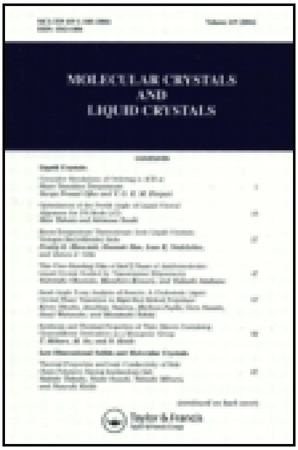

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Optics of Diffuse Light in Nematic Liquid Crystals

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Optics of Diffuse Light in Nematic Liquid Crystals

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This contribution aims to give a theoretical state-of-the-art of a new development in the optics of liquid crystals, namely the one with multiply scattered light, initiated by Refs. 1 and 2. The emphasize will be on its unconventional anisotropic features.

Keywords: Light scattering; liquid crystals; diffusion

OPTICS OF NEMATIC LIQUID CRYSTALS

Light scattering has always been a powerful tool to understand fluctuations in inhomogeneous media. Small inhomogeneities scatter light and the forward transmission profile gives access to the structure function of the fluctuations. Quasi-elastic light scattering also provides details of the dynamics of these fluctuations. In particular, for the physics of a nematic liquid crystal (NLC), light scattering experiments have confirmed theoretical predictions addressing the specificity of the thermal director fluctuations in great detail[3].

It is true that the optics of liquid crystals such as cholesterics is far from conventional. In NLC's however, the fundamentally new physics initially concerned the hydrodynamics of the elastic and viscosity constants that govern the nematic director fluctuations. The optics part of the problem was, though technically involved, straightforward and basically understood. Most observed phenomena were consistent with lowest order perturbation theory of the light-director interaction, in which case light interacts only once and very weakly with a director fluctuation.

Recent work demonstrates new optics in NLC's too. Only writing down the complete set of equations, describing both the hydrodynamics of the NLC and the electromagnetic field is already a *tour de force*[4]. There is no doubt that such equations hide many unexplored phenomena in nonlinear optics. New developments are the possibilities of optical four-wave mixing[5], diffuse transport of light-induced perturbations[6] and enhanced optical nonlinearity[7]. Another one is the possibility of multiple light scattering in oriented NLC's[1,2]. In this process, the light response is linear, but involves many, and even infinitely many director fluctuations. At this point, the physics of liquid crystals and diffuse light start to overlap, and any discussion rapidly becomes technical. This contribution tries to avoid this.

TWO DISCIPLINES MEET ...

The study of light diffusion in the oriented nematic phase has been initiated some 4 years ago by the report of coherent backscattering[8], after earlier work in multi-domain liquid crystals[9]. During the last 10 years, this phenomenon has become a paradigm in the fundamental discussion how wave nature is to affect phenomenological concepts such as "radiative transfer". Coherent backscattering is explained by a constructive *interference* of mutually time-reversed waves, visiting the same scatterers but in opposite order. Reciprocity arguments show that the backscattering from a optically thick sample is enhanced by a factor of (roughly) two compared to the prediction of the equation of radiative transfer[10]. Being an interference effect in multiple scattering, two relevant length scales come to mind: the mean free path ℓ^* of a photon, and its wavelength λ. The expected angular width of coherent backscattering λ/ℓ^* is usually very small. Coherent backscattering has initially been measured in micrometer-sphere suspensions. Not only has the measurement of coherent backscattering ($\ell^* \approx 1$ mm) established the significance of (the role of interference and polarization in) multiple scattering in NLC's, it also demonstrated the possibility to manufacture *optically thick but mono-domain* samples of NLC's.

Well before this first experiment had been done, theoretical studies on higher order light scattering in NLC's had started[11]. In its simplest form, multiple scattering of light can be described by the equation of radiative transfer. As explained in the contribution of H. Stark in this issue, one can acknowledge the birefringent modes E and O in this equation. Like in a Boltzmann equation for particles, one must specify the differential cross-section, which in a NLC depends on the initial and final state of polarization. Former single-scattering studies established that this is basically the structure function associated with the thermal director fluctuations. Hence, a systematic theory of multiple light scattering can be set up.

