Multiple light scattering in nematic liquid crystals

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We present a rigorous treatment of the diffusion approximation for multiple light scattering in anisotropic random media, and apply it to director fluctuations in a nematic liquid crystal. For a typical nematic material, 5CB, we give numerical values of the diffusion constants $D_\parallel$ and $D_\perp$. We also calculate the temporal autocorrelation function measured in Diffusing Wave Spectroscopy.

PACS numbers: 61.30.-v, 42.70.Df, 78.20.Ci

Light transport in random or turbid media has long been treated by radiative transfer theories, the first of which was formulated as early as 1905 by Schuster [1]. For distances large compared to the transport mean-free-path $l^*$, beyond which the direction of light propagation is randomized, these theories can be reduced [2] to a diffusion equation for the light energy density with diffusion constant $D = c^2/\beta$ where $c$ is the speed of light in the medium. In 1984 Kuga and Ishimaru [3] discovered coherent backscattering of light in colloidal suspension, predicted in earlier papers [4], and physicists realized the connection of wave propagation in disordered media to the director. They are birefringent with different velocities of light for ordinary and extraordinary rays. As called the director. They are birefringent with different velocities of light for ordinary and extraordinary rays. As shown in Fig. 1. In this paper, we develop a systematic treatment of the diffusion approximation for multiple light scattering in anisotropic random media (for recent approaches see [17]), which allows us to calculate $D_\parallel$ and $D_\perp$ from known properties of nematics and to obtain the time-dependent response measured in DWS experiments [18]. Fig. 1 shows our calculated values of $D_\parallel$ and $D_\perp$ as a function of external magnetic field $H$ for the compound 5CB. The anisotropy ratio $D_\parallel/D_\perp = 1.45$ is in good agreement with measurements reported in a companion paper [19].

Nematic liquid crystals are strong light scatterers, exhibiting turbidity and coherent backscatter [15]. They differ, however, in significant ways from colloidal suspensions, the most widely studied multiple-scattering media. First, nematic liquid crystals are anisotropic with barlike molecules aligned on average along a unit vector $\mathbf{n}(r,t)$ called the director. They are birefringent with different velocities of light for ordinary and extraordinary rays. As a result the photon energy density, like particle density in an electron system [17], obeys an anisotropic diffusion equation with diffusion coefficients $D_\parallel$ and $D_\perp$ for directions parallel and perpendicular to the equilibrium director $\mathbf{n}_0$. Second, the dominant scattering of visible light is from long-range thermal fluctuations of the director rather than from particles with diameters comparable to the wavelength of light. This leads to a divergent scattering mean-free-path when the external magnetic field $H$ is zero [13,24]. The diffusion constants $D_\parallel$ and $D_\perp$, which in isotropic systems are proportional to the transport mean-free-path, are, however, finite when $H \to 0$ as shown in Fig. 1. In this paper, we develop a systematic treatment of the diffusion approximation for multiple light scattering in anisotropic random media [19], which allows us to calculate $D_\parallel$ and $D_\perp$ from known properties of nematics and to obtain the time-dependent response measured in DWS experiments [18].

![Figure 1](https://via.placeholder.com/50)

**FIG. 1.** The field dependence of the normalized diffusion constants $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ and the anisotropy $(D_\parallel - D_\perp)/D_\perp$ for parameters of a typical nematic liquid crystal 5CB: $K_1/K_3 = 0.79, K_2/K_3 = 0.43$ and $\Delta \varepsilon/\varepsilon_\perp = 0.228$.

We start with the wave equation for the electric light field $E(r,t)$:

$$\left\{ \mathbf{\nabla} \times \mathbf{\nabla} \times + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} [\varepsilon_0 + \delta\varepsilon(r,t)] \right\} E(r,t) = 0 \, . \quad (1)$$

