Orientation ordering of uniaxial molecules in a nematic liquid crystal with respect to director  $\mathbf{n}$  is statistical in character and is characterized by the moments  $\langle P_n(\cos\theta) \rangle$  of the orientation distribution function  $f(\theta)$ . Here,  $\theta$  is the angle between the longitudinal molecular axis  $\mathbf{l}$  and  $\mathbf{n}$ ,  $P_n(\cos\theta)$  represents even Legendre polynomials, and angle brackets denote statistical averaging. The degree of molecular ordering is determined by the  $\langle P_2 \rangle = S$  parameter, and the  $\langle P_n \rangle$  values with  $n \geq 4$  characterize inhomogeneity of the orientation distribution of molecules, which is described by the variances  $\Delta_{mq} = \langle P_m P_q \rangle - \langle P_m \rangle \langle P_q \rangle$ . The influence of the  $\langle P_n \rangle$  parameters on the physical properties of liquid crystals is of great interest for understanding the nature of the liquid crystalline state, development of molecular-statistical theory, and practical applications. For instance, the ratio between  $S$  and  $\langle P_4 \rangle$  determines the anisotropy of elastic deformation moduli of nematics  $K_{ii}$  [1] and the volt-contrast characteristics of liquid crystalline displays, the thresholds and periods of distortions of the director  $\mathbf{n}(\mathbf{r})$  field in nematics under field actions [2], the anisotropy of Leslie viscosity coefficients  $\alpha_i$  [3], the time characteristics of Fredericksz transitions [2], the amplification coefficients and threshold characteristics of lasers with dyes in nematic matrices as active media [4], the intensity of polarized Raman bands [5], the dichroism of two-photon absorption [6], the splitting of polarized absorption bands of

impurity molecules in a nematic phase [7], and other properties of liquid crystals. This has stimulated many works concerned with measuring  $\langle P_{2,4} \rangle$  by various methods and theoretical interpretation of the observed dependences of  $\langle P_4 \rangle$  on  $S$ .

Much less is known about the physical effects in liquid crystals caused by the  $\langle P_n \rangle$  moments with  $n \geq 6$ . The  $\langle P_{2-6} \rangle$  values determine the saturation parameters and the output generation power of lasers with dyes in nematic matrices as active media [4] and also the splitting of polarized Raman bands in nematic liquid crystals [8]. Apart from the physics of liquid crystals, manifestations of the  $\langle P_n \rangle$  moments with  $n \geq 6$  in anisotropic statistically ordered molecular media are of interest because the  $\Delta_{mq}(S)$  dependences at high  $S$  values are sensitive indicators of fine structural distinctions related to the appearance of medium structure anisotropy under external actions or as a result of molecular self-organization [9]. Recently, the first neutron diffraction measurements of  $\langle P_{2-6} \rangle$  in various liquid crystalline phases were performed [10]. The first physical effect caused by  $\langle P_{2-8} \rangle$  moment manifestations, the temperature inversion of the sign of the splitting of polarized electronic absorption bands of impurity dye molecules in a nematic phase, was reported in [11]. The present work is a complete study of this system taking into account the special features of orientation ordering of matrix and impurity molecules, the local field anisotropy for impurity molecules, and other aspects that had not been considered earlier.

## 2. OBJECTS OF STUDY AND EXPERIMENTAL PROCEDURE

The effects caused by  $\langle P_n \rangle$  moments with  $n \geq 4$  can be observed if we select a liquid crystal property that fully depends on the variances  $\Delta_{2n}$  with  $n \geq 2$ , because the  $\Delta_{2n}$  value contains contributions of the  $\sim \langle P_n \rangle$  and  $\langle P_{n \pm 2} \rangle$  terms. Such a property is the difference  $\Delta v = v_{\parallel} - v_{\perp}$  between the  $v_j$  maxima (splitting) of the  $D_j(v)$  electronic absorption bands of uniaxial impurity molecules in a nematic matrix when the light wave is polarized parallel ( $j = \parallel$ ) and normally ( $j = \perp$ ) to  $\mathbf{n}$  [12]. The  $v_j$  value for impurity absorption bands with the transition dipole moment  $\mathbf{d} \parallel \mathbf{l}$  is given by the equation [12]

$$v_j = v_i - S_m \sum_{n \geq 2} A_n(S_m) \left[ \langle P_n \rangle + \frac{C_j \Delta_{2n}}{1 + C_j S} \right], \quad (1)$$