As always in radiative transport, the observable quantity is the time dependent specific intensity at some place $\boldsymbol{r}$ inside or outside the medium, signifying the local current of energy per unit surface, per unit solid angle in the direction $\boldsymbol{k}$. Because Ialso want to study polarization, I shall introduce a second-rank tensor $\Phi_{ij,\mathbf{k}\omega}(\boldsymbol{r},t)$. The usual description in terms of four Stokes variables[12] is inconvenient in birefringent media. In terms of the electric field vector $\boldsymbol{E}$ of the light, the tensor $\Phi_{ij,\mathbf{k}\omega}(\boldsymbol{r},t)$ is defined as the $\boldsymbol{q}$-Fourier transform and Ω- Laplace transform of

$$\Phi_{ij,\mathbf{p}\omega}(\boldsymbol{q},\Omega) \equiv \left\langle E_i(\boldsymbol{p}+\boldsymbol{q}/2,\omega+\Omega/2)\, E_j^*(\boldsymbol{p}-\boldsymbol{q}/2,\omega-\Omega/2)\right\rangle \qquad (1)$$

The brackets $\langle\cdots\rangle$ stand for thermodynamic ensemble averaging. The microscopic variables ω and $\boldsymbol{p}$ are associated with the fast (femtosecond) cycles of the wave packet in space and time. On the other hand, the macroscopic variables Ω and $\boldsymbol{q}$ determine the much slower (nanosecond) propagation of the wave envelope.

Two important observables will be the current density $\boldsymbol{J}_\omega$ and the photon density ρ_ω. In terms of $\Phi_{ij,\mathbf{k}\omega}(\boldsymbol{r},t)$, Maxwell's equations learn that

$$\rho(\boldsymbol{r},t) = \int \frac{d^3p}{(2\pi)^3}\, \varepsilon_{ik}\Phi_{ki,\mathbf{p}}(\boldsymbol{r},t) \tag{2}$$

$$J_m(\boldsymbol{r},t) = \frac{c_0^2}{\omega}\int \frac{d^3p}{(2\pi)^3}\left(p_m\delta_{ik} - \frac{1}{2}p_k\delta_{mi} - \frac{1}{2}p_i\delta_{mk}\right)\Phi_{ki,\mathbf{p}}(\boldsymbol{r},t)\,. \tag{3}$$

Apart from polarization indices, these expressions look very much like the corresponding equations for current and number density in Boltzmann theory (recall the relations $\boldsymbol{v} = \hbar\boldsymbol{p}/m$ and $\hbar\omega = mc_0^2$ so that $\boldsymbol{v} = \boldsymbol{p}c_0^2/\omega$). Later, I will introduce a third observable that captures the polarization in NLC's. It is anticipated that the momentum integrals give contributions only on the two constant-frequency surfaces $\omega_{E,O}(\boldsymbol{p})$ of O and E waves where propagation takes place. Therefore I will drop frequency labels. The current density in Eq. (3) for a *given* polarization n and wavenumber $\boldsymbol{p}$ - called the specific intensity - is directed along the group velocity, which is the direction of energy transport.

An approximate solution of the exact theory

The radiative transfer equation is a balance equation for $\Phi_{ij,\mathbf{k}}(\boldsymbol{r},t)$, in the same way that the Boltzmann equation is a balance equation for the phase space distribution $f(\boldsymbol{r},\boldsymbol{v},t)$ of classical particles. This equation is very difficult to solve, since it involves integrals and derivatives at the same time. Even for the classical Milne problem (randomly distributed scalar point scatterers in a semi-infinite slab geometry) the solution must be obtained numerically[12]. Well, the fluctuations in NLC's are far from point-like (there are even long-range Goldstone-mode correlations), they affect the polarization of the light in a complicated way (recall the inhibition of single OO scattering in a NLC because the O polarization is orthogonal to the optical axis), and the medium is far from semi-infinite (today from several up to ten mean free paths). It is clear that any approach based on a direct solution of the transfer equation is going to be very difficult and will require some *a priori* uncontrolled approximations[11]. The mathematical complexity is the evident price we pay for trying to describe light *and* NLC the best we can, and is likely to obscure the physics. I do not even want to address the question that also the equation of radiative transfer is not an exact equation because it does not predict the coherent backscattering phenomenon mentioned earlier[13].

