

Effective Refractive Index of a Two-Dimensional Polycrystal

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Received March 17, 2015; in final form, April 13, 2015

A relation of the effective refractive index $n^* = \sqrt{n_1 n_2}$ of a two-dimensional polycrystalline dielectric film in the transparency region to the refractive indices n_1 and n_2 of crystallites at the positions of axes 1 and 2, respectively, of refraction ellipsoids of the crystallites in the plane of the film has been obtained. This relation and the relation $L^* = (L_1 + L_2)/2$ between the components of the Lorentz tensors for the film and crystallites have been confirmed by comparison with experimental data for conjugate-polymer films with uniaxial domains.

DOI: 10.1134/S0021364015100033

1. Thin films of conjugated polymers [1–3], their oligomers [4, 5], ferroelectric polymers [6], and organic molecular semiconductors [7, 8] on isotropic substrates used in optoelectronics are uniaxial polycrystals. Their optical axis \mathbf{n} is perpendicular to the plane of the film and is parallel to axes 3 of refraction ellipsoids of crystallites at a random distribution of axes 1 and 2 of crystallites in the plane of the film. Depending on the technology of production of the films, the dimension a of crystallites ranges from tens [1, 2, 7, 8] and hundreds [4, 6] of nanometers to microns [3–5]. For light waves with the wavelength $\lambda \gg a$, wave vector $\mathbf{k}^* \perp \mathbf{n}$, and polarization $\mathbf{E} \perp \mathbf{n}$, such films in the transparency region are two-dimensional composite media with the effective dielectric constant ε^* . For waves with $\mathbf{E} \parallel \mathbf{n}$, $\varepsilon_{\parallel} = \varepsilon_3$. The refractive indices $n_{\parallel} = n_3$ and $n^* = \sqrt{\varepsilon^*}$ are important for control of the morphology, local structure, and physical properties of films [6, 7, 9], as well as for study of the features of the local field [10, 11], anisotropy of interparticle interactions [12], and microscopic nature of these objects. However, the relation between n^* and $n_{1,2}$ in the optical region is still unknown.

A formula for the effective static dielectric constant ε_{st}^* is obtained from the following expression for the effective electrical conductivity of a two-dimensional polycrystal [13]:

$$\sigma^* = \sqrt{\sigma_1 \sigma_2}, \quad (1)$$

with the change $\sigma^* \rightarrow \varepsilon_{st}^*$, $\sigma_i \rightarrow \varepsilon_{i(st)}$. Here, σ_1 and σ_2 are the principal components of the conductivity tensor of a crystallite. Although formula (1) was derived within various theoretical approaches [14–16] and hierarchic models of a polycrystal [17–19], experi-

mental tests of this formula and its analog for ε_{st}^* are absent.

The aims of this work are to obtain the relations $\varepsilon^*(\varepsilon_{1,2})$ and $n^*(n_{1,2})$ and constraints on ε^* and n^* for a two-dimensional polycrystalline film in the optical transparency region and to compare these relations and the previously obtained relation $L^*(L_{1,2})$ between the components of the Lorentz tensor for the film and crystallites [12] with experimental data for quasi-two-dimensional films of conjugated polymers with uniaxial domains.

2. We consider a polycrystalline film consisting of statistically equivalent crystallites, where axes 1 and 2 of the refraction ellipsoids are randomly oriented in the plane of the film. For a light wave with $\lambda \gg a$, amplitude $\mathbf{E} \perp \mathbf{n}$, and $\mathbf{k}^* \perp \mathbf{n}$, we have $\mathbf{E} = \langle \mathbf{E}(\mathbf{r}) \rangle$, where the triangular brackets stand for averaging over the area of the film. The amplitude $\mathbf{D} = \langle \mathbf{D}(\mathbf{r}) \rangle$ of the electric displacement vector is related to \mathbf{E} as