The homogeneous part of the dielectric tensor is $\varepsilon_0$, and the randomly fluctuating part $\delta\varepsilon(r,t)$ is a Gaussian random variable described by the correlation function

$$B^{\varepsilon}(\mathbf{R},t) := \frac{\omega^4}{c^3} \langle \delta\varepsilon(\mathbf{R},t) \otimes \delta\varepsilon(0,0) \rangle^{(N)} \, , \quad (2)$$

where $\langle \ldots \rangle^{(N)}$ denotes the expectation value.
where ω is the frequency of light. The superscript \((N)\) means that we interchange the second and third index in the tensor product \(\delta e_\delta \otimes \delta e_\delta\) to define \(B^\omega: [B^\omega]_{ijk\lambda} \propto \langle \delta e_{i\alpha} \delta e_{j\beta} \rangle\). We call \(B^\omega(R, t)\) the structure factor of the system. It is measured in single light scattering experiments \([2]\), and contains informations about the elastic and dynamic properties of a system. The local uniaxial dielectric tensor can be expressed as

\[
e(r, t) = \varepsilon_\perp 1 + \Delta \varepsilon[n(r, t) \otimes n(r, t)]
\]

Here \(\varepsilon_\perp\) and \(\varepsilon_\parallel\) are the dielectric constants for electric fields, respectively, perpendicular and parallel to the director, and \(\Delta \varepsilon = \varepsilon_\parallel - \varepsilon_\perp\). We assume that the inhomogeneity of the director field only comes from thermal fluctuations of the director around its equilibrium value \(n_0: n(r, t) = n_0 + \delta n(r, t)\), where \(\delta n\) has to be perpendicular to \(n_0\) for small fluctuations. The dominant contribution to \(B^\omega(R, t)\) is proportional to the director correlation function \(\langle \delta n(R, t) \otimes \delta n(0, 0) \rangle\) which we express in momentum space \([2]\):

\[
\langle \delta n(q, t) \otimes \delta n^*(q, 0) \rangle = \sum_{\alpha=1}^{2} \frac{k_\beta T}{K_\alpha(q)} \exp \left[ \frac{K_\alpha(q)}{\eta_\alpha(q)} t \right] \hat{u}_\alpha(q) \otimes \hat{u}_\alpha(q).
\]

Here \(K_\alpha(q) = K_1 q_1^2 + K_2 q_2^2 + \Delta \chi H^2\), where \(K_1, K_2,\) and \(K_3\) are the Frank elastic constants, \(H\) is the external field parallel to \(n_0\), and \(\Delta \chi\) is the anisotropy of the magnetic susceptibility. The quantity \(\eta_\alpha(q)\) is a combination of viscosities which appear in the hydrodynamic equations of the director field, the Leslie-Erickson equations \([21]\).

The unit vectors \(\hat{u}_\alpha(q)\) specify the direction of \(\delta n(q, t)\) in mode \(\alpha = 1\) and \(2\).

In an anisotropic medium with a homogeneous dielectric tensor \(\varepsilon_0\), the electromagnetic field travelling along the unit vector \(\hat{k}\) has two modes with indices of refraction \(n_\alpha(\hat{k})\) and electric polarization \(e_\alpha(\hat{k})\). The polarization \(\mathbf{d}^\omega(\hat{k})\) of the displacement field, \(\mathbf{d}^\omega(\hat{k}) = \varepsilon_0 e_\alpha(\hat{k})\), obeys \(\mathbf{d}^\omega(\hat{k}) \cdot \hat{k} = 0\). After an appropriate normalization, the vectors fulfill the biorthogonality relation \(\mathbf{d}^\omega(\hat{k}) \cdot e_\beta(\hat{k}) = \delta_{\alpha\beta}\). We can now write down the momentum space representation of the averaged retarded and advanced Green’s function of Eq. \([1]\) in the weak-scattering approximation:

\[
\langle G^{R/A}(k, \omega) \rangle(k, \omega) \approx \sum_{\alpha=1}^{2} \left( \frac{\omega/c \pm i/2n_\alpha(k)}{2n_\alpha(k)l_\alpha(k, \omega)} \right)^2 e_\alpha(\hat{k}) \otimes e_\alpha(\hat{k})
\]

with

\[
l_\alpha(k, \omega) = \left[ \frac{\pi}{2} n_\alpha(k) \right] \sum_{\beta=1}^{2} \int_{q^3} [B^\omega_{\alpha\beta}(t = 0)]_{\alpha\beta}^{-1}
\]