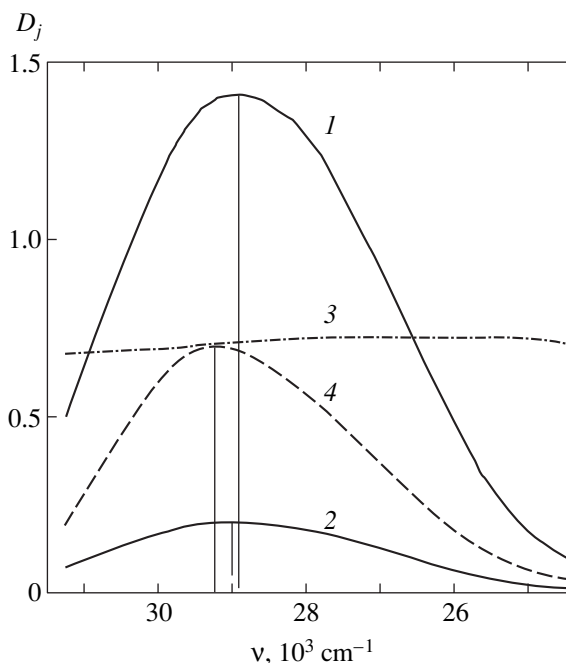
where the summation is over even  $n$ ,  $C_{\parallel} = 2$ ,  $C_{\perp} = -1$ ,  $v_i$  is the  $D_i(v)$  band maximum of impurity absorption in the isotropic liquid crystalline phase, and  $S_m$  is the orientation order parameter of matrix molecules. The  $A_n(S_m) = A_{n0} + A_{n1} S_m$  coefficients [7] characterize changes in the impurity–matrix anisotropic interaction energy under electronic excitation of the impurity, and magnitudes and signs of the  $A_{n0,1}$  parameters are determined by intermolecular interaction contributions of various types.

The splitting  $\Delta v$  is determined by the statistical character of orientation ordering of molecules in the nematic phase, and its value

$$\Delta v = - \frac{3S_m}{(1-S)(1+2S)} \sum_{n \geq 2} A_n(S_m) \Delta_{2n} \quad (2)$$

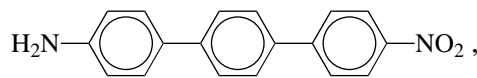
depends on the balance of the contributions of the variances  $\Delta_{2n}$  and the  $\langle P_q \rangle$  values with  $q \geq 4$ . The  $\Delta_{2n}$  parameters change as the temperature of the mesophase varies and are characterized by different dependences on  $S$  [9], which must manifest itself in the character of temperature variations of  $v_j$ . One of the possible effects is the temperature-induced inversion of the sign of  $\Delta v$  caused by mutual balancing of the  $\Delta_{2n}$  contributions to  $\Delta v$ , which is only possible if at least  $n = 2, 4$  and  $\langle P_{2-6} \rangle$  are taken into account. At the inversion point, the condition  $v_{\parallel} = v_{\perp} \neq v_i$  should be satisfied. This effect of the manifestation of the  $\langle P_n \rangle$  moments with  $n \geq 6$  is easy to distinguish from the temperature-induced inversion of the sign of  $\Delta v$  when only one term with  $n = q$  is included in (2) and  $A_q$  vanishes at this point, because the equality  $v_j = v_i$  is then satisfied.

It follows from (2) that the impurity molecules and nematic matrix should satisfy several requirements for the contribution of  $\Delta_{2n}$  with various  $n$  to  $\Delta v$  to be noticeable. For  $A_{n0,1}$  values to be large, the electronic excitation of molecules should be accompanied by strong

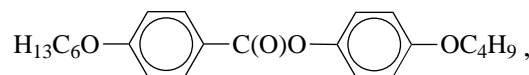


**Fig. 1.** Spectral dependences of the sample optical density components (1)  $D_{\parallel}(v)$  and (2)  $D_{\perp}(v)$  and (3) the dichroic ratio  $0.1D_{\parallel}(v)/D_{\perp}(v)$  in the nematic phase at  $\Delta T = -21.5$  K and (4)  $D_i(v)$  in the isotropic phase at  $\Delta T = 6$  K.