$$\mathbf{D} = \varepsilon^* \mathbf{E}. \quad (2)$$

Inside an individual crystallite, whose position is specified by the vector \mathbf{r}_c , we decompose the vectors $\mathbf{D}(\mathbf{r}_c)$ and $\mathbf{E}(\mathbf{r}_c)$ into the longitudinal and transverse components with respect to the direction \mathbf{k}^* : $\mathbf{D}(\mathbf{r}_c) = \mathbf{D}_l(\mathbf{r}_c) + \mathbf{D}_t(\mathbf{r}_c)$ and $\mathbf{E}(\mathbf{r}_c) = \mathbf{E}_l(\mathbf{r}_c) + \mathbf{E}_t(\mathbf{r}_c)$. Since the wave is transverse, $\langle \mathbf{D}_l(\mathbf{r}_c) \rangle = \langle \mathbf{E}_l(\mathbf{r}_c) \rangle = 0$. Then, we take into account the relation $\mathbf{D}_t(\mathbf{r}_c) = \varepsilon_e(\mathbf{r}_c) \mathbf{E}_t(\mathbf{r}_c)$, where $\varepsilon_e(\mathbf{r}_c)$ is the dielectric constant of the crystallite for an extraordinary wave with the wave vector $\mathbf{k}(\mathbf{r}_c) \parallel \mathbf{k}^*$. Using the representations $\mathbf{D}_t(\mathbf{r}_c) = \mathbf{D} + \delta \mathbf{D}_t(\mathbf{r}_c)$ and $\mathbf{E}_t(\mathbf{r}_c) = \mathbf{E} + \delta \mathbf{E}_t(\mathbf{r}_c)$ and taking into account $\langle \delta \mathbf{D}_t(\mathbf{r}_c) \rangle = \langle \varepsilon_e(\mathbf{r}_c) \delta \mathbf{E}_t(\mathbf{r}_c) \rangle = 0$, we obtain $\mathbf{D} = \langle \varepsilon_e(\mathbf{r}_c) \rangle \mathbf{E}$. The comparison of this expression with Eq. (2) gives

$$\varepsilon^* = \langle \varepsilon_e(\mathbf{r}_c) \rangle. \quad (3)$$

The orientation of the crystallite in the plane of the film is specified by the angle θ between axis 1 of the refraction ellipsoid of the crystallite and the direction \mathbf{k}^* . In the case of an isotropic distribution function $\rho(\theta)$ of crystallites with the same average area, Eq. (3) can be represented in the equivalent form

$$\varepsilon^* = (2/\pi) \int_0^{\pi/2} \varepsilon_e(\theta) d\theta. \quad (4)$$

The substitution of the expression [20]

$$\varepsilon_e(\theta) = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 \cos^2 \theta + \varepsilon_2 \sin^2 \theta}, \quad (5)$$

into Eq. (4) gives the desired relations

$$\varepsilon^* = \sqrt{\varepsilon_1 \varepsilon_2}, \quad n^* = \sqrt{n_1 n_2}. \quad (6)$$

Expressions (6) are independent of the symmetry of the crystal because of the adjustment to the axes of the refraction ellipsoid rather than to the crystallographic axes of crystallites. Macroscopic relations (6) are also independent of the features of the arrangement of structural elements in the unit cell of crystallites. Inequalities $\langle \varepsilon_e(\theta) \rangle \geq \langle \varepsilon_e^{-1}(\theta) \rangle^{-1}$ and $2\sqrt{a_1 a_2} \leq a_1 + a_2$ for $a_{1,2} > 0$ provide a lower and an upper bound for ε^* . As a result,

$$B_l = \frac{2\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \leq \varepsilon^* \leq \frac{\varepsilon_1 + \varepsilon_2}{2} = B_u \quad (7)$$

