Anisotropic Light Diffusion in Oriented Nematic Liquid Crystals

Bart A. van Tiggelen, Roger Maynard, and Anne Heiderich
Laboratoire de Physique Numérique des Systèmes Complexes/CNRS, Université Joseph Fourier, Maison des Magistères, B.P. 166, 38042, Grenoble Cedex 9, France
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Multiple light scattering from thermal fluctuations is studied in single-domain nematic liquid crystals. These systems are uniaxial so that diffusion of light may be anticipated to be anisotropic and polarization to persist in the diffuse regime, in sharp contrast with what is known for light diffusion in isotropic media. Our study enables us to do the first quantitative predictions of light diffusion as a function of the scalar order parameter. [S0031-9007(96)00708-9]

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Since the pioneering work of de Gennes, it is known that fluctuations in the dielectric constant of nematic liquid crystals (and hence light scattering) are dominated by thermal fluctuations of the local director [1–3]. Special aspects of these fluctuations are their tensorial nature as well as their weak but long-range nature. Single light-scattering experiments, both elastic [4] and quasielastic ones [5,6], have successfully been interpreted. Several theoretical studies have been carried out for multiple scattering in nematic states [7–9].

This Letter deals with the theory of light diffusion in single-domain nematic liquid crystals. Diffusion is the extreme limit of multiple scattering and sets in when the medium is much bigger than the mean free path, typically a millimeter in monodomain nematics. Experimentally, it is not easy to create a single-domain nematic sample exceeding this length. Nevertheless, the observation of coherent backscattering in oriented nematics has recently been reported [10] in samples of 1 cm. The data seem consistent with diffusion theory using the de Gennes phase function for light scattering, although anisotropy in the diffusion could not be resolved. Future experiments will certainly try to resolve this interesting feature [11] in more detail. Our diffusion theory, obtained from Maxwell's equations, may offer a theoretical support. In the diffusive regime only few observables remain, such as diffusion tensor D_{ij}, and "Stokes" correlator $\langle E_i E_j^* \rangle$ of the electric polarization vector. In most optical media both diffusion tensor and Stokes correlator are necessarily isotropic (proportional to δ_{ij}). Single-domain nematic liquid crystals offer a unique opportunity to study persisting and controlled anisotropy in multiple scattering.

We first state some rigorous results of light transport theory that we shall mention without derivation, and which serve as a starting point for our calculations. These results find their equivalent in electron transport [12]. Microscopic treatments of scalar-wave transport have been published [13], and a lot of progress has been made concerning the vector character of light [14]. In an infinite random medium the transport of the second-rank tensor $\langle E_i E_k^* \rangle$ in space and time is described by a four-rank tensor $L_{ijkl}(t, \mathbf{r})$ [14]. If energy is conserved, its Laplace-Fourier transform has the asymptotic form

$$L_{ik\mathbf{p},jl\mathbf{p}'}(\Omega, \mathbf{q} \to 0, \omega) = \frac{d_{ik}(\omega, \mathbf{p}, \mathbf{q}) d_{ij}(\omega, \mathbf{p}', \mathbf{q})}{-i\Omega + \mathbf{q} \cdot \mathbf{D}(\omega) \cdot \mathbf{q}}, \quad (1)$$

where d_{ik} represents the eigentensor of the collision operator corresponding to the hydrodynamic eigenvalue close to zero [14]. In time and space coordinates this equation is equivalent to the diffusion equation with an anisotropic diffusion tensor $\mathbf{D}$. The reciprocity principle guarantees the symmetric form of L_{ijkl} on incident and outgoing indices. The diffusion tensor $\mathbf{D}$ is given by a Kubo-Greenwood type formula [12,13],

$$\mathbf{q} \cdot \mathbf{D}(\omega) \cdot \mathbf{q} = \frac{1}{2\pi N(\omega)} \sum_{\mathbf{p}} \{ \boldsymbol{\gamma}(\mathbf{p}, \mathbf{q}) \cdot \mathbf{G}(\omega, \mathbf{p}) \mathbf{G}^*(\omega, \mathbf{p}) \cdot \mathbf{L}(\mathbf{p}, \mathbf{q}) - [\mathbf{q} \cdot \partial_{\mathbf{p}} \, \mathrm{Re}\mathbf{G}(\omega, \mathbf{p})] \cdot \mathbf{L}(\mathbf{p}, \mathbf{q}) \}, \quad (2)$$