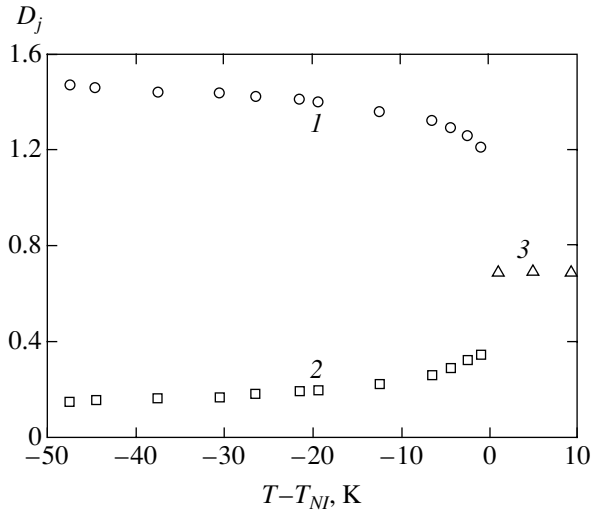
changes in quite a number of their properties, namely, the dipole moment and the anisotropy of linear and nonlinear polarizabilities. Impurity molecules should also have high  $S$  values in the nematic matrix for the  $\Delta_{2n}$  values with  $n \geq 4$  to be large. Such objects are long linear push-pull-type molecules with a system of  $\pi$  conjugated fragments that have terminal  $\pi$  electron-donor and acceptor substituents bound by polar conjugation through a system of molecular fragments. We used the K-6 dye [13],



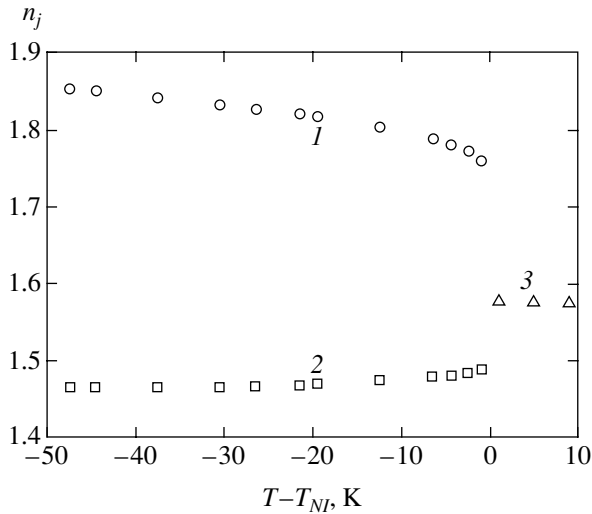
which satisfied the above requirements. The nematic matrix should have high  $S_m$  values for the  $A_n(S_m)$  and  $\Delta v$  values to be maximum and broad nematic phase and  $S_m$  variation ranges. These requirements are satisfied by 4-butoxyphenyl 4'-hexyloxybenzoate liquid crystals (BEHA [13]),



with the crystal–nematic–isotropic liquid (Cr–N–I) phase transition temperatures of 50 and 102.5°C, respectively.



**Fig. 2.** Temperature dependences of the optical densities (1)  $D_{\parallel}$ , (2)  $D_{\perp}$ , and (3)  $D_i$  for dye K-6 in the BEHA matrix.



**Fig. 3.** Temperature dependences of the refractive indices (1)  $n_{\parallel}$ , (2)  $n_{\perp}$ , and (3)  $n_i$  of the BEHA matrix at  $\lambda = 345$  nm.

