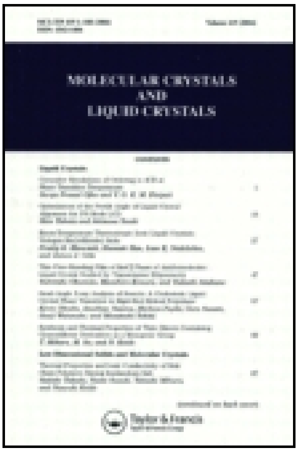

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Orientational Order and Elastic Deformations in Nematic Texturfs of bis-4-Cyanobiphenyloxyalkanes in an Electric Field

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ORIENTATIONAL ORDER AND ELASTIC DEFORMATIONS IN NEMATIC TEXTURES OF BIS-4-CYANOBIPHENYLOXYALKANES IN AN ELECTRIC FIELD

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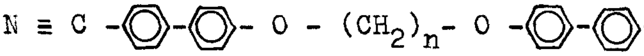
Abstract Using the method of orientational elastic deformations in an electric field we investigated the electrooptical properties of nematics of the bis-4-cyanobiphenyloxyalkanes series differing in the number n of CH_2 groups in the oxymethylene chain of the molecule. The value of optical anisotropy of nematics ($\Delta n = n_e - n_o$) and threshold electric potentials V_0 of deformations of their monodomain planar texture were determined at different temperatures. In both dependences of Δn and I/V_0 on n strong odd-even effect is manifested. However, the characters of alternations observed in them differ greatly. These differences are due to the fact that the value and sign of I/V_0 depend not only on the intermolecular (nematic) orientational order but is determind to a much greater extent by intramolecular order which depends on the polar and polarizing architecture of the molecule.

INTRODUCTION

The molecules of 4,4'-biscyanobiphenyloxyalkanes (BCBOA)

$N \equiv C - C_6H_4 - C_6H_4 - O - (CH_2)_n - O - C_6H_4 - C_6H_4 - C \equiv N$ are very suitable chemical compounds representing fragments of an LC polymer mesogenic in the main chain. They make it possible to follow the effect of the spacer length and flexibility on the mesophase properties. In a recent paper[I] these compounds have been studied by the method of equilibrium electric birefringence in dilute solutions for establishing the relationship between the length of the alkyl chain and the intramolecular polar orientational order in the molecule. In the present work these substan-

ces (BCBOA) are investigated in the nematic phase by the method of threshold elastic deformations in electric fields[2] in order to elucidate the effect of alkyl chain length and flexibility on the degree of intermolecular orientational order and the electrooptical properties of the nematic LC formed in the BCBOA mass.

EXPERIMENTAL

All BCBOA obtained by us form nematic LC, and their isotropization temperatures T_{NI} determined with the aid of a polarizing microscope are given in Table I.

Electrooptical BCBOA characteristics were studied in plane-concave layers up to 5.10^{-3}cm thick by the procedure described in detail.[2] On glass surfaces all investigated BCBOA nematics spontaneously form layers with a planar director orientation, and their homogeneous (monodomain) texture was attained by rubbing the glasses in the desired direction. In order to observe the polarizing-microscope patterns of the samples, a mercury lamp with a green light filter (wavelength $\lambda = 5.46.10^{-5}$cm) was used as the light source. The process of nematics reorientation in the electric field was carried out by the action of sinusoidal fields in the frequency range from 10 to 500kHz at potential difference at the electrodes up to 700V. In the layer of BCBOA nematic n=2 the dielectric anisotropy of which was negative, orientational deformation was accomplished by combined action of electric and magnetic fields by the method developed and used previously.[3]

RESULTS AND DISCUSSION

Optical Anisotropy

The results obtained for BCBOA in the absence of the electric field are illustrated in Figure 1a which shows the polarizing-microscopic pattern of the planar layer of the

BCBOA nematic n=IO at a temperature $\Delta T=30^{\circ}C$ ($\Delta T=T_{NI}-T$) with crossed polarizers. A system of concentric interference bands can be seen in the Figure.

