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. [S0031-9007(96)01115-5]

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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 $D = \bar{c}l^*/3$ where $\bar{c}$ 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 weak localization [5], a precursor of Anderson localization [6]. Since then, our theoretical understanding of light transport in random media has advanced considerably. Detailed studies of multiple scattering of scalar waves [7] was followed by the generalization to include the polarization of light [8], broken time reversal symmetry and optical activity [4,9], and long-range correlations in random scatterers [10,11]. Multiple scattering emerged as a powerful probe of dynamical properties of turbid media with the development of diffusing wave spectroscopy (DWS) [12,13] as an experimental technique capable of measuring dynamic correlations at time scales much shorter [14] than can be probed with single scattering.

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 $\boldsymbol{n}(\boldsymbol{r}, t)$ 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 [16], obeys an anisotropic diffusion equation with diffusion coefficients $D_\parallel$ and $D_\perp$ for directions parallel and perpendicular to the equilibrium director $\boldsymbol{n}_0$. 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 [17,18]. 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 Letter, we develop a systematic treatment of the diffusion approximation for multiple light scattering in anisotropic random media (for recent approaches see [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 [20]. Figure 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 Letter [21].

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

$$\left[\nabla \times \nabla \times + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}[\boldsymbol{\varepsilon}_0 + \delta\boldsymbol{\varepsilon}(\boldsymbol{r}, t)]\right]\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{0}. \quad (1)$$

The homogeneous part of the dielectric tensor is $\boldsymbol{\varepsilon}_0$, and the randomly fluctuating part $\delta\boldsymbol{\varepsilon}(\boldsymbol{r}, t)$ is a Gaussian random variable described by the correlation function

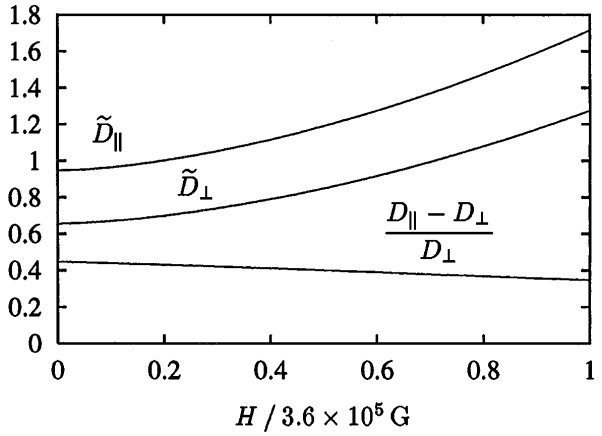

FIG. 1. The field dependence of the normalized diffusion constants $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ and the anisotropy $(D_\parallel - D_\perp)/D_\perp$ for parameters of a typical nematic liquid crystal 5CB: $K_1/K_3 = 0.79$, $K_2/K_3 = 0.43$, and $\Delta\varepsilon/\varepsilon = 0.228$.

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

where ω is the frequency of light. The superscript (N) means that we interchange the second and third index in the tensor product $\delta\boldsymbol{\varepsilon} \otimes \delta\boldsymbol{\varepsilon}$ to define $\boldsymbol{B}^{\omega}$: $[\boldsymbol{B}^{\omega}]_{ijkl} \propto \langle \delta\varepsilon_{ik}\delta\varepsilon_{jl}\rangle$. We call $\boldsymbol{B}^{\omega}(\boldsymbol{R},t)$ the structure factor of the system. It is measured in single light scattering experiments [22] and contains information about the elastic and dynamic properties of a system. The local uniaxial dielectric tensor can be expressed as

$$\boldsymbol{\varepsilon}(\boldsymbol{r},t) = \varepsilon_{\perp}\mathbf{1} + \Delta\varepsilon[\boldsymbol{n}(\boldsymbol{r},t) \otimes \boldsymbol{n}(\boldsymbol{r},t)]. \quad (3)$$

