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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. [S1063-651X(97)12301-7]

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I. INTRODUCTION

Dynamic light scattering (DLS) is one of our most powerful probes [1–3] of the dynamics of materials such as 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 $(qv)^{-1}$ 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 [4]. 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 $l^* = l/\langle 1-\cos\vartheta_s\rangle$, measuring the distance beyond which the direction of propagation is randomized, where ϑ_s is the scattering angle and the angular brackets denote 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 $D=\overline{c}\,\overline{l}^*/3$, where $\overline{c}$ is the speed of light in the medium.

There has been a resurgence of interest in light transport in turbid and random media [5] because of its close connection to the problem of Anderson localization [6] and electron transport in disordered systems [7,8] and because of the development of diffusing wave spectroscopy (DWS) [9–15], 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 [16–22] and discussed in a number of theoretical papers [13,23–28]. DWS has opened up a whole new field of study. It provides heretofore unobtainable information about the dynamics of turbid media, including dense colloids [12], sheared suspensions [29], emulsions [30], and foams [31]. 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 [32] and the measurement of shape fluctuation modes in tense emulsion droplets [30]. Photon diffusion and DWS have also found application in imaging of objects such as tumors in human tissue [33].

With few exceptions [34–37], both theory and experiment have focused 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 [38,39] 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 [34,35], its purpose is to broaden the class of materials to which DWS and its offshoot applications such as 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 Refs. [40,41]. An alternative derivation of the results reported here and a more detailed account of experiments appear in Ref. [42]. A similar treatment of diffusive light transport and DWS was developed by Tiggelen, Maynard, and Heiderich [43].

Anisotropic media differ from isotropic media in two important ways: (1) The speed of light depends in general on both the polarization and direction of light propagation rela-

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tive to anisotropy axes, and (2) scattering cross sections depend not only on the relative direction of incoming and scattered light rays but also on their direction relative to anisotropy axes. Diffusive transport in optically active isotropic media with light speeds depending on the state of circular polarization has been studied [28]. Tiggelen has investigated anisotropic light diffusion induced by a magnetic field H through a series expansion in H [37]. To our knowledge, however, no thorough treatment of multiple light scattering in optically anisotropic media has been published. For electronic systems anisotropic diffusion has been studied both theoretically and experimentally in the context of localization [44,45].

Optical anisotropy leads to anisotropic diffusive light transport. The equation governing the electric-field autocorrelation function $W(\boldsymbol{R},\boldsymbol{T},t)=\langle \boldsymbol{E}(\boldsymbol{R},T+t/2)\cdot\boldsymbol{\varepsilon}_0\boldsymbol{E}(\boldsymbol{R},T-t/2)\rangle$ ($\boldsymbol{\varepsilon}_0$ denotes the dielectric tensor) is

$$\left[\frac{\partial}{\partial T}-\nabla\cdot\boldsymbol{D}\nabla+\mu(t)\right]W(\boldsymbol{R},T,t)=\varrho(\boldsymbol{R},T), \tag{1}$$

where $\boldsymbol{D}$ stands for the anisotropic diffusion tensor. The quantity $\mu(t)$ is the dynamic absorption coefficient measured in DWS experiments [9–15]. It results from an average of short-time dynamic correlations over angle and polarization. We will provide explicit formulas for $\boldsymbol{D}$ and $\mu(t)$ for general anisotropic systems and then concentrate on nematic liquid crystals as one example of a uniaxial system. With the preferred axis along the unit vector $\boldsymbol{n}$, the diffusion tensor $\boldsymbol{D}$ reduces to $\boldsymbol{D}=D_\perp\mathbf{1}+(D_\parallel-D_\perp)\boldsymbol{n}\otimes\boldsymbol{n}$ where $\mathbf{1}$ is the unit element. The dynamic absorption coefficient $\mu(t)$ will turn out to be the angular average of an inverse viscosity.

In this paper, we will restrict ourselves to the weak-scattering limit, and we will treat multiple scattering via the Bethe-Salpeter equation in Sec. III. In isotropic lossless systems, the diffusion equation can be obtained exactly from the Bethe-Salpeter equation by considering only modes associated with the isotropic and ''vector'' spherical harmonics Y_{00} and Y_{1m}. In anisotropic systems, all spherical harmonics couple, and the calculation of diffusion coefficients involves the inversion of an infinite dimensional matrix, which can only be accomplished approximately. We will, therefore, be content with a formal expression for the diffusion tensor in general anisotropic media. We will, however, introduce a sequence of approximations to obtain numerical values for the diffusion constants in nematic liquid crystals. Fortunately, the first term in this sequence undergoes only a very small modification in going to the second in this sequence.

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 $\boldsymbol{n}(\boldsymbol{r})$. Fluctuations in $\boldsymbol{n}(\boldsymbol{r})$ diverge as q^{-2} at small wave number q in the absence of an external aligning magnetic field H. This divergence leads to a vanishing scattering mean free path for extraordinary to extraordinary scattering in the limit $H\to 0$. The diffusion constants are nonetheless well defined and nonzero. For, if l tends to zero, scattering takes place almost entirely in the forward direction. Thus, light has to undergo a large number of scattering events before directional information is lost, and, as a result, l^* is finite.

