

Local-Field Anisotropy in Conjugated Polymers

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The experimental values of the Lorentz tensor components L_j have been obtained for uniaxial optically positive and negative films of conjugated polymers with thicknesses of tens of nanometers. The correlation between the components L_j and the measurable parameter η has been revealed and the analytical dependence $L_j(\eta)$ has been derived in agreement with the experiment.

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1. Thin anisotropic films of conjugated conducting polymers are widely used in electronic and optoelectronic devices such as field-effect diodes and transistors [1, 2], electroluminescence diodes [3], polymer lasers [4, 5], and optical amplifiers [6]. The methods of linear optics and spectroscopy are efficient for investigations of conjugated polymers directed at the understanding of the nature of their physical properties and the optimization of their technological parameters. The interpretation of their data is based on the relation of the components of the linear susceptibility $\chi_j = Nf_j\gamma_j$ with the ensemble-averaged components of the molecular polarizability γ_j [7]. Here, N is the number of molecules in unit volume; $f_j = 1 + L_j(\epsilon_j - 1)$ are the components of the local field tensor for light waves polarized along ($j = \parallel$) and across ($j = \perp$) the optical axis of the film \mathbf{n} , where $\epsilon_j = n_j^2$ and n_j are the refractive indices of the film; and L_j are the Lorentz tensor components ($\text{Tr } L = 1$). The determination of L_j values is one of the key problems of the study and simulation of the properties of conjugated polymers [8–11].

The theory of their optical and spectral properties [8–10] has been developed for close packed linear macromolecules aligned with the optical axis \mathbf{n} . In the model of the Lorentz cylindrical cavity [8, 9], the component L_\perp is given by the formula

$$L_\perp = \frac{l}{2(l^2 + D^2)^{1/2}}, \quad (1)$$

where l and D are the length and diameter of the cylinder containing a macromolecule, respectively. The limiting values $L_\parallel = 0$ and $L_\perp = 0.5$ used in the theory correspond to the condition $l \gg D$ in an infinite chain of conjugated monomer units in the linear macromolecule. However, the rigid-chain macromolecule of a conjugated polymer consists of relatively short linear fragments, conformational subunits [12], which differ

in the orientation and the number of constituent monomer units connected by conjugation. This provides the problem of the applicability region of model (1) for real films with the imperfect axial orientation of macromolecules and their fragments. Films with the predominant location of conformational subunits in the film plane (planar orientation) and the axis \mathbf{n} perpendicular to this plane are also used in practice. In this case, model (1) is inapplicable and other models for calculating L_j are absent.

In this work, the method for experimentally determining L_j for uniaxial films of conjugated polymers is proposed with the use of the data on the dispersion of the refractive indices $n_j(\lambda)$ in the visible range. This method, which was previously implemented for uniaxial liquid crystals with an axial orientation of the molecules [13, 14], is generalized here to the case of polymer films with axial and planar orientations of the macromolecules.

2. Let us consider a uniaxial polymer film with the axis \mathbf{n} parallel (planar orientation) or perpendicular (homeotropic orientation) to the film plane. The film consists of uniaxial macromolecules whose longitudinal axis \mathbf{l} coincides with the vector \mathbf{h} between the ends of the molecule. The polarizability tensor γ of the macromolecule is characterized by the mean value $\bar{\gamma} = (\gamma_\parallel + 2\gamma_\perp)/3$ and anisotropy $\Delta\gamma = \gamma_\parallel - \gamma_\perp$, where γ_\parallel and γ_\perp are the longitudinal and transverse components of γ , respectively. For rigid-chain macromolecules, the linear conformational subunits of the i th kind [12] are characterized by the number of monomer units and values $\Delta\gamma_i$ and $S_i = (3 \overline{\cos^2 \vartheta_i} - 1)/2$. Here, ϑ_i is the angle between the axis \mathbf{l} and the longitudinal axis \mathbf{l}_i of the conformational subunit of the i th kind and the overbar means the averaging over the orientation of the conformational subunits of the i th kind with respect to the axis \mathbf{l} . The anisotropy of the polarizability of the