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 comes only from thermal fluctuations of the director around its equilibrium value $\boldsymbol{n}_0$: $\boldsymbol{n}(\boldsymbol{r},t) = \boldsymbol{n}_0 + \delta\boldsymbol{n}(\boldsymbol{r},t)$, where $\delta\boldsymbol{n}$ has to be perpendicular to $\boldsymbol{n}_0$ for small fluctuations. The dominant contribution to $\boldsymbol{B}^{\omega}(\boldsymbol{R},t)$ is proportional to the director correlation function $\langle \delta\boldsymbol{n}(\boldsymbol{R},t) \otimes \delta\boldsymbol{n}(\boldsymbol{0},0)\rangle$ which we express in momentum space [23]:

$$\langle \delta\boldsymbol{n}(\boldsymbol{q},t) \otimes \delta\boldsymbol{n}^{*}(\boldsymbol{q},0)\rangle = \sum_{\alpha=1}^{2} \frac{k_B T}{K_{\alpha}(\boldsymbol{q})} \exp\left[-\frac{K_{\alpha}(\boldsymbol{q})}{\eta_{\alpha}(\boldsymbol{q})}\, t\right] \hat{\boldsymbol{u}}_{\alpha}(\boldsymbol{q}) \otimes \hat{\boldsymbol{u}}_{\alpha}(\boldsymbol{q}). \quad (4)$$

Here $K_{\alpha}(\boldsymbol{q}) = K_{\alpha} q_{\perp}^2 + K_3 q_{\parallel}^2 + \Delta\chi H^2$, where K_1, K_2, and K_3 are the Frank elastic constants, H is the external field parallel to $\boldsymbol{n}_0$, and $\Delta\chi$ is the anisotropy of the magnetic susceptibility. The quantity $\eta_{\alpha}(\boldsymbol{q})$ is a combination of viscosities which appear in the hydrodynamic equations of the director field, the Leslie-Erickson equations [23]. The unit vectors $\hat{\boldsymbol{u}}_{\alpha}(\boldsymbol{q})$ specify the direction of $\delta\boldsymbol{n}(\boldsymbol{q},t)$ in mode $\alpha = 1$ and 2.

In an anisotropic medium with a homogeneous dielectric tensor $\boldsymbol{\varepsilon}_0$ the electromagnetic field traveling along the unit vector $\hat{\boldsymbol{k}}$ has two modes with indices of refraction $n_{\alpha}(\hat{\boldsymbol{k}})$ and electric polarizations $\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})$. The polarization $\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}})$ of the displacement field, $\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}}) = \boldsymbol{\varepsilon}_0 \boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})$, obeys $\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}}) \cdot \hat{\boldsymbol{k}} = 0$. After an appropriate normalization, the vectors fulfill the biorthogonality relation $\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}}) \cdot \boldsymbol{e}_{\beta}(\hat{\boldsymbol{k}}) = \delta_{\beta}^{\alpha}$ [24]. 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:

$$\langle G^{R/A}\rangle(\boldsymbol{k},\omega) \approx \sum_{\alpha=1}^{2} \{[\omega/c \pm i/2n_{\alpha}(\hat{\boldsymbol{k}})l_{\alpha}(\hat{\boldsymbol{k}},\omega)]^2 - k^2/n_{\alpha}^2(\hat{\boldsymbol{k}})\}^{-1} \boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}}), \quad (5)$$

with

$$l_{\alpha}(\hat{\boldsymbol{k}},\omega) = \left[\frac{\pi}{2} n_{\alpha}(\hat{\boldsymbol{k}}) \sum_{\beta=1}^{2} \int_{\hat{\boldsymbol{q}}^{\beta}} [\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t=0)]_{\alpha\beta}\right]^{-1} \quad (6)$$