The outline of this paper is as follows. In Sec. II, we 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 $\boldsymbol{B}^\omega(\boldsymbol{r},t)$ to describe fluctuations in the dielectric 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 approximation scheme for the diffusion tensor and look at the isotropic limit of our theory. Comments on DWS close Sec. III. Section IV applies the general results of the preceding sections to nematic liquid crystals. A review of relevant properties of nematic liquid crystals and light propagation in uniaxial media is followed by an explanation of dielectric tensor fluctuations in nematics. Finally, we discuss diffusive light transport and DWS in nematics. In particular, we provide explicit numerical calculations of the diffusion coefficients $D_\parallel$ and $D_\perp$ as a function of Frank elastic constants, dielectric anisotropy, and external magnetic field, and point out their relevance for different liquid crystalline systems, such as discotic nematics, smectic-A phases, and polymer liquid crystals. The numerical calculations are summarized in Figs. 7–11. They are in excellent agreement with recent experiments on the nematic compound 5CB by Jester, Kao, and Yodh [41,42]. At the end we address the dynamic absorption coefficient.

II. LIGHT PROPAGATION IN A HOMOGENEOUS MEDIUM WITH DIELECTRIC ANISOTROPY

Light propagation in anisotropic dielectric media is more complicated than it is in isotropic systems. In particular, 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 propagation 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,

$$\mathrm{div}\boldsymbol{D}=4\pi\varrho_{\mathrm{ma}},\quad \mathrm{div}\boldsymbol{B}=0,$$

$$\mathrm{curl}\boldsymbol{E}=-\frac{1}{c}\frac{\partial\boldsymbol{B}}{\partial t},\quad \mathrm{curl}\boldsymbol{H}=\frac{4\pi}{c}\boldsymbol{j}_{\mathrm{ma}}+\frac{1}{c}\frac{\partial\boldsymbol{D}}{\partial t}, \tag{2}$$

where ϱ_{ma} and $\boldsymbol{j}_{\mathrm{ma}}$ are, respectively, the macroscopic charge and current densities. We concentrate on a dielectric medium with

$$\boldsymbol{D}=\boldsymbol{\varepsilon}_0\boldsymbol{E}\quad\text{and}\quad\boldsymbol{B}=\boldsymbol{H}, \tag{3}$$

and we assume that the dielectric tensor $\boldsymbol{\varepsilon}_0$ is real and does not depend on time. Then the energy-balance equation reads

$$\frac{\partial}{\partial t}u+\mathrm{div}\boldsymbol{S}=\boldsymbol{j}_{\mathrm{ma}}\cdot\boldsymbol{E}, \tag{4}$$

where we have introduced the energy density

$$u=\frac{1}{8\pi}(\boldsymbol{E}\cdot\boldsymbol{\varepsilon}_0\boldsymbol{E}+\boldsymbol{H}\cdot\boldsymbol{H}) \tag{5}$$

and the Poynting vector

$$\boldsymbol{S}=\frac{c}{4\pi}\boldsymbol{E}\times\boldsymbol{H}. \tag{6}$$

Both quantities strongly vary in space and time. If we average over one period of oscillation, we obtain their averaged values $\overline{u}$ and $\overline{\boldsymbol{S}}$. In the following we use complex waves $\boldsymbol{E}=\boldsymbol{E}_0 e^{-i\omega t}$ and $\boldsymbol{H}=\boldsymbol{H}_0 e^{-i\omega t}$ whose averaged energy densities and Poynting vectors are given by $\overline{u}=(\boldsymbol{E}_0\cdot\boldsymbol{\varepsilon}\boldsymbol{E}_0+\boldsymbol{H}_0\cdot\boldsymbol{H}_0)/(16\pi)$ and $\overline{\boldsymbol{S}}=c(\boldsymbol{E}_0\times\boldsymbol{H}_0)/(8\pi)$.

For vanishing sources ϱ_{ma} and $\boldsymbol{j}_{\mathrm{ma}}$, we obtain the homogeneous wave equation for the electric light field $\boldsymbol{E}(\boldsymbol{r},t)$:

$$\left[\mathrm{curl}\;\mathrm{curl}+\frac{\boldsymbol{\varepsilon}_0}{c^2}\frac{\partial^2}{\partial t^2}\right]\boldsymbol{E}(\boldsymbol{r},t)=\boldsymbol{0}. \tag{7}$$