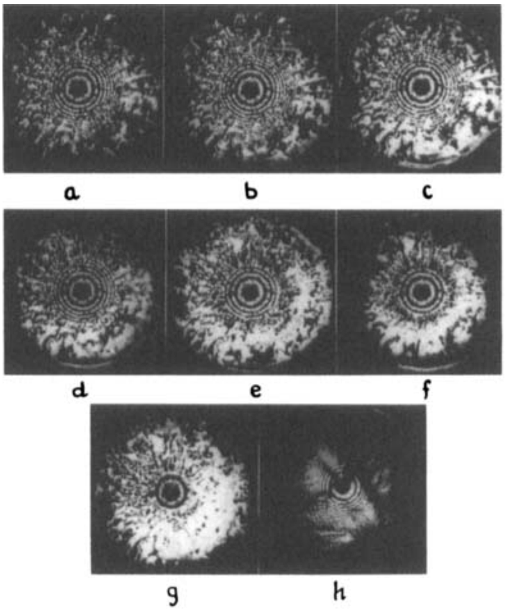


FIGURE I Polarization-microscopic pattern from a plano-concave layer of the BCBOA, n=IO. (a-g) - in the nematic state, h - in the crystalline state, a - the director initial alignment parallel to the preparation plane (E=0), (b - g) - the texture is deformed by an electric field $E.10^{-3}$V/cm=0.7(b), I,27 (c), 2.II(d), 3.24(e), 4.23(f), 7.32(g).

By using them it is possible to plot the dependences of relative retardation $\Delta\lambda/\lambda$ on the layer thickness z ($z=r^2/2R$ where r is the radius of the corresponding dark ring) at a constant temperature T. This plot is illustrated in Figure 2. It is possible to calculate from the slopes $(\Delta\lambda/\lambda)/z$ of straight lines in Figure 2 the value of birefringence Δn of the nematic ($\Delta n=n_e-n_o$ where n_e and n_o are the refractive indices of the extraordinary and ordinary beams, respectively) according to the Equation

$$\Delta n = (\Delta\lambda/\lambda)(\lambda/z) \tag{I}$$

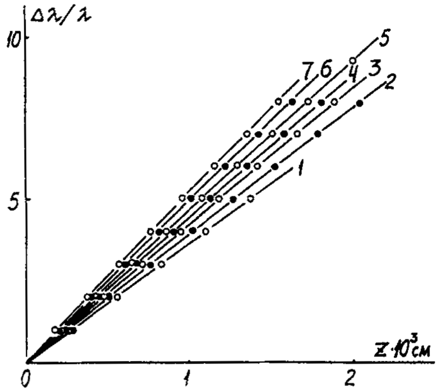


FIGURE 2 Relative retardation $\Delta\lambda/\lambda$ vs layer thickness z for a BCBOA, n=3 at various temperatures $\Delta T^\circ C$=2(I), 5(2), 8.5(3), II(4), I6.5(5), 23(6), 28.5(7).

The values of Δn obtained in this way are shown in Figure 3 in the form of the dependence of Δn on ΔT for some BCBOA. The data in Figure 3 may be considered by using Eq. (2) relating Δn of the nematic to its molecular weight M and density ρ and to the polarizability anisotropy $\Delta\alpha$ of its molecules[4]

$$\Delta n = (2\pi/3n)(n^2 + 2)N_A(\Delta\alpha/M)\rho S \tag{2}$$

where n is the average refractive index of the substance and S is the degree of its nematic orientational order. Among the values contained in the right-hand side of Eq. (2) the values of $(n^2 + 2)/n$, ρ , and S are temperature - dependent. Each of them increases with increasing ΔT. However, the main contribution to this increase is provided by the temperature change in S. Hence, in the first approximation the temperature dependence of the optical anisotropy Δn of the nematic may be identified with that of its orientational order S. The value of $\Delta\alpha$ in Eq. (2) for aromatic compounds (including BCBOA) is determined to a great extent by the anisotropy of polarizability of the mesogenic part of the molecule. Therefore, the value of S determined from the Δn value should be regarded mainly as the characteristic of orientational order of aromatic mesogenic fragments of molecules in the nematic.

As follows from the data in Figure 3, the values of Δn which in the investigated ΔT range (from 2° to 100°) for different BCBOA vary over a wide range from 0.19 to 0.38 not only depend on temperature but are also essentially different for homologues with different lengths of the oxyalkyl spacer.