The spectra of the  $D_j(\nu)$  polarized optical density components of the sample in the region of the electronic absorption of K-6 were measured in a plane-parallel cell of thickness  $d = 40 \mu\text{m}$  with a uniform planar director orientation at a 0.3 wt % dye concentration, which had no noticeable effect on the  $T_{NI}$  value, sample birefringence, and the degree of matrix ordering. The spectra were recorded and automatically processed on a PU-8800 spectrophotometer. Weak background absorption of the pure matrix in the high-frequency wing of the impurity band was subtracted from the absorption of impurity liquid crystals at equal  $d$  and reduced temperature  $\Delta T = T - T_{NI}$  values for each spectrum component. The resulting  $D_j(\nu)$  spectra were used to determine  $D_j(\nu_j)$  and  $\nu_j$  for the nematic and isotropic

phases. Such  $D_j(\nu)$  spectra obtained at  $\Delta T = -21.5$  K and the  $D_j(\nu)$  spectra at  $\Delta T = 6$  K are shown in Fig. 1. The isolated nondegenerate long-wave transition in K-6 is polarized along the long molecular axis, and the dichroism  $N_1(\nu) = D_{\parallel}(\nu)/D_{\perp}(\nu)$  is virtually independent of  $\nu$  within the absorption band, which is evidence of its uniform polarization in the absence of latent unresolved vibronic transitions.

### 3. ORIENTATION ORDERING OF IMPURITY MOLECULES AND LOCAL FIELD ANISOTROPY

The temperature dependences of the  $D_j = D_j(\nu_j)$  and  $D_i = D_i(\nu_i)$  parameters are shown in Fig. 2. At every  $\Delta T$  value, the dichroic ratios  $N_1 = D_{\parallel}/D_{\perp}$  and  $N_2 = D_{\perp}/D_i$  were used to consistently determine  $S$  and the Lorentz tensor  $L_j$  components by the procedure described in [14]. The  $S$  value is given by the equation

$$S = \frac{N_1 g_1 - 1}{N_1 g_1 + 2}, \quad (3)$$

and the  $L_j$  components, by the system of equations

$$N_2 g_2 (N_1 g_1 + 2) = 3, \quad \text{Sp} \mathbf{L} = 1. \quad (4)$$

The correction factors have the form

$$g_1 = \frac{n_{b\parallel} f_{b\perp}^2}{n_{b\perp} f_{b\parallel}^2}, \quad g_2 = \frac{n_{b\perp} \rho_i f_{bi}^2}{n_{bi} \rho f_{b\perp}^2}. \quad (5)$$

Here,  $n_{bj} = n_j(\nu_j)$  is the background refractive indices in the impurity absorption band, which coincide with the refractive indices of the matrix because of the low concentration of impurity molecules,  $f_{bj} = 1 + L_j(n_{bj}^2 - 1)$  is the background components of the light wave local field for impurity molecules in their absorption band, and  $f_{bi} = (n_{bi}^2 + 2)/3$ . The  $n_j$  values at a  $\lambda = 345$  nm wavelength (the mean wavelength of the maxima of the  $D_{i,j}(\lambda)$  bands) obtained as in [7] are shown in Fig. 3. The densities  $\rho$  and  $\rho_i$  for the nematic and isotropic BEHA phases were taken from [13].

The  $S(\Delta T)$  dependence shown in Fig. 4 is well approximated by the equation

$$S = S_0(1 - T/T_1)^\beta \quad (6)$$

with the parameters  $S_0 = 1.047$ ,  $\beta = 0.127$ , and  $T_1 - T_{NI} = 0.18$  K. Figure 4 shows that the experimental  $L_{\perp}$  values in the nematic matrix phase are substantially different from  $L_i = 1/3$  for the isotropic phase, and the  $L_{\perp}(S)$  dependence is well approximated by the equation

$$L_{\perp} = a + bS \quad (7)$$

with the parameters  $a = 0.343$  and  $b = 0.056$ . The  $f_{b\perp} > f_{b\parallel}$  components differ weakly from each other at  $\lambda = 345$  nm over the whole nematic phase interval because of the mutual balancing of the contributions of the tensor  $\mathbf{L}$  anisotropy and matrix birefringence to the anisotropy of the tensor  $\mathbf{f}_b$ . For the K-6 dye, the inclusion of the local field anisotropy insignificantly increases  $S$  compared with the value obtained in [11] by (3) in the isotropic approximation  $f_{b\parallel} = f_{b\perp}$ .