Note that all solutions of Eq. (7) have to fulfill the transversality condition for dielectric field waves, $\mathrm{div}\boldsymbol{\varepsilon}_0\boldsymbol{E}=0$, unless the electric field is static or linear in t. Introduction of the plane-wave ansatz

$$\boldsymbol{E}(\boldsymbol{r},t)=E_0\boldsymbol{e}(\hat{\boldsymbol{k}})\exp[i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)] \tag{8}$$

in wave equation (7) leads to a generalized eigenvalue problem

$$\left[\boldsymbol{P}_t(\hat{\boldsymbol{k}})-\frac{1}{n^2}\boldsymbol{\varepsilon}_0\right]\boldsymbol{e}(\hat{\boldsymbol{k}})=\boldsymbol{0}, \tag{9}$$

where

$$\frac{1}{n^2}=\frac{\omega^2}{c^2k^2} \tag{10}$$

and

$$\boldsymbol{P}_t(\hat{\boldsymbol{k}})=\boldsymbol{1}-\hat{\boldsymbol{k}}\otimes\hat{\boldsymbol{k}} \tag{11}$$

is the projection operator on the space perpendicular to the unit vector $\hat{\boldsymbol{k}}=\boldsymbol{k}/k$. The symbol ⊗ stands for the tensor product: $[\hat{\boldsymbol{k}}\otimes\hat{\boldsymbol{k}}]_{ij}=\hat{k}_i\hat{k}_j$. The solutions to Eq. (9) provide us with the characteristic light modes of the system determined by the refractive index $n_i(\hat{\boldsymbol{k}})=ck/\omega$ and the polarization vector $\boldsymbol{e}_i(\hat{\boldsymbol{k}})$ of the electric field. We define the polarization vector of the displacement field by

$$\boldsymbol{d}^i(\hat{\boldsymbol{k}})=\boldsymbol{\varepsilon}_0\boldsymbol{e}_i(\hat{\boldsymbol{k}}). \tag{12}$$

For each direction $\hat{\boldsymbol{k}}$, there exist two characteristic light modes with associated polarizations $\boldsymbol{d}^i(\hat{\boldsymbol{k}})\perp\hat{\boldsymbol{k}}$. When a plane wave with frequency ω enters the anisotropic medium, it splits into the two characteristic modes that travel with different speeds $c/n_i(\hat{\boldsymbol{k}})$ and wave numbers $k=\omega n_i/c$. We also find a third solution with $n_3=\infty$ and $\boldsymbol{e}_3(\hat{\boldsymbol{k}})\|\hat{\boldsymbol{k}}$, corresponding to a nonpropagating mode with $\omega=0$. It violates $\hat{\boldsymbol{k}}\cdot\boldsymbol{d}^3=0$ but is necessary to construct the complete Green function for wave equation (7) (see below).

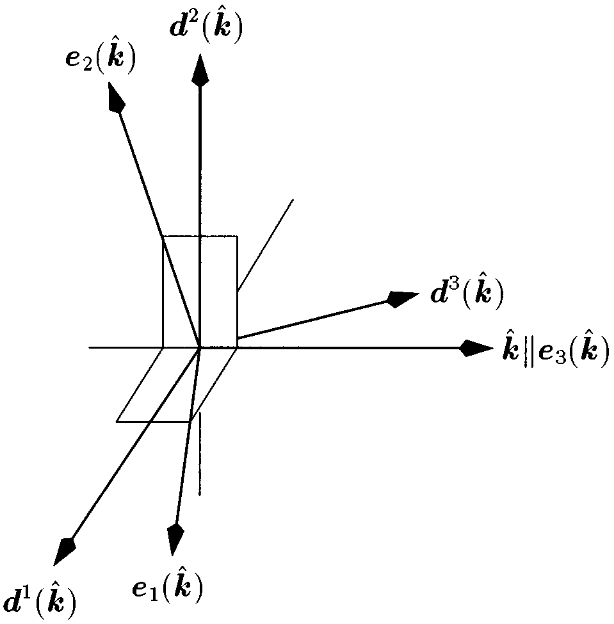

FIG. 1. The polarization vectors $\boldsymbol{e}_i(\hat{\boldsymbol{k}})$ and $\boldsymbol{d}^i(\hat{\boldsymbol{k}})$ for a given propagation direction $\hat{\boldsymbol{k}}$ in an anisotropic media.

The two sets of polarization vectors fulfill the biorthogonality condition

$$\boldsymbol{d}^i(\hat{\boldsymbol{k}})\cdot\boldsymbol{e}_j(\hat{\boldsymbol{k}})=\delta^i_j \quad (i,j=1,2,3), \tag{13}$$

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 this paper. To prove condition (13) we notice that $\boldsymbol{P}_t(\hat{\boldsymbol{k}})$ and $\boldsymbol{\varepsilon}_0$ are symmetric tensors and derive from Eq. (9) the condition

$$\left(\frac{1}{n_i^2}-\frac{1}{n_j^2}\right)\boldsymbol{e}_i\cdot\boldsymbol{d}^j=0, \tag{14}$$

from which Eq. (13) follows after an appropriate normalization. The vectors $\boldsymbol{d}^1$ and $\boldsymbol{d}^2$ are perpendicular to $\hat{\boldsymbol{k}}$. Then, again with Eq. (9) and the biorthogonality condition (13), one can show that $\boldsymbol{d}^1\perp\boldsymbol{d}^2$. In Fig. 1, we summarize the geometry for a given propagation direction $\hat{\boldsymbol{k}}$. Finally, we recall that in general the refractive indices are calculated from Fresnel's equation [47]:

$$\sum_{i=1}^{3}\frac{\overline{\varepsilon_i}\hat{k}_i^2}{n^2-\overline{\varepsilon_i}}=0, \tag{15}$$

where $\overline{\varepsilon_i}$ stands for the principal dielectric constants, the eigenvalues of $\boldsymbol{\varepsilon}_0$, and $\hat{k}_i$ is the component of the unit vector $\hat{\boldsymbol{k}}$ along the ith eigenvector of $\boldsymbol{\varepsilon}_0$. This equation is equivalent to $\det[\boldsymbol{P}_t(\hat{\boldsymbol{k}})-\boldsymbol{\varepsilon}_0/n^2]=0$.