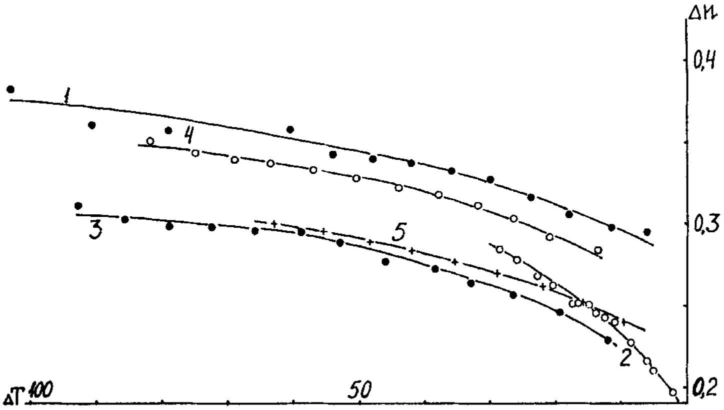


FIGURE 3 Temperature dependence of the birefringence Δn for some BCBOA; n=2(1), 3(2), 5(3), 6(4), 10(5).

In discussing the dependence of Δn on the number of CH_2 groups in the oxyalkyl chain, it is reasonable to compare different BCBOA homologues at the same relative temperature $\tau = \Delta T/T_{NI}$. Table I gives the anisotropies Δn of BCBOA investigated here, which are determined at a relative temperature $\tau = 0.057$. The same data are shown in Figure 4 as the dependence of Δn on the number n of CH_2 groups in the oxyalkyl chain of the molecule.

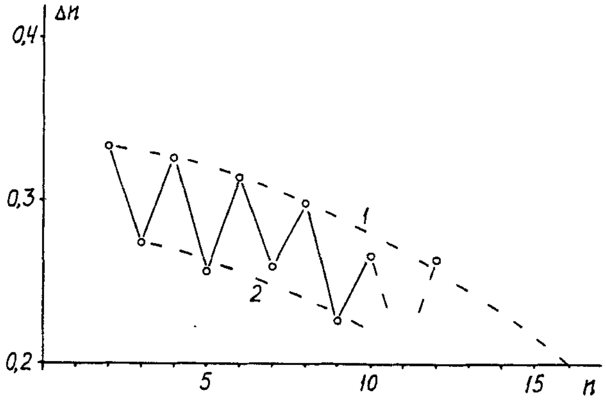


FIGURE 4 Dependence of birefringence Δn for BCBOA on the number of methylene groups n in the oxyalkyl chain, at the relative temperature $\Delta T/T_{NI} = 0.057$. Dashed lines I and 2 characterize this dependence for even n and for odd n respectively.

A characteristic feature of curves in Figure 4 is a very pronounced odd-even effect which is manifested in increasing Δn when an even CH_2 group is added to the oxyalkyl chain and in its decrease when an odd group is introduced. According to Eq. (2), these periodical changes in Δn reflect the alternation in the degree of orientational order S of the mesophase with increasing chain length.

The manifestations of the odd-even effect in optical anisotropy are also well known for other low molecular weight nematics. However, for these LC odd-even alternations of Δn are much less pronounced than those observed by us for BCBOA. This fact may be understood if we take into account the difference in the molecular structure of BCBOA and that of usual low molecular weight LC the molecules of which consist of a rigid mesogenic (usually aromatic) core and one or (more often) two flexible end chains adjoining it. In contrast, in BCBOA molecules flexible methylene chain is the core, whereas rigid mesogenic biphenyl groups are the terminal parts of the molecule. In molecules with the mesogenic core the increase in the length of the alkyl end chain (the predominant conformation of which in a nematic is the trans-chain) per CH_2 group is accompanied by a change in the angle between the para-aromatic axis of the mesogenic core and the C - C end bond of the chain. This, it turn, leads to a change in the orientational order S in the LC medium. In contrast in BCBOA molecules the increase in the length of the core alkyl chain per CH_2 group is accompanied by a change in the angle formed by para-aromatic axes of two mesogenic cyanobiphenyl end parts of the molecule, which leads to a more drastic change in the orientational order in LC than that in the case of the molecules with the mesogenic core.

