Multiple light scattering in anisotropic random media

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In the last decade Diffusing Wave Spectroscopy (DWS) has emerged as a powerful tool to study turbid media. In this article we develop the formalism to describe light diffusion in general anisotropic turbid media. We give explicit formulas to calculate the diffusion tensor and the dynamic absorption coefficient, measured in DWS experiments. We apply our theory to uniaxial systems, namely nematic liquid crystals, where light is scattered from thermal fluctuations of the local optical axis, called director. We perform a detailed analysis of the two essential diffusion constants, parallel and perpendicular to the director, in terms of Frank elastic constants, dielectric anisotropy, and applied magnetic field. We also point out the relevance of our results to different liquid crystalline systems, such as discotic nematics, smectic- $A$ phases, and polymer liquid crystals. Finally, we show that the dynamic absorption coefficient is the angular average over the inverse viscosity, which governs the dynamics of director fluctuations.

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I. INTRODUCTION

Dynamic light scattering (DLS) is one of our most powerful probes of the dynamics of materials like simple liquids, complex fluids, and liquid crystals. In typical experiments, light incident on the sample scatters once, and its intensity is measured at a detector. Motion in the sample, or more generally fluctuations in the local dielectric constant, induce changes in the phase of scattered light, which give rise to temporal fluctuations of the light intensity measured at the detector. Such experiments probe length scales of order the inverse scattering wave vector $q^{-1}$ and hence time scales of order $(vq)^{-1}$ where $v$ is a typical velocity.

There are many materials such as colloids, emulsions, foams, and some liquid crystals that scatter light so strongly that the traditional single scattering analysis of DLS does not apply. In these materials, light undergoes many scattering events before leaving the sample, and the transport of light energy is diffusive rather than ballistic. The study of light transport in random or turbid media dates back to radiative transfer theory, first introduced as early as 1905 by Schuster. These systems are characterized by a scattering mean-free-path $l$, measuring the average distance a photon travels before scattering, and a transport mean-free-path $l^* = l/(1 - \cos \theta_s)$, measuring the distance beyond which the direction of propagation is randomized, where $\theta_s$ is the scattering angle and the angular bracket denotes an average over direction weighted by the differential scattering cross section. For distances greater than $l^*$, light energy transport is described by a diffusion equation with scalar diffusion constant $D = \pi l^*/3$, where $\pi$ is the speed of light in the medium.

There has been a resurgence of interest in light transport in turbid and random media because of its close connection to the problem of Anderson localization and electron transport in disordered systems and electron transport in disordered systems and be-

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cause of the development of Diffusing Wave Spectroscopy (DWS), which permits useful information to be extracted from dynamic correlations of multiply scattered light. Coherent backscattering, a manifestation of weak as opposed to strong or true Anderson localization, has been observed in a number of experiments and discussed in a number of theoretical papers and a number of theoretical papers. DWS has opened up a whole new field of study. It provides heretofore unobtainable information about the the dynamics of turbid media, including dense colloids, sheared suspensions, emulsions, and foams. Because intensity variations measured at the detector arise from phase shifts distributed over many scattering events, DWS detects dynamic phenomena at much shorter time scales than normal DLS. This has permitted the measurement of hydrodynamic interaction contributions to the diffusion constant of colloidal particles and the measurement of shape fluctuation modes in tense emulsion droplets. Photon diffusion and DWS have also found application in imaging of objects such as tumors in human tissue.

With few exceptions, both theory and experiment have focussed on diffusive transport and DWS in isotropic systems. There are, however, many turbid materials such as conventional thermotropic and lyotropic liquid crystals, liquid crystalline colloids and emulsions, and also muscle tissues that are anisotropic. This paper will develop a general treatment of diffusive light transport and DWS in anisotropic media with particular applications to nematic liquid crystals. Though its inspiration is recent experimental work on coherent backscatter, its purpose is to broaden the class of materials to which DWS and its offshoot applications like imaging can be applied. A preliminary account of this work and experiments on multiple scattering in liquid crystals to which it applies were reported in references. An alternative derivation of the results...
studied [28]. Tiggelen has investigated anisotropic light 
rection relative to anisotropy axes. Diffusive transport 
cross sections depend not only on the relative direction 
eral on both the polarization and direction of light prop-
important ways: (1) The speed of light depends in gen-
land, and Heiderich [43].

Anisotropic media differ from isotropic media in two 
from an average of short-time dynamic correlations 
served in DWS experiments [9,10,11,12,13,14,15]. It re-
along the unit vector 
reduces to zero, scattering takes place almost 
mechanism is lost, and, as a result, $l^*$ is finite.

The outline of this paper is as follows. In Sec. II, 
review light propagation including the one-particle 
Green function in homogeneous anisotropic media. In 
Sec. III, we treat diffusive transport of light in general 
anisotropic random media. We introduce the structure 
factor $B_0(r,t)$ to describe fluctuations in the dielec-
tric tensor, discuss electric-field autocorrelation functions 
and their meaning, and relate them to the averaged two-
particle Green function. We, then, discuss the one- and 
two-particle Green functions in the weak-scattering limit 
and derive the diffusion equation for light transport from 
the Bethe-Salpeter equation. We introduce the approxi-
mation scheme for the diffusion tensor and look at the 
isotropic limit of our theory. Comments on DWS close 

II. LIGHT PROPAGATION IN A 
HOMOGENEOUS MEDIUM WITH DIELECTRIC 
ANISOTROPY

Nematic liquid crystals present a difficulty that is 
not generic to anisotropic systems. Light scattering is 
from fluctuations in the direction of the principal axis 
of the dielectric tensor, which is parallel to the local 
Frank director $n(r)$. Fluctuations in $n(r)$ diverge as 
$q^{-2}$ at small wave number $q$ in the absence of an exter-
more complicated than it is in isotropic systems. In par-
cular, the electric field is not always transverse, and 
the speed of light depends on polarization and direction 
of propagation. In this section we review light propaga-
in anisotropic media. Following the work of Nelson
and Lax [46], we will introduce sets of polarization vectors for the electric and dielectric field that will prove to be very useful for our forthcoming considerations. We start with Maxwell’s equations

\[
\begin{align*}
\text{div} D &= 4\pi \varrho_{\text{ma}} \quad \text{div} B = 0 \\
\text{curl} E &= -\frac{1}{c} \frac{\partial B}{\partial t} \quad \text{curl} H = \frac{4\pi}{c} j_{\text{ma}} + \frac{1}{c} \frac{\partial D}{\partial t},
\end{align*}
\]

where \(\varrho_{\text{ma}}\) and \(j_{\text{ma}}\) are, respectively, the macroscopic charge and current densities. We concentrate on a dielectric medium with

\[
D = \varepsilon_0 E \quad \text{and} \quad B = H,
\]

and we assume that the dielectric tensor \(\varepsilon_0\) is real and does not depend on time. Then the energy-balance equation reads:

\[
\frac{\partial}{\partial t} u + \text{div} S = j_{\text{ma}} \cdot E,
\]

where we have introduced the energy density

\[
u = \frac{1}{8\pi} (E \cdot \varepsilon_0 E + H \cdot H)
\]

and the Poynting vector

\[
S = \frac{c}{4\pi} E \times H.
\]

Both quantities strongly vary in space and time. If we average over one period of oscillation, we obtain their averaged values \(\bar{u}\) and \(\bar{S}\). In the following we use complex waves \(\vec{E} = E_0 e^{-i\omega t}\) and \(\vec{H} = H_0 e^{-i\omega t}\) whose averaged energy densities and Poynting vectors are given by \(\bar{u} = \langle E_0 \cdot \varepsilon_0 E_0 + H_0 \cdot H_0 \rangle / (16\pi)\) and \(\bar{S} = c \langle E_0 \times H_0 \rangle / (8\pi)\).

For vanishing sources \(\varrho_{\text{ma}}\) and \(j_{\text{ma}}\), we obtain the homogeneous wave equation for the electric light field \(E(r,t)\):

\[
[\text{curl} \text{curl} + \frac{\varepsilon_0}{c^2} \frac{\partial^2}{\partial t^2}] E(r,t) = 0.
\]

Note that all solutions of Eq. (2.6) have to fulfill the transversality condition for electric field waves, \(\text{div}\varepsilon_0 E = 0\), unless the electric field is static or linear in \(t\). Introduction of the plane-wave ansatz

\[
E(r,t) = E_0 e^{i(k \cdot r - \omega t)}
\]

in wave equation (2.6) leads to a generalized eigenvalue problem

\[
[P_1(\hat{k}) - \frac{1}{n^2} \varepsilon_0] e(\hat{k}) = 0,
\]

where

\[
\frac{1}{n^2} = \frac{\omega^2}{c^2 k^2}.
\]

and

\[
P_1(\hat{k}) = 1 - \hat{k} \otimes \hat{k}
\]

is the projection operator on the space perpendicular to the unit vector \(\hat{k} = k/k\). The symbol \(\otimes\) stands for the tensor product: \([\hat{k} \otimes \hat{k}]_{ij} = \delta_{ij}\). The solutions to Eq. (2.8) provide us with the characteristic light modes of the system determined by the refractive index \(n_1(\hat{k}) = ck/\omega\) and the polarization vector \(e_i(\hat{k})\) of the electric field. We define the polarization vector of the displacement field by

\[
d^i(\hat{k}) = \varepsilon_0 e_i(\hat{k}).
\]

For each direction \(\hat{k}\), there exist two characteristic light modes with associated polarizations \(d^1(\hat{k}) \perp \hat{k}\). When a plane wave with frequency \(\omega\) enters the anisotropic medium, it splits up into the two characteristic modes which travel with different speeds \(c/n_1(\hat{k})\) and wave numbers \(k = \omega n_1/c\). We also find a third solution with \(n_3 = \infty\) and \(e_3(\hat{k})\parallel \hat{k}\), corresponding to a nonpropagating mode with \(\omega = 0\). It violates \(\hat{k} \cdot d^1 = 0\) but is necessary to construct the complete Green function for wave equation (2.6) (see below).

The two sets of polarization vectors fulfill the biorthogonality condition

\[
d^i(\hat{k}) \cdot e_j(\hat{k}) = \delta^i_j \quad (i,j = 1,2,3),
\]

i.e., they are dual to each other like the basis vectors of the real and reciprocal lattice in a crystal. We will use both of them as convenient bases for our tensor quantities throughout the article. To prove condition (2.12) we notice that \(P_1(\hat{k})\) and \(\varepsilon_0\) are symmetric tensors and derive from Eq. (2.8) the condition

\[
\left(\frac{1}{n_1^2} - \frac{1}{n_3^2}\right) e_i \cdot d^j = 0,
\]

from which (2.12) follows after an appropriate normalization. The vectors \(d^1\) and \(d^2\) are perpendicular to \(\hat{k}\). Then, again with Eq. (2.3) and the biorthogonality condition (2.12), one can show that \(d^1 \perp d^2\). In Fig. 1, we summarize the geometry for a given propagation direction \(\hat{k}\). Finally, we recall that in general the refractive indices are calculated from Fresnel’s equation [47]:

\[
\sum_{i=1}^{3} \tilde{\varepsilon}_i \bar{k}_i^2 = 0,
\]

where \(\tilde{\varepsilon}_i\) stands for the principal dielectric constants, the eigenvalues of \(\varepsilon_0\), and \(\bar{k}_i\) is the component of the unit vector \(\hat{k}\) along the \(i\)th eigenvector of \(\varepsilon_0\). This equation is equivalent to \(\det[P_1(\hat{k}) - \varepsilon_0/n^2] = 0\).

Consider now \(E_0 = E_0 e_i(\hat{k})\) and \(H_0\), the amplitude of the magnetic field wave, which are connected via Maxwell’s relations:
\[ k \times E_0 = \frac{\omega}{c} H_0 \quad \text{and} \quad k \times H_0 = -\frac{\omega}{c} \varepsilon_0 E_0 . \]  

The second equation also follows from the first once Eq. (2.8) is solved. For the nonpropagating mode (\( \omega = 0 \)), \( H_0 \), like \( E_0 \), is parallel to \( k \). For the propagating modes, \( H_0 \) is perpendicular to \( \hat{k} \) and \( \varepsilon_0 E_0 \). Using Eqs. (2.8) and (2.15) one shows that electric and magnetic fields carry the same amount of field energy \( \langle H_0 \cdot H_0^* = E_0 \cdot \varepsilon_0 E_0^* \rangle \) and that the averaged energy density \( \bar{\varepsilon} \) of a light wave is

\[ \bar{\varepsilon} = \frac{1}{8\pi} E_0 \cdot \varepsilon_0 E_0^* = \frac{1}{8\pi} |E_0|^2 . \]  

Here the meaning of the scalar \( E_0 \) becomes clear. It is not the magnitude of the electric field, because \( e_i(\hat{k}) \) is not a unit vector, instead it basically stands for the square root of the energy density of the light mode. The poynting vector \( S \) does not generally point along \( \hat{k} \). Its projection on \( \hat{k} \) however fulfills the relation

\[ \hat{k} \cdot S = \frac{c}{n} \bar{\varepsilon} , \]  

which is familiar for isotropic systems.