Consider now $\boldsymbol{E}_0=E_0\boldsymbol{e}_i(\hat{\boldsymbol{k}})$ and $\boldsymbol{H}_0$, the amplitude of the magnetic field wave, which are connected via Maxwell's relations:

$$\boldsymbol{k}\times\boldsymbol{E}_0=\frac{\omega}{c}\boldsymbol{H}_0 \quad\text{and}\quad \boldsymbol{k}\times\boldsymbol{H}_0=-\frac{\omega}{c}\boldsymbol{\varepsilon}_0\boldsymbol{E}_0. \tag{16}$$

The second equation also follows from the first once Eq. (9) is solved. For the nonpropagating mode ($\omega=0$), $\boldsymbol{H}_0$, like $\boldsymbol{E}_0$, is parallel to $\hat{\boldsymbol{k}}$. For the propagating modes, $\boldsymbol{H}_0$ is perpendicular to $\hat{\boldsymbol{k}}$ and $\boldsymbol{\varepsilon}_0\boldsymbol{E}_0$. Using Eqs. (9) and (16) one shows that electric and magnetic fields carry the same amount of field energy ($\underline{\boldsymbol{H}_0 \cdot \boldsymbol{H}_0^*} = \boldsymbol{E}_0 \cdot \boldsymbol{\varepsilon}_0 \boldsymbol{E}_0^*$) and that the averaged energy density $\overline{u}$ of a light wave is

$$\overline{u} = \frac{1}{8\pi}\boldsymbol{E}_0 \cdot \boldsymbol{\varepsilon}_0 \boldsymbol{E}_0^* = \frac{1}{8\pi}|E_0|^2. \tag{17}$$

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

$$\hat{\boldsymbol{k}} \cdot \overline{\boldsymbol{S}} = \frac{c}{n}\overline{u}, \tag{18}$$

which is familiar for isotropic systems.

Finally, we calculate the Green function $\boldsymbol{G}_0(\boldsymbol{r}-\boldsymbol{r}', t-t')$ for the wave equation (7), which has a source term proportional to the time derivative of $\boldsymbol{j}_{\mathrm{ma}}$. In Fourier space we have

$$\left[k^2 \boldsymbol{P}_t(\hat{\boldsymbol{k}}) - \frac{\omega^2}{c^2}\boldsymbol{\varepsilon}_0\right]\boldsymbol{G}_0(\boldsymbol{k},\omega) = -\boldsymbol{1}. \tag{19}$$

We expand $\boldsymbol{G}_0(\boldsymbol{k},\omega)$ in the basis $\{\boldsymbol{e}_i(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_j(\hat{\boldsymbol{k}})\}$ and calculate its components by multiplying the last equation, respectively, from left and right with $\boldsymbol{e}_l(\hat{\boldsymbol{k}})$ and $\boldsymbol{d}^k(\hat{\boldsymbol{k}})$. Using the eigenvalue equation (9) and the algebra for the polarization vectors, described above, we find that $\boldsymbol{G}_0(\boldsymbol{k},\omega)$ is diagonal:

$$\boldsymbol{G}_0(\boldsymbol{k},\omega) = \sum_{\alpha=1}^{2}\left[\frac{\omega^2}{c^2} - \frac{k^2}{n_\alpha^2(\hat{\boldsymbol{k}})}\right]^{-1} \boldsymbol{e}_\alpha(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_\alpha(\hat{\boldsymbol{k}}) + \frac{c^2}{\omega^2}\boldsymbol{e}_3(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_3(\hat{\boldsymbol{k}}). \tag{20}$$

