# **Anisotropic diffusion of light in polymer dispersed liquid crystals**

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Anisotropic diffusion of light was experimentally first observed in bulk nematic liquid crystals. The origin of anisotropy in such a system is due to the optical anisotropy of nematic liquid crystal and to anisotropic scattering of light from nematic orientational fluctuations. In polymer dispersed liquid crystals (PDLCs) anisotropic scatterers—i.e., nematic droplets—are embedded in an optically isotropic polymer. The anisotropy of diffusion of light in this system therefore depends only on the anisotropic scattering from a single nematic droplet and on the average orientation of the droplets in PDLCs. In the absence of an external field droplets in PDLCs are randomly oriented and diffusion of light is isotropic. In an external field the droplets are on average oriented in the direction of the external field and the diffusion of light becomes anisotropic. We calculated the diffusion tensor and expected anisotropy of the diffusion of light as a function of an external field, *kR*, and volume fraction using the theory developed for bulk nematic liquid crystals. Our calculations show that the anisotropy of diffusion tensor can have both the signs and that the magnitude of diffusion constants increase with increasing external field.

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### **I. INTRODUCTION**

Polymer dispersed liquid crystals (PDLCs) are interesting for applications as well as for fundamental research of light scattering in turbid systems. Particular attention is given to the switching behavior of PDLC films for their applicability as light shutters or in electro-optical displays  $[1]$  $[1]$  $[1]$ . Although first produced decades ago  $[2]$  $[2]$  $[2]$ , PDLCs are still of interest for many new applications in photonic devices  $\begin{bmatrix} 3 \end{bmatrix}$  $\begin{bmatrix} 3 \end{bmatrix}$  $\begin{bmatrix} 3 \end{bmatrix}$  and in random lasers  $[4,5]$  $[4,5]$  $[4,5]$  $[4,5]$ . The transmittance of the PDLC films depends on the applied electric field that switches the films from an opaque, multiple-scattering state to a clear, transparent state. The theoretically calculated single-scattering picture gives a good description of the film transmittance  $\lceil 6-8 \rceil$  $\lceil 6-8 \rceil$  $\lceil 6-8 \rceil$ , but is insufficient to explain the angular dependence of the scattered intensity of light where multiple-scattering effects are important  $[9,10]$  $[9,10]$  $[9,10]$  $[9,10]$ . Multiple light scattering can be a hindrance in the production of PDLCs  $[11]$  $[11]$  $[11]$  as well as useful when PDLC's are used as random amplifying media  $[4]$  $[4]$  $[4]$  or switchable screens. Understanding of multiple scattering in PDLCs is important for it is a switchable key property in any application.

Multiple light scattering in bulk nematic liquid crystals has been studied both theoretically  $[12,13]$  $[12,13]$  $[12,13]$  $[12,13]$  and experimentally  $[14,15]$  $[14,15]$  $[14,15]$  $[14,15]$ , and it has been shown that the diffusion of light in such system is anisotropic. The origin of this anisotropy is due to the optical anisotropy of nematic liquid crystals and to anisotropic scattering of light from nematic orientational fluctuations. In PDLCs the situation is different: anisotropic scatterers—i.e., nematic droplets—are embedded in an optically isotropic polymer. The anisotropy of the diffusion of light in this system therefore depends on the anisotropic scattering from a single nematic droplet and on the average orientation of the droplets in PDLCs.

In this paper we address the problem of the anisotropy of the diffusion of light in PDLCs and its dependence on the ordering of the droplets, on the configuration of the director field within the droplets, on the radius, and on the volume fraction of the droplets in the PDLC film. We calculated the diffusion tensor and light diffusion anisotropy using the theory developed for bulk nematic liquid crystals  $[12,13]$  $[12,13]$  $[12,13]$  $[12,13]$ . The discrete dipole approximation  $(DDA)$  [[17](#page-5-14)] was used to calculate the scattering cross section of a single nematic droplet. Our results show that the sign and the size of the anisotropy in the PDLC system is not a trivial function of the relevant parameters. While previous studies of multiple scattering have mostly focused on transmittance of a PDLC film, we calculate a complete diffusion tensor, so multiple scattering in diffusive regime can be calculated for any direction and sample shape.