Finally, we calculate the Green function \( G_0(r-r', t-t') \) for the wave equation (2.6), which has a source term proportional to the time derivative of \( J_{ma} \). In Fourier space we have

\[ [k^2 P_{ij}(\hat{k}) - \frac{\omega^2}{c^2} \varepsilon_0] G_0(k, \omega) = -1 . \]  

We expand \( G_0(k, \omega) \) in the basis \( \{ e_i(\hat{k}) \otimes e_j(\hat{k}) \} \) and calculate its components by multiplying the last equation, respectively, from left and right with \( e_i(\hat{k}) \) and \( d^i(\hat{k}) \). Using the eigenvalue equation (2.8) and the algebra for the polarization vectors, described above, we find that \( G_0(k, \omega) \) is diagonal:

\[ G_0(k, \omega) = \sum_{\alpha=1}^{2} \left[ \frac{\omega^2}{c^2} - \frac{k^2}{n^2_\alpha(k)} \right]^{-1} e_\alpha(\hat{k}) \otimes e_\alpha(\hat{k}) + \frac{c^2}{\omega^2} e_3(\hat{k}) \otimes e_3(\hat{k}) . \]  

The first term on the right-hand side represents the propagating part of the Green function. We will always, throughout this article, indicate it by Greek indices. It acts on the transverse part of the source term. Nelson and Lax [48] have calculated an expression for its far field in coordinate space, which reduces to a spherical wave in an isotropic system. The second term takes into account the longitudinal part of the source. In coordinate space, it is just a non-propagating delta function. As we go on, it will become clear that we can neglect it within our approximation outlined in the next section. We will therefore skip it when we represent tensors through their components. We will also see that for all the tensors involved we can neglect the non-diagonal parts. Thus, whenever we use a Greek superscript or subscript \( \alpha \) it refers, respectively, to the basis “vectors” \( e_\alpha(\hat{k}) \otimes e_\alpha(\hat{k}) \) or \( d^\alpha(\hat{k}) \otimes d^\alpha(\hat{k}) \) \( (\alpha = 1, 2) \). The last quantity we need is the inverse Green function \( [G_0(k, \omega)]^{-1} \), which we conveniently represent as

\[ [G_0(k, \omega)]^{-1} = \sum_{\alpha=1}^{2} \left[ \frac{\omega^2}{c^2} - \frac{k^2}{n^2_\alpha(k)} \right] d^\alpha(\hat{k}) \otimes d^\alpha(\hat{k}) \]

by realizing that the unit tensor can be written as

\[ 1 = \sum_{ij} e_i(\hat{k}) \otimes d^j(\hat{k}) = \sum_{ij} d^i(\hat{k}) \otimes e_j(\hat{k}) . \]  

### III. DIFFUSIVE LIGHT TRANSPORT IN ANISOTROPIC RANDOM MEDIA

We now consider a dielectric medium in which the dielectric tensor possesses a randomly fluctuating part \( \delta \varepsilon(\mathbf{r}, t) \) in addition to the homogeneous term \( \varepsilon_0 \). We treat \( \delta \varepsilon(\mathbf{r}, t) \) as a Gaussian random variable with variance characterized by

\[ B^{\omega}(\mathbf{r}, t) := \frac{\omega^4}{c^4} \left< \delta \varepsilon(\mathbf{r}, t) \otimes \delta \varepsilon(\mathbf{0}, 0) \right>^{(N)} , \]  

where \( \omega \) is the frequency of light. The superscript \( (N) \) means that we interchange the second and third index in the tensor product \( \delta \varepsilon \otimes \delta \varepsilon \) to define \( B^{\omega} \). [\( B^{\omega} \)]_{ijkl} \propto
\( \delta \varepsilon_{ik} \delta \varepsilon_{j\ell} \). We call \( B^\omega(r, t) \) the structure factor of the system. It is measured in single light scattering experiments \( \mathbb{P} \) and contains information about the elastic and dynamic properties of a system.

The electric light field \( \mathbf{E}(r, t) \) obeys the homogeneous wave equation

\[
\text{curl curl} + (\varepsilon_0 + \delta \varepsilon(r, t)) \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}(r, t) = 0 \ , \quad (3.2)
\]

where we have used an adiabatic approximation to pull \( \delta \varepsilon(r, t) \) in front of the time derivatives. It is valid if \( \delta \varepsilon(r, t) \) varies on time scales much longer than the time period of light and the passage time of a light ray through an inhomogeneous medium.

Our task is to calculate measurable quantities from Eq. \( 3.3 \). A very general one is given by the spatial and temporal autocorrelation function for the electric light field, \( \langle \mathbf{E}(r_1, t_1) \otimes \mathbf{E}(r_2, t_2) \rangle \), which we write with the help of center-of-“mass” \((R, T)\) and relative \((r, t)\) coordinates:

\[
W(R, r, T, t) = \langle \mathbf{E}(R + \frac{r}{2}, T + \frac{t}{2}) \otimes \mathbf{E}^*(R - \frac{r}{2}, T - \frac{t}{2}) \rangle . \quad (3.3)
\]

It is a second rank tensor. For \( r = 0 \) we derive the scalar

\[
W(R, T, t) = \langle \mathbf{E}(R, T + \frac{t}{2}) : \varepsilon_0 \mathbf{E}^*(R, T - \frac{t}{2}) \rangle ,
\]

(3.4)

which at \( t = 0 \) is equal to the energy density of the light field when the small fluctuating part \( \delta \varepsilon \) is neglected. The \( T \) dependence is, e.g., due to time-modulated light sources, and \( R \) describes variations of the energy density on long length scales. For \( t \neq 0 \), \( W(R, r, T, t) \) reflects the dynamics of the scattering medium through its dependence on the structure factor \( B^\omega(r, t) \). It is measured in dynamic light scattering experiments either in single scattering \( \mathbb{P} \) or with DWS \( \mathbb{P} \). We will see below that the Fourier transform with respect to \( r \), \( W(R, k, T, t) \), is diagonal in the basis \( \{ e_\alpha(k) \otimes e_\beta(k) \} \) on length \((R)\) and time \((T)\) scales much longer than the wavelength and time period of light,

\[
W(R, k, T, t) \approx \sum_\alpha W^\alpha(R, k, T, t) e_\alpha(k) \otimes e_\alpha(k) .
\]

(3.5)

For \( t = 0 \) we can, therefore, interpret \( W^\alpha(R, k, T, t) \) as the energy density of a light wave with wave vector \( k \) and polarization \( e_\alpha(k) \). To clarify this interpretation we look at the Fourier transform

\[
\int d^3R d^3r dt W(R, r, T, t) e^{i(-K \cdot R - k \cdot r + \Omega T + \omega t)}
\]

\[
= \langle \mathbf{E}(k + \frac{K}{2}, \omega + \frac{\Omega}{2}) \otimes \mathbf{E}^*(k - \frac{K}{2}, \omega - \frac{\Omega}{2}) \rangle . \quad (3.6)
\]

Since \( R \) and \( T \) describe variations on long scales we have \( K \ll k \) and \( \Omega \ll \omega \). Thus the amplitudes \( E(k \pm K/2, \omega \pm \Omega/2) \) are strongly peaked around \( \omega \) and \( k \) which can be identified, respectively, with the frequency and wave vector of the light waves in the medium.

To calculate the autocorrelation function \( W(R, r, T, t) \) for special light sources \( W_0(R, r, T, t) \) and/or given boundary conditions, we need the averaged “two-particle” Green function

\[
\Phi = \langle \mathbf{G}^R \otimes \mathbf{G}^A \rangle^{(N)} , \quad (3.7)
\]

where \( \mathbf{G}^R \) and \( \mathbf{G}^A \) denote the retarded and advanced Green functions with \( \mathbf{G}^A = [{\mathbf{G}^R}]^* \). Then

\[
W(1) = \int d2 \Phi(1, 2) W_0(2) \quad (3.8)
\]

for (notation see Appendix A). In the second subsection we will derive the diffusion pole of \( \Phi \), i.e., that part of \( \Phi \) which corresponds to a diffusion equation for \( W(R, k, T, t) \) in the variables \( R \) and \( T \). It governs the propagation of light energy at long length and time scales. The time \( t \) will appear in an absorption term of the diffusion equation, which is zero for \( t = 0 \). In the first subsection we calculate the averaged retarded and advanced one-particle Green functions, which we need for the derivation of \( \Phi \).

A. The averaged one-particle Green function

The one-particle Green functions \( \langle \mathbf{G}^R \rangle \) and \( \langle \mathbf{G}^A \rangle \) follow from Dyson’s equation \( \mathbb{P} \), which we give in a formal notation,

\[
\langle \mathbf{G}^R / \mathbf{G}^A \rangle = G_0 + G_0 \Sigma^R / \mathbf{G}^A \langle \mathbf{G}^R / \mathbf{G}^A \rangle . \quad (3.9)
\]

We have introduced the retarded and advanced second-rank tensor mass operators \( \Sigma^R \) and \( \Sigma^A \). In the weak-scattering approximation, where the elements of the structure factor \( B^\omega(r, t = 0) \) are assumed to be much smaller than one, these mass operators are proportional to \( B^\omega(r, t = 0) \) and can be written in frequency space as

\[
\Sigma^R / \mathbf{G}^A (r, \omega) = B^\omega(r, t = 0) / \mathbf{G}^R / \mathbf{G}^A (r, \omega) . \quad (3.10)
\]

Figure \( \mathbb{P} \) gives a diagrammatic representation of \( \Sigma^R / \mathbf{G}^A (r, \omega) \). Finally, in momentum space, we get from Eq. \( 3.9 \).
\[ \langle G^{R/A}(k, \omega) \rangle = [G_0^{-1}(k, \omega) - \Sigma^{R/A}(k, \omega)]^{-1} \]  \hspace{1cm} (3.11)

The inverse Green function \( G_0^{-1}(k, \omega) \) is diagonal in the basis \( \{d^a(k) \otimes d^a(k)\} \), and \( \Sigma^{R/A}(k, \omega) \) causes a small perturbation. From perturbation theory we know that to zeroth order in \( \Sigma^{R/A} \) the eigenvectors of \( G_0^{-1}(k, \omega) - \Sigma^{R/A}(k, \omega) \) are unchanged, whereas to first order in

\[ \sum R/A(\mathbf{r}, \omega) = \frac{\omega^2}{c^2} \delta(\mathbf{r}, 0) \frac{\omega^2}{c^2} \delta(\mathbf{r}, 0) \]

\( \langle G^{R/A}(\mathbf{r}, \omega) \rangle \)

**FIG. 2.** The mass operators \( \Sigma^{R/A}(r, \omega) \) in the weak-scattering approximation consist of two scattering events from inhomogeneities in the dielectric medium, which are tied together by spatial correlations \( \text{tering approximation consist of two scattering events from} \) \( \omega \)

We have introduced the scattering vector \( \alpha \) by the wave vector \( \hat{k} \) and polarization \( e_\alpha(\hat{k}) \) of the light modes. The significant effect of \( \Sigma^{R/A} \) comes from its imaginary part. Then, in its final form the propagating part of the Green functions reads

\[ \langle G^{R/A}(k, \omega) \rangle = \sum_{\alpha = 1}^{2} \langle [G^{R/A}(k, \omega)]^\alpha e_\alpha(\hat{k}) \otimes e_\alpha(\hat{k}) \rangle \]

\( \hspace{1cm} (3.13) \)

with

\[ \langle [G^{R/A}(k, \omega)]^\alpha \rangle = \left[ \frac{\omega^2}{c^2} - \frac{k^2}{n_\alpha^2(\hat{k})} + \frac{i\omega}{cn_\alpha(k)l_\alpha(k, \omega)} \right]^{-1}. \]

\( \hspace{1cm} (3.14) \)

We have introduced the scattering mean-free-path \( l_\alpha(\hat{k}, \omega) \) of the light mode \( \{k^\alpha | e_\alpha(\hat{k})\} \) which travels with a wave vector \( k^\alpha = \hat{k} n_\alpha \) and polarization \( e_\alpha(\hat{k}) \):

\[ l_\alpha(\hat{k}, \omega) = -\frac{e}{\omega} n_\alpha(\hat{k}) \text{Im} \Sigma^{R}(\frac{\hat{n}_\alpha}{c} n_\alpha(k, \omega)) l_\alpha(\hat{k}, \omega) \]

\( \hspace{1cm} (3.15) \)

To arrive at \( l_\alpha(\hat{k}, \omega) \), we used a CPA approximation replacing the argument \( k \) of \( \Sigma^{R/A}(k, \omega) \) by the wave vector \( \hat{n}_\alpha \) of the light modes where \( G_0 \) diverges. \( \text{Im} \Sigma^{R} \) follows from Eq. (3.10) using the momentum shell approximation

\[ \text{Im}(G^{R/A}(k, \omega))^\alpha \approx -\frac{\pi e}{2 \omega} n_\alpha(\hat{k}) \delta(\hat{n}_\alpha n_\alpha(\hat{k}) - k) \]

\( \hspace{1cm} (3.16) \)

valid in the limit \( \Sigma^R \to 0 \). We finally obtain

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

\( \hspace{1cm} (3.17) \)

where we sum over all possible scattering events of the incoming light mode \( \{k^\alpha | e_\alpha(\hat{k})\} \) into modes \( \{q^\beta | e_\beta(\hat{k})\} \). The single scattering event is described by the structure factor

\[ |B^{\omega}_{k^\beta q^\beta}(t = 0)|_{\alpha\beta} = \frac{\omega^4}{c^6} \langle |e_\alpha(\hat{k}) \cdot \delta(\mathbf{q}, 0) e_\beta(\hat{q})|^2 \rangle, \]

\( \hspace{1cm} (3.18) \)

which is proportional to the differential scattering cross section. We also introduced the scattering vector

\[ \mathbf{q}_s = \omega c (n_\alpha \hat{k} - n_\beta \hat{q}). \]

\( \hspace{1cm} (3.19) \)

In Eq. (3.17) and throughout the article, we use a shorthand notation for angular integration:

\[ \int \frac{d\Omega}{(2\pi)^3} = \int \frac{d\Omega_1}{(2\pi)^3} \]

\( \hspace{1cm} (3.20) \)

We point out that the scattering mean-free-path \( l_\alpha(\hat{k}, \omega) \) now depends on the direction \( \hat{k} \) and the polarization \( e_\alpha(\hat{k}) \) of the light mode. Following the work of Nelson and Lax [48], it is straightforward to show that \( l_\alpha(\hat{k}, \omega) \) determines an exponential decay of the far field of \( \langle G^{R}(\mathbf{r}, \omega) \rangle \) travelling in the direction \( \hat{k} \) with polarization \( e_\alpha(\hat{k}) \). In our analysis, we have neglected the off-diagonal components of \( \Sigma^{R/A} \), so that the polarization vectors of our light rays are identical to those of the homogeneous medium. We will use this approximation in the next subsection. We conclude with the definition of the two quantities

\[ \Delta \Sigma_\pm(K, \Omega) = \langle G^{R}(\mathbf{k}_+ \pm \mathbf{K}, \omega_+ - \mathbf{k}_-, \omega_-) \rangle, \]

\( \hspace{1cm} (3.21) \)

and

\[ \Delta \Sigma_\pm(\mathbf{k}, \Omega) = \Sigma^{R}(\mathbf{k}_+ \pm \mathbf{K}, \omega_+ - \Sigma^{R}(\mathbf{k}_-, \omega_-) \rangle, \]

\( \hspace{1cm} (3.22) \)

where

\[ \mathbf{k}_\pm = \mathbf{k} \pm \mathbf{K}/2 \text{ and } \omega_\pm = \omega \pm \Omega/2. \]

\( \hspace{1cm} (3.23) \)

We will use both of these in the following, especially when \( \mathbf{K} = 0 \) and \( \Omega = 0 \):

\[ \Delta \Sigma_\pm(\mathbf{0}, 0) = 2i\text{Im}(G^{R}(\mathbf{k}, \omega) \rangle \]

\( \hspace{1cm} (3.24) \)

and

\[ \Delta \Sigma_\pm(\mathbf{0}, 0) = 2i\text{Im}(G^{R}(\mathbf{K}, \omega) \rangle = -\frac{\omega}{c} n_\alpha(\hat{k})\Delta(\hat{n}_\alpha k, \omega) \]