Another feature distinguishing the properties of BCBOA from those of the molecules with the mesogenic core is the relatively slight change in the amplitude of periodical changes in Δn (and, hence, in S) in Figure 4 with increasing length of the alkyl chain. This difference in properties may imply that in a medium with the nematic order the presence of two mesogenic cyanobiphenyl fragments at the ends of the methylene chain of the molecule favours the retention in it of a considerable number of trans-conformers not only for the lower but also for the higher members of the BCBOA homologous series. It can be clearly seen in Figure 4 that the values of Δn in the series of both even and odd homologues decrease with increasing number of CH_2 groups in the alkyl chain. This decrease in Δn illustrated by broken curves I and 2 in Figure 4 may be associated with the fact that the increase in the length of the alkyl chain in the BCBOA molecule leads to a decrease in its specific anisotropy

$\Delta\alpha/M$ contained in Eq. (2). In fact, the main contribution to the difference between two main polarizabilities $\Delta\alpha$ of the BCBOA molecule is provided by two optically anisotropic cyanobiphenyl groups. This contribution does not change with increasing length of the alkyl chain in the homologous series, whereas molecular weight M increases. This should result in a decrease in $\Delta\alpha/M$ and, correspondingly, in Δn of the nematic with retention of invariable S value.

The samples of some of BCBOA investigated after crystallization retain a relatively distinct system of interference rings (Figure I), which makes it possible to determine the value of Δn for these BCBOA in the crystalline phase by using the plot in Figure 2 and Eq. (I). Thus, for BCBOA n=IO, Δn was found to be equal to 0.453. Now if it is assumed that BCBOA n=IO after crystallization forms a uniaxial crystal, it is possible to estimate the absolute value of the degree of intermolecular orientational order S of its mesophase (strictly speaking S is proportional to the ratio of anisotropies of molar refractions of the nematic and the crystal) from the ratio of Δn of the nematic phase to that of the crystal. For BCBOA n=IO the value of S determined in this way ranges from 0.52 to 0.66 at temperature variation, ΔT, from IO°C to 63°C. These values of S may be considered quite reasonable if they are compared to the value of S = 0.5 determined by the NMR method for BCBO-decane at T_{NI}.[5]

The value of S for BCBOA n=IO at a temperature τ = 0.057 may be estimated from the data in Figure 3 (curve 5) and the value of Δn=0.453 for the crystal. This value of S was found to be 0.59. The same value may also be taken for all other even homologues at τ = 0.057 if the considerations applied above in discussing the data in Figure 4 are taken into account. The values for odd homologues at the same temperature were estimated from the values of jumps in Δn at the odd-even transitions (Figure 4). The values of S are given in Table I.

TABLE I Isotropization temperatures T_{NI} and electrooptical characteristics of the BCBOA nematics (with the number of methylene groups n in the alkyl chain) at the relative temperature $\tau = T/T_{NI} = 0.057$.

n	2	3	4	5	6	7	8	9	IO	I2
T_{NI}(K)	543	442	528	459	49I	455	478	449	460	444
Δn	0.333	0.274	0.326	0.256	0.3I5	0.260	0.298	0.227	0.266	0.264
S	0.59	0.49	0.59	0.47	0.59	0.50	0.59	0.47	0.59	0.59
V_0(V)	-6.I	2	8.45	2.48	3.48	I.63	2.I2	I.75	I.76	I.66
I/V_0^2S	-0.043	0.5I	0.024	0.34	0.I4	0.74	0.37	0.69	0.55	0.6I

Elastic Deformations and Dielectric Properties

The results obtained for BCBOA by the method of elastic deformations in electric fields are illustrated in Figure I (b-g) which shows the polarizing microscopic patterns of the initally planar layer of the BCBOA n=IO nematic the texture of which is deformed by electric fields with different strengths E. It can be seen from Figure I that the appearing deformation is the splay at which the system of interference rings is distorted in the range of layer thickness $z \geqslant z_k$. This fact corresponds to the threshold character of the BCBOA molecules reorientation and indicates that the molecular longitudional axes are aligned along the electric field direction, according to the positive dielectric anisotropy of the nematic. Similar results were obtained for all other BCBOA (except BCBOA n=2). From the patterns obtained it is possible to calculate the value of critical (threshold) potential V_0 of BCBOA by measuring the radius r_k of the critical boundary from the Equations

$$V_0 = z_k E \quad ; \quad z_k = r_k^2/2R \qquad (3)$$

where R is the radius of the lens curvature, z_k is the thickness of the layer corresponding to the critical boundary, and E is the electric field strength. The values of V_o determined in this way for BCBOA n= 3 - I2 at temperature $\tau = T/T_{NI} = 0.057$ are listed in Table I.