The  $S_m(\Delta T)$  dependence is not known for the BEHA matrix. In [11, 13], it was identified with the  $S(\Delta T)$  dependence for impurity molecules of dye K-2 (4-dimethylamino-4'-nitroazobenzene,  $\lambda_{\max} = 490$  nm) obtained from the dichroism  $N_1$  of its absorption band without taking the tensor  $\mathbf{f}_b$  anisotropy into account. Let us show that the experimental  $S(\Delta T)$  and (7) dependences can be used to determine the  $S_m(\Delta T)$  function for the nematic matrix. The dependence [14]

$$L_{\perp} = 1/3 + CS_m \quad (8)$$

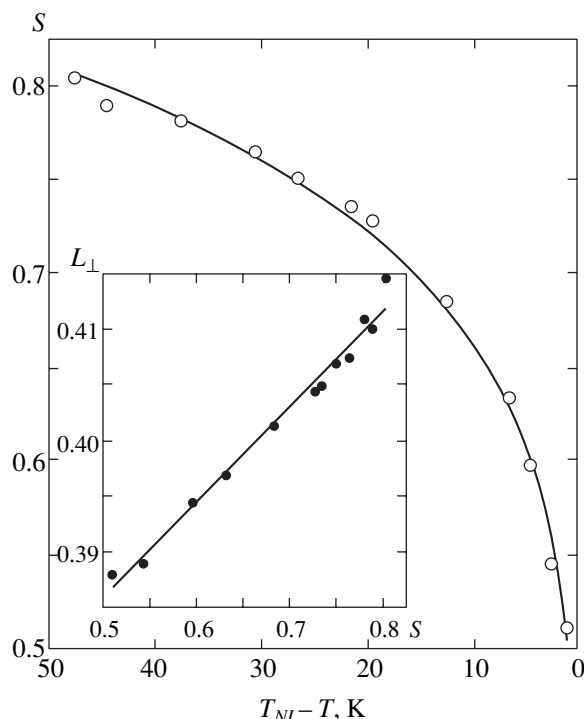
and the universal relation [15]

$$S(\Delta T) = AS_m(\Delta T) + B, \quad (9)$$

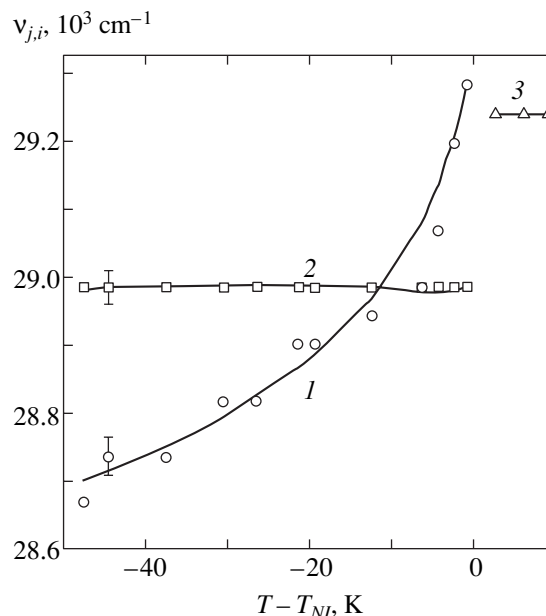
which is valid for all impurity nematics studied thus far, lead to empirical formula (7) with the parameters  $a = 1/3 - bB$  and  $b = C/A$ . Substituting  $B = (1 - 3a)/3b$  into (9) yields the dependence  $AS_m(\Delta T)$ . Approximating this dependence by (6) allows the  $AS_{m0}$  product and  $A$  parameter to be determined (in the natural limit  $S_{m0} = 1$ ). Applying this procedure to BEHA yields  $B = 0.116$ ,  $AS_{m0} = 1.191$ ,  $\beta = 0.118$ , and  $T_1 - T_{NI} = 0.6$  K. At  $S_{m0} = 1$  and  $A = 1.191$ , the  $S_m(\Delta T)$  values insignificantly differ from  $S(\Delta T)$  for K-6 molecules over the whole range of the existence of the nematic phase and are somewhat underestimated at  $\Delta T > 35$  K, whereas the  $S$  values for K-2 molecules [11, 13] are slightly higher than those for K-6 in this temperature range.