\( \hspace{1cm} (3.25) \)
B. The averaged two-particle Green function

The averaged two-particle Green function \( \Phi \) obeys the Bethe-Salpeter equation [64]:

\[
\Phi = \left[ (G^R \otimes (G^A))^\otimes (N) + (G^R \otimes (G^A))^{(N)} \right] U \Phi ,
\]

where \( U \) stands for the irreducible vertex function or intensity operator, which in the weak-scattering approximation, equals the structure factor: \( U \approx B \). The Bethe-Salpeter equation is best handled in momentum and frequency space. With all arguments, the Green function is \( \Phi_{kk'}(K, \Omega, t) \) (see Appendix A). The variables \( K, \Omega \) correspond to the center of “mass” coordinates \( R, T \), introduced in the introduction to this section, and the wave vectors \( k, k' \) to relative coordinates \( r, r' \). The superscript \( \omega \) is the light frequency, and the \( t \) dependence explicitly comes from the structure factor \( B_{kk'}(t) \). The explicit form of the Bethe-Salpeter equation reads

\[
\int \frac{d^3k_1}{(2\pi)^3} \left( [1_{kk'}^{(4)} - f_k^\omega(K, \Omega) B_{kk'}^{(4)}(t)] \Phi_{kk'}(K, \Omega, t) \right) = f_k^\omega(K, \Omega) 1_{kk'}^{(4)},
\]

(3.27)

which we derive in Appendix A. In the last equation we have introduced an abbreviation for the tensor product of the averaged one-particle Green functions:

\[
f_k^\omega(K, \Omega) = \left[ (G^R)^\otimes (G^A)^\otimes \right]^{(N)},
\]

(3.28)

and a shorthand notation \( 1_{kk'}^{(4)} \) for the combination of the delta function and the unit element of fourth rank tensors:

\[
[1_{kk'}^{(4)})_{ijkl} := (2\pi)^3 \delta(k - k') (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})/2 .
\]

(3.29)

Here \( i, j, k \) and \( l \) are Cartesian indices. The Bethe-Salpeter equation (3.27) can be solved formally by iteration, leading to a sum of ladder diagrams (see Fig. B). \( \Phi \) is represented as a sum of vertex diagrams, each of which is a product of a vertex function and a structure factor. The multiple integrals and the sum can be done analytically for \( \delta \)-function correlations for \( K \rightarrow 0 \), and the solution represents the diffusion pole of \( \Phi \). For the anisotropic, long-range correlations of our problem this procedure is not possible. Vollhardt and Wölffe [63] derived the diffusion pole for isotropic electron transport directly from Eq. (2.27), and MacKintosh and John [13] applied their method to light. In the anisotropic case one has to be more careful, as Wölffe and Bhatt showed for electrons [14]. We will proceed a similar way, however our problem is more difficult because we have to deal with the different polarizations of light.

Let \( \Psi_{k}^{(n)}(K, \Omega, t) \) and \( \lambda^{(n)}(K, \Omega, t) \) be, respectively, the \( n \)th eigenvector and eigenvalue of the integral operator of Eq. (3.27),

\[
\int \frac{d^3k_1}{(2\pi)^3} [1_{kk'}^{(4)} - f_k^\omega(K, \Omega) B_{kk'}^{(4)}(t)] \Psi_{kk'}^{(n)} = \lambda^{(n)} \Psi_{kk'}^{(n)},
\]

(3.30)

and \( \overline{\Psi}_{k}^{(n)}(K, \Omega, t) \) the eigenvectors of the Hermitian adjoint operator [22]. Then it is straightforward to show [13], with the aid of the completeness relation

\[
\sum_n \Psi_{k}^{(n)} \otimes \overline{\Psi}_{k'}^{(n)} = 1_{kk'}^{(4)},
\]

(3.31)

that

\[
\Phi_{kk'}(K, \Omega, t) = \sum_n \Psi_{k}^{(n)} \otimes \overline{\Psi}_{k'}^{(n)} \lambda^{(n)} f_k^\omega(K, \Omega)
\]

(3.32)

solves the Bethe-Salpeter equation. When \( K = 0 \) and \( \Omega = t = 0 \), the quantity \( \Delta G_{kk'}^{\omega}(0, 0) \), defined in Eq. (3.21), is an eigenvector of Eq. (3.31) with eigenvalue \( \lambda^{(0)}(0, 0, 0) = 0 \). To prove this statement we use a special case of one of the Ward identities (see Appendix B1),

\[
\Delta \Sigma_{kk'}^{\omega}(0, 0) = \int \frac{d^3k_1}{(2\pi)^3} B_{kk'}^{\omega}(t = 0) \Delta G_{kk'}^{\omega}(0, 0) ,
\]

(3.33)

and the relation

\[
\Delta G_{kk'}^{\omega}(0, 0) = f_k^\omega(0) \Delta \Sigma_{kk'}^{\omega}(0, 0) .
\]

(3.34)

The first equation is obvious from the definition of the mass operator, and the second one is given in Appendix B2. We have identified the diffusion pole, as we shall explicitly see soon. The result is valid beyond the weak-scattering approximation and based on the Ward identities [1]. All other eigenvalues of Eq. (3.31) are positive, and in the real-space coordinate \( R \) they give exponentially decaying contributions to \( \Phi_{kk'}^{\omega}(K, \Omega, t) \), which are not important at long length scales [13].

The diffusion approximation follows when we calculate \( \lambda^{(0)}(K, \Omega, t) \) with the help of perturbation theory in the limit \( K, \Omega, t \rightarrow 0 \). First, we have to get the eigenvalue

FIG. 3. The two-particle Green function \( \Phi \) as a sum of ladder diagrams. \( f_k^{\omega}(K, \Omega) \) propagates two electric field modes with wave vectors \( k \) and \( k' \). Both are scattered by the same inhomogeneity in the dielectric tensor described by the structure factor \( B_{kk'}^{\omega}(t) \) for the scattering vector \( k - k' \).
equation in this limit. To proceed we need the following equation, which we derive in Appendix B.

\[
[\Delta G^\omega_k(K, \Omega)]^\alpha \approx \left[ f^\omega(0,0)]^{\alpha}\right) \left[ \Delta \Sigma^\omega_k(0,0)\right]_\alpha
- \frac{\partial [G^\omega_0]^{\alpha}}{\partial k} \cdot K - \frac{2\omega}{c^2} \Omega .
\] (3.35)

As usual, the Greek superscript or subscript \( \alpha \) refers, respectively, to the basis “vectors” \( e_\alpha(\hat{k}) \otimes e_\alpha(\hat{k}) \) or \( d^\alpha(\hat{k}) \otimes d^\alpha(\hat{k}) \). Equation 3.35 gives \( [\Delta G^\omega_k(K, \Omega)]^\alpha \) to first order in \( K, \Omega \) and \( \Delta \). It shows clearly where the wave vector \( K \) and the frequency \( \Omega \) come in. Now, we formulate eigenvalue equation 3.35 in components, choosing \( K = 0 \) and \( \Omega = 0 \). Then, we apply the last equation and arrive at

\[
\left( [\Delta \Sigma^\omega_k(0,0)]_\alpha - \frac{\partial [G^\omega_0]^{\alpha}}{\partial k} \cdot K - \frac{2\omega}{c^2} \Omega \right) \Psi_k^\alpha
- [\Delta G^\omega_k(0,0)]^\alpha \sum_\beta \int \frac{d^3k_1}{(2\pi)^3} [B^\omega_{k_1k}(t)]_{\alpha \beta} \Psi_k^\beta
= \lambda(K, \Omega, t) [\Delta \Sigma^\omega_k(0,0)]_\alpha \Psi_k^\alpha
\] (3.36)

as the eigenvalue equation, which is correct up to first order in \( K, \Omega, \) and \( \Sigma \). In a next step we use the ansatz

\[
\Psi_k^\alpha \propto [\Delta G^\omega_k(0,0)]^\alpha \left[ \tilde{\Psi}_0 \pi + \sum_{i=1} \tilde{\Psi}_i \varphi^\alpha_i(\hat{k}) \right] ,
\] (3.37)

for the eigenfunctions to turn the eigenvalue equation into a matrix equation. The first factor on the right-hand side forces \( k \) to equal \( \frac{\omega}{c} n_\alpha(\hat{k}) \) in the momentum shell approximation:

\[
[\Delta G^\omega_0(0,0)]^\alpha \approx -i\pi \frac{c}{\omega} n_\alpha(\hat{k}) \delta(\hat{k} - k) .
\] (3.38)

The amplitude \( \tilde{\Psi}_0 \) represents the zeroth eigenvector, and the second term involves a complete set of real basis functions \( \varphi^\alpha(\hat{k}) \) on the unit sphere and for the space spanned by \( e_1 \otimes e_1 \) and \( e_2 \otimes e_2 \). We will comment below on how to choose these basis functions. Keeping only dominant terms in the small \( \Omega, K, \) and \( t \) limit, we can write the eigenvalue equation in matrix form as

\[
\begin{bmatrix}
\frac{2\pi^3}{\pi^2} [-i\Omega + \mu(\omega, t)] - i[G(K)]^\dagger \\
-iG(K) \\
\end{bmatrix}
\begin{bmatrix}
\tilde{\Psi}_0 \\
\cdots \\
\end{bmatrix}
= \lambda(K, \Omega, t)
\begin{bmatrix}
\Sigma \\
S_0 \\
S_1 \\
\end{bmatrix}
\begin{bmatrix}
\tilde{\Psi}_0 \\
\cdots \\
\end{bmatrix} .
\] (3.39)

The matrix \( \Sigma \) basically represents the structure factor \( [B^\omega_{k^\alpha \cdot q^\beta}(0)]_{\alpha \beta} \) in our chosen basis,

\[
[B]_{ij} = \pi \sum_{\alpha,\beta} \left\{ \int_k \int_q \varphi^\alpha_i(\hat{k}) \varphi^\beta_j(\hat{q}) \right\},
\] (3.40)

The vector \( \mathcal{G} \) with components

\[
[\mathcal{G}(K)]_i = \pi \sum_{\alpha} \int_k n_\alpha(\hat{k}) \frac{\partial [G^\omega_0]^{\alpha}}{\partial k} \cdot K \varphi^\alpha_i(\hat{k})
\] (3.41)

is linear in \( K \). Another important term is the coefficient

\[
\mu(\omega, t) = \frac{c\pi^3}{2\omega} \sum_{\alpha,\beta} \int_k \int_q [B^\omega_{k^\alpha \cdot q^\beta}(0) - B^\omega_{k^\alpha \cdot q^\beta}(t)]_{\alpha \beta} ,
\] (3.42)

where

\[
\frac{n^3}{\pi^2} = \frac{1}{8\pi} \sum_{\alpha} \int dQ n^3_{\alpha}(\hat{k}) .
\] (3.43)

For reasons that will become clear shortly, we will refer to \( \mu(\omega, t) \) as a dynamic absorption coefficient. It is an average over dynamical modes that decay in time. As a result, it increases from zero with increasing \( t > 0 \). Finally we have the constant

\[
\Sigma = \pi^3 \sum_{\alpha,\beta} \int_k \int_q [B^\omega_{k^\alpha \cdot q^\beta}(0)]_{\alpha \beta}
= -\frac{c^2}{\omega^2} \sum_{\alpha} \int \frac{d^3k}{(2\pi)^3} [\Delta \Sigma^\omega_k(0,0)]_{\alpha \beta} [\Delta G^\omega_k(0,0)]^{\alpha} .
\] (3.44)

The second equation above states that \( \Sigma \) is proportional to the normalization factor of the eigenfunction \( \Delta G^\omega_k(0,0) \), with \( \Delta \Sigma^\omega_k(0,0) \) being the eigenfunction of the Hermitian conjugated problem. The vector \( S_0 \) and the matrix \( S_1 \) are irrelevant here. For \( K, \Omega, t = 0 \) the zeroth eigenvalue \( \lambda(0) \) corresponds to the eigenvector \( [1,0,0,\ldots] \) in the eigenvalue equation 3.35, and we want to know how it evolves for small \( K, \Omega, \) and \( t \). The vector \( \mathcal{G}(K) \) couples the zeroth element with the matrix \( \mathcal{B} \). The coupling can be removed by an orthogonal transformation with

\[
U = \begin{bmatrix}
\frac{1}{1 - iB^{-1} \mathcal{G}(K)} & i\mathcal{G}(K) \cdot B^{-1} \\
1 & 1
\end{bmatrix} ,
\] (3.46)

which essentially renormalizes the zeroth element. Calculating \( \lambda(0) \) from the transformed system to leading order in \( K^2 \) shows that \( \lambda(0) \Sigma \) is equal to the renormalized zeroth element, or

\[
\lambda(0) = \frac{2\pi^3}{c\Sigma} \left[-i\Omega + \mu(\omega, t) + \frac{c\pi}{2\omega^2} \mathcal{G}(K) \cdot B^{-1} \mathcal{G}(K) \right] .
\] (3.47)

Now, we are ready to write down the diffusion approximation of the averaged two-particle Green function from Eq. 3.32,

\[
\Phi^\omega_{kk}(K, \Omega, t) \approx \frac{1}{N} \frac{\Delta G^\omega_k(0,0) \otimes \Delta G^\omega_k(0,0)}{\Delta \omega + \mu(\omega, t) + K \cdot D(\omega) K} .
\] (3.48)
with

\[ \mathbf{K} \cdot \mathbf{D}(\omega) \mathbf{K} = \frac{c}{2n^3} \mathcal{G}(\mathbf{K}) \cdot \mathbf{B}^{-1} \mathcal{G}(\mathbf{K}) \]  \hspace{1cm} (3.49) \]

and \( N = -2n^2 \omega^2 / (\pi c^3) \). When \( \mu(\omega, t) = 0 \) the Green function \( \Phi_{kk}(\mathbf{K}, \Omega, t) \) possesses a simple diffusion pole with an anisotropic diffusion tensor \( \mathbf{D}(\omega) \). In addition we have the dynamic absorption coefficient \( \mu(\omega, t) \), which forms the basis of DWS. We will discuss it in the next subsection.

From Eq. (3.49), it is clear that the problem of calculating \( \mathbf{D}(\omega) \) reduces to the problems of calculating \( \mathcal{G}(\mathbf{K}) \) and the inverse matrix \( \mathbf{B}^{-1} \). If we had at our disposal a few small, \( \mathbf{B}^{-1} \) does not depend on the special choice we make.