In contrast to all other homologues of the BCBOA series investigated, the dielectric anisotropy of the BCBO-ethane nematic (n=2) was negative. Hence, to carry out deformation in the planar layer of this LC, a method was applied in which the sample was subjected to the combined action of the magnetic and electric fields which were directed normal to the plane (and, correspondingly, to the director) of the sample. With this geometry of the experiment the condition of the equilibrium threshold deformation is determined from Eq. (4)[3]

$$\Delta\chi H^2 + (\Delta\varepsilon /4\pi)E^2 = \pi^2 K_I/z_k^2 \qquad (4)$$

where H and E are the strength of the magnetic and electric fields, respectively, $\Delta\chi$ is the difference between the main magnetic susceptibilities (magnetic anisotropy) of the nematic, $\Delta\varepsilon$ is the difference between its main dielectric permittivities (dielectric anisotropy) and K_I is the splay elasticity constant for LC.

In the first experimental stage the sample texture was deformed by the magnetic field in the absence of the electric field (E=0 in Eq. (4)) and the Freedericksz constant F[6] was determined from the experimental values of H and z_k

$$F = z_k H = \pi(K_I/\Delta\chi)^{I/2} \qquad (5)$$

The value of F for BCBOA n=2 at a temperature ΔT = 3I°C (τ = 0.057) was equal to 4.5cm.Gauss.

In the second experimental stage by using the combined effect of H and E values of the fields we determined the electric field strength E at which the initial planar texture of the nematic deformed by the magnetic field H was restored by the opposing field E (in Eq. (4) $I/z_k=0$). For BCBOA n=2 (at $\tau=0.057$) the ratio of these "equivalent" fields was $E/H = 2(\pi\Delta\chi/|\Delta\varepsilon|)^{I/2} = I.36 V.cm^{-I}.Gauss^{-I}$. The substitution of $H = F/z_k$ gives $z_k E = I.36 V.cm^{-I}.Gauss^{-I}$ or, applying the numerical value of F obtained by us, we find the threshold deformation potential of BCBOA n=2 in the electric field

$$V_o = z_k E = 2\pi^{3/2}(K_I/|\Delta\varepsilon|)^{I/2} = -6.IV \qquad (6)$$

The negative sign of V_o denotes conditionally that $\Delta\varepsilon$ of the nematic is negative.

The dependence of V_o on ΔT is shown in Figure 5 for some BCBOA.

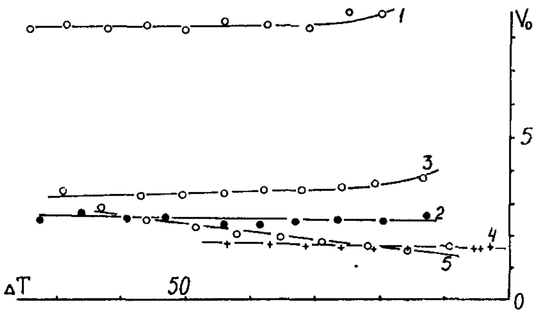


FIGURE 5 Dependence of the threshold potential V_o on the temperature ΔT for some BCBOA at n=4(I), 5(2), 6(3), 7(4), I0(5).

The curves in Figure 5 show that for lower homologues this dependence is very slight but is considerable for higher homologues, e.g. at n=I0 (curve 5) and is manifested as a decrease in V_o with increasing temperatures. If Eq. (6) is applied, this means that the elasticity constant K_I for this nematic decreases more markedly with increasing T than its dielectric anisotropy $\Delta\varepsilon$, which is typical of low molecular weight unassociated nematics the molecules of which contain a rigid mesogenic core.[2,3] The alkyl chain core of the BCBOA n=IO molecule is not rigid and not mesogenic. However, just its high length and flexibility can favor the similarity of some properties of this molecule in the mesophase and those of a system of two molecules with rigid mesogenic cores. The temperature dependence of $K_I/\Delta\varepsilon$ described by curve 5 in Figure 5 is probably one of these properties.