The exact solution of an approximate theory

Fortunately, the equation of radiative transfer contains an simple asymptotic limit in the regime of infinite multiple scattering. This means we look on time and length scales very large compared to the typical times and distance between two photon collisions. In this regime the motion of the photon cloud is anticipated to be diffuse, and governed by a diffusion equation,

$$\partial_t \rho(\boldsymbol{r}, t) - \boldsymbol{\nabla} \cdot \boldsymbol{D} \cdot \boldsymbol{\nabla} \rho(\boldsymbol{r}, t) = \text{source} . \tag{4}$$

This equation defines the diffusion tensor $\boldsymbol{D}$. Equation (4) is equivalent* to saying that the current density (3) and photon energy (2) are related by Fick's Law $\boldsymbol{J}(\boldsymbol{r}, t) = -\boldsymbol{D} \cdot \boldsymbol{\nabla} \rho(\boldsymbol{r}, t)$. The diffusion equation is rather straightforward to solve, even for complex geometries and complicated scattering mechanisms. Extensive numerical studies[12] as well as experiments[15] have established that the scaling relations predicted by diffusion theory, at least in normal isotropic media, mimic "reality" very well, all the way down to scales of only one mean free path! It remains to be seen whether the diffusion approximation also applies to anisotropic media such as liquid crystals and magneto-active media[16], being much richer and less evident than the one in isotropic media.

Some 4 years ago, no concrete example was known where the concept of "anisotropic photon diffusion", i.e. $D_{ij} \neq D\delta_{ij}$, had been necessary to invoke. Light diffusion, even in media with strong angular-dependent scattering, always turned out isotropic. A well-known consequence of isotropic diffusion is the universal law $T \sim 1 + (3/2)\cos\theta$ for the angular diffuse

*In the diffusion equation the ***antisymmetric*** part of the diffusion tensor is lost, and one needs to fall back to Fick's law. An example is the Photon Hall Effect[14]. In a NLC one does not expect such a part.

transmission coefficient of a slab. This law explains for instance the "limb darkening" of the light emerging from the Solar photosphere[17]. Due to their broken rotational symmetry, NLC's seem to be an ideal candidate to exhibit anisotropic diffusion, next to light scattering in an external magnetic field. The question is than whether all features can be understood qualitatively and quantitatively from the well-known laws of single scattering. Very recently, anisotropic light scattering has been verified in a magnetic field (the "Photon Hall Effect" [14]) as well as in the liquid crystal 5CB[18].

ANISOTROPIC LIGHT DIFFUSION IN LIQUID CRYSTALS

In diffusion theory one wishes to express $\Phi_{ij,\mathbf{p}}(\boldsymbol{r},t)$ in terms of *only* the photon density ρ and the total current density $\boldsymbol{J}$. The latter are related by Fick's law, and the photon density is obtained by solving the diffusion equation. Key issue is the relation between the microphysics of scattering and fluctuations and the macroscopic motion of the diffuse wave packet, as described by $\boldsymbol{D}$. To understand experiments, one has to make the link with the principal observable $\Phi_{ki,\mathbf{p}}(\boldsymbol{r},t)$ that contains information on photon flux, photon density and polarization. In isotropic media this relation is intuitively obvious and can be found in classical textbooks[19],

$$\begin{aligned}\Phi_{ij,\mathbf{k}} &\sim \left(\delta_{ij} - \hat{\boldsymbol{k}}_i\hat{\boldsymbol{k}}_j\right) \times \delta(\omega/c_0 - k) \times \left[\rho\, v_E + 3\hat{k}_n J_n + \cdots\right] \\ &= v_E \left(\delta_{ij} - \hat{\boldsymbol{k}}_i\hat{\boldsymbol{k}}_j\right) \delta(\omega/c_0 - k) \left[\rho - \ell^* \hat{k}_n \partial_n \rho\right] . \qquad (5)\end{aligned}$$

The second equality follows by applying Fick's law and the familiar identity $D_{ij} = \frac{1}{3} v_E \ell^* \delta_{ij}$. Equation (5) expresses that in isotropic media the polarization is transverse but unpolarized, the waves are concentrated on the frequency shell $k = \omega/c_0$ and finally that the local specific intensity can be obtained from the local photon density and its first derivative. This last aspect suggests that higher order derivatives have apparently been neglected on long length scales.