Unfortunately these functions are difficult to find in anisotropic systems, and we are reduced to using us-

able approximation schemes. One scheme is to choose functions \( \varphi_i(\mathbf{k}) \) so that only a few components of \( \mathcal{G}(\mathbf{K}) \), say \( \mathcal{G}(\mathbf{K}) \), for \( i = 1, 2 \), are non-zero. Then, if the off-

diagonal components of \( \mathbf{B}_{ij} \), coupling \( j \) for \( j > 2 \) to \( \mathcal{G}(\mathbf{K}) \), are small, \( \mathcal{G}(\mathbf{K}) \) can be approximated by using only the upper half of \( \mathbf{B}_{ij} \) to calculate \( \mathbf{B}^{-1} \). We will use this approach in numerical calculations of \( \mathbf{D}(\omega) \) in nematic liquid crystals in the next section.

To test our theory we apply it to isotropic systems. We concentrate on the case, where fluctuations in the polarization vectors are small, \( \mathcal{G}(\mathbf{K}) \), for a fixed scattering angle \( \theta_\alpha \). Then the components of the structure factor assume the form

\[ \mathcal{G}(\mathbf{K})_{ij} = \left[ \mathcal{G}(\mathbf{K})_{ii} \right]_{ij} \mathcal{G}(\mathbf{K})_{jj} \]

where \( \mathcal{G}(\mathbf{K})_{ij} \) only depends on the scattering angle \( \theta_\alpha \). In isotropic systems, all the polarization vectors perpendicular to \( \hat{k} \) are equivalent, and the diffusion constant should not depend on the special choice we make. Therefore, to treat the factor \( \left[ \mathcal{G}(\mathbf{K})_{ij} \right]_{ij} \), we can add integrations \( \int d\phi_1/(2\pi) \int d\phi_2/(2\pi) \) in the formula for \( \mathbf{B} \). The angles \( \phi_1 \) and \( \phi_2 \), respectively, describe rotations of the polarization vectors about \( \hat{k} \) and \( \hat{q} \). The integration over \( \phi_1 \) and \( \phi_2 \) for a fixed scattering angle \( \theta_\alpha \) is straightforward, and we obtain

\[ \int \frac{d\phi_1}{2\pi} \int \frac{d\phi_2}{2\pi} \mathcal{G}(\mathbf{K})_{ij} = \mathbf{B}^{-1}(\mathbf{K}, \mathbf{K}) \]

where the dielectric constant \( \varepsilon_0 \) comes in through the normalization of the polarization vectors. With this trick we are able to show that \( \mathbf{B} \) is essentially diagonalized by spherical harmonics. As basis functions we choose \( \varphi_{1,m}(\mathbf{k}) = [Y_{1,m}(\theta, \varphi), Y_{1,-m}(\theta, \varphi)] \) and \( \varphi_{2,m}(\mathbf{k}) = [Y_{2,m}(\theta, \varphi), -Y_{2,-m}(\theta, \varphi)] \) with \( \delta_{\theta} = \hat{k} \cdot \mathbf{K} \). We also use the addition theorem, \( \mathcal{P}_{lm}(\theta_\alpha) \propto \sum_m Y_{lm}(\theta, \varphi)Y_{lm}(\theta', \varphi') \) \( \{3\} \), and arrive at \( \mathbf{B}_{1,m}^{\gamma l m} \propto \delta_{\gamma l} \delta_{\theta_m} \delta_{\theta_{mm'}} \). The only non-zero element of \( \mathcal{G} \) is \( \mathcal{G}_{110} = -2\varepsilon_0 K / \sqrt{3} \), and we finally find the formula for the diffusion constant:

\[ D = \frac{16}{3^n} \pi \varepsilon_0 \]

where \( \varepsilon_0 \) stands for the speed of light in the system. Compared to the scalar case we have an additional factor \( 1 + \cos^2 \theta_\alpha \). \( \mathcal{P}_{lm}(\theta_\alpha) \) alone describes a scattering process with incoming and outgoing polarization perpendicular to the scattering plane (VV scattering), whereas \( \cos^2 \theta_\alpha \mathcal{P}_{lm}(\theta_\alpha) \) belongs to scattering with polarizations in the plane (HH scattering) \( \{1\} \). In a group theoretical language \( \mathcal{B}_{1,m}^{\gamma l m} \propto \delta_{\theta_m} \delta_{\theta_{mm'}} \) transforms under a high-dimensional identity representation of \( \text{SO}(3) \). The relation \( \mathcal{B}_{1,m}^{\gamma l m} \propto \delta_{\theta_m} \delta_{\theta_{mm'}} \) then means that we have decomposed \( \mathcal{B}_{1,m}^{\gamma l m} \) via the irreducible representations of \( \text{SO}(3) \). For less symmetric phases, like a nematic liquid crystal, we can at least partially diagonalize \( \mathbf{B} \) with the help of group theory. The relevant symmetry group \( \text{D}_{sch} \) has only one-dimensional representations induced by the functions \( \exp(i m \varphi) \), where \( \varphi \) is the azimuthal angle around the symmetry axis. Decomposing \( \mathbf{B} \) with the help of spherical harmonics gives \( \mathcal{B}_{1,m}^{\gamma l m} \propto \delta_{\theta_m} \delta_{\theta_{mm'}} \), where different \( l \) and \( l' \) now couple to each other (see next section).

The numerator of the Green function in Eq. (3.48) contains an interesting effect. The second factor \( \Delta \mathcal{G}_{\mathbf{k}}(0, 0) \) collects the light sources or the incoming light waves. The first factor determines the energy density \( W_{\alpha}(\mathbf{R}, \mathbf{k}, \mathbf{T}, \mathbf{Z}) \) of an outgoing light wave independent of the sources. This means that in the diffusion approximation the outgoing light looses all its correlations with the light sources. Integrating \( \Delta \mathcal{G}_{\mathbf{k}}(0, 0) \) over the wavenumber \( k \) shows that the ratio for the energy densities \( W_{\alpha}(\mathbf{R}, \mathbf{k}, \mathbf{T}, \mathbf{Z}) \) in polarization states 1 and 2 is \( |n_1(\mathbf{k})/n_2(\mathbf{k})|^2 \). Experiments measure light intensities. In Eq. (2.17) we learned that only the projection of the Poynting vector \( S^\alpha \) on \( \mathbf{k} \) is simple: \( S^\alpha \cdot \mathbf{k} = (c/n_\alpha) W_{\alpha}(\mathbf{R}, \mathbf{k}, \mathbf{T}, \mathbf{Z}) \). Its ratio for the output polarizations 1 and 2 is \( |n_1(\mathbf{k})/n_2(\mathbf{k})|^2 \) which gives the ratio of the output intensities when the Poynting vectors are parallel to \( \mathbf{k} \). This is the case for light travelling along the prim-
cipal axes of the dielectric tensor. The Green function in Eq. (3.48) suggests that there is a diffusion equation for each light wave with direction $k$ and polarization $\alpha$. This does not mean that we have additional conserved quantities besides the energy density. It only means that after randomizing the incoming light the distribution of the light modes in the light field stays the same.

C. Diffusing Wave Spectroscopy

If we sum over the two polarization states and integrate over all wave vectors $k$ the Green function in Eq. (3.48) is equivalent to a diffusion equation for the scalar time-correlation function $W(\mathbf{R}, T, t)$, introduced in Eq. (3.4),

$$\frac{\partial}{\partial T} - \nabla \cdot \mathbf{D}(\omega) \nabla + \mu(\omega, t) W(\mathbf{R}, T, t) = \varrho(\mathbf{R}, T) .$$

(3.54)

This equation is the basis of Diffusing Wave Spectroscopy (DWS) [3]. Solving it for special boundary conditions and sources $\varrho(\mathbf{R}, T)$, which depend on experimental arrangements, gives $W(\mathbf{R}, T, t)$ in terms of the dynamic absorption coefficient $\mu(\omega, t)$. In our derivation of the diffusion pole we had to restrict ourselves to times $t$ with $[\mathbf{B}_{k=\mathbf{q}=0}(0) - \mathbf{B}_{k=\mathbf{q}=0}(t)]_{\alpha\beta} \ll [\mathbf{B}_{k=\mathbf{q}=0}(0)]_{\alpha\beta}$. In this time range we expect being able to perform a Taylor expansion in $t$ which gives a linear time dependence for $\mu(\omega, t)$:

$$\mu(\omega, t) = \mu_0 t .$$

(3.55)

The constant $\mu_0$ reflects some averaged dynamical properties of the system. For the diffusion of particles in colloidal suspensions, $\mu_0 = 2D_0\omega^2/(4\pi\epsilon)$, where $D_0$ is the self-diffusion constant of the particles [13]. The condition just imposed on $t$ means that DWS probes the dynamics of a system on a much shorter time scale than single light scattering does, which requires times where $[\mathbf{B}_{k=\mathbf{q}=0}(t)]_{\alpha\beta}$ has already decayed considerably. DWS, therefore, offers the possibility of studying the validity of the dynamical description of the system on short time scales. This was done for colloidal suspensions by Kao et al. [32], who studied the Brownian motion of single particles at short times where the mean-square displacement is not simply proportional to $t$. As a result, $\mu(\omega, t)$ does not follow the linear time law of Eq. (3.55). Experimentalists prefer a different picture for DWS which they have developed for colloidal suspensions [13]. They sum up all possible light paths in the scattering medium to arrive at the time-correlation function after some averaging procedure. We will show here that our approach is totally equivalent to this picture. However it has the advantage that it automatically tells us how to perform this averaging procedure.

DWS experiments are usually performed with continuous light sources and the diffusion equation reduces to

$$[\mu(\omega, t) - \nabla \cdot \mathbf{D}(\omega) \nabla] W(\mathbf{R}, \ldots, t) = \varrho(\mathbf{R}, \ldots) .$$

(3.56)

We can rewrite this equation as the Laplace transform of a problem, where the source is a light pulse,

$$\left[ \frac{\partial}{\partial T} - \nabla \cdot \mathbf{D}(\omega) \nabla \right] P(\mathbf{R}, \tau) = \delta(\tau) \varrho(\mathbf{R}, \ldots)$$

(3.57)

with

$$W(\mathbf{R}, \ldots, t) = \int_0^\infty P(\mathbf{R}, \tau) \exp[-\mu(\omega, t)\tau] d\tau .$$

(3.58)

The lower limit $-0$ means a small negative time $\tau$ in order to pick up the delta function. We can interpret $P(\mathbf{R}, \tau)$ (after an appropriate normalization) as the probability that light, emitted by the source at time $\tau = 0$, arrives at the detector point $\mathbf{R}$ after a time $\tau$. Then the time-correlation function $W(\mathbf{R}, \ldots, t)$ follows after a summation over all light paths where each light path contributes a factor $\exp[-\mu(\omega, t)\tau]$ to the decay of $W(\mathbf{R}, \ldots, t)$. For isotropic systems $\tau$ is directly connected to the path length $s = c\tau/n$ of light and $P(\mathbf{R}, \tau)$ also represents the path-length distribution. The exact form of $P(\mathbf{R}, \tau)$ depends on the choice of the light source and the boundary conditions [19] which we don’t address here.

IV. LIGHT DIFFUSION IN NEMATIC LIQUID CRYSTALS

The nematic liquid crystalline phase consists of rodlike organic molecules which tend to align parallel to each other but which show no long-range positional order of their centers of mass. The local average direction of the molecules is described by a unit vector $\mathbf{n}(r, t)$ called director. It appears in the local dielectric tensor

$$\epsilon(r, t) = \epsilon_\perp \mathbf{1} + \Delta \epsilon [\mathbf{n}(r, t) \otimes \mathbf{n}(r, t)] ,$$

(4.1)

where $\epsilon_\perp$ and $\epsilon_\parallel$ are the dielectric constants for electric fields, respectively, perpendicular and parallel to the director and where $\Delta \epsilon = \epsilon_\parallel - \epsilon_\perp$ stands for the dielectric anisotropy. The energetically favored state of a nematic phase is a uniform director field $\mathbf{n}(r, t) = \mathbf{n}_0$ throughout the sample. Its distortion costs energy which can be calculated from the Frank-Oseen-Zöcher free energy [54]:

$$F[\mathbf{n}] = \frac{1}{2} \int [K_1(\text{div}\mathbf{n})^2 + K_2(\mathbf{n} \cdot \text{curl}\mathbf{n})^2$$

$$+ K_3(\mathbf{n} \times \text{curl}\mathbf{n})^2 - \Delta \chi(\mathbf{n} \cdot \mathbf{H})^2] d^3r .$$

(4.2)

There are three characteristic distortions, called splay ($K_1$), twist ($K_2$) and bend ($K_3$), where $K_1$, $K_2$ and $K_3$ are the Frank elastic constants. We also include a magnetic-field term with $\Delta \chi = \chi_\parallel - \chi_\perp$ the anisotropy of the magnetic susceptibility. If $\Delta \chi > 0$ an alignment of the director parallel to the field $\mathbf{H}$ is favored. Even in a uniformly aligned sample there exist thermally induced fluctuations of the director:
They lead to fluctuations in the local dielectric tensor and hence scatter light. This is the physical phenomenon for which we want to calculate the diffusion approximation of light. In order to apply the formulas of the preceding section, we have to look first at the light propagation in an uniaxial medium. Then we need to calculate the structure factor, associated with the director fluctuations, which governs the single-light-scattering event.

A. Light propagation in uniaxial media

Let us, for a moment, suppress the fluctuations of the director and look at a homogeneous medium with a uniaxial dielectric tensor

\[ \varepsilon_0 = \varepsilon_\perp 1 + \Delta \varepsilon [n_0 \otimes n_0] . \]  

The equilibrium value \( n_0 \) of the director is also called the optical axis because it establishes a special axis for light propagation. The two light modes follow from solving the eigenvalue equation (2.8). They are well described in the literature [17]. We mainly summarize the results here, introduce some notation, and perform an interesting variable transformation for later use.