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

$$\int_{\hat{\boldsymbol{q}}^{\beta}} \cdots = \int \frac{d\Omega_q}{(2\pi)^3}\, n_{\beta}^3(\hat{\boldsymbol{q}}) \cdots . \quad (7)$$

To the order of our calculations $\langle G^{R/A}\rangle(\boldsymbol{k},\omega)$ is diagonal in the polarization $\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})$. In what follows, a Greek index will refer to the "basis vectors" $\boldsymbol{e}_{\alpha} \otimes \boldsymbol{e}_{\alpha}$ or $\boldsymbol{d}^{\alpha} \otimes \boldsymbol{d}^{\alpha}$. The scattering mean free path $l_{\alpha}(\hat{\boldsymbol{k}},\omega)$ in the nematic phase has been calculated [17,18]. For small H for the extraordinary ray it has the form $l_{\alpha}^{-1} \propto (\frac{\omega}{c})^2 \frac{k_B T}{K} \ln(\frac{\Delta\chi H^2}{K(\omega/c)^2})$, where K is an appropriately averaged elastic constant.

Let us look at the spatial and temporal autocorrelation function for the electric light field: $\langle \boldsymbol{E}(\boldsymbol{R} + \frac{\boldsymbol{r}}{2}, T + \frac{t}{2}) \otimes \boldsymbol{E}^{*}(\boldsymbol{R} - \frac{\boldsymbol{r}}{2}, T - \frac{t}{2})\rangle$, where we have already introduced the center of "mass" $(\boldsymbol{R},T)$ and relative $(\boldsymbol{r},t)$ coordinates. From this quantity, others follow as special cases: the energy density of light at time T is $W_1(\boldsymbol{R},T) = \langle \boldsymbol{E}(\boldsymbol{R},T) \cdot \boldsymbol{\varepsilon}_0 \boldsymbol{E}^{*}(\boldsymbol{R},T)\rangle$, where the T dependence is, e.g., due to time-dependent sources; the temporal correlation function of a steady-state light field is $W_2(\boldsymbol{R},t) = \langle \boldsymbol{E}(\boldsymbol{R},\frac{t}{2}) \cdot \boldsymbol{\varepsilon}_0 \boldsymbol{E}^{*}(\boldsymbol{R},-\frac{t}{2})\rangle$, which reflects the dynamics of the scattering media measured in DWS experiments. The Fourier transform with respect to $\boldsymbol{r}$ gives the energy density with wave vector $\boldsymbol{k}$ [2]. To calculate the autocorrelation function for special light sources and/or given boundary conditions, we need the "two particle" Green's function $\boldsymbol{\Phi} = \langle \boldsymbol{G}^{R} \otimes \boldsymbol{G}^{A}\rangle^{(N)}$. Our goal is to derive the diffusion pole of $\boldsymbol{\Phi}$ in momentum and frequency space. With all arguments, Green's function is $\boldsymbol{\Phi}^{\omega}_{\boldsymbol{k}\boldsymbol{k}'}(\boldsymbol{K},\Omega,t)$. $\boldsymbol{K},\Omega$ correspond to the center of mass coordinates $\boldsymbol{R},T$ and $\boldsymbol{k},\boldsymbol{k}'$ to the relative coordinates $\boldsymbol{r},\boldsymbol{r}'$. The superscript ω is the light frequency, and the t dependence explicitly comes from the structure factor $\boldsymbol{B}^{\omega}_{\boldsymbol{k}\boldsymbol{k}'}(t)$. In the weak-scattering approximation, $\boldsymbol{\Phi}^{\omega}_{\boldsymbol{k}\boldsymbol{k}'}(\boldsymbol{K},\Omega,t)$ can be represented as a sum of ladder diagrams, which is equivalent to the Bethe-Salpeter equation:

$$\int \frac{d^3k_1}{(2\pi)^3} [\mathbf{1}^{(4)}_{\boldsymbol{k}\boldsymbol{k}_1} - \boldsymbol{f}^{\omega}_{\boldsymbol{k}}(\boldsymbol{K},\Omega)\boldsymbol{B}^{\omega}_{\boldsymbol{k}\boldsymbol{k}_1}(t)]\boldsymbol{\Phi}^{\omega}_{\boldsymbol{k}_1\boldsymbol{k}'}(\boldsymbol{K},\Omega,t) = \boldsymbol{f}^{\omega}_{\boldsymbol{k}}(\boldsymbol{K},\Omega)\mathbf{1}^{(4)}_{\boldsymbol{k}\boldsymbol{k}'}, \quad (8)$$

where

$$\boldsymbol{f}^{\omega}_{\boldsymbol{k}}(\boldsymbol{K},\Omega) = [\langle \boldsymbol{G}^{R}\rangle(\boldsymbol{k}_{+},\omega_{+}) \otimes \langle \boldsymbol{G}^{A}\rangle(\boldsymbol{k}_{-},\omega_{-})]^{(N)}, \quad (9)$$

with $\boldsymbol{k}_{\pm} = \boldsymbol{k} \pm \boldsymbol{K}/2$, $\omega_{\pm} = \omega \pm \Omega/2$, and $[\mathbf{1}^{(4)}_{\boldsymbol{k}\boldsymbol{k}'}]_{ijkl} := (2\pi)^3\delta(\boldsymbol{k} - \boldsymbol{k}')(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})/2$. The multiple integrals and the sum can be done analytically for δ-function correlations but not for the anisotropic, long-range correlations of our problem. Volhardt and Wölfle [25] derived the diffusion pole for isotropic electron transport directly from Eq. (8), and MacKintosh and John [11] applied their

method to light. In the anisotropic case one has to be more careful [16]. If $\mathbf{\Psi}_{\boldsymbol{k}}^{(n)}(\boldsymbol{K},\Omega,t)$ and $\lambda^{(n)}(\boldsymbol{K},\Omega,t)$ are, respectively, the nth eigenvector and eigenvalue of the integral operator of Eq. (8),

$$\int \frac{d^3k_1}{(2\pi)^3}[\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}_1}^{(4)} - \boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega}(t)]\mathbf{\Psi}_{\boldsymbol{k}_1}^{(n)} = \lambda^{(n)}\mathbf{\Psi}_{\boldsymbol{k}}^{(n)}, \tag{10}$$

with $\overline{\mathbf{\Psi}}_{\boldsymbol{k}}^{(n)}(\boldsymbol{K},\Omega,t)$ the eigenvectors of the Hermitian adjoint operator, it is straightforward to show that

$$\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega,t) = \sum_n \frac{\mathbf{\Psi}_{\boldsymbol{k}}^{(n)} \otimes \overline{\mathbf{\Psi}}_{\boldsymbol{k}'}^{(n)}}{\lambda^{(n)}} \boldsymbol{f}_{\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega) \tag{11}$$

solves the Bethe-Salpeter equation [11]. In the case of $\boldsymbol{K} = \mathbf{0}$ and $\Omega = t = 0$, it can be shown that the quantity $\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)$, where

$$\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega) = \langle\boldsymbol{G}^{R}\rangle(\boldsymbol{k}_+,\omega_+) - \langle\boldsymbol{G}^{A}\rangle(\boldsymbol{k}_-,\omega_-), \tag{12}$$

is an eigenvector with eigenvalue $\lambda^{(0)}(\mathbf{0},0,0) = 0$. This is a very general result, based on the Ward identities valid beyond the weak-scattering approximation [25]. 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 $\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega,t)$ of Eq. (11), which are not important at long length scales [11]. To establish the diffusion approximation we have to apply perturbation theory to calculate $\lambda^{(0)}(\boldsymbol{K},\omega,t)$ for small $\boldsymbol{K}$, Ω, 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 α of $\mathbf{\Psi}_{\boldsymbol{k}}$ we use the ansatz