in which the second-rank tensor $\mathbf{L}(\mathbf{p}, \mathbf{q}) = 2(\mathbf{p} \cdot \mathbf{q})\mathbf{I} - \mathbf{pq} - \mathbf{qp}$ and $N(\omega)$ represents the density of states per unit volume with frequency ω. The bilinear second-rank tensor $\boldsymbol{\gamma}$ describes angular correlations in multiple scattering as expressed by the integral equation,

$$\boldsymbol{\gamma}(\mathbf{p}, \mathbf{q}) = \mathbf{L}(\mathbf{p}, \mathbf{q}) + \sum_{\mathbf{p}'} \boldsymbol{\gamma}(\mathbf{p}', \mathbf{q}) \cdot \mathbf{G}(\omega, \mathbf{p}') \mathbf{G}^*(\omega, \mathbf{p}') \cdot U_{\mathbf{p}'\mathbf{p}}(\omega), \quad (3)$$

in terms of the Bethe-Salpeter vertex $U_{\mathbf{p}'\mathbf{p}}(\omega)$ and the Dyson Green's tensor $\mathbf{G}(\omega, \mathbf{p})$ [15]. For isotropic media with weak disorder the famous Boltzmann result applies, $\gamma_{ij} = 2(\mathbf{p} \cdot \mathbf{q})\delta_{ij}/(1 - \langle \cos\theta \rangle)$, which makes the extinction length different from the mean free path.

The polarization of diffuse radiation is described by the second-rank tensor d_{ij} in Eq. (1). Perturbation theory gives us

$$\mathbf{d}(\mathbf{p}, \mathbf{q}) = -\frac{2\omega}{\pi c_0} \mathrm{Im}\mathbf{G}(\omega, \mathbf{p}) - \frac{i\omega}{\pi c_0} \boldsymbol{\gamma}(\mathbf{p}, \mathbf{q}) \cdot \mathbf{G}(\omega, \mathbf{p}) \mathbf{G}^*(\omega, \mathbf{p}). \quad (4)$$

The first term is known as the spectral function in electron transport theory. The second term represents a small anisotropy in the specific intensity profile arising from a net energy flow, which in turn is due to a small energy-density gradient (in Fourier space expressed by $\mathbf{q}$).

We shall apply these results to single-domain nematic liquid crystals, relying on four realistic assumptions to be mentioned below. The uniaxiality of the nematic host state (one optical axis $\mathbf{n}$ with dielectric tensor $\boldsymbol{\varepsilon} = \varepsilon_\perp \mathbf{I} + \varepsilon_a \mathbf{nn}$) gives rise to two modes of propagation, defined as *ordinary* (O) and *extraordinary* (E). They possess orthonormal polarization vectors $\mathbf{o}$ and $\mathbf{e}$ with separate dispersion laws $\omega_{e/o}(\mathbf{k})$ and group velocities $\mathbf{v}_{e/o}$ [16]. In the first Born approximation (our first approximation), scattering of light is described by the static (four rank) correlation tensor [2,3],

$$U_{\mathbf{p},\mathbf{p}+\mathbf{f}}(\omega) = \left(\frac{\omega}{c_0}\right)^4 \langle \delta\boldsymbol{\varepsilon}(\mathbf{0}) \delta\boldsymbol{\varepsilon}(\mathbf{f}) \rangle = \frac{(\omega/c_0)^4 \varepsilon_a^2 kT/K_1}{\mathbf{f}^2 + A(\mathbf{f}\cdot\mathbf{n})^2 + 1/\xi^2} \times \sum_{\alpha=1,2} (\mathbf{e}_\alpha \mathbf{n} + \mathbf{n}\mathbf{e}_\alpha)(\mathbf{e}_\alpha \mathbf{n} + \mathbf{n}\mathbf{e}_\alpha). \quad (5)$$