What will change in this expression for light diffusion in NLC's? In isotropic media the diffusion constant D is a simple product of a (dynamic) transport velocity v_E and a (static) transport mean free path ℓ^*. This is the first property that is not expected in nematics: Not only the group velocity depends on the direction of propagation (due to birefringence), but also the mean free path[20–22]. We expect the diffusion constant to be some weighted angular average of their product, that is not going to factorize in the products of their individual averages. Secondly, as opposed to isotropic media, we might expect the local radiation to be polarized. And finally, in uniaxial media *two different* dispersion laws E/O exist, of which E has a longitudinal component in its electric polarization vector.

Outline of Calculation

The detailed derivation of the generalization of Eq. (5) to liquid crystals will not be given here. The basic technique is to find the eigenvalue closest to zero of the exact transport equation[23,24]. The actual calculation has been carried out with numerical iteration [2] and with spherical harmonic expansion[1]. On infinite length scales - that is $\boldsymbol{q} = 0$ in Eq. (1) - this hydrodynamic eigenvalue must be exactly zero because of conservation laws[25]. The eigenfunction associated with this eigenvalue is the so-called spectral function of the system. For the NLC it reads, in terms of the two electric polarization vectors $\boldsymbol{e}^n$,

$$\Phi_{ij,\mathbf{p}}(\boldsymbol{q}=0) \sim S_{ij}(\omega,\boldsymbol{p}) = \sum_{n=E,O} e_i^n e_j^n \, \delta\left[\omega - \omega_n(\boldsymbol{p})\right] . \tag{6}$$

This function must be the generalization of the term ρ in Eq. (5). The spectral function counts the number density of microstates in phase space $(\boldsymbol{r},\boldsymbol{p})$. Thus, Eq. (6) expresses that diffusion stives after equipartition of the energy over the microstates given the constraint of constant frequency, perhaps an intuitively clear outcome.

Second-order Rayleigh-Schrödinger perturbation theory in $\boldsymbol{q}$ provides the eigenvalue in the bilinear form $D_{ij}q_iq_j$, where D_{ij} can be identified with the diffusion tensor. It turns out to be given by a vector generalization of the Kubo-Greenwood formula, known for electrons in disordered semiconductors[25]. At the same time, the eigenfunction associated with the diffuse mode achieves a perturbational term $\Gamma_{nm}k_n q_m$, which provides the generalization of the term $k_n \partial_n$ in Eq. (5). The actual calculation of the tensors D_{ij} and Γ_{ij} is enabled by the following *controlled* approximations:

1. Interference between E and O waves can be neglected. In multiple scattering this holds true when the dephasing of O and E modes between two successive collisions is large, i.e. $(k_E - k_O)\ell \gg 1$, with k the wave number. Because the scattering mean free path ℓ is so much larger than the wavelength (roughly $100\,\mu$m), this inequality is obeyed even near the NI phase transition. This transition is weakly first order so that a finite order parameter (and thus a finite value for $k_E - k_O$) remains near T_c.

2. The correlation length ξ of the director fluctuations is less than the mean free path. Most NLC's are put in a magnetic field (typically 1 Tesla) to get them aligned and this inequality ($\xi \approx 5\ \mu$m, and $\ell \approx 0.1$ mm) is well satisfied.

Especially the first approximation makes NLC's very attractive as a multiple scattering medium. It implies that the two polarization modes are unaware of each other during propagation and only couple during the scattering from a thermal fluctuation. Moreover, since such scatterings prohibit OO-transitions, any O-wave must change abruptly into an E-wave after each scattering event. The second approximation implies that multiple light scattering in a NLC can actually be envisaged as subsequent single scattering, as is true in milk and fog. When $\xi > \ell$ this familiar picture will be lost and theory becomes more involved[23].