#### **II. THEORY AND RESULTS**

Polymer dispersed liquid crystal materials consist of randomly distributed nematic liquid crystal droplets embedded in an isotropic solid polymer matrix. The refractive index of the polymer is usually chosen so that it is close to the ordinary index of refraction of the liquid crystal. Actual values of the indices of refraction of a polymer and liquid crystal in PDLCs differ from the values in the bulk, since both the liquid crystal and solid polymer matrix are contaminated by each other. The configuration of the nematic director (the direction of the optical axis) in the droplet in PDLCs is usually bipolar in the absence of a field and close to uniform in a strong field (Fig. [1](#page-1-0)). So in both cases in the interior of a droplet the director points predominantly along one direction **n**. In the absence of an external electric or magnetic field the average orientations of different droplets are random and the system is optically isotropic. In an external field the droplets partially orient along the direction of the field and the scattering properties, and consequently propagation of light through such a system becomes anisotropic.

The scattering properties of PDLCs depend on the configuration of the director field within a single droplet, the size

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FIG. 1. Schematic presentation of bipolar and uniform configurations of a nematic droplet.

and the shape of the droplet, and the interdroplet properties, such as concentration of the droplets, relative orientation of droplets, and size and shape polydispersity. In our study we assume the droplets are spherical and identical; i.e., we neglect the shape and size polydispersity.

The transmittance of the direct beam through the PDLC film is governed by the scattering mean free path  $l_{sca}$  which is related to the scattering cross section  $\sigma$  of an average droplet,  $l_{sca}^{-1} = \rho_N \sigma$ , where  $\rho_N$  is the number density of the scatterers. It depends on the average orientation of the droplets and their positional and orientational correlations. When the sample is thick enough and almost no direct beam is transmitted through the sample, propagation of light can be treated with the diffusion approximation. The transmittance of diffuse light in isotropic media depends on the transport mean free path *l* \* , which is in isotropic systems related to the scattering mean free path through a simple relation

$$
l^* = l_{scd}/(1 - \langle \cos \vartheta \rangle),\tag{1}
$$

<span id="page-1-1"></span>where  $\vartheta$  is the scattering angle and  $\langle \cdots \rangle$  denotes the average (weighted by the normalized differential cross section) over all scattering angles. The transport mean free path is related to the light diffusion constant  $D = v l^* / 3$ , where *v* is the transport velocity of light in a medium (usually group velocity). In optically anisotropic media diffusion of light is no longer isotropic and its propagation is described by the diffusion tensor  $[12,13]$  $[12,13]$  $[12,13]$  $[12,13]$ . The transport mean free path as a scalar pa-rameter given in Eq. ([1](#page-1-1)) has no longer any meaning. The direction-dependent transport mean free path can be obtained from the diffusion tensor  $[16]$  $[16]$  $[16]$ .

The theory for diffusing light in bulk nematic liquid crystals was independently developed by Stark and Lubensky [[13](#page-5-11)] and van Tiggelen, Maynard, and Heiderich [[12](#page-5-10)]. The part of the results of this theory used in our calculation can be summarized in an anisotropic diffusion equation for the electric field autocorrelation function at  $t=0$ ,  $W(\mathbf{R},T,t)$  $=\langle \mathbf{E}(\mathbf{R}, T+t/2) \cdot \varepsilon_0 \mathbf{E}(\mathbf{R}, T-t/2) \rangle$ :

$$
\left[\frac{\partial}{\partial T} - D_{\parallel} \nabla_{\parallel}^2 - D_{\perp} \nabla_{\perp}^2 \right] W(\mathbf{R}, T, 0) = 0, \tag{2}
$$

where **R** and *T* are space and time variables. The two independent diffusion constants  $D_{\parallel}$  and  $D_{\perp}$ , which correspond to directions along and perpendicular to the optical axis, respectively, can be expressed with the extended matrix elements  $B_{\alpha\beta}$  of the optical structure function:

$$
B_{\alpha\beta}(\mathbf{k}_{in}, \mathbf{k}_{out}) = \frac{\omega^4}{c^4} \langle \delta \varepsilon_{\alpha\beta}(\mathbf{q}) \delta \varepsilon_{\alpha\beta}^*(\mathbf{q}) \rangle, \tag{3}
$$