and $\varepsilon^* = \sqrt{B_l B_u}$. The quantity n^* is in the interval

$$b_l \leq n^* \leq b_u, \quad (8)$$

where $b_{l(u)} = \sqrt{B_{l(u)}}$ and $n^* = \sqrt{b_l b_u}$. The intervals $\Delta B = B_u - B_l = (\varepsilon_1 - \varepsilon_2)^2 / [2(\varepsilon_1 + \varepsilon_2)]$ and $\Delta b = b_u - b_l = \Delta B / (b_u + b_l)$ significantly depend on λ . If the dipole moment of the transition for the long-wavelength electron absorption band of the crystallite is oriented along one of the axes $i = 1, 2$ of the refraction ellipsoid of the crystallite, the approach of λ to this absorption band with the maximum λ_i will be accompanied by a resonance increase in the difference $\varepsilon_1 - \varepsilon_2$ and quantities ΔB and Δb .

According to the expression for ε^* in Eq. (6), ε^* satisfies the equation

$$\frac{\varepsilon_1 - \varepsilon^*}{\varepsilon_1 + \varepsilon^*} + \frac{\varepsilon_2 - \varepsilon^*}{\varepsilon_2 + \varepsilon^*} = 0, \quad (9)$$

of the form corresponding to the theory of the effective two-dimensional medium consisting of disklike crystallites. This coincidence of exact formula (6), which is independent of the shape of crystallites, with the result of the theory of the effective medium is an accidental consequence of approximations underlying this theory and assumption of a disklike shape of crystal-

lites. Equation (9) and the results of the theory of the effective medium for three-dimensional polycrystals with spherical crystallites [15, 21, 22] can be written in the form of the common equation

$$\sum_{i=1}^D \frac{\varepsilon_i - \varepsilon^*}{\varepsilon_i + (D-1)\varepsilon^*} = 0 \quad (10)$$

for the effective dielectric constant ε^* of a D -dimensional polycrystal with optical anisotropic crystallites in the form of D -dimensional spheres.

3. In addition to the conditions of applicability of Eqs. (6), a number of requirements should be satisfied for their experimental verification: quite smooth variation of $\varepsilon(\mathbf{r})$ at the edges of crystallites for the minimization of scattering effects at these edges; preferable uniaxiality of crystallites with their optical axes 1 parallel to the plane of the film for the fixation of axes 1 and 2 of crystallites in this plane; and the existence of n_1 and n_2 for single-crystal samples and their independence of the dimension of a crystallite. Monocrystalline (monodomain) and polycrystalline (polydomain) films of conjugated polymers mostly satisfy these requirements.

Macromolecules of these polymers consist of linear fragments—conformational subunits [23], which differ in the orientation and number of their monomer units connected by π -electron conjugation. For a monodomain uniaxial film of a conjugated polymer on the orienting substrate, optical axis 1 is parallel to the plane of the film and conformational subunits have an axial (A) orientation with respect to axis 1. The film is characterized by the refractive indices $n_{eA} = n_1$ and $n_{oA} = n_2 = n_3$. For the polydomain film of a conjugated polymer obtained by the spin-coating method [9], the orientation of conformational subunits is planar (P) and the optical axis \mathbf{n} is perpendicular to the plane of the film. The extraordinary ($n_{eP} = n_{\parallel}$) and effective ordinary ($n_{oP} = n^*$) refractive indices of the film are measured by the ellipsometry method. If the effect of intermolecular interactions on the local structure and optical properties of domains is decisive as compared to the effect of the orienting substrate, the relation $n_{oA} = n_{eP}$ should be satisfied together with the following analog of Eq. (6) for uniaxial domains:

$$n^* = \sqrt{n_{eA} n_{oA}} = n_{oP}. \quad (11)$$

Another consequence of the polycrystal property of the two-dimensional film is the relation [12]

$$L^* = (L_1 + L_2)/2 \quad (12)$$

between the components of Lorentz tensors L^* and L_i ($i = 1, 2, 3$) for the film and crystallite, respectively. The quantities L^* and L_i specify the components $f^* = 1 + L^*(\varepsilon^* - 1)$ and $f_i = 1 + L_i(\varepsilon_i - 1)$ of the local field tensors for the film and crystallite, respectively. These components relate the local fields $E_{\text{loc}}^* = f^* E$ and