The two approximations simplify the analysis enormously. What remains is a relatively simple expression for the diffusion tensor involving a wave number integral over the E and O constant-frequency surfaces. I would like to point out one final subtlety. In conventional isotropic media it is well known that the transport mean free path ℓ^* (appearing in the diffusion constant) and the scattering mean free path ℓ (determining the exponential decay of the coherent signal) are related by the "Boltzmann" relation $\ell^* = \gamma\ell$, where $\gamma = (1 - \langle\cos\theta\rangle)^{-1}$. The number $\langle\cos\theta\rangle$ denotes the cosine of the scattering angle in one collision event, averaged over directions. It emphasizes that for diffusion, forward scattering ($\theta \approx 0$) is not counted as real scattering. For a NLC this factor is definitely going to be important since - due to the hydrodynamic Goldstone modes - the scattering function is proportional to $1/(\boldsymbol{k}_{\text{in}} - \boldsymbol{k}_{\text{out}})^2$. However, it is not necessarilly a number, but depends on polarization. The final Kubo formula reads,

$$\boldsymbol{q} \cdot \boldsymbol{D} \cdot \boldsymbol{q} = \frac{1}{(2\pi)^3 N(\omega)} \sum_{n=\text{E,O}} \int \frac{\mathrm{d}^2 S^n}{v^n}\, \ell^n(\boldsymbol{k}^n)\,(\boldsymbol{v}^n \cdot \boldsymbol{q})\,\gamma^n\left(\hat{\boldsymbol{k}}^n, \boldsymbol{q}\right) . \quad (7)$$

In this equation, $\mathrm{d}^2 S^n$ denotes the surface element on the frequency surface n and $\boldsymbol{v}^n$ the group velocity of mode n, being the normal vector of the constant-frequency surface S^n; $\ell^n(\boldsymbol{k}^n)$ is the scattering mean free path for the polarization n in the direction $\mathbf{k}^n$[20-22]. The factor $N(\omega)$ stands for the number of microstates per unit volume, and equals the wave number integral of Eq. (6),

$$N(\omega) = \int \frac{\mathrm{d}^3 p}{(2\pi)^3}\, \mathrm{Tr}\, \boldsymbol{S}(\omega, \boldsymbol{p}) = \frac{1}{(2\pi)^3} \sum_{n=\text{E,O}} \int \frac{\mathrm{d}^2 S^n}{v^n} . \quad (8)$$

The bilinear form $\gamma^n(\boldsymbol{k}^n, \boldsymbol{q})$ can be calculated from the phase functions and the polarization selection rules[23]. In isotropic media γ^n cannot depend on polarization so that $\gamma^n(\boldsymbol{k}, \boldsymbol{q}) = \gamma \times (\boldsymbol{k} \cdot \boldsymbol{q})$, where the scalar number γ is given by an expression announced earlier. The uniaxiality and the special selection rules in a NLC lead to,

$$\gamma^n(\boldsymbol{k}, \boldsymbol{q}) = A_n(c)\,(\boldsymbol{k} \cdot \boldsymbol{q}) + \frac{1}{2} B_n(c)\,(\boldsymbol{n}_0 \cdot \boldsymbol{k})\,(\boldsymbol{n}_0 \cdot \boldsymbol{q}) - (\boldsymbol{k} \cdot \boldsymbol{e}^n)\,(\boldsymbol{q} \cdot \boldsymbol{e}^n) . \quad (9)$$

This involves four even functions of $c = \left(\boldsymbol{n}_0 \cdot \hat{\boldsymbol{k}}\right)$, rather than one scalar number γ. Apparently, a considerable amount of information survives after the multiple scattering process.