where  $\mathbf{q} = \mathbf{k}_{out} - \mathbf{k}_{in}$  is the scattering vector, indices  $\alpha$  and  $\beta$ denote the polarization vectors  $\mathbf{e}_{\alpha}(\mathbf{k}_{in})$  and  $\mathbf{e}_{\beta}(\mathbf{k}_{out})$  of the incoming and scattered light, respectively, and  $\delta \varepsilon_{\alpha\beta}(\mathbf{q})$  $= \mathbf{e}_{\beta}(\mathbf{k}_{out}) \cdot \delta(\mathbf{q}) \mathbf{e}_{\alpha}(\mathbf{k}_{in})$  with  $\delta \epsilon(\mathbf{q})$  being the fluctuating part of the dielectric tensor. The two diffusion constants are

$$
D_{\gamma} = \frac{(4\pi)^3 c}{18n_p^5} \frac{B_{oo}^{\gamma} + B_{ee}^{\gamma} - 2B_{ee}^{\gamma}}{B_{ee}^{\gamma}B_{oo}^{\gamma} - (B_{ee}^{\gamma})^2},
$$
(4)

<span id="page-1-2"></span>where  $\gamma$  denotes  $\parallel$  or  $\perp$ . In the above expression it has already been taken into account that the polymer matrix is isotropic with refractive index  $n_p$  and  $c$  is the velocity of light in vacuum. The matrix elements are given by

$$
\mathcal{B}_{ee}^{\gamma} = \int_{\mathbf{k}_{in}} \int_{\mathbf{k}_{out}} [(\varphi_{\gamma,\mathbf{k}_{in}}^{2} - \varphi_{\gamma,\mathbf{k}_{in}} \varphi_{\gamma,\mathbf{k}_{out}}) B_{ee}(\mathbf{k}_{in}, \mathbf{k}_{out}) \n+ \varphi_{\gamma,\mathbf{k}_{in}}^{2} B_{eo}(\mathbf{k}_{in}, \mathbf{k}_{out})],
$$
\n
$$
\mathcal{B}_{oo}^{\gamma} = \int_{\mathbf{k}_{in}} \int_{\mathbf{k}_{out}} [(\varphi_{\gamma,\mathbf{k}_{in}}^{2} - \varphi_{\gamma,\mathbf{k}_{in}} \varphi_{\gamma,\mathbf{k}_{out}}) B_{oo}(\mathbf{k}_{in}, \mathbf{k}_{out}) \n+ \varphi_{\gamma,\mathbf{k}_{in}}^{2} B_{oe}(\mathbf{k}_{in}, \mathbf{k}_{out})],
$$
\n
$$
\mathcal{B}_{eo}^{\gamma} = - \int_{\mathbf{k}_{in}} \int_{\mathbf{k}_{out}} \varphi_{\gamma,\mathbf{k}_{in}} \varphi_{\gamma,\mathbf{k}_{out}} B_{eo}(\mathbf{k}_{in}, \mathbf{k}_{out}), \qquad (5)
$$

where the functions  $\varphi_{\gamma,k}$  are the first eigenfunctions of the structure factor (for details see Ref. [[16](#page-5-15)]). In the case of bulk nematic liquid crystals it has been shown that modified spherical harmonics with  $l=1$  defined in Ref.  $[16]$  $[16]$  $[16]$  are a good approximation for the eigenfunctions. If the medium is not birefringent, these functions simplify to normal real spherical harmonics with  $l=1$  and  $|m|=0,1$  for  $D_{\parallel}$  and  $D_{\perp}$ , respectively. In our case the spherical harmonics are also sufficiently good if we take into account the functions up to *l*=7. The error of all the obtained values for  $D_{\parallel}$  and  $D_{\perp}$  is then smaller than 2% and 0.2%, respectively. The error is smaller for smaller droplet radii and lower fields. For small volume fractions we have neglected that in the external field the PDLC becomes slightly birefringent. For values of the volume fraction of 10% and more we have used the modified spherical harmonics as eigenfunctions as in Ref.  $[16]$  $[16]$  $[16]$ , which account for the average birefringence of the system.