We introduced unit vectors $\mathbf{e}_1 = (\mathbf{f}\times\mathbf{n})/|\mathbf{f}\times\mathbf{n}|$, $\mathbf{e}_2 = \mathbf{n}\times\mathbf{e}_1$, and $A = K_3/K_1 - 1$; the elastic constants for splay, twist, and bend deformations are K_1, K_2, and K_3 [17]. In the adopted "two-constant approximation" of the free energy ($K_1 = K_2$), our second approximation, polarization, and spatial aspects decouple conveniently. A magnetic field induces a finite correlation length $\xi = \sqrt{K_1/\chi_a B^2}$, typically 1 μm for 1 T, and much less than the mean free path. This guarantees that a pointlike treatment of the fluctuations (our third approximation) can be used. This approximation has explicitly been verified by us in the scalar-wave picture [18], for which it is even possible to treat the long-range $1/r$ structure factor of the thermal fluctuations in the absence of magnetic field. All these parameters can be related to the scalar order parameter $S = \frac{1}{2}\langle 3\cos^2\theta - 1\rangle$ of the nematic phase. One expects that on varying the order parameter, ε_a, $\chi_a \sim S$, and $K_i \sim S^2$ [3], so that $\xi \sim \sqrt{S}$ and $A \sim$ const. Although exceptions to these scaling relations exist, they can serve to understand our numerical results as a function of the order parameter.

The Dyson Green's tensor is

$$\mathbf{G}(\mathbf{p}) = \frac{\mathbf{ee}}{\cos^2\delta_e[k_e^2(\hat{\mathbf{p}}) - p^2] + ik_e(\hat{\mathbf{p}})/\ell_e(\hat{\mathbf{p}})} + \frac{\mathbf{oo}}{k_o^2 - p^2 + ik_o/\ell_o(\hat{\mathbf{p}})}; \quad (6)$$

$\delta_e(\hat{\mathbf{p}})$ is the angle between phase and group velocity of the E wave. The polarization- and angle-dependent extinction length $\ell_{e/o}(\hat{\mathbf{p}})$ can be calculated numerically [8,19]. The longitudinal field as well as its coupling to the E mode are beyond the first Born approximation.

Our main task is to solve Eq. (3) for a nematic liquid crystal. EO interference can be neglected since $\varepsilon_a \omega \ell_{e,o}/c_0 \approx 10^3 \gg 1$, implying that E and O waves dephase between two collisions. This is our fourth and final approximation. Reciprocity imposes that $\gamma_{ij}(\mathbf{p},\mathbf{q}) = \gamma_{ji}(-\mathbf{p},-\mathbf{q})$. Uniaxial symmetry allows the presence of two vectors $\mathbf{n}$ and $\mathbf{o} \propto \mathbf{n}\times\hat{\mathbf{p}}$ in this tensor. However, a selection rules for OO polarization transitions guarantees the ordinary vector $\mathbf{o}$ to be absent. This leaves us with

$$\gamma(\mathbf{p},\mathbf{q}) = 2(\mathbf{p}\cdot\mathbf{q})\mathbf{A}(p,c) - \mathbf{pq} - \mathbf{qp} + (\mathbf{n}\cdot\mathbf{p})(\mathbf{q}\cdot\mathbf{n})\mathbf{B}(p,c). \quad (7)$$

The symmetric tensors $\mathbf{A}$ and $\mathbf{B}$ are even functions of $c = \mathbf{n}\cdot\hat{\mathbf{p}}$. From Eq. (3) we obtain a closed set of coupled equations for the scalar variables $\mathbf{e}\cdot\gamma(\hat{\mathbf{p}},\mathbf{q})\cdot\mathbf{e} \equiv \gamma_e(\hat{\mathbf{p}},\mathbf{q})$, $\mathbf{o}\cdot\gamma(\hat{\mathbf{p}})\cdot\mathbf{o} \equiv \gamma_o(\hat{\mathbf{p}},\mathbf{q})$,