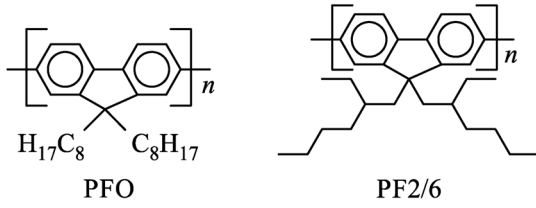


Fig. 1. Structural formulas of monomers for the PFO and PF2/6 polymers.

$E_i^{(\text{loc})} = f_i E_i$ for the film and crystallite to the respective macroscopic fields E and E_i for them. For light with the polarization $\mathbf{E} \parallel \mathbf{n}$ at $\varepsilon_{\parallel} = \varepsilon_3$, we have $E_{\parallel} = E_3$ and $L_{\parallel} = L_3$. Relation (12) follows from the equality $\varepsilon_{\parallel} = \varepsilon_3$ [12].

For uniaxial crystallites with optical axes 1 parallel to the plane of the film, and $L_{2A} = L_{3A} = [1 - L_{1A}]/2$, it follows from Eq. (12) that

$$L^* = [1 - L_{2A}]/2 = L_{\perp P}, \quad (13)$$

where the component $L_{\perp P}$ for the film corresponds to the polarization of the light wave $\mathbf{E} \perp \mathbf{n}$. Formula (13) follows from the equality $n_{oA} = n_{eP} = n_{\parallel}$, which established a relation between Eqs. (11) and (13). The quantity L_{2A} lies in the interval $1/3 \leq L_{2A} < 1/2$ [10, 11], where the inaccessible upper bound corresponds to monodomain uniaxial films of conjugated polymers with the orientation of all conformational subunits of the polymer chain along optical axis 1 and the metallic conductivity of the chain in this direction in the absence of conductivity in the perpendicular directions. The quantity L^* is in the range $1/4 < L^* \leq 1/3$, where the lower bound corresponds to the blocking of the conductivity of the two-dimensional polycrystal when σ_1 or σ_2 in Eqs. (1) tends to zero [21]. Since quantities in Eqs. (11) and (13) refer to the axes of the refraction ellipsoids of the film and crystallites (domains), respectively, these expressions (under the conditions of their applicability) are independent of the details of the structure of the polymers, the orientation of the axes of their conformational subunits, and other structural features of films.

Conjugated polymers PFO [24, 25] and PF2/6 [26] have known values $n_{j(A,P)}(\lambda)$ ($j = e, o$) in the visible transparency regions that were measured by the ellipsometry method [25, 26] and known values $L_{1,2(A)}$ and $L_{\perp P}$ [10, 11] obtained from these data. Structural formulas of monomer units for these rigid-chain polymers are shown in Fig. 1. Macromolecules PFO [25] with the contour length $l_c \approx 96$ nm contained 116 monomers on average and had conformational subunits with the mean length $\xi \approx 10$ nm, close to the persistent length $l_p \approx 8.6$ nm [24]. Rodlike molecules PF2/6 with the length $l_c \approx 2l_p$ nm included 20 monomers [26]. Both polymers can exist in the thermotropic nematic, glassy, and crystalline phases [2, 24].