being the scattering mean-free-path of a light mode \(\{k^\alpha | e_\alpha(\hat{k})\}\) with wave vector \(k^\alpha = \hat{e}_\alpha n_\alpha \hat{k}\) and polarization \(e_\alpha(\hat{k})\). The structure factor \([B^\omega_{k^\alpha q^\beta}(t = 0)]_{\alpha\beta}\) describes the scattering of \(\{k^\alpha | e_\alpha(\hat{k})\}\) into \(\{q^\beta | e_\beta(\hat{k})\}\). The symbol \(\int_{q^3} \cdots \int_{q^3} \) always stands for an angular integration:

\[
\int_{q^3} \cdots \int_{q^3} = \int_{\Omega} \frac{d\Omega}{(2\pi)^3} n_\parallel^3(q) \cdots
\]

To the order of our calculations \(\langle G^{R/A}(k, \omega) \rangle(k, \omega)\) is diagonal in the polarization \(e_\alpha(\hat{k})\). In what follows, a Greek index will refer to the “basis vectors” \(e_\alpha \otimes e_\alpha\) or \(\mathbf{d}^\alpha \otimes \mathbf{d}^\alpha\). The scattering mean-free-path \(l_\alpha(k, \omega)\) in the nematic phase has been calculated \([23, 24]\). For small \(H\) for the extraordinary ray it has the form \(l_\alpha^{-1} \sim (\hat{q})^2 \frac{\gamma H}{\omega K_{\alpha}(\hat{k})}\), where \(K\) is an appropriately averaged elastic constant.

Let us look at the spatial and temporal autocorrelation function for the electric light field: \(\langle E(R + \hat{\omega}, T + \frac{1}{2}) \otimes E^*(R - \frac{1}{2}, T - \frac{1}{2}) \rangle\), where we have already introduced center of “mass” \((R, T)\) and relative \((r, t)\) coordinates. From this quantity, others follow as special cases: the energy density of light at time \(T\) is \(W_1(R, T) = \langle E(R, T) \cdot e_\alpha E^*(R, T) \rangle\), where the \(T\) dependence is, e.g., due to time dependent-sources; the temporal correlation function of a steady-state light field is \(W_2(R, t) = \langle E(R, \frac{3}{2}) \cdot e_\alpha E^*(R, -\frac{1}{2}) \rangle\), which reflects the dynamics of the scattering media measured in DWS experiments. The Fourier transform with respect to \(r\) gives the energy density with wave vector \(k\) \([2]\). To calculate the autocorrelation function for special light sources and/or given boundary conditions, we need the “two-particle” Green’s function \(\Phi = \langle G^{R} \otimes G^{A}\rangle(N)\). Our goal is to derive the diffusion pole of \(\Phi\) in momentum and frequency space. With all arguments, the Green’s function is \(\Phi_{kk}(K, \Omega, t)\), \(K, \Omega\) correspond to the center of “mass” coordinates \(R, T\) and \(k, k’\) to the relative coordinates \(r, r’\). The superscript \(\omega\) is the light frequency, and the \(t\) dependence explicitly comes from the structure factor \(B_{kk}(t)\). In the weak-scattering approximating, \(\Phi_{kk}(K, \Omega, t)\) can be represented as a sum of ladder diagrams, which is equivalent to the Bethe-Salpeter equation:

\[
\int \frac{d^3k_1}{(2\pi)^3} [1_{kk, k_1}^{(4)} - f_k^{(4)}(K, \Omega) B_{kk_1}(t)] \Phi_{kk_1}(K, \Omega, t) = f_k^{(4)}(K, \Omega) 1_{kk}^{(4)},
\]

where

\[
f_k^{(4)}(K, \Omega) = \langle \langle G^{R}(k_+, \omega+) \otimes \langle G^{A}(k_-, \omega-) \rangle \rangle(K, \omega, \omega)\rangle^{(N)}
\]

with \(k_\pm = k \pm K/2\) and \(\omega_\pm = \omega \pm \Omega/2\) and, \(1_{kk}^{(4)}\) \(\delta_{ij} \delta_{\alpha \beta} + \delta_{i\beta} \delta_{\alpha j}\). The multiple integrals and the sum can be done analytically for \(\delta\)-function correlations but not for the anisotropic, long-range correlations of our problem. Vollhardt and Wölfle \([23]\) derived
the diffusion pole for isotropic electron transport directly from Eq. (10) and MacKintosh and John [11] applied their method to light. In the anisotropic case one has to be more careful [1]. If \( \Psi_k^{n}(K, \Omega, t) \) and \( \lambda^{n}(K, \Omega, t) \) are, respectively, the \( n \)th eigenvector and eigenvalue of the integral operator of Eq. (10):