One light mode is immediately obvious. Its polarization vectors \( \varepsilon_2(\hat{k}) \) and \( d^2(\hat{k}) \) are both perpendicular to \( n_0 \) and \( \hat{k} \) with a refractive index \( n_2 = \sqrt{\varepsilon_\perp} \). Since it behaves as in an isotropic system, it is named the ordinary light mode. We choose \( n_0 \) parallel to the \( z \) axis and write \( \hat{k} \) in spherical coordinates:

\[
\begin{bmatrix}
0 \\
0 \\
1
\end{bmatrix}
\quad \text{and} \quad
\begin{bmatrix}
\sin \theta \cos \varphi \\
\sin \theta \sin \varphi \\
\cos \theta
\end{bmatrix}.
\]

Then the ordinary light ray is represented by

\[
n_2 = \sqrt{\varepsilon_\perp} \quad \text{and} \quad e_2(\hat{k}) = \frac{1}{n_2} \begin{bmatrix}
-\sin \varphi_k \\
\cos \varphi_k \\
0
\end{bmatrix} .
\]

In the extraordinary light mode, the refractive index depends on \( \hat{k} \), and the polarization vector \( e_1(\hat{k}) \) is generally not perpendicular to \( \hat{k} \). It follows from \( e_1(\hat{k}) = \varepsilon_0^{-1} d^1(\hat{k}) \) where the polarization vector \( d^1(\hat{k}) \) is determined by \( d^1(\hat{k}) \perp d^2(\hat{k}) \) and \( d^1(\hat{k}) \perp \hat{k} \). The refractive index can be calculated from the eigenvalue equation (2.8) or Fresnel’s equation (2.14):

\[
n_1(\hat{k}) = n_2 \tilde{n}_1(\hat{k}) \quad \text{with} \quad \tilde{n}_1(\hat{k}) = \left[ \frac{1 + \alpha}{1 + \alpha \cos^2 \delta_k} \right]^{1/2} ,
\]

where we have introduced the relative dielectric anisotropy of the system,

\[
\alpha = \Delta \varepsilon / \varepsilon_\perp .
\]

Equation (4.17) does not represent the most symmetric form for \( n_1(\hat{k}) \), but we found it useful for our calculations. Finally, we get

\[
e_1(\hat{k}) = \frac{n_1(\hat{k})}{n_2} \begin{bmatrix}
\cos \varphi_k \\
\sin \varphi_k \\
0
\end{bmatrix} \begin{bmatrix}
\cos \delta_k & - \sin \delta_k \\
\sin \delta_k & \cos \delta_k \\
0 & 1
\end{bmatrix} .
\]

In the structure factor, \( \hat{k} \) always appears together with the refraction index. For the extraordinary mode we, therefore, replace the angular variables \( \cos \varphi_k \) and \( \sin \varphi_k \) by an equivalent set,

\[
C_k := \tilde{n}_1(\hat{k}) \cos \varphi_k \quad \text{and} \quad S_k := \tilde{n}_1(\hat{k}) \sin \varphi_k ,
\]

in which the refraction index takes the form

\[
\tilde{n}_1(\hat{k}) = \sqrt{1 + \alpha (1 - C_k^2)} .
\]

The “trigonometric” identity,

\[
S_k^2 + (1 + \alpha) C_k^2 = 1 + \alpha
\]

is valid. The new coordinate \( C_k \) ranges from \(-1\) to \(1\) and contains the same information as \( \cos \varphi_k \). Thus, we can choose, e.g., spherical harmonics as basis functions on the unit sphere with \( \cos \varphi_k \) replaced by \( C_k \). The usefulness of \( C_k \) appears when we calculate its differential with respect to \( \cos \varphi_k \):

\[
(1 + \alpha) dC_k = \tilde{n}_1(\hat{k}) d\cos \varphi_k .
\]

The differential \( dC_k \) absorbs the factor \( \tilde{n}_1(\hat{k}) \) which always comes with our integrals thus making them easier:

\[
\int_{C_k} = \int \frac{dC_k}{(2\pi)^3} n_1^3(\hat{k}) \ldots
\]

\[
= n_2^3 (1 + \alpha) \int \frac{dC_k d\varphi_k}{(2\pi)^3} \ldots .
\]

Alternatively, we can say that we have constructed a complete set of basis functions in the variables \( \cos \varphi_k \) and \( \varphi_k \) with respect to the weight function \( \tilde{n}_1(\hat{k}) \). In the following we will also use the notation \( C_k = \cos \varphi_k \) and \( S_k = \sin \varphi_k \) for ordinary light modes.

B. Structure factor for director fluctuations

Here we will derive the structure factor for director fluctuations. To begin we recall the general form of the time-dependent structure factor for light scattering in an anisotropic dielectric,

\[
[B_{k^* q^*}(t)]_{\alpha \beta} = \frac{\omega^4}{c^4} \left< e_\alpha(\hat{k}) \cdot \delta \varepsilon(q, t) e_\beta(\hat{q}) \right> \left[ e_\alpha(\hat{k}) \cdot \delta \varepsilon(q, 0) e_\beta(\hat{q}) \right]^* ,
\]
where \( \mathbf{q}_s = \hat{n}_0 \cdot \mathbf{k} - n_\beta \mathbf{q} \) stands for the scattering vector. We first need the Fourier component \( \delta \mathbf{e}(\mathbf{q}_s, t) \) of the fluctuating part of the dielectric tensor. We insert \( \mathbf{n}(r, t) = \mathbf{n}_0 + \delta \mathbf{n}(r, t) \) into \( \varepsilon(r, t) \) of Eq. (4.14) and collect the first-order terms in \( \delta \mathbf{n}(r, t) \). After a Fourier transformation we arrive at

\[
\delta \varepsilon(\mathbf{q}_s, t) = \Delta \varepsilon [\mathbf{n}_0 \otimes \delta \mathbf{n}(\mathbf{q}_s, t) + \delta \mathbf{n}(\mathbf{q}_s, t) \otimes \mathbf{n}_0] \quad (4.16)
\]

with \( \delta \mathbf{n}(\mathbf{q}_s, t) \) the amplitude of a director mode. It has only two components since the director is a unit vector. Furthermore, for small fluctuations, \( \delta \mathbf{n} \) is perpendicular to \( \mathbf{n}_0 \). An appropriate basis for \( \delta \mathbf{n}(\mathbf{q}_s, t) \), as shown in Fig. 4, consists of a unit vector \( \hat{u}_1(\mathbf{q}_s) \), lying in the plane defined by \( \mathbf{n}_0 \) and \( \mathbf{q}_s \), and a second one, \( \hat{u}_2(\mathbf{q}_s) \), perpendicular to this plane:

\[
\delta \mathbf{n}(\mathbf{q}_s, t) = \delta n_1(\mathbf{q}_s, t) \hat{u}_1(\mathbf{q}_s) + \delta n_2(\mathbf{q}_s, t) \hat{u}_2(\mathbf{q}_s) \quad (4.17)
\]

**FIG. 4.** Basis vectors \( \hat{u}_1(\mathbf{q}_s) \) and \( \hat{u}_2(\mathbf{q}_s) \) for given director \( \mathbf{n}_0 \) and wave vector \( \mathbf{q}_s \).

Next we need the temporal autocorrelation function of the director modes \( [54,55] \), which is rather complex. We state it and then explain the individual terms:

\[
\langle \delta \mathbf{n}(\mathbf{q}_s, t) \otimes \delta \mathbf{n}^*(\mathbf{q}_s, 0) \rangle =
\sum_{\delta=1}^{2} \frac{k_B T}{K_\delta(\mathbf{q}_s)} \exp \left[ \frac{-K_\delta(\mathbf{q}_s)}{\eta_\delta(\mathbf{q}_s)} \right] \hat{u}_\delta(\mathbf{q}_s) \otimes \hat{u}_\delta(\mathbf{q}_s) \quad (4.18)
\]

with

\[
K_\delta(\mathbf{q}_s) = K_{\delta 1} q_\perp^2 + K_{\delta 2} q_\parallel^2 + \Delta \chi H^2 \quad ,
\]

where \( q_\perp \) and \( q_\parallel \) are the components of \( \mathbf{q}_s \), perpendicular and parallel to \( \mathbf{n}_0 \). The chosen basis for \( \delta \mathbf{n}(\mathbf{q}_s, t) \) provides the normal coordinates, because the correlation function is already diagonal. The free energy of one director mode follows from the Frank-Oseen-Zocher free energy \( [12] \) as \( K_\delta(\mathbf{q}_s) )|\delta n_\delta(\mathbf{q}_s, t)|^2 \). For a general vector \( \mathbf{q}_s \) it is either composed of splay and bend (\( \delta = 1 \)) or twist and bend distortions (\( \delta = 2 \)). The factor \( k_B T / K_\delta(\mathbf{q}_s) \) results from the equipartition theorem giving the mean-square amplitude of each mode. The dynamics of the director modes is described by the Leslie-Erickson equations \( [54] \). They combine the Navier-Stokes equation for a uniaxial media with dynamical equations for the director. A detailed analysis \( [54] \) shows that director modes are diffusive with a relaxation frequency given by the quotient of elastic \( [K_\delta(\mathbf{q}_s)] \) and viscous \( [\eta_\delta(\mathbf{q}_s)] \) forces. This is the origin of the exponential factor in the correlation function. The viscosity \( \eta_\delta(\mathbf{q}_s) \) is a combination of several Leslie viscosities which we will address in subsection IV D. We neglect a fast relaxing part which comes from the coupling between the director and the fluid motion.

Finally, we are able to write down the structure factor

\[
[B^\omega_{k\cdot \mathbf{q}^\beta}(t)]_{\alpha\beta} = (\Delta \varepsilon)^2 k_B T \omega^4
\]

\[
\times \sum_{\delta=1}^{2} \frac{N(e_\alpha, e_\beta, \hat{u}_\delta)}{K_\delta(\mathbf{q}_s)} \exp \left[ -\frac{K_\delta(\mathbf{q}_s)}{\eta_\delta(\mathbf{q}_s)} t \right] \quad (4.20)
\]

\[
N(e_\alpha, e_\beta, \hat{u}_\delta) = [(\mathbf{n}_0 \cdot e_\beta)(\hat{u}_\delta \cdot e_\alpha) + (\hat{u}_\delta \cdot e_\beta)(\mathbf{n}_0 \cdot e_\alpha)]^2 \quad (4.21)
\]

a geometrical factor. It has two interesting implications. First, there exists no scattering of an ordinary light ray into an ordinary light ray because \( e_2 \perp \mathbf{n}_0 \), and therefore \( N = 0 \). Second, forward and backward scattering along \( \mathbf{n}_0 \) is always forbidden. The other terms of \( [B^\omega_{k\cdot \mathbf{q}^\beta}(t)]_{\alpha\beta} \) are familiar from the previous discussion. The scattering mean-free-path \( l_0(\mathbf{k}, \omega) \), defined in Eq. (4.17), has been already discussed in detail by two groups \( [56,57] \). Its dependence on \( \mathbf{k} \) and polarization was calculated by Val’kov and Romanov \( [56] \). We just stress one point. The structure factor diverges for \( H = 0 \) and \( \mathbf{q}_s \to 0 \), and we expect the scattering mean-free-paths to be zero in an infinite medium. But \( \mathbf{q}_s = 0 \) can only occur in scattering events where the extraordinary polarization is preserved. Hence, only the scattering mean-free-path \( l_1(\mathbf{k}, \omega) \) of an extraordinary light-ray is zero. In Appendix C we give \( [B^\omega_{k\cdot \mathbf{q}^\beta}(0)]_{11} \) for \( \mathbf{q}_1 \to \mathbf{k}^1 \) (the notation we use there is explained below). From this form it is obvious that \( l^{-1}_1(\mathbf{k}, \omega) \) diverges weakly like \( -\ln(2\Delta \chi H^2 / (K_{\Delta} q_0^2 c^2)) \) for magnetic fields much smaller than \( \sqrt{K_3 / 2\Delta \chi H^2 c} \). The explicit form of the structure factor is very complex because it explicitly depends on the direction of the incoming \( \mathbf{k} \) and outgoing \( \mathbf{q} \) light through the geometrical factor. We mention some symmetry properties which facilitate its handling. The first two are quite obvious:

\[
[B^\omega_{k\cdot \mathbf{q}^\beta}(0)]_{1\alpha} = [B^\omega_{q^\beta k^\alpha}(0)]_{1\alpha} \quad (4.22)
\]

and

\[
[B^\omega_{k\cdot \mathbf{q}^\beta}(0)]_{\alpha\beta} = [B^\omega_{-k\cdot \mathbf{q}^{-\beta}}(0)]_{\alpha\beta} \quad . (4.23)
\]

The structure factor has to reflect the symmetry of the nematic phase described by the group \( D_{\infty h} \). For example, it has to exhibit the rotational symmetry about the director \( \mathbf{n}_0 \). If we choose
\[ \hat{k} = \begin{bmatrix} \sin \theta_k \cos \varphi_k \\ \sin \theta_k \sin \varphi_k \\ \cos \theta_k \end{bmatrix} \quad \text{and} \quad \hat{q} = \begin{bmatrix} \sin \theta_q \cos \varphi_q \\ \sin \theta_q \sin \varphi_q \\ \cos \theta_q \end{bmatrix}, \] (4.24)

then \( [B^\omega(0)]_{\alpha \beta} \) depends only on the relative azimuthal angle \( \varphi = \varphi_q - \varphi_k \). The existence of mirror planes, containing \( n_0 \), implies that the structure factor should be invariant under \( \varphi \rightarrow -\varphi \). In appendix C we give its explicit form in terms of \( \alpha, \beta, \epsilon, q, T, \) and \( \varphi \). We also introduce a scaled structure factor \( \tilde{B}^\omega(0) \) through

\[ [B^\omega(0)]_{\alpha \beta} = (\Delta \varphi) \frac{\omega^2}{c^2} \frac{k_B T}{K_3 n_0^2} [B^\omega(0)]_{\alpha \beta}, \] (4.25)

which depends on scaled parameters: the relative dielectric anisotropy \( \Delta \varphi = \Delta \varepsilon / \varepsilon \perp \), the Frank elastic constants

\[ \overline{K} = K_1 / K_3 \quad \text{and} \quad \overline{K}_2 = K_2 / K_3, \] (4.26)

and the magnetic field

\[ H = H / H_0 \quad \text{with} \quad H_0 = n_2 \frac{\omega}{c} \sqrt{K_3 / \Delta \chi}. \] (4.27)

If we introduce the magnetic coherence length

\[ \xi_3 = \sqrt{\frac{K_3}{\Delta \chi H^2}}, \] (4.28)

which gives the length scale over which director fluctuations are correlated, we obtain for the scaled magnetic field

\[ \hat{h} = \frac{\lambda}{2 \pi \xi_3}, \] (4.29)

where \( \lambda = n_2 \omega / c \). Thus, \( \hat{h} = 1 \) corresponds to a very short coherence length of \( \lambda / 2 \pi \).

In the next subsection we will use the material parameters of a typical nematic compound CB25 to discuss the diffusion constants. 5CB is liquid crystalline at room temperature, and we use the parameters for 5 K below the nematic-isotropic transition \[ [18]. \] The bend elastic constant is \( K_3 = 5.3 \times 10^{-7} \) dyne and is, as usual for conventional thermotropic nematic liquid crystals, larger than the splay and twist constants: \( K_1 = 0.79 \) and \( K_2 = 0.43 \). For green light \( (\omega / c = 1.15 \times 10^{15} \text{cm}^{-1}) \) the dielectric constant is \( \varepsilon \perp = 2.381 \) and the anisotropy is \( \alpha = 0.228 \) corresponding to the refractive indices \( n_1(\varphi = 90^\circ) = 1.710 \) and \( n_2 = 1.543 \). Finally, the magnetic anisotropy is \( \Delta \chi = 0.95 \times 10^{-7} \), from which we obtain a characteristic magnetic field of \( H_0 = 3.6 \times 10^6 \text{G} \).