macromolecule is given by the expression $\Delta\gamma = \sum_i N_i \Delta\gamma_i S_i > 0$, where N_i is the number of subunits of the i th kind in the macromolecule. The orientational ordering of the macromolecules with respect to the axis \mathbf{n} is characterized by the order parameter $S = \langle 3 \cos^2 \theta - 1 \rangle / 2$, where θ is the angle between the axes \mathbf{l} and \mathbf{n} and $\langle \dots \rangle$ means the averaging over the ensemble of the macromolecules. The sign of S determines the axial ($0 < S \leq 1$, type O_A) or planar ($-0.5 \leq S < 0$, type O_P) orientations of the macromolecules. The procedure for determining the components L_{\perp} and $L_{\parallel} = 1 - 2L_{\perp}$ depends on the sign of $\Delta n = n_{\parallel} - n_{\perp}$; this sign coincides with the sign of the product $\Delta\gamma S$ [15] and depends on the type of orientation of the macromolecules.

Optically positive films with the planar orientation of \mathbf{n} on orienting substrates and the axial orientation of macromolecules [16, 17]. Let us introduce the parameters $\bar{\varepsilon} = (\varepsilon_{\parallel} + 2\varepsilon_{\perp})/3$ and $Q = (\varepsilon_{\parallel} - \varepsilon_{\perp})/(\bar{\varepsilon} - 1)$ in the visible range and the quantities

$$r_0 = 1 - \frac{2Q^2(\bar{\varepsilon} - 1)}{3(3 + Q)(\bar{\varepsilon} + 2)}, \quad b = \frac{3(\bar{\varepsilon} - 1)}{4\pi N\bar{\gamma}(\bar{\varepsilon} + 2)} - r_0, \quad (2)$$

$$b_1 = \frac{2r_0Q^2}{(3 - Q)(3 + 2Q)}, \quad b_2 = b_1[(6 + Q)/Q]^2.$$

The desired L_{\perp} values are given by the expression [15]

$$L_{\perp} = L_{\perp k} - \frac{(\bar{\varepsilon} + 2)}{12(\bar{\varepsilon} - 1)} \times [(b_1 b_2)^{1/2} - b - [(b_1 - b)(b_2 - b)]^{1/2}], \quad (3)$$

where $L_{\perp k} = (3 + 2Q)/3(3 + Q)$. With known $n_j(\lambda, T)$ values for a discrete set of values λ_i ($i = 1 - p$) in the visible range, an unknown function $b(\lambda, T)$ in the range $\lambda_1 - \lambda_p$ is approximated by the polynomial

$$b(\lambda, T) = a_0(T) + a_1(T)\lambda + \dots + a_m(T)\lambda^m. \quad (4)$$

The quantity $L_{\perp}(T)$ is independent of λ and $m + 2$ unknowns ($L_{\perp}^{(m)}$, $a_0 - a_m$) correspond to each temperature. They are determined from the system of $m + 2 = p$ equations (3) each corresponding to one of the values λ_i . A higher approximation in Eq. (4) implies a higher accuracy of values $n_j(\lambda, T)$; otherwise, the physical solutions of the system of equations for ($L_{\perp}^{(m)}$, $a_0 - a_m$) can be absent. The criterion of the adequacy of the used approximation in Eq. (4) is the agreement of the $L_{\perp}^{(m)}$ values with the quantities $\langle L_{\perp}^{(m-1)} \rangle$ averaged over the $L_{\perp}^{(m-1)}$ values corresponding to $p - 1$ reference values λ_i from the set $\lambda_1 - \lambda_p$ [13, 14].