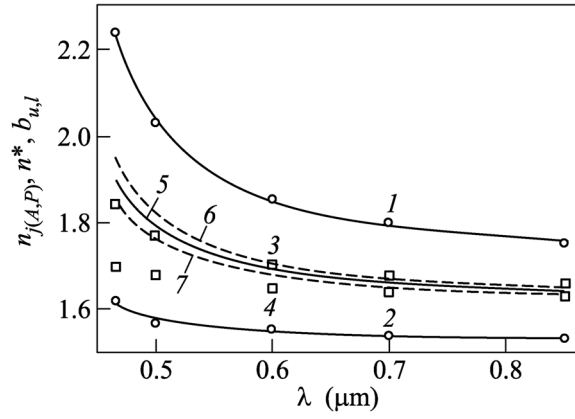


Fig. 2. Dispersion dependences of the refractive indices (1) n_{eA} , (2) n_{oA} , (3) n_{oP} , (4) n_{eP} , and (5) n^* and boundary values (6) b_u and (7) b_l in Eq. (8) for PFO polymer films. Solid lines 1 and 2 are the approximations by function (14).

However, the last phase can exist at the molecular weight $M_n > M_n^* \approx 10$ kg/mol [2]. The relation $M_n^* > M_n = 7.6$ kg/mol for the PF2/6 polymer [26] excluded this phase for this polymer.

Uniaxial monodomain PFO (PF2/6) films on a rubbed polyimide substrate with the nematic orientational ordering of conformational subunits (molecules) with a thickness $d = 100$ nm [25] (51 nm [26]) and an area of about 1 cm^2 were obtained by the transformation of the thermotropic nematic phase to a glassy state under fast cooling. For polydomain PFO (PF2/6) films with the planar orientation of conformational subunits and $d = 48$ nm [25] (46 nm [26]), the mean dimension of domains $a \approx 30$ nm ($a < 10$ nm) [2] is typical of polyfluorene and other conjugate-polymer films obtained by spin-coating [1]. The absence of the crystalline phase (crystallites) in monodomain and polydomain PFO and PF2/6 films [25, 26] was manifested in the absence of the absorption band characteristic of this phase with $\lambda_{\text{max}} \approx 425$ nm on the wing of the long-wavelength electron absorption band [24]. The morphology of PF2/6 films [26] with the planar orientation of molecules and $l_p \approx a < l_c$ corresponded to an intermediate state between the polydomain structure with randomly oriented nematic domains whose optical axes lie in the plane of the film and the homogeneous monodomain state with a random distribution of molecular axes in the plane of the film.

Figure 2 shows the refractive indices $n_{j(A,P)}(\lambda_k)$ for five λ_k values in the visible transparency region of PFO films as obtained from $n_{j(A,P)}(\lambda)$ plots [25]. The $n_{j(A,P)}(\lambda_k)$ values are well approximated by the function [27]

$$n_j(\lambda) = n_{o_j} + G_j \lambda^2 \lambda_j^2 / (\lambda^2 - \lambda_j^2), \quad (14)$$

where λ_j corresponds to the maximum of the single long-wavelength electron absorption band of the PFO film, G_j is proportional to the integral absorption coefficient of the film within this band, and the contribution n_{0j} comes from shorter wavelength transitions. The relation $G_{eA} \gg G_{oA}$ represents the polarization of the absorption band along the optical axis of the film and ensures strong dispersion of $n_{eA}(\lambda)$. The value $\lambda_{eA} = (0.406 \pm 0.005) \mu\text{m}$ obtained from approximation (14) coincides with the experimental value $\lambda_{eA} = (0.402 \pm 0.005) \mu\text{m}$ [25]. The functions $n_{jA}(\lambda)$ given by Eq. (14) were used to calculate the dependences $n^*(\lambda)$ and $b_{u,l}(\lambda)$ shown in the figure, where it is seen that relation (11) is satisfied throughout the entire visible range within the accuracy $\delta n_{oP} \approx 0.03$ [25]. The aforementioned resonance increase in the width Δb of interval (8) from 0.01 to 0.1 is observed with a decrease in λ approaching λ_{eA} . In this case, $n_{oP}(\lambda_k)$ values are monotonically shifted from the upper to lower bound of interval (8) because of the relation $\lambda_{eA} > \lambda_{oP} = (0.389 \pm 0.009) \mu\text{m}$ (experimental value is $\lambda_{oP} = (0.385 \pm 0.005) \mu\text{m}$ [25]) and a weaker dispersion $n_{oP}(\lambda)$ as compared to $n_{eA}(\lambda)$ and $b_{l,u}(\lambda)$.