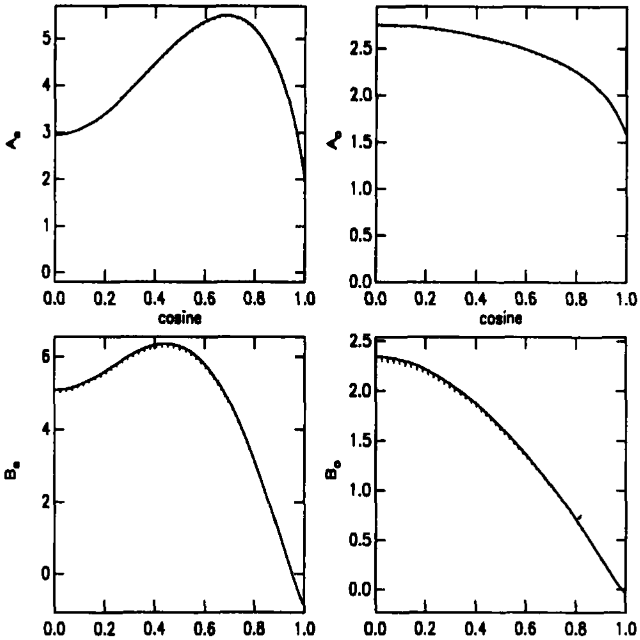

FIGURE 1 Numerical solution for the four functions in equation 9 as a function of (the cosine of) the angle with respect to the optical axis. We choose $\varepsilon_a/\varepsilon_\perp = 0.228$, equal elastic constants for splay and twist distortions but a larger bend elastic constant $K_3/K_2 = 2.3$. The correlation length amounts $\xi = 1.8\,\mu$m. These values correspond roughly to the compound 5CB. The diffusion anisotropy is calculated to be $D_\parallel/D_\perp = 1.51$.

Discussion of Results

In Figure 1 I show the four functions A_E, A_O, B_E and B_O calculated for the liquid crystal 5CB. The calculation relies on a "two-constant approximation" that adopts equal Frank elastic constants for splay (K_1) and twist (K_2) distortion. For $K_3/K_{1,2} = 2.3$ the theoretical prediction is $D_\parallel/D_\perp = 1.51$. For the actual values $K_3/K_2 = 2.3$ and $K_3/K_1 = 1.27$ of 5CB one finds the value $D_\parallel/D_\perp = 1.45$[1,27]. These values are all consistent with the experimental result 1.60 ± 0.25[18]. The enhanced diffusion along the optical axis is caused by both the positive uniaxial symmetry (see below) and the relatively big bend elastic constant. The absolute value of the diffusion constant is expressed as $D_{\parallel,\perp} = d_{\parallel,\perp} \times 8\pi c_0^3 K_1 \sqrt{\varepsilon_\perp}/3kT\varepsilon_a^2\omega^2$

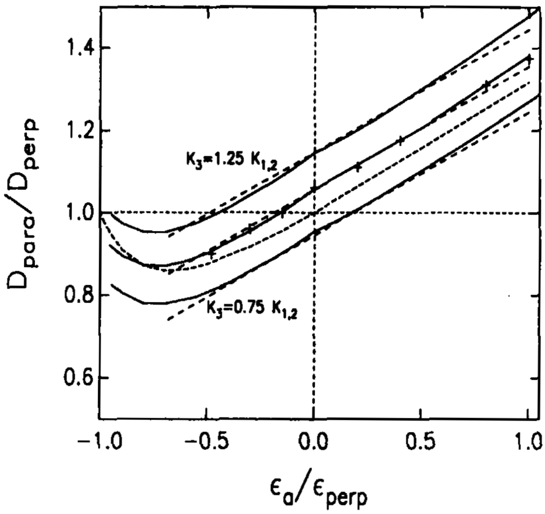

FIGURE 2 Diffusion anisotropy as a function of the uniaxial dielectric anisotropy. The middle solid line corresponds to an isotropic free energy; the other two assume equal strength of play and twist distortion, but a different bend strength. The fine dashed line through the center represents the kinematic anisotropy $(3-2p)/p$ predicted by equation (10). The points are taken from Figure 8 of Stark and Lubensky [1]. The 3 parallel dashed lines denote the Taylor expansion given in Eq. (123) of Ref. [1]. The correlation length is fixed at 5 μm but a change would hardly affect the figure.

where the unit comes from a simple scaling argument[26]. For 5CB the value $d_{\|} = 2.18$ is obtained, leading to $D_{\|} = 1.95 \cdot 10^9$ cm^2/sec. So far, the four functions in Figure 1 only show up inside the angular integration of Eq. (7), but we will see that they determine diffuse angular transmission.