Optically negative films with the homeotropic orientation of \mathbf{n} and the planar orientation of macromolecules

obtained at the centrifugal spreading of a drop of the polymer solution over the surface of a rotating substrate (the spin-coating technique) [16, 17]. Let us introduce the parameter $Q_d = (\varepsilon_{\perp} - \varepsilon_{\parallel})/(\bar{\varepsilon} - 1)$ in the visible range and the quantities

$$r_{0d} = 1 - \frac{2Q_d^2(\bar{\varepsilon} - 1)}{3(3 - Q_d)(\bar{\varepsilon} + 2)},$$

$$d = \frac{3(\bar{\varepsilon} - 1)}{4\pi N\bar{\gamma}(\bar{\varepsilon} + 2)} - r_{0d}, \quad (5)$$

$$d_1 = \frac{2r_{0d}Q_d^2}{(3 + Q_d)(3 - 2Q_d)}, \quad d_2 = d_1[(6 - Q_d)/Q_d]^2.$$

They are related to the component L_{\perp} as [15]

$$L_{\perp} = L_{\perp k} + \frac{(\bar{\varepsilon} + 2)}{12(\bar{\varepsilon} - 1)} \times [(d_1 d_2)^{1/2} + d - [(d_1 - d)(d_2 - d)]^{1/2}], \quad (6)$$

where $L_{\perp k} = (3 - 2Q_d)/3(3 - Q_d)$. Approximation (4)

is used for the function $d(\lambda, T)$. The values ($L_{\perp}^{(m)}$, $a_0 - a_m$) are determined as discussed above using the known values $n_j(\lambda, T)$ for the values λ_i ($i = 1 - p$) from the system of equations (6) each corresponding to one of the values λ_i .

3. The structural formulas of the monomer units of the macromolecules for the investigated polymers are shown in Fig. 1. The table presents the references with $n_j(\lambda)$ dependences for uniaxial films of these polymers with various orientations of the macromolecules and thicknesses in the range of 50–150 nm. With the use of the values $n_j(\lambda_i)$ obtained from these dependences for five wavelengths λ_i in the visible transparency range of each film, the $L_{\perp}^{(3)}$ values presented in the table and $\langle L_{\perp}^{(2)} \rangle$ are determined. The $L_{\perp}^{(3)}$ and $\langle L_{\perp}^{(2)} \rangle$ values for all films and the various relations between them are in agreement with each other within the accuracy of the determination of $\langle L_{\perp}^{(2)} \rangle$; this accuracy for the PFO (O_A) and TFB (O_P) [PFB (O_P)] films reaches 0.0001 (0.001).

The $L_{\perp}^{(3)}$ value is very sensitive to the accuracy of the values $n_j(\lambda_i)$ and $n_j(\lambda)$ dependences presenting changes in the chemical structure of monomers, molecular weight of the polymer, and technology of the production and thermal treatment of the films. For the PPV [18], BuEH-PPV [19], and MEH-PPV [20] films with close thicknesses and the planar orientation of macromolecules, the difference of the $L_{\perp}^{(3)}$ values presents the effect of the lateral substituent of the monomer unit on the electronic structure of polymer molecules, their orientational order, and $n_j(\lambda)$ dependence. For the MEH-PPV polymer, the sixfold