#### 4. THE SPECIAL FEATURES OF CHANGES IN $v_j(\Delta T)$ AND THEIR INTERPRETATION

The  $v_j(\Delta T)$  dependences shown in Fig. 5 are characterized by certain features that have not been observed earlier for transitions with  $\mathbf{d} \parallel \mathbf{l}$  [7, 12]. The temperature inversion of the sign of  $\Delta v$  is observed at  $\Delta T = \Delta T^* = -6.5$  K, while the  $v_{\perp}$  value is constant over the whole mesophase interval, and a strong  $v_{\parallel}(\Delta T)$  dependence is observed with a change in the ratio between  $v_j$  and  $v_{\parallel}$  close to  $T_{NI}$ . The  $D_{\parallel}(v)$  and  $D_{\perp}(v)$  bands correspond to the absorption of orthogonally polarized normal light waves in the liquid crystal that do not interact with each other. As a result, exact degeneracy  $v_{\parallel} = v_{\perp}$  is observed at  $\Delta T^*$ . The difference  $v_i > v_j$  at  $\Delta T^*$  is evidence of the presence in (2) of contributions of variances  $\Delta_{2n}$  with several values of  $n \geq 2$  and mutual balancing of these

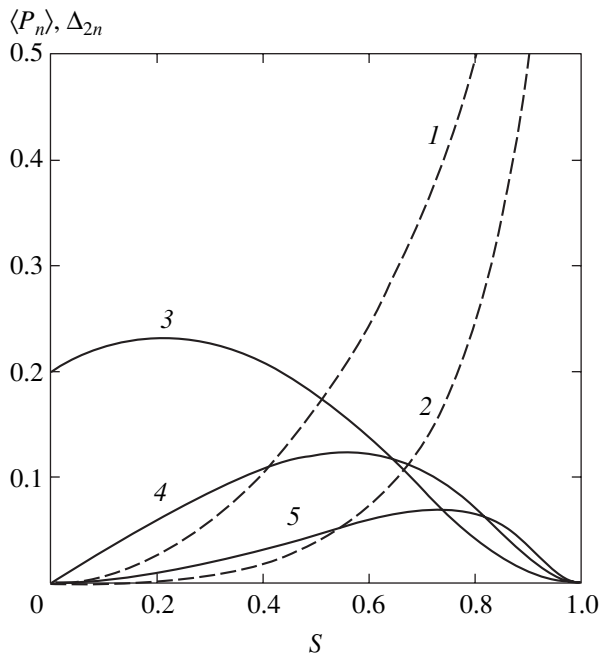


**Fig. 4.** Dependences  $S(\Delta T)$  and  $L_{\perp}(S)$  for K-6 molecules in the BEHA matrix (open and solid circles) and their approximations by (6) and (7) (lines).



**Fig. 5.** Temperature dependences of the maxima (1)  $v_{\parallel}$ , (2)  $v_{\perp}$ , and (3)  $v_i$  of  $D_{j, i}(v)$  absorption bands of dye K-6 in the BEHA matrix. Solid lines were obtained by interpolating the  $v_j$  values calculated by (1) with the parameters given in text.

contributions to  $\Delta v$  at this point. Figures 2–4 show that the  $S(\Delta T)$  dependence and the ensuing temperature dependences of the  $D_j$ ,  $n_j$ , and  $L_{\perp}$  parameters do not have singularities in the neighborhood of the  $\Delta T^*$  point,



**Fig. 6.** Dependences of (1)  $\langle P_4 \rangle$ , (2)  $\langle P_6 \rangle$ , (3)  $\Delta_{22}$ , (4)  $\Delta_{24}$ , and (5)  $\Delta_{26}$  on  $\langle P_2 \rangle = S$  for distribution function (10).

which is also evidence of contributions of moments  $\langle P_n \rangle$  with  $n \geq 4$  to  $\Delta v$  sign change. Note that, for many impurity systems studied thus far, taking into account terms with  $n = 2$  in (1) was sufficient for describing  $v_j(S)$  dependences (see [7, 12] and the references therein).