As is seen in the figure, $n_{eP} - n_{oA} \approx 0.1$ at all λ_k values, which corresponds to a higher orientational ordering of conformational subunits of the polymer in a monodomain film on an orienting substrate than that in a polydomain film on an isotropic substrate. However, this difference is not critical for the satisfaction of Eq. (13). The substitution of the value $L_{2A} = 0.4530 \pm 0.0002$ [11], which is obtained from the $n_{jA}(\lambda_k)$ values shown in Fig. 2, into Eq. (13) gives $L^* = 0.274$. This value coincides with the value $L_{\perp P} = 0.270 \pm 0.007$ [11], which is obtained from the $n_{jP}(\lambda_k)$ values. Thus, Eqs. (11) and (13) are satisfied for PFO films.

In the visible transparency region of PF2/6 films with the planar orientation of macromolecules [26], a higher value $n_{eP} - n_{oA} \approx 0.15$ is manifested in the difference $n_{oP} > b_u$ exceeding the experimental error δn_{oP} . The substitution of the value $L_{2A} = 0.472 \pm 0.006$ [11] into Eq. (13) gives the value $L^* = 0.264$, which is above the experimental value $L_{\perp P} = 0.220 \pm 0.020$ [11]. The latter equalities in Eqs. (11) and (13) are violated for PF2/6 films.

The relation $L_{\perp P} < 1/4$ is also valid for polydomain (polycrystalline) films of many other known conjugated polymers with the planar orientation of macromolecules [10, 11]. This can indicate the unblocking of the conductivity of these films with a decrease in the dimensions of crystallites and with an increase in the conductivity of intercrystallite regions. In contrast to low-molecular polycrystals [4, 5, 7, 8], amorphous intercrystallite regions for films of conjugated polymers include the middle parts of macromolecules whose ends are located in neighboring crystallites [1]. This ensures the π -electron conjugation of neighbor-

ing crystallites and the unblocking of the conductivity in intercrystallite regions. The necessity of allowance for the conductivity of intercrystallite amorphous regions in bulk polycrystals of conjugated polymers was mentioned in [28], where the electric and spectral properties of these objects were described within the theory of the effective medium. The relation $L_{\perp P} < 1/4$ is independent evidence of this fact for two-dimensional polydomain (polycrystalline) films of conjugated polymers with the planar orientation of macromolecules.

4. Thus, Eqs. (6)–(8) and (11) provide the relation $n^*(n_{1,2})$ in the optical region for a two-dimensional polycrystal. The confirmation of these relations for polydomain films of the conjugated PFO polymer solves the important problem of the relation between $n_j(A)$ and $n_j(P)$ for films with the axial (A) and planar (P) orientations of macromolecules [9, 25, 26]. In contrast to the static values σ^* and ε_{st}^* , the allowable ranges of quantities $\varepsilon^*(\lambda)$ and $n^*(\lambda)$ in the optical transparency region significantly depend on λ and vary in a resonance manner near polarized absorption bands of a polycrystal. Relation (13) confirmed for PFO-polymer films is a criterion, additional to Eqs. (6)–(8) and (11), that films are polydomain. The constraint $1/4 < L^*$ corresponds to the blocking of the conductivity of a two-dimensional polycrystal when σ_1 or σ_2 in Eq. (1) tends to zero [21]. The violation of the latter equalities in Eqs. (11) and (13) for films of conjugated polymers can indicate the amorphization of intercrystallite regions, and the relation $L_{\perp P} < 1/4$ indicates the unblocking of the conductivity of a polycrystal that is characteristic of these objects at the amorphization of intercrystallite regions.

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Translated by R. Tyapaev