\[
\int \frac{d^{3}k}{(2\pi)^{3}} [1_{Kk}^{(4)}] f_k^{\omega}(K, \Omega) B_{kK}^{\omega}(t) \Psi_k^{n} = \lambda^{n}(K, \Omega) \Psi_k^{n},
\]

(10)

and \( \Psi_k^{(n)}(K, \Omega, t) \) the eigenvectors of the Hermitian adjoint operator, it is straightforward to show that

\[
\Psi_k^{\omega}(K, \Omega, t) = \sum_{n} \Psi_k^{n} \otimes \Psi_k^{n} \lambda^{n}(K, \Omega) f_k^{\omega}(K, \Omega)
\]

(11)

solves the Bethe-Salpeter equation [1]. In the case of \( K = 0 \) and \( \Omega = t = 0 \), it can be shown that the quantity \( \Delta G_k^{\omega}(0, 0) \), where

\[
\Delta G_k^{\omega}(K, \Omega) = \langle \mathbf{G}^{(0)}(k_{+}, \omega_{+}) - \mathbf{G}^{(0)}(k_{-}, \omega_{-}) \rangle,
\]

(12)

is an eigenvector with eigenvalue \( \lambda^{(0)}(0, 0, 0) = 0 \). This is a very general result, based on the Ward identities valid beyond the weak-scattering approximation [2]. We have identified the diffusion pole, as we shall explicitly see soon. All other eigenvalues are positive, and, in real space, they give exponentially decaying contributions to \( \Phi_{\omega k}(K, \Omega, t) \) of Eq. (11), which are not important at long length scales [1]. To establish the diffusion approximation we have to apply perturbation theory to calculate \( \lambda^{(0)}(K, \omega, t) \) for small \( K \), \( \Omega \), and \( t \). Therefore, we expand the eigenvectors into a set of basis functions and turn the eigenvalue equation (10) into a matrix equation. For the component \( \alpha \) of \( \Psi_k \) we use the ansatz

\[
\Psi_k^{\omega} \propto [\Delta G_k^{\omega}(0, 0)]^{\alpha} \bar{\Psi}_0 \pi + \sum_{i} \bar{\Psi}_i^{\omega} \varphi_i^{\omega}(\hat{k})
\]

(13)

where

\[
[\Delta G_k^{\omega}(0, 0)]^{\alpha} \approx -i\pi c^{-1} n_{\alpha}(\hat{k}) \delta\left(\frac{\omega}{c} n_{\alpha}(\hat{k}) - k\right).
\]

(14)

The first factor on the right-hand side of Eq. (13) is due to the momentum shell approximation, Eq. (14); it is strongly peaked around the wave numbers of the light modes. The amplitude \( \bar{\Psi}_0 \pi \) then represents the zeroth eigenvector. The second term corresponds to the space of all other eigenvectors where the \( \varphi_i^{\omega}(\hat{k}) \) are basis functions on the unit sphere, e.g. spherical harmonics, which can in general depend on polarization \( \alpha \). We also use the relation

\[
[\Delta G_k^{\omega}(K, \Omega)]^{\alpha} \approx [f_k^{(0)}(0, 0)]^{\alpha} \times \left[ \Delta \Sigma_k^{\omega}(0, 0) - \frac{\partial G_0^{-1}}{\partial K} - \frac{\partial G_0^{-1}}{\partial \omega} \right]_{\alpha},
\]

(15)

which gives \( [\Delta G_k^{\omega}(K, \Omega)]^{\alpha} \) correctly to first order in \( K \), \( \Omega \) and \( \Sigma \) (see [20]). \( \Sigma \) is the mass operator and \( \Delta \Sigma_k^{\omega} \) is defined the same way as \( \Delta G_k^{\omega} \). \( G_0 \) stands for the Green’s function of the homogeneous medium. The coupling between the zeroth and the other eigenvectors then produces the diffusion tensor, and, finally, the Green’s function takes the form:

\[
\Phi_{\omega k}(K, \Omega, t) \approx \frac{1}{N} \frac{\Delta G_k^{\omega}(0, 0) \otimes \Delta G_k^{\omega}(0, 0)}{-i\omega + \mu(\omega, t) + K \cdot D(\omega) K}
\]