The first term on the right-hand side represents the propagating part of the Green function. We will always, throughout this paper, 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 nonpropagating δ 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 nondiagonal parts. Thus, whenever we use a Greek superscript or subscript α it refers, respectively, to the basis ‘‘vector’’ $\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})$ or $\boldsymbol{d}^\alpha(\hat{\boldsymbol{k}}) \otimes \boldsymbol{d}^\alpha(\hat{\boldsymbol{k}})$ ($\alpha=1,2$). The last quantity we need is the inverse Green function $[\boldsymbol{G}_0(\boldsymbol{k},\omega)]^{-1}$, which we conveniently represent as

$$[\boldsymbol{G}_0(\boldsymbol{k},\omega)]^{-1} = \sum_{\alpha=1}^{2}\left[\frac{\omega^2}{c^2} - \frac{k^2}{n_\alpha^2(\hat{\boldsymbol{k}})}\right]\boldsymbol{d}^\alpha(\hat{\boldsymbol{k}}) \otimes \boldsymbol{d}^\alpha(\hat{\boldsymbol{k}}) + \frac{\omega^2}{c^2}\boldsymbol{d}^3(\hat{\boldsymbol{k}}) \otimes \boldsymbol{d}^3(\hat{\boldsymbol{k}}) \tag{21}$$

by realizing that the unit tensor can be written as

$$\boldsymbol{1} = \sum_{ij} \delta_j^i \boldsymbol{e}_i(\hat{\boldsymbol{k}}) \otimes \boldsymbol{d}^j(\hat{\boldsymbol{k}}) = \sum_{ij} \delta_i^j \boldsymbol{d}^i(\hat{\boldsymbol{k}}) \otimes \boldsymbol{e}_j(\hat{\boldsymbol{k}}). \tag{22}$$

III. DIFFUSIVE LIGHT TRANSPORT IN ANISOTROPIC RANDOM MEDIA

We now consider a dielectric medium in which the dielectric tensor possesses a randomly fluctuating part $\delta\boldsymbol{\varepsilon}(\boldsymbol{r},t)$ in addition to the homogeneous term $\boldsymbol{\varepsilon}_0$. We treat $\delta\boldsymbol{\varepsilon}(\boldsymbol{r},t)$ as a Gaussian random variable with variance characterized by

$$\boldsymbol{B}^\omega(\boldsymbol{r},t) := \frac{\omega^4}{c^4}\langle \delta\boldsymbol{\varepsilon}(\boldsymbol{r},t) \otimes \delta\boldsymbol{\varepsilon}(\boldsymbol{0},0)\rangle^{(N)}, \tag{23}$$

where ω is the frequency of light. The superscript (N) means that we interchange the second and third indices in the tensor product $\delta\boldsymbol{\varepsilon} \otimes \delta\boldsymbol{\varepsilon}$ to define $\boldsymbol{B}^\omega$: $[\boldsymbol{B}^\omega]_{ijkl} \propto \langle \delta\varepsilon_{ik}\delta\varepsilon_{jl}\rangle$. We call $\boldsymbol{B}^\omega(\boldsymbol{r},t)$ the structure factor of the system. It is measured in single light scattering experiments [1] and contains information about the elastic and dynamic properties of a system.

The electric light field $\boldsymbol{E}(\boldsymbol{r},t)$ obeys the inhomogeneous wave equation

$$\left[\mathrm{curl}\ \mathrm{curl} + [\boldsymbol{\varepsilon}_0 + \delta\boldsymbol{\varepsilon}(\boldsymbol{r},t)]\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{0}, \tag{24}$$

where we have used an adiabatic approximation to pull $\delta\boldsymbol{\varepsilon}(\boldsymbol{r},t)$ in front of the time derivatives. It is valid if $\delta\boldsymbol{\varepsilon}(\boldsymbol{r},t)$ 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. (24). A very general one is given by the spatial and temporal autocorrelation function for the electric light field, $\langle \boldsymbol{E}(\boldsymbol{r}_1,t_1) \otimes \boldsymbol{E}(\boldsymbol{r}_2,t_2)\rangle$, which we write with the help of center of ‘‘mass’’ $(\boldsymbol{R},T)$ and relative $(\boldsymbol{r},t)$ coordinates:

$$\boldsymbol{W}(\boldsymbol{R},\boldsymbol{r},T,t) = \left\langle \boldsymbol{E}\left(\boldsymbol{R}+\frac{\boldsymbol{r}}{2}, T+\frac{t}{2}\right) \otimes \boldsymbol{E}^*\left(\boldsymbol{R}-\frac{\boldsymbol{r}}{2}, T-\frac{t}{2}\right)\right\rangle. \tag{25}$$

It is a second rank tensor. For $\boldsymbol{r}=\boldsymbol{0}$ we derive the scalar

$$W(\boldsymbol{R},T,t) = \left\langle \boldsymbol{E}\left(\boldsymbol{R}, T+\frac{t}{2}\right) \cdot \boldsymbol{\varepsilon}_0 \boldsymbol{E}^*\left(\boldsymbol{R}, T-\frac{t}{2}\right)\right\rangle, \tag{26}$$

which at $t=0$ is equal to the energy density of the light field when the small fluctuating part $\delta\boldsymbol{\varepsilon}$ is neglected. The T dependence is, e.g., due to time-modulated light sources, and $\boldsymbol{R}$ describes variations of the energy density on long length scales. For $t \neq 0$, $W(\boldsymbol{R},T,t)$ reflects the dynamics of the scattering medium through its dependence on the structure factor $\boldsymbol{B}^\omega(\boldsymbol{r},t)$. It is measured in dynamic light scattering experiments either in single scattering [1] or with DWS [49]. We will see below that the Fourier transform with respect to $\boldsymbol{r}$, $\boldsymbol{W}(\boldsymbol{R},\boldsymbol{k},T,t)$, is diagonal in the basis $\{\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\beta(\hat{\boldsymbol{k}})\}$ on length ($\boldsymbol{R}$) and time (T) scales much longer than the wavelength and time period of light,

$$\boldsymbol{W}(\boldsymbol{R},\boldsymbol{k},T,t)\approx\sum_\alpha W^\alpha(\boldsymbol{R},\boldsymbol{k},T,t)\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}}). \tag{27}$$