It can be seen from the data in Figure 5 that at a constant temperature the value of V_0 for different BCBOA depends markedly and non-monotonically on the length of the alkyl spacer in their molecules, this dependence being even accompanied by a change in the V_0 sign. This fact shows that the main contribution to the dependence of V_0 on the number of CH_2 groups in the alkyl chain is provided by the change in the value and sign of the dielectric anisotropy $\Delta\varepsilon$ of the nematic (although according to Eq. (6) variations in its elasticity constant K_I can also be of some importance). Therefore, subsequently the data obtained will be discussed in terms of the dependence of I/V_0 on n because it is this dependence that can (at least qualitatively) characterize the corresponding change in $\Delta\varepsilon$.

Table I gives the values of V_0, and curve I in Figure 6 describes the dependence of the corresponding values of I/V_0 on the number n of CH_2 groups in the alkyl chain of the molecule.

Curve I in Figure 6 clearly illustrates the pronounced periodicity of the change in I/V_0 with increasing n, which is accompanied by a change in sign on passing from n=2 to n=3. This periodicity is a manifestation of the odd-even effect in dielectric properties of the BCBOA nematics and is related to the change in the degree of order in the system under investigation. However, if the curve in Figure 6 is compared to that in Figure 4 which describes the dependence of anisotropy Δn (and, correspondingly, of the degree of nematic order S) on n, essential difference eetween these two curves can be seen. The value of Δn (and, hence, that of S) is higher for BCBOA homologues with even n, and, conversely, the value of I/V_0 (and, hence, that of $\Delta\varepsilon$) is higher for homologues with odd n.

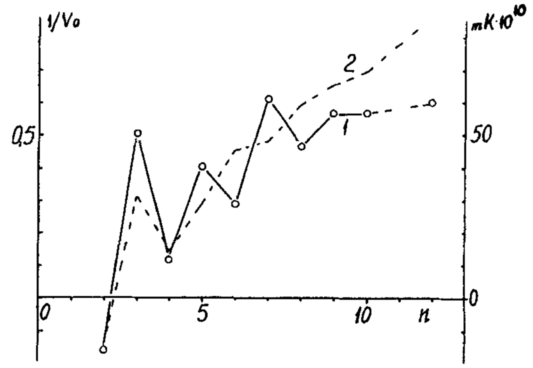


FIGURE 6 Dependence of the inverse threshold potential I/V_0 for the BCBOA nematics (I) and the molar Kerr constant ${}_mK$ for BCBOA benzene solutions (2) on number of methylene groups n in the oxyalkane chain.

Moreover, for both even and odd series of homologues the increase in n in Figure 6 is accompanied on the average by an increase in positive dielectric anisotropy $\Delta\varepsilon$, whereas positive optical anisotropy Δn for the same homologues decreases with increasing n.

These differences in the curves of the dependence of Δn on n and I/V_0 (or those of $\Delta\varepsilon$) on n illustrate clearly the differences in molecular characteristics determining the optical and dielectric properties of the nematics being investigated. According to Eq. (2), the optical anisotropy Δn of the mesophase is determined by the anisotropy $\Delta\alpha$ of its molecules and the degree of intermolecular orientational (nematic) order S, whereas dielectric anisotropy $\Delta\varepsilon$ depends not only on these two structural characteristics but is also determined to a considerable extent by the dipole architecture of the molecules, i.e. their intramolecular orientational-polar order (IOPO).

The theory of dielectric properties of nematic LC[7] in which a rigid particle with the axial symmetry of dielectric properties, the anisotropy of polarizability $\Delta\gamma$, and dipole moment μ forming an angle θ with the symmetry axis is a model for the molecule, leads to Eq. (7) for the dielectric anisotropy of the nematic

$$\Delta\varepsilon = 4\pi N_A PQXS \tag{7}$$

where P and Q are the constant factors of the internal field, S is the degree of nematic order, and function