The optical structure function of a single droplet is proportional to the differential cross section and was calculated using the DDA  $\lceil 17 \rceil$  $\lceil 17 \rceil$  $\lceil 17 \rceil$ . We modified the FORTRAN DDA code written by Draine and Flatau  $[18]$  $[18]$  $[18]$  to enable calculations of the scattering cross section for the inhomogenuous director configuration of the droplets. In the DDA a scatterer is described by a number of dipoles with a given polarizability tensor placed on a discrete lattice. In the DDA code written by Draine and Flatau the input parameters for the polarizability tensor for a given lattice point are the eigenvalues of the polarizability tensor. In our modification we added in the description of the polarizability tensor at a given lattice point

two angles that describe the rotation of the polarizability tensor with respect to the droplet's coordinate system. The simplified configuration of the director within the bipolar droplet was taken as shown in Fig. [1.](#page-1-0) It corresponds to strong planar anchoring at the droplet' surface; i.e., the director at the surface is tangential to the surface everywhere on the surface but on the poles, where point defects are located. On the axis of the droplet the director is parallel to the axis, and in the region from the axis to the surface the curvature of the nematic field gradually increases as shown in Fig. [1.](#page-1-0) Refractive indices used in our calculations were polymer  $n_p = 1.55$ and ordinary and extraordinary indices of liquid crystal  $n<sub>o</sub>$  $= 1.52$  and  $n_e = 1.70$ , respectively. The number of dipoles used in the DDA was 15 515, which was enough to satisfy the criteria for applicability of the DDA  $[19]$  $[19]$  $[19]$ . We have chosen the DDA since the range of parameters of the droplets  $(kR,$  indices of refraction) is such that neither the Rayleigh-Gans nor anomalous diffraction approximation fully applies [[20](#page-5-18)]. The optical structure function can be written approximately as the product of the single-droplet optical structure function (the form factor) and droplet structure factor. A Percus-Yevick approximation for hard spheres has been used as a structure factor  $[21]$  $[21]$  $[21]$ .

The orientational distribution function of the droplets in the external field was taken to correspond to a simple case of the thermal distribution of spherical droplets embedded in an isotropic medium. In the external electric field *E* the potential energy of an anisotropic droplet with volume *V* is  $\epsilon_a \epsilon_0 E^2 V \cos^2(\theta)$ , where  $\epsilon_a$  is the average anisotropy of the dielectric tensor of the droplet and  $\vartheta$  is the angle between the direction of the field and the orientation of the droplet. In thermal equilibrium in the absence of other potentials the orientational distribution function is  $f(\theta) = Ae^{b_0^2 \cos^2(\theta)}$ , where *A* is the normalization factor and  $b_0 = \sqrt{\frac{\epsilon_a \epsilon_0 E^2 V}{k_B T}}$  the normalized field.

The anisotropy of the diffusion tensor  $(D_{\parallel}-D_{\perp})/D_{\perp}$  is shown in Fig. [2](#page-2-0) as a function of normalized applied field  $b_0$ for uniform and bipolar configurations of the director inside the droplet and for different volume fractions  $\rho$  of the droplets. Surprisingly, the sign of the anisotropy differs for different configurations of the director field inside the droplets. For the uniform configuration the anisotropy is first slightly negative up to the value of the normalized field  $b_0=2$  and then increases with  $b_0$  and reaches the maximum value of 0.22 at  $b_0$ =6 for dilute samples and value of 0.21 at  $b_0$ =7 for volume fraction  $\rho = 0.5$  and then decreases with  $b_0$  but remains positive. For the bipolar configuration, which has a smaller anisotropy of the scattering cross section, the anisotropy is always negative.

In Fig. [3](#page-2-1) the anisotropy as a function of volume fraction of liquid crystals in the PDLC sample is shown for the value of normalized field  $b_0$ = 17 and radius  $R$ = 250 nm. While for bipolar droplets the anisotropy does not depend much on volume fraction, it increases with the volume fraction for uniform droplets.