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

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

$$=\left\langle\boldsymbol{E}\left(\boldsymbol{k}+\frac{\boldsymbol{K}}{2},\omega+\frac{\Omega}{2}\right)\otimes\boldsymbol{E}^*\left(\boldsymbol{k}-\frac{\boldsymbol{K}}{2},\omega-\frac{\Omega}{2}\right)\right\rangle. \tag{28}$$

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

To calculate the autocorrelation function $\boldsymbol{W}(\boldsymbol{R},\boldsymbol{r},T,t)$ for special light sources $\boldsymbol{W}_0(\boldsymbol{R},\boldsymbol{r},T,t)$ and/or given boundary conditions, we need the averaged ''two-particle'' Green function

$$\boldsymbol{\Phi}=\langle\boldsymbol{G}^R\otimes\boldsymbol{G}^A\rangle^{(N)}, \tag{29}$$

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

$$\boldsymbol{W}(1)=\int d2\,\Phi(1,2)\boldsymbol{W}_0(2) \tag{30}$$

(for notation see Appendix A). In the second subsection we will derive the diffusion pole of $\boldsymbol{\Phi}$, i.e., that part of $\boldsymbol{\Phi}$ that corresponds to a diffusion equation for $\boldsymbol{W}(\boldsymbol{R},\boldsymbol{k},T,t)$ in the variables $\boldsymbol{R}$ 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 $\boldsymbol{\Phi}$.

A. The averaged one-particle Green function

The one-particle Green functions $\langle\boldsymbol{G}^R\rangle$ and $\langle\boldsymbol{G}^A\rangle$ follow from Dyson's equation [50], which we give in a formal notation:

$$\langle\boldsymbol{G}^{R/A}\rangle=\boldsymbol{G}_0+\boldsymbol{G}_0\boldsymbol{\Sigma}^{R/A}\langle\boldsymbol{G}^{R/A}\rangle. \tag{31}$$

(In the following we will use a coordinate-free representation for tensors and their contractions which we explain here. If $\boldsymbol{A}$ and $\boldsymbol{B}$ are second rank tensors so is $\boldsymbol{AB}$ with components $[\boldsymbol{AB}]_{ij}=\Sigma_k A_{ik}B_{kj}$. For fourth rank tensors $\boldsymbol{C}$ and $\boldsymbol{D}$ we form a tensor $\boldsymbol{CD}$ of the same rank with components $[\boldsymbol{CD}]_{ijkl}=\Sigma_{mn}C_{ijmn}D_{mnkl}$. Finally, $\boldsymbol{CA}$ is a second rank tensor and has components $[\boldsymbol{CA}]_{ij}=\Sigma_{kl}C_{ijkl}A_{kl}$.) We have introduced the retarded and advanced second-rank-tensor mass operators $\boldsymbol{\Sigma}^R$ and $\boldsymbol{\Sigma}^A$. In the weak-scattering approximation, where the elements of the structure factor $\boldsymbol{B}^\omega(\boldsymbol{r},t=0)$ are assumed to be much smaller than 1, these mass operators are proportional to $\boldsymbol{B}^\omega(\boldsymbol{r},t=0)$ and can be written in frequency space as

$$\boldsymbol{\Sigma}^{R/A}(\boldsymbol{r},\omega)=\boldsymbol{B}^\omega(\boldsymbol{r},t=0)\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{r},\omega). \tag{32}$$

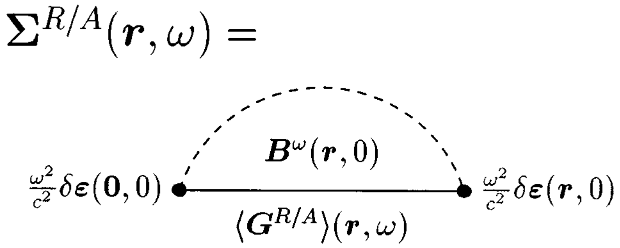

FIG. 2. The mass operators $\boldsymbol{\Sigma}^{R/A}(\boldsymbol{r},\omega)$ in the weak-scattering approximation consist of two scattering events from inhomogeneities in the dielectric tensor, which are tied together by spatial correlations $[\boldsymbol{B}^\omega(\boldsymbol{r},t=0)]$ and the averaged propagators $\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{r},\omega)$ of the electric field.

Figure 2 gives a diagrammatic representation of $\boldsymbol{\Sigma}^{R/A}(\boldsymbol{r},\omega)$.