### C. Light diffusion — results

In this subsection we discuss the two essential components \( D_{||} \) and \( D_{\perp} \) of the diffusion tensor \( D(\omega) \), which describe the diffusion of light, respectively, parallel and perpendicular to the director \( n_0 \). Before we can apply our general formulas from Eqs. (3.40), (3.41) and (3.49) we need to be more specific about our basis functions \( \varphi_i(\hat{k}) \), and we need the derivatives of the components \( [G_0^{-1}]_{\alpha \beta} \) which we gave in Eq. (2.21). The calculations for the latter are straightforward and the results for the extraordinary and ordinary light mode are, respectively,

\[ \tilde{n}_i(\hat{k}) \frac{\partial [G_0^{-1}]_{\alpha \beta}}{\partial \hat{k}} \cdot \hat{K} = - \frac{2}{\varepsilon_\perp} \left( \frac{1 - C_\ell^2 \cos \varphi_k}{\sqrt{1 + \alpha}} K_\perp + C_k K_\parallel \right) \] (4.30)

and

\[ \frac{\partial [G_0^{-1}]_{\alpha \beta}}{\partial \hat{k}} \cdot \hat{K} = - \frac{2}{\varepsilon_\perp} \left( \frac{1 - C_\ell^2 \cos \varphi_k}{\sqrt{1 + \alpha}} K_\perp + C_k K_\parallel \right). \] (4.31)

In the above, we chose

\[ \hat{K} = [K_\perp, 0, K_\parallel], \] (4.32)

wrote \( \hat{k} \) in spherical coordinates as in Eq. (4.24) and then switched to the new \( C \) coordinate. The right-hand side of Eq. (4.31) for the ordinary mode is the isotropic result \( \hat{k} \cdot \hat{K} \), which is modified in the extraordinary case. As our basis functions, we choose

\[ \varphi_i^\alpha(\hat{k}) \rightarrow \varphi_i^{\alpha \text{lm}}(\hat{k}) = \delta^\alpha_{\gamma \text{lm}} \varphi_{\text{lm}}(C_k, \varphi_k), \] (4.33)

where \( \varphi_{\text{lm}}(C_k, \varphi_k) \) stands for a real combination of the spherical harmonics \( Y_{lm}(C_k, \varphi_k) \) and \( Y_{-m}(C_k, \varphi_k) \) in the new coordinate \( C_k \) instead of \( \cos \theta_k \). It will soon become clear why the coordinate \( C_k \) is so helpful. The index \( \gamma \) stands for the basis of the tensor space, which we identify here with our basis \( e_{\alpha} \otimes e_{\alpha} \). Only functions with odd parity, i.e., with odd \( l \), contribute to the diffusion. This is immediately obvious from the parity of the structure factor and the derivative of \( [G_0^{-1}]_{\alpha \beta} \). Furthermore, we only need functions \( \varphi_{\text{lm}}(C_k, \varphi_k) \) containing \( \cos m \varphi_k \) \((m \geq 0) ; \sin m \varphi_k \) is not necessary because of the symmetry of \([B^\omega(0)]_{\alpha \beta}\). With this choice of basis functions the matrix \( B \) is decomposed into submatrices for each \( m \), \( [B]^{\gamma \delta} \), \( \delta \equiv \gamma \equiv \delta \equiv \gamma \), because a term \( \cos \gamma \delta \cos \varphi_k \), \((n \neq m) \), is not compatible with the rotational symmetry of the structure factor. Then, the diffusion constants \( D_{||} \) and \( D_{\perp} \) are, respectively, related to \( m = 0 \) and \( m = 1 \):

\[ D_{||} K_{\parallel}^2 = \frac{c}{2n^2} \sum_{\gamma \delta \text{lm}} [\mathcal{G}(K)]_{\gamma \text{lm}} [B^{-1}]_{\text{lm}} \mathcal{G}(K)_{\gamma \text{lm}} \mathcal{G}(K)_{\gamma \text{lm}} \] (4.34)

\[ D_{\perp} K_{\perp}^2 = \frac{c}{2n^2} \sum_{\gamma \delta \text{lm}} [\mathcal{G}(K)]_{\gamma \text{lm}} [B^{-1}]_{\text{lm}} \mathcal{G}(K)_{\gamma \text{lm}} \mathcal{G}(K)_{\gamma \text{lm}} \] (4.35)

with
Only elements of \( G(K) \) with \( l = 1 \) are non-zero, \( [G(K)]_{1m} \neq 0 \), since the right-hand sides of Eqs. (4.30) and (4.33) depend only on \( l = 1 \) basis functions. This rises the question of how important higher \( l \) contributions are. For our calculations we, therefore, choose \( l = 1 \) and \( l = 3 \) functions:

\[
\begin{align*}
\varphi_{10}(C, \varphi) &= \sqrt{3\pi} C \\
\varphi_{11}(C, \varphi) &= \sqrt{3\pi} \sqrt{1 - C^2} \cos\varphi \\
\varphi_{30}(C, \varphi) &= \sqrt{7\pi} C(5C^2 - 3) \\
\varphi_{31}(C, \varphi) &= \sqrt{2\pi} \sqrt{1 - C^2} (1 - 5C^2) \cos\varphi
\end{align*}
\] (4.37a-d)

with the normalization

\[
\int \frac{dC \, d\varphi \varphi \psi m(C, \varphi) \varphi \psi m'(C, \varphi)}{(2\pi)^3} = \delta_{mm'} \delta_{\varphi \varphi'}.
\] (4.38)

The non-zero components of \( G(K) \) turn out to be

\[
\begin{align*}
|G(K)|_{110} &= -2\varepsilon_\perp (1 + \alpha) K_1 / \sqrt{6} \\
|G(K)|_{210} &= -\varepsilon_\perp K_1 / \sqrt{6} \\
|G(K)|_{111} &= -2\varepsilon_\parallel \sqrt{1 + \alpha} K_1 / \sqrt{6} \\
|G(K)|_{211} &= -2\varepsilon_\perp K_1 / \sqrt{6}.
\end{align*}
\] (4.39a-d)

The integrations to obtain the matrix elements of \( B \) requires more effort. We were able to calculate the integrals over \( \varphi \) analytically with the help of

\[
\begin{align*}
\int d\varphi_k d\varphi_q \phi(\varphi - \varphi_k) &= 2\pi \int d\varphi \phi(\varphi) \\
\int d\varphi_k d\varphi_q \phi(\varphi) \cos\varphi_k \cos\varphi_q &= \pi \int d\varphi \phi(\varphi) \cos\varphi \\
\int d\varphi_k d\varphi_q \phi(\varphi) \cos^2\varphi_k &= \pi \int d\varphi \phi(\varphi).
\end{align*}
\] (4.40a-c)

The results are listed in appendix C. The remaining integrations were performed numerically.

To discuss the diffusion constants we collect the prefactors of the quantities involved to obtain an “averaged” transport mean-free-path

\[
l_0^* = 9\pi \frac{c^2}{\omega^2} \frac{K_3}{k_B T} \frac{1}{\alpha^2}
\] (4.41)

and write the diffusion constant \( D_\parallel \) and \( D_\perp \) in the form

\[
D_\parallel = c_\perp l_0^* D_\parallel / 3, \quad D_\perp = c_\parallel l_0^* D_\perp / 3.
\] (4.42)

reminiscient of isotropic systems. The numerical factors \( \tilde{D}_\parallel \) and \( \tilde{D}_\perp \) only depend on the scaled parameters \( \alpha, h, K_1, \) and \( K_2, \) and \( c_\perp \) is the speed of light of the ordinary light ray. We stress that \( l_0^* \) is an averaged quantity and that our theory does not give a procedure to construct transport mean-free-paths for different light directions (see however Ref. [12]). The factor \( 9\pi \) in \( l_0^* \) is chosen such that \( \tilde{D}_\parallel \) and \( \tilde{D}_\perp \) are approximately 1 in the limit of an “isotropic” nematic with \( K_1 = K_2 = 1, \alpha = 0, \) and \( h = 0. \) We find \( \tilde{D}_\parallel = 1.053 \) and \( \tilde{D}_\perp = 0.998 \) with a small anisotropy of \( D_\parallel / D_\perp = 1.06 \) because of the inherent anisotropy in the nematic structure factor, which is represented by the geometrical factor \( N(e_\omega, e_\beta, u_\delta) \) of Eq. (4.21). We, at least qualitatively, understand why \( D_\parallel \) is larger than \( D_\perp \). The diffusion constants grow when the system’s ability to scatter light decreases. From equations (4.34) and (4.33) for \( D_\parallel \) and \( D_\perp, \) we recognize that the diffusion constants are, respectively, determined by scattering around the director \( (m = 0) \) or perpendicular to it \( (m = 1). \) However, forward and backward scattering along the director is suppressed by the geometrical factor and we expect \( D_\parallel \) to be larger than \( D_\perp. \) In a completely isotropic system, where we set \( N(e_\omega, e_\beta, u_\delta) \) equal to 1, the transport mean-free-path is easy to calculate. It is a factor of 4/9 smaller than \( l_0^* \), which again demonstrates the effect of the geometrical factor. For temperature \( T = 300 \) K and the parameters of the compound 5CB, which we summarized in the last subsection, we obtain \( l_0^* = 2.3 \) mm in agreement with experiments [11, 12].

In the following we will explore the dependence of the diffusion constants on the scaled parameters \( K_1, K_2, \alpha, \) and \( h. \) In Figs. [3] and [4] we plot the relative changes of the diffusion constants when spherical harmonics of \( l = 3, \) in addition to \( l = 1, \) are included in the calculations. The field dependence in Fig. [5] shows that the changes are around 1 % or smaller and that \( D_\perp \) is more strongly affected by higher spherical harmonics. The same is valid for the \( K_1 \) and \( K_2 \) dependence in Fig. [3]. Only for extreme situations \( K_1 < 0.1 \) or \( K_2 < 0.1 \) do the changes grow to 3 %. We conclude that the restriction to spherical harmonics of \( l = 1 \) gives a good approximation for the diffusion constants. The following graphs will, however, all be presented with the \( l = 3 \) contributions included.

For the nematic compound 5CB we show in Fig. [5] how the diffusion constants \( \tilde{D}_\parallel \) and \( \tilde{D}_\perp \) and the relative anisotropy \( (D_\parallel - D_\perp) / D_\parallel \) behave in a magnetic field. \( \tilde{D}_\parallel \) and \( \tilde{D}_\perp \) grow with \( H \) because the magnetic field suppresses director fluctuations. The field dependence of the relative anisotropy in the diffusion is weak. For ordinary magnetic fields up to \( 5 \times 10^4 \) G, which corresponds to a magnetic coherence length \( \xi \) of approximately 1 \( \mu \)m, the changes in \( \tilde{D}_\parallel \) and \( \tilde{D}_\perp \) are small. The values for \( H = 0 \) read \( \tilde{D}_\parallel = 0.95 \) and \( \tilde{D}_\perp = 0.65 \) with a ratio \( \tilde{D}_\parallel / \tilde{D}_\perp = 1.45 \) in good agreement with experiments [11, 12]. The reason why we plot \( \tilde{D}_\parallel, \tilde{D}_\perp \) and \( (D_\parallel - D_\perp) / D_\parallel \) on a large field range is to show that the quantities smoothly approach finite values at \( h = 0. \) This is not obvious since the structure factor possesses a singularity at \( h = 0 \) and for vanishing scattering vector \( q. \) Strictly speaking, our weak-scattering approximation is not applicable here. However, in a completely isotropic model we understand why the quantities are finite. The
familiar factor $1 - \cos \theta_\alpha$ in formula \( \theta \) cancels the singularity from $q_2^2 \propto 1 - \cos \theta_\alpha$.

In Fig. 6 we explore the anisotropy in the diffusion depending on the anisotropy $\alpha$ in the dielectric constants. As already discussed, when $\alpha = 0$ there is a small non-zero value of $(D_\parallel - D_\perp)/D_\perp$. This grows with $\alpha$ because the speed of light of the extraordinary light ray is larger along the director than perpendicular to it. On the other hand, for $\alpha < -0.15$, the anisotropy $(D_\parallel - D_\perp)/D_\perp$ changes sign, and light diffuses faster perpendicular to the director. This effect and the inversion point $D_\parallel = D_\perp$ should be observable in discotic nematics where $\alpha$ is negative.

![FIG. 5. Field dependence of the relative changes of diffusion constants after $l = 3$ spherical harmonics (superscript 13) are included in addition to $l = 1$ spherical harmonics (superscript 1). Parameters are $K_1 = K_2 = 1$ and $\alpha = 0$. The magnetic field is given relative to the characteristic field $H_0 = 3.6 \times 10^5$ G of the nematic compound 5CB.](image)

![FIG. 6. Relative difference as a function of $K_1$ and $K_2$ at $\alpha = 0$, $h = 0.1$ between $D_\parallel^{(13)}$ calculated with $l = 1$ spherical harmonics only and $D_\parallel^{(13)}$ calculated with both $l = 1$ and $l = 3$.](image)

Finally, we discuss the dependence of the diffusion on the elastic constants $K_1$ and $K_2$. In Fig. 7 we show that $D_\perp$ decreases with the elastic constants since the light scattering from the director modes increases. At the extreme values $K_1 = K_2 = 0.01$, we find $\bar{D}_\perp = 0.07$. The contour lines reveal an asymmetry between the splay ($\bar{K}_1$) and the twist ($\bar{K}_2$) distortions. $\bar{D}_\perp$ decreases more strongly with $\bar{K}_2$. The diffusion constant $\bar{D}_\parallel$ shows a similar behavior. Fig. 8 gives the anisotropy $(\bar{D}_\parallel - \bar{D}_\perp)/\bar{D}_\perp$ for the same range. It grows with decreasing elastic constants showing that $\bar{D}_\perp$ is more affected by splay and twist distortions than $\bar{D}_\parallel$. The asymmetry be-

![FIG. 7. Field dependence of the diffusion constants $\bar{D}_\parallel$ and $\bar{D}_\perp$ and the relative anisotropy $(\bar{D}_\parallel - \bar{D}_\perp)/\bar{D}_\perp$ for the nematic compound 5CB ($\overline{K}_1 = 0.79$, $\overline{K}_2 = 0.43$, $\alpha = 0.228$ and $H_0 = 3.6 \times 10^5$ G).](image)

![FIG. 8. Relative anisotropy $(\bar{D}_\parallel - \bar{D}_\perp)/\bar{D}_\perp$ depending on the dielectric anisotropy $\alpha$ for $\overline{K}_1 = \overline{K}_2 = 1$ and $h = 0.01$.](image)
 tween splay and twist is clearly visible. The last two graphs cover the range of conventional thermotropic nematics where usually $K_1 < 1$ and $K_2 < 1$. In Fig. 11 we extend this range to $K_1 = K_2 = 10$ and observe that the anisotropy $(D_\parallel - D_\perp)/D_\parallel$ changes sign. The contour line on the base of the coordinate system indicates where $D_\parallel = D_\perp$. Roughly speaking, $D_\parallel < D_\perp$ if $K_1 > 0.6$ and $K_2 > 1.4$. In a smectic-A phase (SmA) twist and bend deformations are expelled by the layered structure [54], hence $K_1 \ll 1$. Unfortunately, this also means that certain scattering vectors show very weak scattering so that the diffusion approximation cannot be achieved for reasonably sized samples. However, in the vicinity of a SmA-nematic phase transition, where the layered structure softens (SmA) or starts to form (nematic phase), the diffusion approximation of light could be used to study the behavior of the Frank elastic constants close to the transition. A second interesting system is a polymer nematic liquid crystal. For long rigid rods one expects a large splay constant [60,61]. Taratuta et al. [62] determined the Frank elastic constants for a special system and found the ratios $K_1 = 0.85$ and $K_2 = 0.07$ with an absolute value of $K_3 = 4.7 \times 10^{-7}$ dyne, which is suitable for the diffusion approximation of light. From these parameters we predict a “large” ratio of $D_\parallel/D_\perp = 2.8$. The reported system has a very low dielectric anisotropy $\alpha$ and scatters light only weakly. However, it should be possible to find systems which are more favorable regarding $\alpha$.