Figure [4](#page-3-0) shows the dependence of both diffusion constants on the normalized field. The diffusion constant in the direction parallel to external field—i.e., to the average orientation of the droplets—first slightly  $(\sim 1\%)$  decreases with

<span id="page-2-0"></span>

FIG. 2. (Color online) Anisotropy of the diffusion constant  $(D_{\parallel}-D_{\perp})/D_{\perp}$  versus applied external field for uniform and bipolar configurations and for volume fractions 0.01 and 0.5. *R*= 250 nm  $(kR = 4.73)$ .

the external field and for values of the normalized field more than 2 (when also the anisotropy becomes positive) it increases as expected. An increase of both diffusion constants with field is expected in the dilute system because of increasing orientational correlations between the droplets and, at higher volume fraction of nematic droplets in the polymer, also because of positional correlations between the droplets.

In Fig. [5](#page-3-1) the dependence of the diffusion constants on volume fraction is shown for uniform and bipolar configurations. The diffusion constants of bipolar droplets are smaller than for uniform configuration for both zero field and  $b_0$ = 17. For zero field the diffusion constants decrease as for uncorrelated scattering, where  $D \propto 1/\rho$  (structure factor=1). This is expected, as there are no orientational correlations between neighboring droplets. Since the refractive indices of neighboring droplets for the incoming light with a given

<span id="page-2-1"></span>

FIG. 3. (Color online) Anisotropy of diffusion constant  $(D_{\parallel}-D_{\perp})/D_{\perp}$  versus volume fraction for uniform and bipolar configurations for  $R = 250$  nm  $(kR = 4.73)$  and  $b_0 = 17$ .

<span id="page-3-0"></span>

FIG. 4. (Color online) Diffusion constants normalized by the value at  $b0=0$  versus applied field for uniform and bipolar configurations and for volume fractions 0.01 and 0.5. *R*= 250 nm  $(kR = 4.73)$ .

wave vector are independent, the scattering from neighboring droplets is independent and, consequently, also the positional correlations have no effect on the scattering. For high fields the droplets are orientationally correlated and also positional correlation effects become clearly visible. In Fig. [5](#page-3-1) the calculated dependences of the diffusion constants on volume fraction differ from the lines that correspond to uncorrelated scattering.

Figure [6](#page-3-2) shows the dependence of the diffusion constants for zero field and at  $b_0=17$  on the radius of the droplets for uniform and bipolar configurations for the dilute system. In Fig. [7](#page-3-3) the corresponding anisotropies of the diffusion constant at  $b_0 = 17$  for both configurations are shown. While in the absence of the field the smallest value of the diffusion constant for the bipolar configuration is at *R*= 150 nm  $(kR = 2.84)$ , in the field it is at  $R = 250$  nm  $(kR = 4.73)$ . For

<span id="page-3-1"></span>

FIG. 5. (Color online) Diffusion constants versus volume fraction for uniform and bipolar configurations for *R*= 250 nm  $(kR=4.73)$ . Lines correspond to uncorrelated scattering-i.e., the structure factor= 1.

<span id="page-3-2"></span>

FIG. 6. (Color online) Dependences of diffusion constants on radius for  $b_0=0$  and  $b_0=17$  for bipolar and uniform configurations. Volume fraction is 0.01.

uniform droplets no minima are observed in the region of our calculations; it is, however, interesting that the anisotropy of the diffusion constant changes sign at approximately *R*  $= 250$  nm ( $kR = 4.73$ ). In this case the increase of the diffusion constants in the field is slightly larger than in the bipolar case; the values at  $b_0=17$  are by a factor of 2.2–4.3 larger than the values at  $b0=0$  for volume fraction  $\rho = 0.01$  and 4.9–5.8 for  $\rho$ =0.5, while for bipolar droplets this factor is in the range from 1.3–2.8 for  $\rho = 0.01$  and 2.2–3.7 for  $\rho = 0.5$ .

## **III. DISCUSSION**

Our results show two main interesting features. First, the values of both diffusion constants  $D_{\parallel}$  and  $D_{\perp}$  increase with

<span id="page-3-3"></span>

FIG. 7. (Color online) Dependence of anisotropy on the radius at  $b_0 = 17$  for bipolar and uniform configurations for two different volume fractions.

the field and the values in high fields are by a factor of about 2–6 larger than in the absence of the field. This means the system becomes less scattering, more transparent in all directions. Consequently, also the transport mean free path in any direction is in the field larger than in the absence of the field. On the other hand, the contrast between the refractive index of the polymer and the average refractive index of the liquid crystal droplets for the light wave vector perpendicular to the field increases with the field, so that for the dilute system the scattering mean free path  $l_{\text{sea},\perp}$  decreases by a factor of about 10 in high fields and along the field the contrast between the indices decreases so that the  $l_{\text{scal}}$  increases. And second, the anisotropy of the diffusion of light is small and in some cases even negative.