(16)

with \( N = \frac{2\pi\omega^2}{(\pi c^3)} \) and \( \pi \) being the angular and arithmetic average of the two refractive indices. The denominator represents a diffusion pole, which also contains an “absorption” coefficient \( \mu(\omega, t) \). The diffusion tensor follows from

\[
K \cdot D(\omega) K = \frac{c}{2n^3} [\mathcal{G}(K)]^* \cdot B^{-1} \mathcal{G}(K)
\]

(17)

with

\[
[\mathcal{G}(K)]_{\alpha i} = \pi \int_{k^0} n_{\alpha}(\hat{k}) \left[ \frac{\partial G_0^{-1}}{\partial K} \right]_{\alpha i} \varphi_i^{\omega}(\hat{k})^*\varphi_i^{\omega}(\hat{k})
\]

and

\[
[B]^{\alpha i}_{\beta j} = \sum_{\gamma} \pi \int_{k^0} \int_{q^0} ([\varphi_i^{\omega}(\hat{k})^*\varphi_j^{\omega}(\hat{q})]_{\beta \gamma} - [\varphi_i^{\omega}(\hat{k})^*\varphi_j^{\omega}(\hat{q})]_{\beta \gamma}) [B_{k^0}^{\omega q}(0)]_{\alpha \gamma}.
\]

In principal all \( \varphi_i^{\omega}(\hat{k}) \) of odd parity contribute to \( D(\omega) \). For isotropic systems we choose spherical harmonics:

\[
\varphi_i^{\omega}(\hat{k}) \rightarrow Y_{lm}(\hat{\theta}, \varphi).
\]

Only the components \( [\mathcal{G}(K)]_{m = 1} \) are nonzero and \( [B]^{m i}_{lm} \propto \delta_{l1} \). Therefore, only spherical harmonics of \( l = 1 \) contribute to \( D(\omega) \) and we get the familiar formula

\[
D = \frac{1}{c} \varpi^2 \propto [(1 - \cos \theta)^{-1}]^{-1}
\]

The absorption coefficient reads

\[
\mu(\omega, t) = \frac{c^{2} n^3}{2n^3} \sum_{\alpha \beta} \int_{k^0} \int_{q^0} [B_{k^0 q^0}^{\omega}(0) - B_{k^0 q^0}^{\omega}(t)]_{\alpha \beta}.
\]

It represents an angular average over all the dynamical modes of the system. For \( t = 0 \), it is zero and then increases due to the decaying temporal correlations in \( \langle \delta \varphi(\hat{k}) \rangle \) and the concomitant decrease in \( \langle \delta \varphi(\hat{k}) \rangle \). The numerator in Eq. (14) indicates which initial and final polarization states have a nonzero overlap with the diffusion pole. The second factor \( \Delta G_k^{\omega}(0, 0) \) depends only on the input wave. The first factor \( \Delta G_k^{\omega}(0, 0) \) involves only the output wave and determines the ratio of densities of photons in the two output polarization states 1 and 2 independent of the state of the input wave. An integration over \( k \) (\( j k^2 dk \)) shows that this ratio is \( [n_1^{\omega}(\hat{k})/n_2^{\omega}(\hat{k})]^3 \) for the wave direction \( \hat{k} \). This effect should be measurable. Finally, the Green’s function corresponding to \( W_{m}(R, t) \) follows from \( \Phi_{\omega k}(K, \Omega, t = 0) \) by integrating over \( k \) and applying the appropriate trace operation.
The diffusion tensor $\mathbf{D}(\omega)$ has the same uniaxial form as the dielectric tensor in Eq. (3). We express the diffusion coefficients $D_{||}$ and $D_{\perp}$ in terms of a typical length $l_0^\ast = 9\pi c^2 / k_BT \Delta \varepsilon / \Delta \varepsilon_{||} (c_\| = c / \sqrt{\varepsilon_{||}})$ times unitless numerical factors $\tilde{D}_{||}$ and $\tilde{D}_{\perp}$ via

$$D_{||} = c_\| l_0^\ast \tilde{D}_{||} / 3 \ , \ D_{\perp} = c_\perp l_0^\ast \tilde{D}_{\perp} / 3 \ ,$$