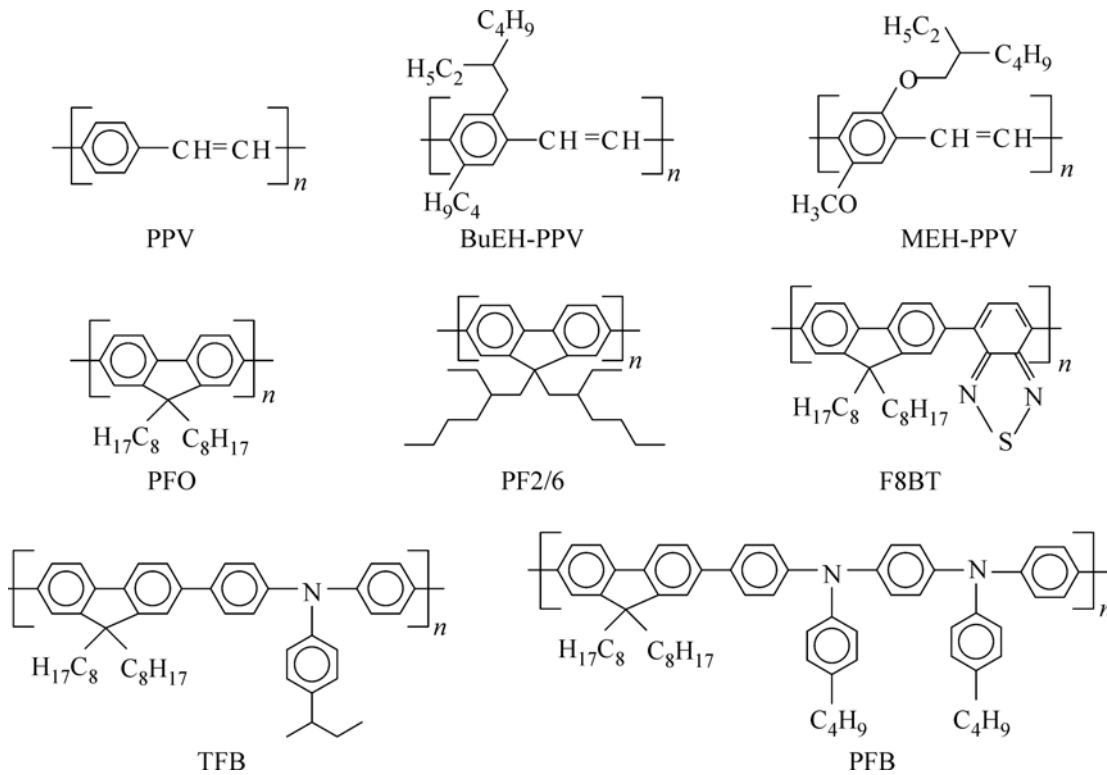


Fig. 1. Structure formulas of monomers for the conjugated polymers under investigation.

decrease in the molecular weight from 1.6×10^6 g/mole [20] to 2.8×10^5 g/mole [21] results in the doubling of $L_{\perp}^{(3)}$. For the PFO [16] and PF2/6 [17] films with the axial or planar orientation of the macromolecules, the small difference in the lateral substituents of the monomer unit is manifested in the difference between the $L_{\perp}^{(3)}$ values. At the same time, the differences in the chemical structures of the TFB and PFB monomers [23] do not affect the $L_{\perp}^{(3)}$ values. The $L_{\perp}^{(3)}$ values for the same unannealed (O_p) and annealed (O_p^*) F8BT film are different [23].

The relation of L_{\perp} with the molecular properties is characterized by the correlation of L_{\perp} with the parameter $L_{\perp k}$. For the films of different polymers with the same orientation of the macromolecules, it follows from the expressions $L_{\perp k} - 1/3 = Q/[3(3 + Q)]$ and $Q \propto \Delta\gamma S/\bar{\gamma}$ [15] that $L_{\perp k} - 1/3$ is an indicator of the anisotropy of the polarizability $\Delta\gamma$ and the orientation ordering of the macromolecules and their subfragments. The function $L_{\perp k}(\lambda)$ for films with the axial (planar) orientation of macromolecules decreases (increases) monotonically and weakly with an increase in λ in the visible range. This is due to the normal dispersion $\Delta n(\lambda)$ and the polarization of the long-wavelength electron $\pi-\pi^*$ transition along the conjugation

chain of the monomer units entering into the conformational subunit, i.e., along the longitudinal axis of the conformational subunit l , and the longitudinal axis