In Figure 2 I show the anisotropy $D_{\|}/D_{\perp}$ of the diffusion constant parallel and perpendicular to the optical axis as a function of the dielectric anisotropy ε_a of the host medium. A convenient property of this ratio is that $\varepsilon_a = \varepsilon_{\|} - \varepsilon_{\perp}$ is the only parameter really sensitive to the order parameter of the nematic phase (the dependency of the elastic constants $K_i \sim S^2$ cancels). As a result this graph gives a good impression of what is predicted for the anisotropy in diffusion when the order parameter changes.

Since most people know that the diffusion constant is a product of velocity and mean free path, an often posed question is which one is the primary responsible. In fact, *three* sources exist for the anisotropy in diffusion: the birefringence of the host medium, the director fluctuations perpendicular to the optical axis and the anisotropic free energy of the NLC. The latter has been discussed in great detail by Stark and Lubensky[27]. The first can be traced by ignoring both the angular dependence of the mean free path in Eq. (7) and the scattering phase functions (i.e. $\gamma^n\left(\hat{\boldsymbol{k}}^n, \boldsymbol{q}\right) \to (\boldsymbol{v}^n \cdot \boldsymbol{q})$ which would correspond to $A_n = 1$ and $B_n = 0$). This would make the diffusion tensor proportional to the *kinematic anisotropy* associated with the birefringence,

$$D_{ij} \to \sum_{n=\mathrm{E,O}} \int \frac{\mathrm{d}^2 S^n}{v^n}\, \hat{v}_i^n \hat{v}_j^n / \sum_{n=\mathrm{E,O}} \int \frac{\mathrm{d}^2 S^n}{v^n}$$
$$= \frac{1}{3}\, p\delta_{ij} + (1-p)\, \hat{n}_i \hat{n}_j \,. \tag{10}$$

The fine dashed line in Figure 2 shows what is obtained in this approximation. One infers that the tendency is well reproduced. Discrepancies are due to the anisotropic director fluctuations and/or nonequal elastic constants. In particular, I obtain the nontrivial value $D_{\|}/D_{\perp} = 1.056$ when $\varepsilon_a \approx 0$ and $K_1 = K_2 = K_3$, a value that is exactly reproduced by Stark and Lubensky[1]. The diffusion is isotropic for $\varepsilon_a/\varepsilon_{\perp} = -0.15$. The 3 parallel dashed lines confirm the good quantitative agreement between Refs. [1] and [2].

The overal tendency in Figure 2 can be understood in the following way. From Eq. (6) it is easy to calculate the total number of photons E or O. Their ratio is a measure of the amount of polarization in the medium. We find,

$$\frac{\rho_E}{\rho_O} = \int \frac{\mathrm{d}^2 S_E}{v^E} / \int \frac{\mathrm{d}^2 S_O}{v^O} = \frac{\varepsilon_{\|}}{\varepsilon_{\perp}} \,. \tag{11}$$

For positive dielectric anisotropies ($\varepsilon_{\|} > \varepsilon_{\perp}$), extraordinary photons dominate. For $\varepsilon_{\|} > \varepsilon_{\perp}$ the E constant-frequency surface is flattened along the optical axis, and consequently we may expect that light scattering is favoured *along* the optical axis, suggesting that $D_{\parallel} > D_{\perp}$. For very negative dielectric anisotropy, O waves dominate and the diffusion anisotropy is supressed. This cheap argument does not acknowledge the inhibition of direct OO scattering, that needs an intermediate - and hence anisotropic - E wave.

Finally I mention that the anisotropy in diffusion is hardly influenced by the correlation length ξ[27]. This can be verified easily in a simplified model that ignores polarization[23]. Only when $\xi \geq \ell$ (corresponding to a magnetic field of only 100 Gauss) significant modifications start to appear since our approximation 2 is violated. One then looses the familiar picture of subsequent single scattering for multiple scattering.