$$X = \Delta\gamma + (\mu^2/2kT)(3\cos^2\theta - 1) \tag{8}$$

characterizes the IOPO in the nematic. The expression for X (in which the factor $\Delta\alpha$ is neglected) is identical with the equation of the molar Kerr constant ${}_mK$ for an assembly of polar anisotropic molecules in solution. This expression has been discussed[1] in the analysis of data on electric birefringence (EB) in BCBOA solutions. It has been shown that in the BCBOA homologous series both the equilibrium and the kinetic flexibility of the oxyalkane chain of the molecule are of considerable importance in the EB phenomenon. In this case for lower homologues the equilibrium flexibility plays the most important role, whereas for higher homologues the kinetic flexibility of the oxyalkane spacer is of major importance. As a result, mutual correlation in the orientations of anisotropic polar units of the molecule (characterizing the degree of the IOPO of the molecule) becomes weaker with increasing length of the oxyalkane chain. This is manifested in a gradual disappearance of the odd-even effect and in the general rise of the curve expressing the dependence of ${}_mK$ on n. According to the data[1], this dependence is described by broken curve 2 in Figure 6. Curve 2 and curve I in this figure exhibit many similar features.

However, it is more logical to compare the value of

${}_{m}K$ characterizing the IOPO of the molecule in solution with that of X which characterizes the IOPO of the molecule in the nematic phase. According to Eq. (7), we have $X = (\Delta\varepsilon/S)/4\pi N_A PQ$ or, applying Eq. (6) $X=(\pi^2/N_A PQ)K_I/V_0^2 S$. Consequently, the value of $I/V_0^2 S$ (in which only the factor K_I is neglected) may be regarded as the characteristic of the IOPO of BCBOA molecules in the nematic phase. The values of $I/V_0^2 S$ are calculated according to the data on V_0 and S given in Table I and are listed in the last line of this Table. The same values of $I/V_0^2 S$ depending on n are described by curve I in Figure 7. The same Figure (just like Figure 6) shows for comparison curve 2 of the dependence of ${}_{m}K$ on n for BCBOA in solutions.[I]

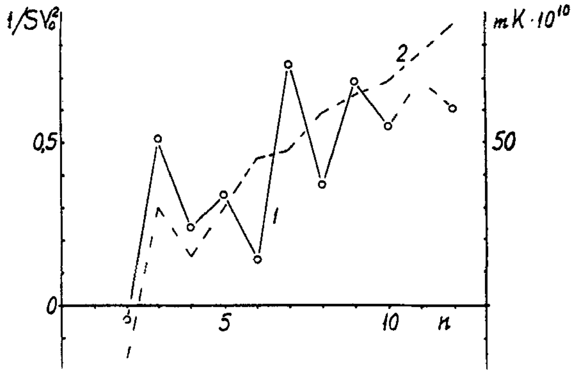


FIGURE 7 Dependence of the I/SV_0^2 value for BCBOA nematics (I) and the molar Kerr constant ${}_{m}K$ for BCBOA benzene solutions (2) on number of methylene groups n in the oxyalkane chain.

A property characteristic of both curves in Figure 7 is an increase in the positive value of ${}_{m}K$ and $I/V_0^2 S$ with increasing n, which is accompanied by a change in the sign of these values (and, correspondingly, of EB sign in solution and $\Delta\varepsilon$ sign in the nematic) from the negative to the positive on passing from n=2 to n=3. Moreover, in the range of $n \leqslant 5$ the shape of these curves is virtually identical, and they exhibit drastic odd-even changes at which the maximum of the ${}_mK$ and I/V_0^2S values is observed at odd n. This fact implies that for lower homologues of the BCBOA series the increase in the length of the alkyl chain leads to conformational transformations in the molecule, which change its IOPO in a similar way both in solution and in the mesophase. These transformations are determined to a considerable extent by equilibrium chain flexibility.

For higher BCBOA homologues ($n > 5$) the shapes of curves I and 2 in Figure 7 differ greatly. The odd-even effect manifested in the alternations of the ${}_mK$ values with increasing n virtually disappears at $n \geqslant 5$, whereas the value of I/V_0^2S (and, hence, that of IOPO) continues to change periodically over the entire series investigated (up to $n=I2$).

As already pointed out above, the disappearance of the odd-even effect reflected in the shape of curve 2 for higher BCBOA homologues may be explained by the disturbance of their IOPO in solution as a result of increasing kinetic flexibility of the chains when they become longer. In accordance with this, the presence of alternations in curve I in Figure 7 should mean that IOPO is retained in the molecules of higher BCBOA homologues in the nematic phase. Consequently, although the anisotropy of dielectric properties of the nematic phase depends to a considerable extent on the intramolecular dipolar structure of molecules, the existence of the nematic potential plays an important role in the maintenance of order in this structure.

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