We also calculated a Taylor expansion for the relative anisotropy $(D_\parallel - D_\perp)/D_\perp$ around $K_1 = K_2 = 1$ and $\alpha = 0$ and found

$$
\frac{D_\parallel - D_\perp}{D_\perp} = 0.06 - 0.1(K_1 - 1) - 0.3(K_2 - 1) + 0.3\alpha
+ 0.1(K_1 - 1)^2 + 0.2(K_2 - 1)^2
- 0.08(K_1 - 1)(K_2 - 1),
$$

(4.43)

where the coefficients are material independent. Second order terms in $\alpha$ and couplings to $K_1$ and $K_2$ are negligible. The expansion summarizes the whole discussion.

D. Diffusing Wave Spectroscopy

With Diffusing Wave Spectroscopy it is possible to measure the averaged dynamical properties of a system through the dynamic absorption coefficient $\mu(\omega, t)$ of Eq. (4.20). Director modes are purely diffusive, as described by Eq. (4.20) for the structure factor, and possess a viscosity $\eta(q_s)$ which we specify here [54]:

$$
\eta_1(q_s) = \gamma - \frac{(\mu_3 q_\perp^2 - \mu_2 q_\parallel^2)^2}{\eta_0 q_\perp^4 + \eta_1 q_\perp^2 q_\perp^2 + \eta_0 q_\parallel^4}
$$

(4.44a)

$$
\eta_2(q_s) = \gamma - \frac{\mu_2 q_\parallel^2}{\eta_0 q_\parallel^2 + \eta_0 q_\parallel^2}
$$

(4.44b)

FIG. 9. Diffusion constant $D_\perp$ depending on $K_1$ and $K_2$.
Parameters are $\alpha = 0$ and $h = 0.1$.

FIG. 10. Relative anisotropy $(D_\parallel - D_\perp)/D_\parallel$ depending on $K_1$ and $K_2$, which range from 0.01 to 1. Parameters are $\alpha = 0$ and $h = 0.1$.

FIG. 11. Relative anisotropy $(D_\parallel - D_\perp)/D_\parallel$ depending on $K_1$ and $K_2$, which range from 0.1 to 10. Parameters are $\alpha = 0$ and $h = 0.1$. 

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with
\[ \eta_a = \mu_4/2 \]  
(4.45a)
\[ \eta_b = (-\mu_2 + \mu_4 + \mu_5)/2 \]  
(4.45b)
\[ \eta_c = (\mu_3 + \mu_4 + \mu_6)/2 \]  
(4.45c)
\[ \eta_m = \mu_1 + \eta_b + \eta_c . \]  
(4.45d)

The Leslie viscosities \( \mu_4 \) govern the viscous flow of the fluid and couple it to the director motion. The Miesowicz viscosities \( \eta_a, \eta_b, \) and \( \eta_c \) can be measured in pure flow experiments. The rotational viscosity \( \gamma \) characterizes viscous forces due to rotations of the director. With the explicit formula for the structure factor and small enough times to expand the exponential time factor, \( \mu(\omega, t) \) becomes proportional to time \( t \),
\[ \mu(\omega, t) \approx (\Delta c)^2 k_B T \frac{\pi^3 \omega^4}{2n^2 c_s^2} \sum_{\alpha, \beta, \delta} \int k^2 \int q^3 \frac{N(e_{\alpha}, e_{\beta}, \eta \delta(q_s))}{\eta \beta(q_s)} t . \]  
(4.46)

If we collect all the prefactors, the dynamic absorption coefficient can be written as
\[ \mu(\omega, t) = \mu_0 t \]  
(4.47)
with \( \mu_0 = \alpha^2 \frac{2 k_B T \omega^4}{9 \pi} \frac{\bar{\mu}}{c_\perp} \gamma . \]

Here the numerical factor \( \bar{\mu} \) is a dimensionless angular average involving the geometrical factor and the viscosities of the director modes. It depends on the Leslie viscosities relative to \( \gamma \) and the dielectric anisotropy \( \alpha \), and it is equal 1 if \( \eta_s(q_s) = \gamma \) and \( \alpha = 0. \) When \( \eta_s(q_s) = \gamma \), \( \bar{\mu} \) can be evaluated analytically even when \( \alpha \neq 0 \), and we find \( \bar{\mu} = \frac{1 + \alpha/4}{1 + \alpha/2} \). For the compound 5CB, \( \gamma/\bar{\mu} = 0.60 \pm 0.20 \) poise was experimentally determined by Kao et al. using DWS and the last formula. This value is in good agreement with the rotational viscosity \( \gamma = 0.81 \) poise of 5CB and shows the validity of the theory. Of course \( \gamma \) is larger than the measured \( \gamma/\bar{\mu} \) since \( \eta_s(q_s) \) is smaller than \( \gamma \) [see Eqs. (4.37)] so that \( \bar{\mu} \) exceeds 1. However, the values of the Leslie viscosities are such that \( \bar{\mu} \) is of order 1 in usual thermotropic nematics. Furthermore, the Leslie-Erickson theory seems to describe the director modes properly down to \( 4 \times 10^{-8} \) s, the time resolution of the experiments, since there is no indication for a deviation from the linear time dependence of \( \mu(\omega, t) \) predicted by the diffusive director modes. Materials where the viscosities \( \eta_s(q_s) \) for different values of \( q_s \) differ by factors of \( 10^2 \) to \( 10^3 \) are polymer nematic liquid crystals [12] with some director modes relaxing on a much larger time scale than they do in ordinary nematics. It would be interesting to study such systems to see if they exhibit a deviation from the Leslie-Erickson theory for short times which, e.g., would show up in a different temporal power law for \( \mu(\omega, t) \).

Finally, we point out an important difference between nematic liquid crystals and colloidal suspensions. The dynamic absorption coefficient only contains the viscosities of the director modes. The Frank elastic constants cancel because they determine both the light scattering and the dynamics. On the other hand, in colloidal suspensions \( 19 \), \( \mu_0 = 2D_B g \omega^2/(l^*)^2 \), where the transport mean-free-path \( l^* \) characterizes light propagation and the diffusion constant \( D_B \) the Brownian motion of the colloidal particles.

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**APPENDIX A: THE BETHE-SALPETER EQUATION**

The averaged two-particle Green function with all space-time coordinates looks like
\[ \Phi(x_1, y_1, \tau_1; x_2, y_2, \tau_2) = (G R(x_1, x_2; \tau_1, \tau_2) \otimes G A(y_2, y_1; \tau_2, \tau_1))^N . \]  
(A1)

After an integration over electric field sources \( W_0(2) \) at points \( (x_2, \tau_2) \) and \( (y_2, \tau_2) \) it provides us with the full autocorrelation function \( W(1) \) of the electric light field at \( (x_1, \tau_1) \) and \( (y_1, \tau_1) \):
\[ W(1) = \int d2 \Phi(1, 2) W_0(2) . \]  
(A2)

In the following we abbreviate a pair of points in space time by its index \( i \). The two-particle Green function follows from the Bethe-Salpeter equation [50]
\[ \Phi(1, 4) = f(1, 4) + \int d2 d3 f(1, 2) U(2, 3) \Phi(3, 4) . \]  
(A3)

The quantity \( f(1, 2) \) stands for the tensor product of the averaged one-particle Green functions:
\[ f(1, 2) = [(G R)(x_1 - x_2; \tau_1 - \tau_2) \otimes (G A)(y_2 - y_1; \tau_2 - \tau_1)]^N . \]  
(A4)

It propagates two electric field modes from 2 to 1 between their scattering events. The irreducible vertex function \( U \) describes different characteristic sets of scattering events [50]. We will soon specify it. Note, that there is no preferred point in time and that the scattering medium on average is homogeneous in space. Therefore, all our averaged quantities do not change under translations in space time, and hence they can only depend on differences of the coordinates. We now introduce center-of-“mass” \((R_i, T_i)\) and relative \((r_i, t_i)\) coordinates:
\[ x_i = R_i + r_i/2 \quad y_i = R_i - r_i/2 \]
\[ \tau_i = T_i + t_i/2 \quad \bar{\tau}_i = T_i - t_i/2 . \]
\[ (A5) \]

It is straightforward to show that the Jacobian determinant for this coordinate transformation is 1, and we have
\[ di = d^3R_i \; d^3r_i \; dT dt . \]
\[ (A6) \]

All of our quantities only depend on differences in the center-of-"mass" coordinates \( R_i \) and \( T_i \) because of the homogeneity in space-time. To discuss the Bethe-Salpeter equation we perform a Fourier transformation,
\[ \int d^3(R_i - R_j) \; d^3(T_i - T_j) \; d^3r_i \; d^3r_j \; dt_i \; dt_j \]
\[ \times \exp[-i(\mathbf{K} \cdot (R_i - R_j) + k_i \cdot r_i - k_j \cdot r_j)] \]
\[ \times \exp[i(\Omega(T_i - T_j) + \omega_i t_i - \omega_j t_j)] \]
\[ (A7) \]

which transforms our quantities as follows:
\[ \Phi(1, 4) \rightarrow \Phi_{\omega_1 \omega_2}(K, \Omega) \]
\[ U(2, 3) \rightarrow U_{\omega_2 \omega_3}(K, \Omega) \]
\[ f(1, 2) \rightarrow f_{\omega_1 \omega_2}(K, \Omega) \delta_{\omega_1 \omega_2} 1^{(4)}_{k_1 k_2} , \]
\[ (A8) \]

where \( \omega_1, \omega_2 = 2\pi \delta(\omega_1 - \omega_2) \), and \( 1^{(4)}_{k_1 k_2} \), defined in Eq. \((3.29)\), contains \( \delta(k_1 - k_2) \). The delta functions appear since \( f(1, 2) \) only depends on the differences of the relative coordinates. In Eq. \((3.28)\) we give the explicit form of \( f_{k_1}^{(4)}(K, \Omega) \). The Fourier transformed Bethe-Salpeter equation finally takes the form
\[ \int \frac{d^3 k_1}{(2\pi)^3} \frac{d\omega_1}{2\pi} \left[ 1^{(4)}_{k k k} \delta_{\omega_1} - f_{k}^{(4)}(K, \Omega) U_{\omega k k}(K, \Omega) \right] \]
\[ \times \Phi_{\omega \omega')(K, \Omega) = f_{k}^{(4)}(K, \Omega) 1^{(4)}_{k k k} \delta_{\omega \omega'} \]
\[ (A9) \]

and the autocorrelation function \( W(K, k, \Omega, \omega) \) for light with frequency \( \omega \) and wave vector \( k \) follows from
\[ W(K, k, \Omega, \omega) = \int \frac{d^3 k'}{(2\pi)^3} \frac{d\omega'}{2\pi} \Phi_{\omega \omega'}(K, \Omega) \]
\[ \times W_{\omega}(K, k', \Omega, \omega') . \]
\[ (A10) \]

So far, our manipulations are generally valid. Now we introduce some approximations. We use the weak-scattering approximation in which the irreducible vertex function is given by
\[ \Phi_{\omega}(k, \Omega) \]
\[ (A11) \]

\[ \delta \varepsilon(x_1, \tau_1) \frac{1}{c^2} \frac{\partial^2}{\partial \tau_1^2} \delta(\tau_1 - \tau_2) \delta(y_1 - y_2) \]
\[ (A12) \]

It only considers scattering events of the two electric field modes which are tied together through the structure factor. Fig. 12 gives a graphic representation of \( U(1, 2) \).

\[ \Phi_{\omega}(k, \Omega) = \int \frac{d^3(\omega - \omega')}{c^2} \delta \varepsilon(x_1, \tau_1) e^{-i(\omega - \omega')t} , \]
\[ (A13) \]

where the time derivative of a delta function was handled with its representation via plane waves. Since the temporal correlations in the dielectric tensor decay on a time scale much longer than the time period of light, \( U_{\omega}(k, \Omega) \) is strongly peaked around \( \omega = \omega' \). The Bethe-Salpeter equation \((A9)\) then implies the same behavior for \( \Phi_{\omega}(k, \Omega) \), i.e., \( \Phi_{\omega}(k, \Omega) \propto p(\omega - \omega') g(\omega) \), where \( p(\omega - \omega') \) stands for the strongly peaked part around \( \omega = \omega' \) and \( g(\omega) \) for the remaining smooth function in \( \omega' \). The Green function \( \Phi_{\omega}(k, \Omega) \) picks up a source term of frequency \( \omega' \) and produces an autocorrelation function with frequencies \( \omega \) centered narrowly around \( \omega' \). In the time domain this corresponds to \( \exp(i\omega \tau) \) times a slowly varying factor in \( t \). It is therefore appropriate to introduce the Green function
\[ \Phi_{\omega}(k, \Omega, \omega') = \int d^3(\omega - \omega') \Phi_{\omega}(k, \Omega) e^{-i(\omega - \omega')t} , \]
\[ (A14) \]

which gives this factor for light sources of frequency \( \omega' \). If we multiply Eq. \((A9)\) by \( \exp[-i(\omega - \omega')t] \), rewrite the argument of \( \delta \varepsilon \delta \varepsilon^{(N)} \) as \( \omega - \omega_1 = \omega - \omega' - (\omega_1 - \omega') \), and integrate over \( t \) we finally arrive at the Bethe-Salpeter equation of subsection III.1 for \( \Phi_{\omega}(k, \Omega) \):
\[ \int \frac{d^3 k}{(2\pi)^3} [1^{(4)}_{k k_1}] - f_k'(K, \Omega) B^{\prime}_{k k_1}(t) \Phi_{k k_1}(K, \Omega, t) = f_k'(K, \Omega) 1^{(4)}_{k k_1} \]  
with

\[ B^{\prime}_{k k_1}(t) = \frac{(\omega')^4}{e^4} \langle \delta \varepsilon \otimes \delta \varepsilon \rangle^{(N)}(k - k_1, t) . \]

In deriving the last equation we replaced the argument \( \omega \) in \( f_k'(K, \Omega) \) in Eq. (A3) by \( \omega' \) and the factor \( \omega_1^4 \) in \( U_{\omega \omega_1}^{(4)}(K, \Omega) \) by \( (\omega')^4 \).