$$\mathbf{\Psi}_{\boldsymbol{k}}^{\alpha} \propto [\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha}\left[\tilde{\mathbf{\Psi}}_0\pi + \sum_i \tilde{\mathbf{\Psi}}_i^{\alpha}\varphi_i^{\alpha}(\hat{\boldsymbol{k}})\right], \tag{13}$$

where

$$[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha} \approx -i\pi\frac{c}{\omega}n_{\alpha}(\hat{\boldsymbol{k}})\delta\left(\frac{\omega}{c}n_{\alpha}(\hat{\boldsymbol{k}}) - k\right). \tag{14}$$

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 $\tilde{\mathbf{\Psi}}_0$ then represents the zeroth eigenvector. The second term corresponds to the space of all other eigenvectors where the $\varphi_i^{\alpha}(\hat{\boldsymbol{k}})$ are basis functions on the unit sphere, e.g., spherical harmonics, which can in general depend on polarization α. We also use the relation

$$[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)]^{\alpha} \approx [\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha\alpha} \times\left[\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0) - \frac{\partial\boldsymbol{G}_0^{-1}}{\partial\boldsymbol{k}}\boldsymbol{K} - \frac{\partial\boldsymbol{G}_0^{-1}}{\partial\boldsymbol{\omega}}\Omega\right]_{\alpha}, \tag{15}$$

which gives $[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)]^{\alpha}$ correctly to first order in $\boldsymbol{K}$, Ω, and $\mathbf{\Sigma}$ (see [26]). ($\mathbf{\Sigma}$ is the mass operator and $\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}$ is defined the same way as $\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}$. $\boldsymbol{G}_0$ stands for Green's function of the homogeneous medium.) The coupling between the zeroth and the other eigenvectors then produces the diffusion tensor, and, finally, Green's function takes the form

$$\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega,t) \approx \frac{1}{N}\frac{\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0) \otimes \Delta\boldsymbol{G}_{\boldsymbol{k}'}^{\omega}(\mathbf{0},0)}{-i\Omega + \mu(\omega,t) + \boldsymbol{K}\cdot\boldsymbol{D}(\omega)\boldsymbol{K}}, \tag{16}$$

with $N = -2\overline{n^3}\omega^2/(\pi c^3)$ and $\overline{n^3}$ 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

$$\boldsymbol{K}\cdot\boldsymbol{D}(\omega)\boldsymbol{K} = \frac{c}{2\overline{n^3}}[\mathcal{G}(\boldsymbol{K})]^{*}\cdot\mathcal{B}^{-1}\mathcal{G}(\boldsymbol{K}), \tag{17}$$

with

$$[\mathcal{G}(\boldsymbol{K})]_{\alpha i} = \pi\int_{\hat{\boldsymbol{k}}^{\alpha}} n_{\alpha}(\hat{\boldsymbol{k}})\left[\frac{\partial\boldsymbol{G}_0^{-1}}{\partial\boldsymbol{k}}\right]_{\alpha}(\hat{\boldsymbol{k}})[\varphi_i^{\alpha}(\hat{\boldsymbol{k}})]^{*}$$

and

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

In principle all $\varphi_i^{\alpha}(\hat{\boldsymbol{k}})$ of odd parity contribute to $\boldsymbol{D}(\omega)$. For isotropic systems we choose spherical harmonics: $\varphi_i^{\alpha}(\hat{\boldsymbol{k}}) \rightarrow Y_{lm}(\vartheta,\varphi)$. Only the components $[\mathcal{G}(\boldsymbol{K})]_{\alpha l=1 m}$ are nonzero and $[\mathcal{B}]_{\beta l' m'}^{\alpha l m} \propto \delta_{ll'}$. Therefore, only spherical harmonics of $l = 1$ contribute to $\boldsymbol{D}(\omega)$ and we get the familiar formula $D = \frac{1}{3}cl^{*} \propto [\langle 1 - \cos\vartheta\rangle]^{-1}$. The absorption coefficient reads