The negative anisotropy of the diffusion of light is somehow surprising when one misleadingly thinks of the scattering mean free path—i.e., the total scattering cross sections. Negative anisotropy namely means that the light diffuses faster in the direction for which the scattering mean free path is smaller. When talking about the scattering mean free path, we think of the light intensity that propagates in a given direction *unscattered*, while when talking about the transport mean free path or diffusion constant, we think of *scattered* light intensity that propagates in that direction. In the isotropic case this significant difference is not that obvious, since the transport mean free path is proportional to the scattering mean free path. However, its importance can already be seen also for the isotropic case. Although the scattering mean free path of the randomly oriented bipolar droplet is larger than of the randomly oriented uniform droplet of the same radius, the transport mean free path of the bipolar droplet is smaller (Fig. [5](#page-3-1)). In expression (1) this means the effect of  $1/\langle 1-\cos \vartheta \rangle$  prevails over  $l_{sca}$ . In the case of optically anisotropic scatterers the weighted average  $\langle 1 - \cos \vartheta \rangle$  over the scattering angle  $\vartheta$  is in a way replaced by a combination of the averages of certain angular functions (the eigenfunctions) over both incoming and scattered wave vectors with different combinations of polarizations  $\alpha$  and  $\beta$  (i.e., the matrix elements  $\mathcal{B}_{\alpha\beta}^{\gamma}$  as is given by expression ([4](#page-1-2)) [[16](#page-5-15)].

The question arises as to what are the conditions for negative anisotropy of the diffusion of light in the case of optically uniaxial scatterers for positive optical anisotropy  $n_{\parallel}$ − $n_{\perp}$  > 0. For small droplets *(kR* < 1) the angular dependence of the differential cross section is weak  $\lceil 20 \rceil$  $\lceil 20 \rceil$  $\lceil 20 \rceil$  and, consequently,  $l^*(\mathbf{k}_{in}) \approx l_{sca}(\mathbf{k}_{in})$ , so the negative anisotropy of diffusion is achieved only when the index of refraction of the polymer is close or equal to the extraordinary index of refraction of liquid crystal,  $n_{\parallel}/n_p \approx 1$ . For larger droplets the angular dependence of the differential cross section of an average droplet becomes significant. It depends on *kR*, indices of refraction, the configuration of the nematic droplet, and wave vector of the incoming light. In Fig. [8](#page-4-0) the dependence of the scattered intensity on the scattering angle  $\vartheta$  is shown for average scattering (averaged over the azimuthal scattering angle  $\varphi$  and over orientations of the spheres for  $b_0$ =6) for the sphere with uniform configuration and with bipolar configuration. The shape (number, position, and the depths of the minima) of the differential cross section depends on *kR* and configuration of the droplet, while its mag-

<span id="page-4-0"></span>

FIG. 8. (Color online) Dependence of differential cross section averaged over the azimuthal scattering angle  $\varphi$  and over orientations of the sphere for  $b_0=6$  for bipolar and uniform configurations with the anisotropy of the diffusion constant −0.18 and 0.23, respectively.  $\parallel$  and  $\perp$  stand for the incoming wave vector parallel and perpendicular to the droplet's axis, respectively. *R*= 250 nm.