**APPENDIX B: TWO IDENTITIES**

1. **Ward Identity**

The Ward identity establishes a linear relation between the irreducible vertex function \( U \) and the mass operator. In the weak-scattering approximation it says

\[ \Delta \Sigma_k^\alpha(K, 0) = \int \frac{d^3 k'}{(2\pi)^3} B^{\alpha *}_{k k'}(t = 0) \Delta G^{\alpha}_{k'}(K, 0) , \]  

which can be proven by a variable transformation of the integrand. For the general case see Vollhardt and Wölfle [1].

2. **A useful identity**

In this subsection we derive a useful relation between \( \Delta G_{k}^{\alpha}(K, \Omega) \), \( f_{k}^{\alpha}(K, \Omega) \), and \( \Delta \Sigma_{k}^{\alpha}(K, \Omega) \). We start with the definition

\[ \Delta G_{k}^{\alpha}(K, \Omega) = \langle G^{R}\rangle(k_+, \omega_+) - \langle G^{A}\rangle(k_-, \omega_-) \]  
and insert the unit tensor \( 1 \) in the appropriate representation:

\[ \Delta G_{k}^{\alpha}(K, \Omega) = \langle G^{R}\rangle(k_+, \omega_+) \left( \langle G^{A}(k_-, \omega_-) \right) - \left( \langle G^{R}(k_+, \omega_+) \right) \langle G^{A}(k_-, \omega_-) \right) . \]  

Then we introduce \( \langle G^{R/A}(k_+, \omega_+) \) from Eq. (B.1), use definitions (B.12) and (B.28) for \( \Delta \Sigma_{k}^{\alpha}(K, \Omega) \) and \( f_{k}^{\alpha}(K, \Omega) \), and arrive at the final equation:

\[ \Delta G_{k}^{\alpha}(K, \Omega) = f_{k}^{\alpha}(K, \Omega) \left[ \Delta \Sigma_{k}^{\alpha}(K, \Omega) \right. 
- \left. \{ G_{0}^{-1}(k_+, \omega_+) - G_{0}^{-1}(k_-, \omega_-) \} \right] . \]  

In the case \( K \to 0, \Omega \to 0 \), we get to first order in \( K \) and \( \Omega \):

\[ \Delta G_{k}^{\alpha}(K, \Omega) \approx f_{k}^{\alpha}(0, 0) \left[ \Delta \Sigma_{k}^{\alpha}(0, 0) \right. 
- \left. \frac{\partial G_{0}^{-1}}{\partial k} - K - \frac{\partial G_{0}^{-1}}{\partial \omega} \Omega \right] , \]

where we neglect first-order terms from \( f_{k}^{\alpha} \) and \( \Delta \Sigma_{k}^{\alpha} \) because the components of \( \Delta \Sigma_{k}^{\alpha} \) are already much smaller than one. The last equation shows that the leading order in \( K \) and \( \Omega \) comes solely from \( G_{0}^{-1}(k, \omega) \).

In subsection [II A] we calculated the Green functions \( \langle G^{R/A}(k, \omega) \) to first order in \( \Sigma^{R/A}(k, \omega) \). They were diagonal and only involved the diagonal elements of \( \Sigma^{R/A}(k, \omega) \). Let us look at Eq. (B.5) under this approximation concentrating on the propagating part of our quantities only. As explained in the main text a Greek superscript or subscript corresponds, respectively, to the basis vector \( e_{\alpha}(\hat{k}) \otimes e_{\alpha} \) or \( d^\alpha(\hat{k}) \otimes d^\alpha \). For \( f_{k}^{\alpha}(0, 0) \) we find

\[ f_{k}^{\alpha}(0, 0) = \left[ \langle G^{R}\rangle(k, \omega) \otimes \langle G^{A}\rangle(k, \omega) \right]^{(N)} \approx \sum_{\alpha, \beta} \left[ \langle G^{R}\rangle(k, \omega) \right]^\alpha \left[ \langle G^{A}\rangle(k, \omega) \right]^\beta \]  

\[ e_{\alpha}(\hat{k}) \otimes e_{\beta}(\hat{k}) \otimes e_{\alpha}(\hat{k}) \otimes e_{\beta}(\hat{k}) . \]  

Terms like \( e_1 \otimes e_2 \otimes e_3 \otimes e_4 \) appear because the subscript \( (N) \) tells us to interchange the second and third basis vector in \( \langle G^{R} \rangle \otimes \langle G^{A} \rangle \). They only couple non-diagonal components to each other. Since non-diagonal elements are beyond our approximation we don’t have to consider them. Then we are able to write

\[ \left[ f_{k}^{\alpha}(0, 0) \right]^{(N)} = \left\{ \begin{array}{cc} \left[ \langle G^{R}\rangle(k, \omega) \right]^\alpha \left[ \langle G^{A}\rangle(k, \omega) \right]^\alpha, & \alpha = \beta \\
0, & \alpha \neq \beta \end{array} \right. \]  

The component 0 refers to terms like \( e_1 \otimes e_1 \otimes e_2 \otimes e_2 \). The derivation of \( G_{0}^{-1} \) with respect to \( \omega \) just gives

\[ \frac{\partial G_{0}^{-1}}{\partial \omega} \Omega = \frac{2\omega}{e^4} \Omega . \]  

The derivation with respect to \( k \) contains two contributions:

\[ \frac{\partial G_{0}^{-1}}{\partial k} = \sum_{\alpha} \left[ \frac{\partial G_{0}^{-1}}{\partial k} d_{\alpha}(\hat{k}) \otimes d_{\alpha}(\hat{k}) \right] + \left[ G_{0}^{-1} \right]^{\alpha} \frac{\partial}{\partial k} \left[ d^\alpha(\hat{k}) \otimes d^\alpha(\hat{k}) \right] . \]  

The second one only produces non-diagonal elements which we do not have to consider. This statement seems to be obvious because a small rotation of \( k \) rotates \( d_{\alpha}(\hat{k}) \). But our basis vectors are more general and we have to look at it more carefully. We have to show that \( e_{i}(\hat{k}) \cdot \frac{\partial}{\partial k_i} d^i(\hat{k}) \) is zero. With \( d^i = \varepsilon_0 e_i \) and \( \varepsilon_0 \) being symmetric and independent of \( k \) we can write

\[ e_{i}(\hat{k}) \cdot \frac{\partial}{\partial k_i} d^i(\hat{k}) = -e_{i}(\hat{k}) \cdot \frac{\partial}{\partial k_i} e_{i}(\hat{k}) . \]  

From the biorthogonality relation it is clear that

\[ d^i(\hat{k}) \cdot \frac{\partial}{\partial k_i} e_{i}(\hat{k}) = -e_{i}(\hat{k}) \cdot \frac{\partial}{\partial k_i} d^i(\hat{k}) . \]
which verifies the statement. We are now able to write down Eq. (B3) within our approximation:

\[
[\Delta G_k^\omega (K, \Omega)]^\alpha \approx [f_k^\omega (0, 0)]^{\alpha \alpha} \left( [\Delta \Sigma_k^\omega (0, 0)]_\alpha 
- \frac{\partial [G_0^{-1}]}{\partial k} \cdot \mathbf{K} - \frac{2\omega}{c^2} \right). \quad (B12)
\]
APPENDIX C: STRUCTURE FACTOR

We give the two important structure factors in scaled form and in the coordinates $C_k, S_k, C_q, S_q$ and $\varphi$. From the notation it is clear whether they belong to an extraordinary or ordinary light ray. The parameters are $\alpha, K_1, K_2$ and $h$.

\[
[B_{k, q}^\omega(0)]_{12} = \frac{1}{(1+\alpha)Q_1^2} \left[ \frac{S_k^2 \sin^2 \varphi}{\bar{K}_1 Q_1^2 + Q_1^2 + h^2} + \frac{(S_k \cos \varphi - S_q)^2}{\bar{K}_2 Q_1^2 + Q_1^2 + h^2} \right]
\]

\[
[B_{k, q}^\omega(0)]_{11} = \frac{1}{(1+\alpha)Q_1^2} \left[ \frac{\cos^2 \varphi N_1 + 2\cos \varphi N_2 + N_3}{\bar{K}_1 Q_1^2 + Q_1^2 + h^2} + \frac{\sin^2 \varphi N_4}{\bar{K}_2 Q_1^2 + Q_1^2 + h^2} \right]
\]

with

\[
Q_1^2 = (C_k - C_q)^2, \quad Q_2^2 = S_k^2 - 2S_k S_q \cos \varphi + S_q^2
\]

and

\[
N_1 = (S_k^2 C_q - S_q^2 C_k)^2, \quad N_2 = S_k S_q (S_k^2 C_q - S_q^2 C_k) (C_k - C_q)
\]

\[
N_3 = S_k^2 S_q^2 (C_k - C_q)^2, \quad N_4 = (S_k^2 C_q + S_q^2 C_k)^2
\]

The integration over $\varphi$ gives:

\[
\int [B_{k, q}^\omega(0)]_{12} d\varphi = \frac{\pi}{2} \frac{S_k}{S_q^2} \left[ \frac{C(h) + 2\bar{K}_2(S_k^2 - S_q^2)}{K_2^2} - C(h) \frac{2\bar{K}_2(S_k^2 + S_q^2)}{K_1^2} \right]
\]

\[
\int \cos \varphi [B_{k, q}^\omega(0)]_{12} d\varphi = \frac{\pi}{4} \frac{S_k}{S_q^2} \left[ \left( \frac{S_k^2}{S_q^2} C_q + \frac{S_q^2}{S_k^2} C_k \right) \left( \frac{1}{K_1} \frac{1}{K_2} \right) - 2C_k C_q \left( \frac{1}{K_1} + \frac{1}{K_2} \right) \right. \]

\[
+ \frac{1}{C(h)} \left( \frac{S_k}{S_q} C_q + \frac{S_q}{S_k} C_k \right)^2 \left( \frac{1}{K_2} \frac{1}{K_1} \right) - \frac{C(h) + \bar{K}_1(S_k^2 + S_q^2) + \bar{K}_2(S_k^2 + S_q^2)}{I(\bar{K}_2)} \right]
\]

\[
\int [B_{k, q}^\omega(0)]_{11} d\varphi = \frac{\pi}{2} \left[ \left( \frac{S_k^2}{S_q^2} C_q - \frac{S_q^2}{S_k^2} C_k \right)^2 \left( \frac{1}{K_1} \frac{1}{K_2} \right) - 2C_k C_q \left( \frac{1}{K_1} + \frac{1}{K_2} \right) \right. \]

\[
+ \frac{1}{C(h)} \left( \frac{S_k}{S_q} C_q + \frac{S_q}{S_k} C_k \right)^2 \left( \frac{1}{K_2} \frac{1}{K_1} \right) + \frac{4S_k^2}{K_2} \frac{I(\bar{K}_1)}{K_1} \left( \frac{K_1(S_k^2 C_q + S_q^2 C_k)(C_k + C_q) + C(h)C_k C_q}{K_1} \right) \left] \right)
\]

\[
\int \cos \varphi [B_{k, q}^\omega(0)]_{11} d\varphi = \frac{\pi}{4} \frac{1}{S_k S_q^3} \left[ \frac{1}{K_1} \left( \frac{S_k^2 C_q - S_q^2 C_k}{S_q} \right)^2 \left( \frac{1}{K_1} \frac{1}{K_2} \right) + 2\bar{K}_1(S_k^4 C_q^2 - S_q^4 C_k^2)(S_k^2 - S_q^2) \right]
\]

\[
- \frac{1}{K_2} \left( S_k^2 C_q + S_q^2 C_k \right)^2 \left( \frac{C(h) + 2\bar{K}_2(S_k^2 + S_q^2)}{K_2^2} \right)
\]

\[
+ \frac{1}{C(h)} \left( S_k^2 C_q + S_q^2 C_k \right)^2 \left( \frac{C(h) + \bar{K}_1(S_k^2 + S_q^2) + \bar{K}_2(S_k^2 + S_q^2)}{K_2^2 I(\bar{K}_2)} \right)
\]

\[
+ 4S_k^2 S_q^2 I(\bar{K}_1) \left( \frac{C(h) + \bar{K}_1(S_k^2 + S_q^2) + \bar{K}_2(S_k^2 + S_q^2)}{I(\bar{K}_1)} \right) \left( K_1(S_k^2 C_q + S_q^2 C_k)(C_k + C_q) + C(h)C_k C_q \right) \]

with

\[
C(h) = (C_k - C_q)^2 + h^2 \quad \text{and} \quad I(\bar{K}_1) = \left\{ \left[ K_1(S_k - S_q)^2 + C(h) \right] \left[ K_1(S_k + S_q)^2 + C(h) \right] \right\}^{-1/2}.
\]
The structure factor \( [\mathcal{B}_{k_1q_1}^\omega(0)]_{11} \) diverges for \( h = 0 \) and \( q_1 \rightarrow k_1 \). We give \( [\mathcal{B}_{k_1q_1}^\omega(0)]_{11} \) for small \( \varphi \) and \( \delta C = C_k - C_q \):

\[
[\mathcal{B}_{k_1q_1}^\omega(0)]_{11} = \frac{1}{(1 + \alpha)^2 (1 + \alpha)^2 C_k^2 \delta C^2 / S_k^2 + S_k^2 \varphi^2}
\times \left[ \frac{(1 + \alpha)^2 C_k^2 \delta C^2}{K_1(1 + \alpha)^2 C_k^2 / S_k^2 + 1} \delta C^2 + K_1 S_k^2 \varphi^2 + h^2 \right]^{1/2} + \frac{S_k^2 C_k^2 \varphi^2}{[K_2(1 + \alpha)^2 C_k^2 / S_k^2 + 1] \delta C^2 + K_2 S_k^2 \varphi^2 + h^2}.
\]

\[\tag{C6}\]

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