Finally, in momentum space, we get from Eq. (31),

$$\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)=[\boldsymbol{G}_0^{-1}(\boldsymbol{k},\omega)-\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)]^{-1}. \tag{33}$$

The inverse Green function $\boldsymbol{G}_0^{-1}(\boldsymbol{k},\omega)$ is diagonal in the basis $\{\boldsymbol{d}^i(\hat{\boldsymbol{k}})\otimes\boldsymbol{d}^j(\hat{\boldsymbol{k}})\}$, and $\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)$ causes a small perturbation. From perturbation theory we know that to zeroth order in $\boldsymbol{\Sigma}^{R/A}$ the eigenvectors of $\boldsymbol{G}_0^{-1}(\boldsymbol{k},\omega)-\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)$ are unchanged, whereas to first order in $\boldsymbol{\Sigma}^{R/A}$ the diagonal elements

$$[\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)]_i=\boldsymbol{e}_i(\hat{\boldsymbol{k}})\cdot\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)\boldsymbol{e}_i(\hat{\boldsymbol{k}}) \tag{34}$$

contribute to the eigenvalues of $\langle\boldsymbol{G}^{R/A}\rangle$ renormalizing the wave numbers $(\omega/c)n_i$ of the light modes. The significant effect of $\boldsymbol{\Sigma}^{R/A}$ comes from its imaginary part. Then, in its final form the propagating part of the Green functions reads

$$\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)\approx\sum_{\alpha=1}^{2}[\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)]^\alpha\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}}) \tag{35}$$

with

$$[\langle\boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)]^\alpha=\left[\frac{\omega^2}{c^2}-\frac{k^2}{n_\alpha^2(\hat{\boldsymbol{k}})}\mp\frac{i\omega}{cn_\alpha(\hat{\boldsymbol{k}})l_\alpha(\hat{\boldsymbol{k}},\omega)}\right]^{-1}. \tag{36}$$

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

$$l_\alpha(\hat{\boldsymbol{k}},\omega)=\left[-\frac{c}{\omega}n_\alpha(\hat{\boldsymbol{k}})[\mathrm{Im}\boldsymbol{\Sigma}^R(\boldsymbol{k}^\alpha,\omega)]_\alpha\right]^{-1}. \tag{37}$$

To arrive at $l_\alpha(\hat{\boldsymbol{k}},\omega)$, we used a coherent potential approximation replacing the argument $\boldsymbol{k}$ of $[\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)]_\alpha$ by the wave vector $\boldsymbol{k}^\alpha=(\omega/c)n_\alpha\hat{\boldsymbol{k}}$ of the light modes where $\boldsymbol{G}_0$ diverges. $\mathrm{Im}\boldsymbol{\Sigma}^R$ follows from Eq. (32) using the momentum shell approximation

$$[\mathrm{Im}\langle \boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)]^\alpha \approx -\frac{\pi}{2}\frac{c}{\omega}n_\alpha(\hat{\boldsymbol{k}})\,\delta((\omega/c)n_\alpha(\hat{\boldsymbol{k}})-k) \tag{38}$$

valid in the limit $\boldsymbol{\Sigma}^R\to\boldsymbol{0}$. We finally obtain

$$l_\alpha(\hat{\boldsymbol{k}},\omega)=\left[\frac{\pi}{2}n_\alpha(\hat{\boldsymbol{k}})\sum_{\beta=1}^{2}\int_{\hat{\boldsymbol{q}}^\beta}[\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(t=0)]_{\alpha\beta}\right]^{-1}, \tag{39}$$

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

$$[\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(t=0)]_{\alpha\beta}=\frac{\omega^4}{c^4}\langle|\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\cdot\delta\boldsymbol{\varepsilon}(\boldsymbol{q}_s,0)\boldsymbol{e}_\beta(\hat{\boldsymbol{q}})|^2\rangle, \tag{40}$$

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

$$\boldsymbol{q}_s=\frac{\omega}{c}(n_\alpha\hat{\boldsymbol{k}}-n_\beta\hat{\boldsymbol{q}}). \tag{41}$$

In Eq. (39) and throughout this paper, we use a shorthand notation for angular integration:

$$\int_{\hat{\boldsymbol{q}}^\beta}\cdots=\int\frac{d\Omega_{\boldsymbol{q}}}{(2\pi)^3}n^3_\beta(\hat{\boldsymbol{q}})\cdots. \tag{42}$$

We point out that the scattering mean free path $l_\alpha(\hat{\boldsymbol{k}},\omega)$ now depends on the direction $\hat{\boldsymbol{k}}$ and the polarization $\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})$ of the light mode. Following the work of Nelson and Lax [48], it is straightforward to show that $l_\alpha(\hat{\boldsymbol{k}},\omega)$ determines an exponential decay of the far field of $\langle\boldsymbol{G}^R\rangle(\boldsymbol{R},\omega)$ traveling in the direction $\hat{\boldsymbol{k}}$ with polarization $\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})$. In our analysis, we have neglected the off-diagonal components of $\boldsymbol{\Sigma}^{R/A}$, 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

$$\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)=\langle\boldsymbol{G}^R\rangle(\boldsymbol{k}_+,\omega_+)-\langle\boldsymbol{G}^A\rangle(\boldsymbol{k}_-,\omega_-), \tag{43}$$

and

$$\Delta\boldsymbol{\Sigma}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)=\boldsymbol{\Sigma}^R(\boldsymbol{k}_+,\omega_+)-\boldsymbol{\Sigma}^A(\boldsymbol{k}_-,\omega_-), \tag{44}$$

where

$$\boldsymbol{k}_\pm=\boldsymbol{k}\pm\boldsymbol{K}/2 \quad\text{and}\quad \omega_\pm=\omega\pm\Omega/2. \tag{45}$$