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

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 $\langle\delta\boldsymbol{\varepsilon}\otimes\delta\boldsymbol{\varepsilon}\rangle$.) The numerator in Eq. (16) indicates which initial and final polarization states have a nonzero overlap with the diffusion pole. The second factor $\Delta\boldsymbol{G}_{\boldsymbol{k}'}^{\omega}(\mathbf{0},0)$ depends only on the input wave. The first factor $\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)$ 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 ($\int k^2\,dk$) shows that this ratio if $[n_1(\hat{\boldsymbol{k}})/n_2(\hat{\boldsymbol{k}})]^3$ for the wave direction $\hat{\boldsymbol{k}}$. This effect should be measurable. Finally, Green's function corresponding to $W_2(\boldsymbol{R},t)$ follows from $\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega = 0,t)$ by integrating over $\boldsymbol{k},\boldsymbol{k}'$ and applying the appropriate trace operation.

The diffusion tensor $\boldsymbol{D}(\omega)$ has the same uniaxial form as the dielectric tensor in Eq. (3). We express the diffusion coefficients $D_{\parallel}$ and $D_{\perp}$ in terms of a typical

length $l_0^* = 9\pi \frac{c_\perp^2}{\omega^2} \frac{K_3}{k_B T} \frac{\varepsilon_\perp^2}{\Delta\varepsilon^2} (c_\perp = c/\sqrt{\varepsilon_\perp})$ times unitless numerical factors $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ via

$$D_\parallel = c_\perp l_0^* \tilde{D}_\parallel / 3, \qquad D_\perp = c_\perp l_0^* \tilde{D}_\perp / 3, \tag{18}$$

where $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ depend on K_1/K_3, K_2/K_3, and $\Delta\varepsilon/\varepsilon_\perp$. For the material 5CB, $K_3 = 5.3 \times 10^{-7}$ dyne, $\varepsilon_\perp = 2.381$, and $\Delta\varepsilon/\varepsilon_\perp = 0.228$. With $T = 300$ K and green light ($\omega/c = 1.15 \times 10^5$ cm^{-1}) we get $l_0^* = 2.3$ mm, which is in agreement with experiments [15,21]. As basis functions we choose spherical harmonics depending on a new "polar angle" ϑ'_α (see [26]). For the ordinary light ray $\vartheta'_2 = \vartheta$, for the extraordinary one ϑ'_α is given by $\cos\vartheta'_1 = n_1(\hat{\boldsymbol{k}}) \cos\vartheta/\sqrt{\varepsilon_\perp}$. The basis functions $\varphi^\alpha_{lm}(\hat{\boldsymbol{k}}) = Y_{lm}(\vartheta'_\alpha(\vartheta), \varphi)$ are orthogonal with respect to the weight $n_\alpha^3(\hat{\boldsymbol{k}}) d\Omega_k \propto d\cos\vartheta'_\alpha \, d\varphi$. Then, only $[\mathcal{G}(\boldsymbol{K})]_{\alpha l=1 m}$ is nonzero [26]. We studied the contributions of different l to $D_\parallel$ 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 $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ for 5CB with $K_1/K_3 = 0.79$, $K_2/K_3 = 0.43$, and $\Delta\chi = 0.95 \times 10^{-7}$. At $H = 0$, $\tilde{D}_\parallel = 0.95$ and $\tilde{D}_\perp = 0.65$ 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 $K_1 = K_2 = K_3$, $D_\parallel/D_\perp = 1.06$ 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

$$\mu(\omega, t) \approx t\mu_0, \qquad \mu_0 = \frac{2k_B T}{9\pi} \frac{\omega^4}{c_\perp^3} \frac{\Delta\varepsilon^2}{\varepsilon_\perp^2} \frac{\tilde{\mu}}{\gamma}, \tag{19}$$

where γ is the rotational viscosity and $\tilde{\mu}$ a numerical factor depending on all other viscosities and $\Delta\varepsilon/\varepsilon_\perp$. Note that unlike scattering in colloids, μ_0 depends only on viscosities and is independent of the static structure factor ($\propto k_B T/Kq^2$). 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]:

$$W_2 \propto \int_0^\infty d\tau P(\tau) \exp(-\mu_0 t\tau), \tag{20}$$

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 τ. (Note that this integral is over time τ rather than path length because the light velocity is not a constant along an arbitrary path.)