Diffusion Approximation

The eigenfunction corresponding to the eigenvalue with long-range diffusion gives all information on long length scales. The diffusion approximation consists of using this eigenfunction on *all* length scales. In real space, the macroscopic wave number $\boldsymbol{q}$ becomes a space gradient. The specific intensity tensor becomes[23],

$$\Phi_{ij,\mathbf{p}} \sim \sum_{n=E,O} e_i^n e_j^n \delta\left[\omega - \omega_n(\boldsymbol{p})\right] \left\{1 - \ell^n(\boldsymbol{p})\gamma^n(\hat{\boldsymbol{p}}, \boldsymbol{\nabla})\right\} \rho(\boldsymbol{r}) \,. \tag{12}$$

This is the desired generalization of Eq. (5) to a NLC. Upon solving the diffusion equation (4) for a slab of length L, one can find the polarization and current at any point in the medium. In Figure 3 I show a calculation for the transmission coefficient, adopting the parameters known for 5CB. It is confirmed that the emergent radiation deviates from the universal law in isotropic media and is polarized since, at any angle, more E than O photons emerge.

The total transmission coefficient, summed over polarization and angle can easily be obtained from Eq. (12) and the Kubo formula (7). If the z-axis is chosen to be the normal of the slab, the result is,

$$T(L) = \frac{4D_{zz}/v_E}{L + 4D_{zz}/v_E} \,. \tag{13}$$

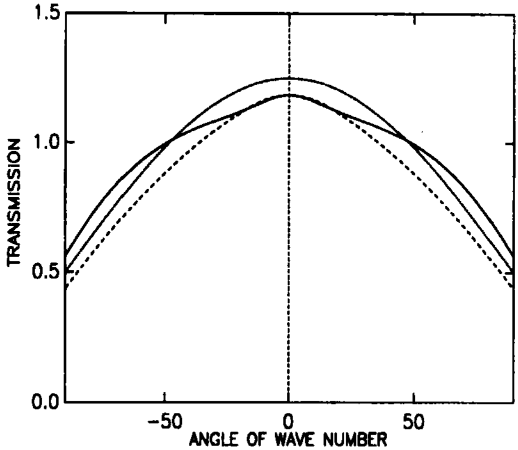

FIGURE 3 Angular- and polarization-dependent transmission profiles $dT^n/d\Omega$ of diffuse light from a slab filled with the compound 5CB (see Figure 1), as predicted by the diffusion approximation. Bold are the transmitted E photons, dashed the O photons. The angle Ω corresponds to the angle of the wave number with respect to the slab normal (and not the one of the group velocity which would be different for different polarizations!). The slab normal is chosen normal to the slab. The dotted line shows the universal law $0.5+0.75\cos\theta$ for isotropic media. No birefringent refraction has been treated at the interface.

For dimensional reasons, D_{zz} must be divided a "transport" velocity v_E, but its choice is *a priori* not clear due to birefringence. The rigorous prediction of Eq. (12) is,

$$v_E(\boldsymbol{n}_0\cdot\hat{\boldsymbol{z}}) = 2\sum_{n=E,O}\int\frac{d^2S_+^n}{v^n}(\boldsymbol{v}^n\cdot\hat{\boldsymbol{z}})/\sum_{n=E,O}\int\frac{d^2S_+^n}{v^n}, \tag{14}$$

i.e. a weighted average of the group velocities over the frequency surfaces $d^2S_+^n$ corresponding to the *transmitted* photons. Since this result depends on the direction of the optical axis with respect to the slab normal one gets the interesting and perhaps unexpected prediction that $T_\perp/T_\parallel \neq D_\perp/D_\parallel$. This is a fingerprint of birefringence in light diffusion. We could call $3D_{zz}/v_E$ *the* transport mean free part ℓ^* of the light, so that Eq. (13) takes the form of Ohm's classical law $T = 4\ell^*/3L$. Another geometry-independent definition is also possible, and uses the final displacement of the center of gravity of a diffuse cloud[28].

PROSPECTS

This contribution dealt with anisotropic diffusion of light in nematic liquid crystals. Other interesting aspects of multiple light scattering in such systems are coherent backscattering and diffuse wave spectroscopy. The first has so far only been investigated numerically[22]. Experimentally, this phenomenon has been measured but its anisotropic features have not yet been revealed[8]. Diffuse wave spectroscopy is the generalization of quasi-elastic light scattering to multiple scattering and recent experimental studies[18] have confirmed theoretical predictions[23,28] using the laws of single scattering. Future studies may use this technique to monitor director fluctuations on time scales beyond hydrodynamic continuum theory. Also, liquid crystals may be useful to study multiple light scattering in the presence of long-range dielectric fluctuations.

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