(18)

where $\tilde{D}_{||}$ and $\tilde{D}_{\perp}$ depend on $K_1/K_3$, $K_2/K_3$ and $\Delta \varepsilon / \varepsilon_{||}$. For the material 5CB, $K_3 = 5.3 \times 10^{-7}$ dyne, $\varepsilon_{||} = 2.381$ and $\Delta \varepsilon / \varepsilon_{||} = 0.228$. With $T = 300$ K and green light ($\omega / c = 1.15 \times 10^9$ cm$^{-1}$) we get $l_0^\ast = 2.3$ mm, which is in agreement with experiments [2,3]. As basis functions we choose spherical harmonics depending on a new “polar angle” $\vartheta'$ (see [20]). For the ordinary light ray $\vartheta' = \vartheta$, for the extraordinary one $\vartheta'$ is given by $\cos \vartheta' = n_1(\hat{k}) \cos \vartheta / \sqrt{\varepsilon_{||}}$. The basis functions $\varphi_{\alpha}^{m}(\hat{k}) = Y_{\alpha m}^{\prime}(\vartheta', \varphi)$ are orthogonal with respect to the weight $n_1^3(\hat{k}) d\vartheta' d\varphi$. Then, only $[\mathcal{G}(K)]_{\alpha l=\lambda m}$ is nonzero [20]. We studied the contributions of different $l$ to $D_{||}$ and $D_{\perp}$ and found that $l = 3$ in addition to $l = 1$ gives changes of less than 2%. In Fig. 1 we plot our results for $D_{||}$ and $D_{\perp}$ for 5CB with $K_1/K_3 = 0.79$, $K_2/K_3 = 0.43$ and $\Delta \chi = 0.95 \times 10^{-7}$. At $H = 0$, $D_{||}$ = 0.95 and $D_{\perp}$ = 0.65 are finite even though, as noted earlier, the scattering mean-free-path for the extraordinary light ray is infinite. The anisotropy in the diffusion constants decreases with both $\Delta \varepsilon$ and anisotropy in the Frank elastic constants. In the limit $\Delta \varepsilon = 0$ and $K_1 = K_2 = K_3$, $D_{||}/D_{\perp}$ = 1.06 is not unity because of the inherent anisotropy in the structure factor. The diffusion approximation is valid only for times $t$ much smaller than characteristic relaxation times of the director modes. In this case we get

$$\mu(\omega, t) \approx t \mu_0 \ , \ \mu_0 = \frac{2 k_BT}{9\pi} \frac{\omega^4 \Delta \varepsilon_{\perp}^2}{c_\perp^2} \tilde{\mu},$$

(19)

where $\gamma$ is the rotational viscosity and $\tilde{\mu}$ a numerical factor depending on all other viscosities and $\Delta \varepsilon / \varepsilon_{||}$. Note that unlike scattering in colloids, $\mu_0$ depends only on viscosities and is independent of the static structure factor ($\propto k_BT/kq^2$). This is because the same fluctuations determine scattering and dynamics. Finally, we point out that the appropriate Laplace-Fourier transform of Eq. (16) leads to a temporal autocorrelation function $W_2$ that can be expressed in a form reminiscent of the average over light paths used in isotropic systems [12,13]:

$$W_2 \sim \int_0^\infty d\tau P(\tau) \exp(-\mu_0 t \tau) \ , $$

(20)

where $P(\tau)$ is the probability that an anisotropic random walker enters the medium at a prescribed point and leaves it at another point after a time $\tau$. (Note that this integral is over time $\tau$ rather than path length because the light velocity is not a constant along an arbitrary path.)

This work was supported in part by the Deutsche Forschungsgemeinschaft under Grant No. Sta 352/2-1 and by NSF under Grant No. DMR 91-20688. We wish to thank Ming Kao, Kristen Jester and Arjun Yodh for helpful discussions.

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