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Note added— While this paper was being processed for publication, a very similar paper [Bart A. van Tiggelen, Roger Maynard, and Anne Heiderich, Phys. Rev. Lett. **77**, 639 (1996)] was published. Its results are in good agreement with ours.

[1] A. Schuster, Astrophys. J. **21**, 1 (1905).
[2] A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic Press, New York, 1978), Vol. 1/2.
[3] Y. Kuga and A. Ishimaru, J. Opt. Soc. Am. A **1**, 831 (1984).
[4] See A. A. Golubentsev, Zh. Eksp. Teor. Fiz. **86**, 47 (1984) [Sov. Phys. JETP **86**, 26 (1984)].
[5] P.-E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985); M. P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).
[6] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).
[7] *Scattering and Localization of Classical Waves in Random Media,* edited by P. Sheng (World Scientific, Singapore, 1990).
[8] M. J. Stephen and G. Cwilich, Phys. Rev. B **34**, 7564 (1986); E. Akkermans, P. E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49**, 77 (1988).
[9] F. C. MacKintosh and S. John, Phys. Rev. B **37**, 1884 (1988).
[10] S. John and M. J. Stephen, Phys. Rev. B **28**, 6358 (1983).
[11] F. C. MacKintosh and S. John, Phys. Rev. B **40**, 2383 (1989).
[12] G. Maret and P. E. Wolf, Z. Phys. B **65**, 409 (1987).
[13] D. J. Pine, D. A. Weitz, P. M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[14] M. H. Kao, A. G. Yodh, and D. J. Pine, Phys. Rev. Lett. **70**, 242 (1993).
[15] H. K. M. Vithana, L. Asfaw, and D. L. Johnson, Phys. Rev. Lett. **70**, 3561 (1993); D. V. Vlasov, L. A. Zubkov, N. V. Orekhova, and V. P. Romanov, Pis'ma Zh. Eksp.Teor. Fiz. **48**, 86 (1988) [JETP Lett. **48**, 91 (1988)].
[16] P. Wölfle and R. N. Blatt, Phys. Rev. B **30**, 3542 (1984).
[17] A. Y. Val'kov and V. P. Romanov, Zh. Eksp. Teor. Viz. **90**, 1264 (1986) [Sov. Phys. JETP **63**, 737 (1986)].
[18] D. Langevin, and M.-A. Bouchiat, J. Phys. (Paris), Colloq. **36**, C1-197 (1975).
[19] B. A. van Tiggelen, Phys. Rev. Lett. **75**, 422 (1995); S. Ramaswany, J. Phys. Chem. **98**, 9318 (1994).
[20] M. J. Stephen, Phys. Rev. B **37**, 1 (1988).
[21] M. Kao, K. Jester, A. Yodh, and P. Collings, following Letter, Phys. Rev. Lett. **77**, 2233 (1996).
[22] B. J. Berne and R. Pecora, *Dynamic Light Scattering—With Applications to Chemistry, Biology, and Physics* (John Wiley & Sons, Inc., New York, 1976).
[23] P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1975).
[24] M. Lax and D. F. Nelson, Phys. Rev. B **4**, 3694 (1971).
[25] D. Vollhardt and P. Wölfle, Phys. Rev. B **22**, 4666 (1980).
[26] T. Lubensky and H. Stark (to be published).