The  $\langle P_n \rangle$  parameters and  $\Delta_{2n}$  variances in (1) were calculated using the distribution function

$$f(\theta) = \frac{1}{Z} \exp[\lambda_2(S)P_2(\cos\theta)],$$

$$Z = \int_{-1}^1 \exp[\lambda_2(S)P_2(\cos\theta)] d\cos\theta, \quad (10)$$

which corresponds to experimental distributions when  $S$  values are high and native and impurity molecules are fairly long and do not contain terminal alkyl chains [7, 10, 12]. The  $\lambda_2(S)$  parameters were found by the equation  $S = \partial \ln Z / \partial \lambda_2$  and then used to calculate the  $\langle P_n \rangle(S)$  and  $\Delta_{2n}(S)$  dependences shown in Fig. 6. Because of the closeness of the  $S$  and  $S_m$  values at equal  $\Delta T$  values (see above), it was assumed that  $S_m = S$  in (1).

An analysis showed that including terms with  $n = 2$  and 4 in (1) and the determination of the  $A_{n0,1}$  adjustment parameters from two pairs of  $v_{\parallel, \perp}$  values at two  $\Delta T$  points or from  $v_{\perp}$  values at four  $\Delta T$  points gave a linear  $v_{\parallel}(\Delta T)$  dependence with  $\Delta v$  sign change at  $\Delta T$  close to the middle of the mesophase temperature interval. Including terms with  $n = 2-6$  into (1) substantially

improved agreement between theory and experiment. The  $v_j(\Delta T)$  dependences corresponding to the smallest mean-square deviation of the calculated  $v_j(\Delta T)$  values from experimental ones with the parameters  $A_{20} = 1855$ ,  $A_{21} = 2348$ ,  $A_{40} = -20123$ ,  $A_{41} = 13867$ ,  $A_{60} = 36773$ , and  $A_{61} = -35260 \text{ cm}^{-1}$  are shown in Fig. 5. Agreement between theory and experiment is evidence of mutual consistency of all special features of changes in  $v_j(\Delta T)$  and the ratios between them and  $v_i$  mentioned above. Even an insignificant deviation of the calculated  $v_{\perp}$  values at  $\Delta T = -6.5$  and  $-4.5$  K toward lower values compared with experiment corresponds to substantial exaggeration of calculated  $v_{\parallel}$  values at the same temperatures. On the whole, agreement between the calculated and experimental  $v_j(\Delta T)$  dependences obtained in this work is similar to that reported [11], where  $S$  values for dye K-2 were used as  $S_m$  [13] and the local field anisotropy was ignored in determining  $S$ . The  $A_{n0,1}$  parameters given above are also close to those obtained earlier [11]. These observations are evidence of a weak influence of the characteristics of changes in  $S_m(\Delta T)$  and  $S(\Delta T)$  on  $\Delta v$  sign change for impurity molecules.

The largest discrepancy between the theoretical and experimental  $v_j(\Delta T)$  values is observed at the inversion point  $\Delta T^*$ ; the discrepancy rapidly decreases as the system departs from this point. Figure 6 shows that, in the interval  $S = 0.5-0.8$ ,  $\Delta_{22}(S)$  monotonically decreases as  $S$  increases, whereas the  $\Delta_{24}(S)$  and  $\Delta_{26}(S)$  dependences are nonmonotonic and pass maxima at  $S = 0.55$  and  $0.74$ , respectively. As the experimental  $S(\Delta T^*) = 0.633$  value coincides with the value  $0.638$  at which the  $\Delta_{22}(S) - \Delta_{24}(S)$  difference changes sign from positive to negative as  $S$  increases, the discrepancy between theory and experiment in the vicinity of  $\Delta T^*$  can be related to a small difference between the real distribution function and model (10). Such differences have been reported for low  $S$  values [5, 7, 14].

To summarize, the temperature-induced inversion of the sign of  $\Delta v$  and the other observed characteristics of changes in  $v_j(\Delta T)$  are caused by the statistical character of orientation ordering of impurity molecules and manifestations of the  $\langle P_n \rangle$  moments of the orientation distribution function with  $n = 2-8$ . Distribution function (10) can serve as a good approximation for theoretically modeling the physical properties of nematics in the region of high  $S$  values. The spectral characteristics of polarized impurity absorption and luminescence [9] are effective and are the only indicators of fine structural features of anisotropic statistically ordered molecular media known thus far.

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