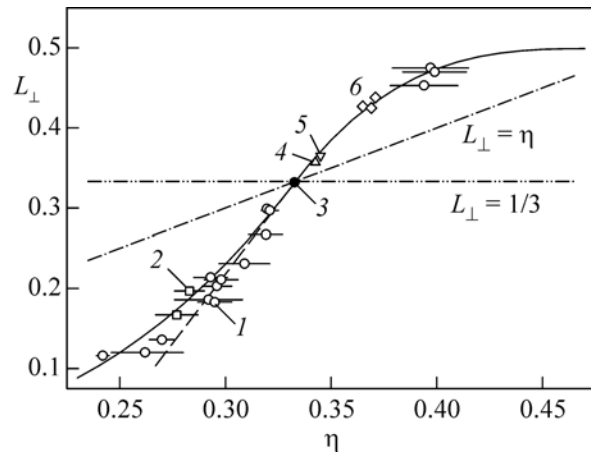


Fig. 2. Correlation of the $L_{\perp} = L_{\perp}^{(3)}$ values with the η values for (1) conjugated polymers, (2) Py and Bp discotic liquid crystals, (3) PVA film, (4) Cd-A Langmuir-Blodgett film [24], and the smectic phase A for the (5) ECh8 [14] and (6) MHPOBC liquid crystals [24]. The solid and dashed lines correspond to Eqs. (9) and (10) and Eq. (7), respectively, with the coefficients presented in the main text and $L_{\perp} = \eta$ and $L_{\perp} = 1/3$.

Values λ_i (in nanometers), $L_{\perp}^{(3)}$, and η for the films of the indicated polymers with the axial (O_A) and planar (O_P and O_P^*) orientations of the macromolecules for unannealed (O_P) and annealed (O_P^*) samples

Polymer	Orientation	λ_1	λ_2	λ_3	λ_4	λ_5	$L_{\perp}^{(3)}$	η
PPV	O_P [18]	550	600	700	800	900	0.116	0.242
BuEH-PPV	O_P [19]	525	535	545	560	590	0.136	0.270
MEH-PPV	O_P^* [20]	590	620	663	775	1000	0.120	0.262
MEH-PPV	O_P [21]	633	700	800	900	1000	0.203	0.296
PFO	O_A [16]	466	500	600	700	850	0.453	0.394
PFO	O_P [16]	466	500	600	700	850	0.267	0.319
PF2/6	O_A [17]	425	450	500	600	750	0.475	0.397
PF2/6	O_P^* [17]	450	475	500	600	750	0.231	0.309
F8BT	O_A [16]	550	576	650	750	850	0.470	0.399
F8BT	O_P^* [22]	532	615	696	777	858	0.186	0.292
F8BT	O_P [23]	565	646	727	808	889	0.183	0.295
F8BT	O_P^* [23]	565	646	727	808	889	0.214	0.293
TFB	O_P [23]	484	565	646	727	808	0.297	0.321
PFB	O_P [23]	484	565	646	727	808	0.299	0.320

of the macromolecule **1**. Taking into account a weak change in $L_{\perp k}(\lambda)$, the value $\eta = \langle L_{\perp k}(\lambda) \rangle$ averaged over the range $\lambda_1 - \lambda_5$ is used.

Figure 2 shows the L_{\perp} and η values for conjugated polymers and some other materials [24] such as discotic liquid crystals of the derivatives of pyrene (Py) and benzoperylene (Bp), film of polyvinyl alcohol (PVA), multilayer Cd-A (cadmium arachidate) Langmuir–Blodgett film, and films of smectic phase A for the MHPOBC and ECh8 liquid crystals [14]. The spread of the η values corresponds to the variation range of $L_{\perp k}(\lambda_i)$.

The inequalities $L_{\perp} < \eta < 1/3$ are satisfied for polymer films with the planar orientation of the macromolecules and discotic liquid crystals. The inequalities $L_{\perp} > \eta > 1/3$ are satisfied for smectics A, Langmuir–Blodgett films, and polymer films with the axial orientation of the macromolecules. An increase in η corresponds to an increase in L_{\perp} and the linear correlation

$$L_{\perp} = A_p \eta + B_p \quad (7)$$

is valid near $\eta = 1/3$. In the range $0.28 < \eta < 1/3$, $A_p = 3.473$ and $B_p = -0.826$; therefore, $(1 - A_p)/3 = -0.824 \approx B_p$ and

$$A_p = (L_{\perp} - 1/3)/(\eta - 1/3) = \text{const.} \quad (8)$$

It is expected that Eqs. (7) and (8) are satisfied for conjugated polymers at $\eta > 1/3$, similar to the Cd–A, ECh8, and MHPOBC films with the coefficients $A_i =$