$$\gamma_e(\hat{\mathbf{p}},\mathbf{q}) = 2(\hat{\mathbf{v}}_e\cdot\mathbf{q})\cos\delta_e + \int \frac{d^2\hat{\mathbf{p}}'}{4\pi}\Phi_{EE}(\mathbf{k}_e,\mathbf{k}_e')\gamma_e(\hat{\mathbf{p}}',\mathbf{q}) + \int \frac{d^2\hat{\mathbf{p}}'}{4\pi}\Phi_{EO}(\mathbf{k}_e,\mathbf{k}_o')\gamma_o(\hat{\mathbf{p}}',\mathbf{q}), \quad (8a)$$

$$\gamma_o(\hat{\mathbf{p}},\mathbf{q}) = 2(\hat{\mathbf{v}}_o\cdot\mathbf{q}) + \int \frac{d^2\hat{\mathbf{p}}'}{4\pi}\Phi_{OE}(\mathbf{k}_o,\mathbf{k}_e')\gamma_e(\hat{\mathbf{p}}',\mathbf{q}), \quad (8b)$$

with phase functions

$$\Phi_{EE}(\mathbf{k}_e,\mathbf{k}_e') = \frac{k_e'\ell_e(\mathbf{k}_e')}{4\pi k_e\cos^2\delta_e'}\mathbf{e}'\mathbf{e}'\cdot U_{\mathbf{k}_e,\mathbf{k}_e'}(\omega)\cdot\mathbf{ee}, \quad (9a)$$

$$\Phi_{EO}(\mathbf{k}_e,\mathbf{k}_o') = \frac{k_o'\ell_o(\mathbf{k}_o')}{4\pi k_e}\mathbf{o}'\mathbf{o}'\cdot U_{\mathbf{k}_e,\mathbf{k}_o'}(\omega)\cdot\mathbf{ee}, \quad (9b)$$

$$\Phi_{OE}(\mathbf{k}_o,\mathbf{k}_e') = \frac{k_e'\ell_e(\mathbf{k}_e')}{4\pi k_o\cos^2\delta_e'}\mathbf{e}'\mathbf{e}'\cdot U_{\mathbf{k}_o,\mathbf{k}_e'}(\omega)\cdot\mathbf{oo}, \quad (9c)$$

and the selection rule $\Phi_{OO}(\mathbf{k}_o,\mathbf{k}_o') = 0$. Two choices for $\mathbf{q}$ ($\hat{\mathbf{p}}$ and $\mathbf{n}$) allow us to determine the four functions $A_{e/o}(c)$, $B_{e/o}(c)$ numerically (Fig. 1).

Uniaxial symmetry imposes that $D_{ij} = D_\perp\delta_{ij} + (D_\parallel - D_\perp)n_i n_j$. Neglecting again EO interference, Eq. (2) simplifies to

$$D_\perp q^2 + (D_\parallel - D_\perp)(\mathbf{q}\cdot\mathbf{n})^2 = \frac{c_0^3}{4\pi\omega^2(\varepsilon_\parallel + \varepsilon_\perp)\sqrt{\varepsilon_\perp}} \times \left[\int d^2S_e(\hat{\mathbf{v}}_e\cdot\mathbf{q})\ell_e(\hat{\mathbf{p}})\frac{1}{2}\gamma_e(\hat{\mathbf{p}},\mathbf{q}) + \int d^2S_o(\hat{\mathbf{v}}_o\cdot\mathbf{q})\ell_o(\hat{\mathbf{p}})\frac{1}{2}\gamma_o(\hat{\mathbf{p}},\mathbf{q})\right]. \quad (10)$$

$D_\perp$ and $D_\parallel$ follow from Eq. (10) by separately choosing $\mathbf{q} = \mathbf{n}$ and integration over all angles of $\mathbf{q}$.