nitude depends on the indices of refraction. For negative anisotropy of the diffusion constant the scattering in the forward direction for the incoming light perpendicular to the optical axis must dominate more over the scattering in other directions than for the incoming light parallel to the optical axis. This is shown in Fig. [8.](#page-4-0) For the case where the wave vector of incoming light is parallel to the optical axis, the intensity scattered by the bipolar droplet is still mainly in forward directions ( $0^{\circ} \le \theta \le 45^{\circ}$ ); however, relatively more intensity is scattered in other directions ( $\vartheta \ge 45^{\circ}$ ) than for the case when the wave vector of incoming light is perpendicular to the optical axis. The corresponding anisotropy of the diffusion constant is negative,  $(D_{\parallel} - D_{\perp})/D_{\perp} = -0.18$ . For a uniform sphere the situation is different; for both parallel and perpendicular wave vectors of incoming light, the ratio of scattered intensity in forward directions and other directions is approximately the same (the shape of both curves is similar) and the anisotropy of the diffusion constant is positive,  $(D_{\parallel} - D_{\perp})/D_{\perp} = 0.23$ —i.e., reflects the optical anisotropy. However, the differences in the scattered intensities for both cases are small (note that the intensity axis in Fig. [8](#page-4-0) is logarithmic) and it is difficult to tell from the dependences of the differential cross sections on the scattering angle  $\vartheta$ whether the anisotropy of the diffusion of light is positive or negative. Not only the differential cross sections for the directions of the wave vector of incoming light other than parallel and perpendicular to the optical axis matter, also the probability that the light is scattered away from the perpendicular or parallel direction, when it already propagates in that direction, is important; i.e., the total scattering cross section for a given direction of incoming light is important  $\left[l_{sca}\right]$ in Eq.  $(1)$  $(1)$  $(1)$  for the isotropic case]. The sensitivity of the anisotropy of the diffusion tensor on the details of differential cross sections is reflected also in its nonmonotonic behavior as functions of the normalized external field and radius (kR).

What do our results tell about the PDLCs? First we have to note that the dependences on the normalized field (Figs. [2](#page-2-0))

and [4](#page-3-0)) do not show the switching behavior of the PDLC film, since the normalized field in our case is a parameter in the orientational distribution function, which is not directly connected to the applied voltage; the switching in a PDLC film is more abrupt and the orientational distribution function as a function of applied voltage more complicated, since neither are the droplets perfectly spherical nor is the polymer around the droplets perfectly isotropic. Usually there is a preferred orientation for each droplet, and in order to orient the droplet along the field—i.e., away from the preferred orientation the torque on the droplet due to the electric field must prevail over the elastic torque associated with the preferred orientation, which usually occurs above some treshold field (see, for example, Ref.  $[22]$  $[22]$  $[22]$ ). For fields  $b_0 \le 10$  a bipolar configuration is expected and, consequently, also the negative anisotropy of the diffusion constant in the case of refractive indices used in our calculations. This means the system becomes more transparent in the direction perpendicular to the external field than in the direction along the field, which is somewhat contradictory to what was previously believed. However, the calculated transport mean free path for our parameters in the absence of the field at volume fraction 0.5 is 20–50  $\mu$ m, which is comparable to the usual thickness of a PDLC film, but is, on the other hand, much less than the lateral dimensions of a usual PDLC film. That means that already in moderate external fields multiple scattering is no longer in the diffusion limit, and for its description a ballistic approach (Monte Carlo) is suitable [[10](#page-5-8)]. At high enough fields the transport mean free path increases by a factor of about 5 and is considerably larger than the thickness, so almost no multiple scattering is present and at least the transmittance can be well described by single scattering  $(6,7,23)$  $(6,7,23)$  $(6,7,23)$  $(6,7,23)$  $(6,7,23)$ . Such a situation is necessary for optimal performance of light shutters made of PDLC films; i.e., in the absence of the applied voltage the transport mean free path must be smaller than the thickness of the film and when the voltage is applied it should be much larger, so that the light scattering goes from multiple scattering where the transport mean free path (the diffusion constant) is the relevant parameter to single scattering where the scattering mean free path is important. PDLCs for applications in random lasers and the H-PDLCs are usually thicker  $\lceil 4 \rceil$  $\lceil 4 \rceil$  $\lceil 4 \rceil$  and the diffusion approximation in many cases applies.

In this paper we have not studied the dependence of diffusion constants and its anisotropy on the refractive indices of liquid crystals and polymers. The values of refractive indices strongly affect the differential cross section, and the dependences are certainly not trivial.

In conclusion, we have calculated the diffusion of light and its anisotropy in polymer dispersed liquid crystals. Our results show that the anisotropy of the diffusion constants depends on the configuration on the nematic director, *kR*, and external field in a very subtle way, and it is difficult to predict even its sign without actually carrying out the calculations.

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