2.770, $B_i = -0.591$, and $(1 - A_i)/3 = -0.590$ in the range of $1/3 < \eta < 0.37$. In this case, $L_{\perp} \rightarrow 1/3$ for Q , $Q_d \rightarrow 0$, and $\eta \rightarrow 1/3$. With the use of the relation $A = (L_{\perp} - 1/3)/(L_{\perp k} - 1/3)$, the anisotropy $\Delta f = f_{\parallel} - f_{\perp}$ for optically negative films can be represented in the form $\Delta f_d = Q_d(\bar{\epsilon} - 1)(A - 1)/3$. In the visible transparency range of these films, the inequalities $L_{\perp} < L_{\perp k} < 1/3$, $A > 1$, and $\Delta f_d > 0$ are satisfied. The trend $Q_d \rightarrow 0$ owing to a change in the chemical structure of the monomer units of the macromolecules with a small variation of A corresponds to the trend $\Delta f_d \rightarrow 0$. The $L_{\perp}(\eta)$ dependence is nonlinear at $\eta < 0.28$ and a decrease in η corresponds to a decrease in $A_p(\eta)$; the $L_{\perp}(\eta)$ values for conjugated polymer films are in good agreement with the respective values for Py and Bp discotic liquid crystals. Under the condition $(\epsilon_{\perp} - 1) \gg (\epsilon_{\parallel} - 1)$, Q_d is close to $Q_d^{\text{max}} = 1.5$, which corresponds to $L_{\perp k} = \eta = 0$. Under the conditions $0 < L_{\perp} < \eta$ and $\eta \rightarrow 0$, $L_{\perp}(\eta) \rightarrow 0$; $A_p(\eta)$, $A \rightarrow 1$, and $\Delta f_d \rightarrow 0$. The $\eta = 0$ ($\epsilon_{\parallel} = 1$) is inaccessible, because it corresponds to the polarization of all molecular transitions along the axes \mathbf{l}_i of the conformational subunits, coaxiality $\mathbf{l}_i \parallel \mathbf{l}$ of all the conformational subunits, and their perfect ordering with $\mathbf{l} \perp \mathbf{n}$.

The inequalities $L_{\perp} > L_{\perp k} > 1/3$, $A > 1$, and $\Delta f = Q(\bar{\epsilon} - 1)(1 - A)/3 < 0$ are satisfied in the transparency range for the Cd–A, ECh8, and MHPOBC films and conjugated polymers with the axial orientation of the

macromolecules. The trend $Q \rightarrow 0$ owing to a change in the chemical structure of the monomer units of the macromolecules with a small variation of A corresponds to the trend $\Delta f \rightarrow 0$. The $L_{\perp}(\eta)$ dependence is nonlinear at $\eta > 0.35$ and $A_p(\eta)$ decreases with an increase in η . Under the condition $(\varepsilon_{\parallel} - 1) \gg (\varepsilon_{\perp} - 1)$, Q is close to $Q_{\max} = 3$, which corresponds to $L_{\perp k} = \eta = 0.5$. Under the conditions $0.5 > L_{\perp} > \eta$ and $\eta \rightarrow 0.5$, $L_{\perp}(\eta) \rightarrow 0.5$; $A_p(\eta), A \rightarrow 1$; and $\Delta f \rightarrow 0$. The value $\eta = 0.5$ ($\varepsilon_{\perp} = 1$) is inaccessible owing to the mentioned polarization conditions of the molecular transitions, the coaxiality $\mathbf{l}_i \parallel \mathbf{l}$ of all the conformational subunits, and their perfect ordering with $\mathbf{l} \parallel \mathbf{n}$.