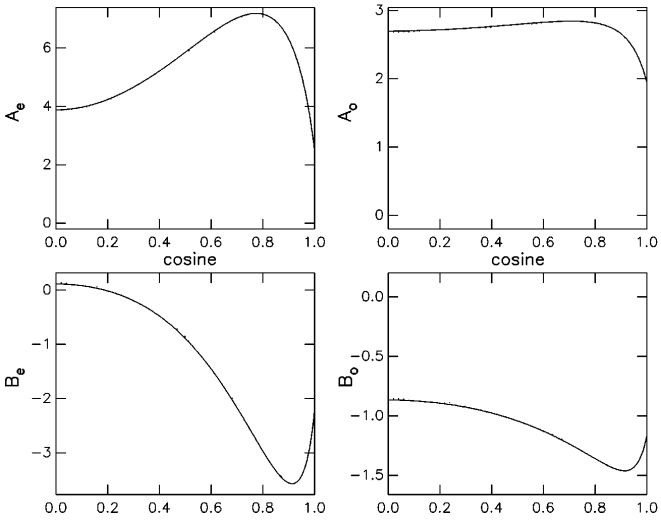

FIG. 1. Solution of Eq. (8) in a nematic liquid crystal, obtained after 10 iterations. Shown are $A_o(c)$, $A_e(c)$, $B_o(c)$, and $B_e(c)$, defined in Eq. (7) ($\varepsilon_a = -0.2$, $\varepsilon_\perp = 1.0$, $\omega\xi/c_0 = 63.7$, and $a = -0.254$). We obtained for the diffusion tensor $D_\parallel = 0.905 D_{\rm scal}$ and $D_\perp = 1.021 D_{\rm scal}$, with $D_{\rm scal}$ the diffusion constant obtained in the scalar theory.

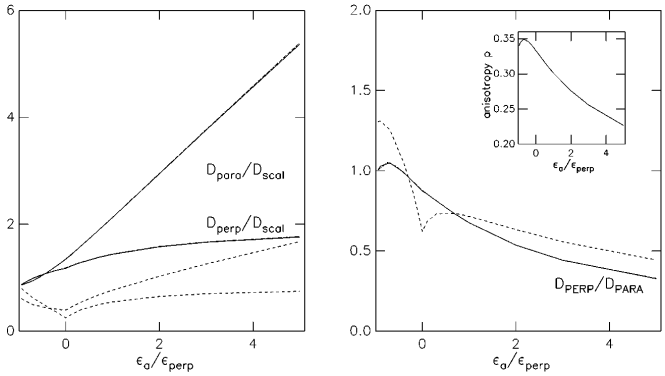

FIG. 2. Numerical solution of Eqs. (8) and (10). Left: Diffusion constant parallel and perpendicular to optical axis, expressed in scalar units, as a function of the dielectric anisotropy $\varepsilon_a/\varepsilon_\perp$ ($\varepsilon_\perp = 1$ and $A = 0.254$). The curves for $\omega\xi/c_0 = 63.7$ and $\omega\xi/c_0 = 32.3$ (dashed) essentially overlap. The dashed lines have been obtained by ignoring angular correlations in multiple scattering. For positive anisotropies they differ considerably from the exact ones. Upon varying the order parameter S, the only relevant change in diffusion can be argued to come from $\varepsilon_a \sim S$ (see text). Right: the ratio $D_\perp/D_\parallel$ as a function of the same dielectric anisotropy. Inset for comparison: kinematic anisotropy p of the group velocity tensor $\langle v_i v_j\rangle = p\delta_{ij} + (1 - 3p)n_i n_j$ versus dielectric anisotropy.

In Fig. 2 we display $D_\perp/D_{\rm scal}$, $D_\parallel/D_{\rm scal}$, and $D_\perp/D_\parallel$ calculated numerically as a function of $\varepsilon_a/\varepsilon_\perp$, with ξ and A kept fixed; $D_{\rm scal} = \frac{1}{3} v_o \ell_B^*$ is the diffusion constant obtained in the scalar theory [20], with $\ell_B^* = 8\pi c_0^2 K_1 \varepsilon_\perp / kT\varepsilon_a^2\omega^2$. For $\varepsilon_a/\varepsilon_\perp \gg 1$, the E states can be verified to dominate, and, since their frequency surface becomes flattened along the optical axis, this corresponds to a *quasi-1D case*. This explains why $D_\parallel \gg D_\perp$. For $\varepsilon_a < 0$, the O waves take the upper hand and we infer $D_\perp \approx D_\parallel$. The free energy influences the anisotropy of the diffusion tensor as well: $A > 0$ tends to lower scattering along the optical axis, enhancing the relative importance of $D_\parallel$. This is particularly evident for the nematic PAA (Table I).