4. Let us obtain the polynomial approximations $P_1(\eta)$ ($0 \leq \eta \leq 1/3$) and $P_2(\eta)$ ($1/3 \leq \eta \leq 0.5$) for the function $L_{\perp}(\eta)$. The equalities $P_r(\eta) = L_{\perp}(\eta) = \eta$ at $\eta = 0, 1/3$, and 0.5 give

$$P_1(\eta) = \eta + \eta(\eta - 1/3)F_1(\eta),$$

$$P_2(\eta) = \eta + (\eta - 1/3)(0.5 - \eta)F_2(\eta).$$

At $\eta = 1/3$, $dP_r/d\eta = A_p$; hence, $F_1(1/3) = 3(A_p - 1)$ and $F_2(1/3) = 6(A_p - 1)$. In view of these conditions for the functions $F_r(\eta) = \alpha_r + \beta_r\eta$ of the lowest order,

$$P_1(\eta) = \eta + \eta(\eta - 1/3) \times [\alpha_1 + 3\eta(3A_p - \alpha_1 - 3)], \quad (9)$$

$$P_2(\eta) = \eta + (\eta - 1/3)(0.5 - \eta) \times [\alpha_2 + 3\eta(6A_p - \alpha_2 - 6)]. \quad (10)$$

The requirements $1 > dP_r/d\eta \geq 0$ at $\eta = 0$ and 0.5 impose the constraints $0 < F_1(0) \leq 3$ and $0 < F_2(0.5) \leq 6$; since $A_p = 3.473$, they correspond to the inequalities $0 < \alpha_1 \leq 3$ and $32.514 \leq \alpha_2 < 44.514$. Function (9) has a minimum of -0.0025 ($\eta_{\min} \approx 0.072$) at $\alpha_1 = 3$ and a maximum and a minimum at $2.724 = \alpha_1^* < \alpha_1 < 3$ and increases monotonically at $\alpha_1 \leq \alpha_1^*$ ($\eta_1^* \approx 0.047$). Function (10) has a maximum of 0.5012 ($\eta_{\max} \approx 0.464$) at $\alpha_2 = 32.514$ and a maximum and a minimum at $32.514 < \alpha_2 < 33.617 = \alpha_2^*$ ($\eta_2^* \approx 0.477$) and increases monotonically at $\alpha_2 \geq \alpha_2^*$. In view of the requirement of the monotonicity of the functions $P_r(\eta)$, $\alpha_1 = \alpha_1^*$ and $\alpha_2 = \alpha_2^*$ can be accepted in Eqs. (9) and (10). As is seen in Fig. 2, functions (9) and (10) with these parameters well approximate the $L_{\perp}(\eta)$ dependences for conjugated polymer films and low-molecular compounds with the layered smectic structure.

The L_{\perp} values for the PFO, PF2/6, and F8BT (O_A) films are close to 0.5 in model (1) at $l \gg D$. Although the correlation $L_{\perp}(\eta)$ revealed above and the real inaccessibility of the equality $L_{\perp} = \eta = 0.5$ are disregarded in model (1), Fig. 2 demonstrates that the limiting val-

ues $L_{\parallel} = 0$ and $L_{\perp} = 0.5$ [8, 9] can be used for objects with $\eta > 0.45$.

5. The above consideration shows the possibility of determining the components L_j with a high accuracy for polymer nanofilms with the planar and homeotropic orientations of the optical axis. This made it possible to reveal fine features of a change in L_j in dependence on the chemical structure of the monomers, the molecular weight of the polymer, and the technology of the production and thermal treatment of the films. Dependences (9) and (10) provide the determination of $L_{\perp}(\eta)$ from the experimental values of $n_j(\lambda)$ and η without any information on the structure of the macromolecules, their orientation order, and conformation.

The experimental L_j values provide new possibilities: (i) the determination of S values for the macromolecules from the dichroism of polarized bands $\alpha_j(\lambda)$ of the electronic absorption coefficient of the films [7]; (ii) the reconstruction of the dispersion dependences of the real, $\gamma_j'(\lambda)$, and imaginary, $\gamma_j''(\lambda)$, parts of the components $\gamma_j(\lambda)$ for the investigation of the electronic structure of conjugated polymer molecules [12]; (iii) the explanation of the difference $\lambda_{\parallel} \neq \lambda_{\perp}$ between the maxima of the components $\alpha_j(\lambda)$ for the films with the axial and planar orientations of the macromolecules [16, 17, 20]; and (iv) the analysis of the effects of the mixing of electronic excitations in conjugated polymers owing to the intermolecular resonance dipole-dipole interactions (local-field effects) [7, 25].

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