It is convenient to discuss our results as a function of the scalar order parameter S, relying on the scaling relations of K_i, ε_a, and ξ discussed earlier. The calculations displayed in Fig. 2 show that the anisotropy in diffusion is rather insensitive to the correlation length ξ as long as it remains less than the mean free path. As A is supposed not to depend on S, the only sensitive parameter is the dielectric anisotropy $\varepsilon_a \sim S$. In this way, Fig. 2 represents the diffusion tensor as a function of the scalar order parameter. We notice that the same scaling arguments lead to the prediction that $D_{\rm scal}$ will not be sensitive to the order parameter S either, just as was predicted 25 years ago for the single scattering transmission [21]. It is interesting to note that upon approaching the phase transition ($S \to 0$) none of our four assumptions seems to be violated. However, since the mean free path diverges as $\xi \to 0$, other scattering mechanisms than thermal fluctuations may take over.

The polarization of the light can be represented by its Stokes correlator $\langle E_i(\mathbf{p} + \mathbf{q}/2)E_k^*(\mathbf{p} - \mathbf{q}/2)\rangle$. In the diffuse domain it equals, by Eq. (4), the second-rank tensor $d_{ik}(\mathbf{p}, \mathbf{q})$. The total polarization can be defined as the ratio between the average electromagnetic energy density from E modes and the one from O modes. Since the electromagnetic energy density is proportional to $\varepsilon \cdot \langle \mathbf{E}\mathbf{E}^*\rangle$, we get

$$\rho(\omega, \mathbf{p}) \equiv \varepsilon \cdot \mathbf{d}(\boldsymbol{\omega}, \mathbf{p}) = \mathbf{ee}\,\delta[\omega - \omega_e(\mathbf{p})] + \mathbf{oo}\,\delta[\omega - \omega_o(\mathbf{p})] + \mathcal{O}(\mathbf{p}, \mathbf{q}). \quad (11)$$

Upon integration over $\mathbf{p}$, the degree of polarization becomes

$$\frac{\rho_e}{\rho_o} = \int \frac{d^2 S_e}{|\mathbf{v}_e|} \Big/ \int \frac{d^2 S_o}{|\mathbf{v}_o|} = \frac{\varepsilon_\parallel}{\varepsilon_\perp}, \quad (12)$$

TABLE I. Adopted input data for two common nematic liquid crystals (taken from Tables 3.1 and 3.2 of Ref. [2]), and numerical values for the diffusion tenor of the light ($A \equiv K_3/K_{1,2} - 1$); ℓ_B^* denotes the transport mean free path predicted by the scalar theory given in the text; p is the kinematic anisotropy defined in Fig. 2. We assumed that $K_1 = K_2$. All calculations are done using a vacuum wavelength of 500 nm.

	$\varepsilon_\perp$	ε_a	$K_{1,2}$	A	ξ	T	p	$D_\perp/D_{\rm scal}$	$D_\parallel/D_{\rm scal}$	$D_\perp/D_\parallel$	ℓ_B^*
PAA	2.47	+0.88	3.0 pN	2.17	2.2 μm	398 K	0.3214	1.61	2.82	0.57	0.28 mm
MBBA	5.40	−0.70	3.2 pN	0.91	1.5 μm	299 K	0.3376	1.28	1.64	0.78	1.36 mm

i.e., the ratio of number of microstates available. This shows that long-range diffusion finally leads to equipartition [22], as anticipated heuristically. For $\varepsilon_a < 0$ O radiation dominates in the diffuse regime: For positive anisotropies most radiation will be E.

In summary, we have calculated bulk diffusion characteristics of light scattered from thermal fluctuations in oriented nematic liquid crystals. We calculated the uniaxial anisotropy in diffusion tensor and the degree of polarization in the diffusive domain. Both unfamiliar properties, the functions $A_{e/o}(\theta)$ and $B_{e/o}(\theta)$ introduced in this work, in particular, are observable in the diffuse transmission. Within the diffusion approximation they determine the angular and polarization dependent transmission. The outcome may be compared to multiple scattering experiments in nematic liquid crystals, as a function of the order parameter.

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