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

$$\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)=2i\ \mathrm{Im}\langle\boldsymbol{G}^R\rangle(\boldsymbol{k},\omega) \tag{46}$$

and

$$\Delta\boldsymbol{\Sigma}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)=2i\ \mathrm{Im}\boldsymbol{\Sigma}^R(\boldsymbol{k},\omega)=-\frac{\omega}{c}\frac{2i}{n_\alpha(\hat{\boldsymbol{k}})l_\alpha(\hat{\boldsymbol{k}},\omega)}. \tag{47}$$

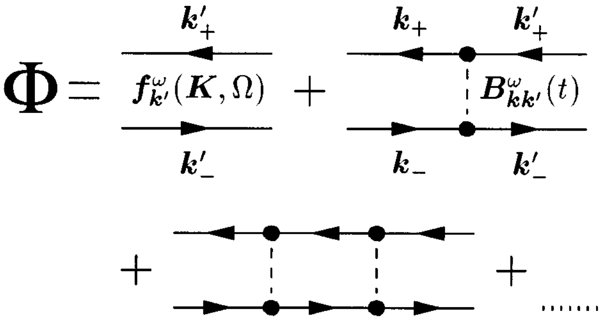

FIG. 3. The two-particle Green function $\boldsymbol{\Phi}$ as a sum of ladder diagrams. $\boldsymbol{f}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)$ propagates two electric field modes with wave vectors $\boldsymbol{k}'_+$ and $\boldsymbol{k}'_-$ Both are scattered by the same inhomogeneity in the dielectric tensor described by the structure factor $\boldsymbol{B}^\omega_{\boldsymbol{kk}'}(t)$ for the scattering vector $\boldsymbol{k}-\boldsymbol{k}'$.

B. The averaged two-particle Green function

The averaged two-particle Green function $\boldsymbol{\Phi}$ obeys the Bethe-Salpeter equation [50]

$$\boldsymbol{\Phi}=[\langle\boldsymbol{G}^R\rangle\otimes\langle\boldsymbol{G}^A\rangle]^{(N)}+[\langle\boldsymbol{G}^R\rangle\otimes\langle\boldsymbol{G}^A\rangle]^{(N)}\boldsymbol{U}\boldsymbol{\Phi}, \tag{48}$$

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

$$\int\frac{d^3k_1}{(2\pi)^3}[\boldsymbol{1}^{(4)}_{\boldsymbol{kk}_1}-\boldsymbol{f}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)\boldsymbol{B}^\omega_{\boldsymbol{kk}_1}(t)]\boldsymbol{\Phi}^\omega_{\boldsymbol{k}_1\boldsymbol{k}'}(\boldsymbol{K},\Omega,t)$$
$$=\boldsymbol{f}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)\boldsymbol{1}^{(4)}_{\boldsymbol{kk}'}, \tag{49}$$

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:

$$\boldsymbol{f}^\omega_{\boldsymbol{k}}(\boldsymbol{K},\Omega)=[\langle\boldsymbol{G}^R\rangle(\boldsymbol{k}_+,\omega_+)\otimes\langle\boldsymbol{G}^A\rangle(\boldsymbol{k}_-,\omega_-)]^{(N)}, \tag{50}$$

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

$$[\boldsymbol{1}^{(4)}_{\boldsymbol{kk}'}]_{ijkl}:=(2\pi)^3\delta(\boldsymbol{k}-\boldsymbol{k}')(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})/2. \tag{51}$$

Here i, j, k, and l are Cartesian indices. The Bethe-Salpeter equation (49) can be solved formally by iteration, leading to a sum of ladder diagrams (see Fig. 3). The multiple integrals and the sum can be done analytically for δ-function correlations for $\boldsymbol{K}\to\boldsymbol{0}$, and the solution represents the diffusion pole of $\mathbf{\Phi}$. For the anisotropic, long-range correlations of our problem this procedure is not possible. Vollhardt and Wölfle [51] derived the diffusion pole for isotropic electron transport directly from Eq. (49), and MacKintosh and John [15] applied their method to light. In the anisotropic case one has to be more careful, as Wölfle and Bhatt showed for electrons [44]. We will proceed in a similar way, however, our problem is more difficult because we have to deal with the different polarizations of light.

Let $\mathbf{\Psi}_{\boldsymbol{k}}^{(n)}(\boldsymbol{K},\Omega,t)$ and $\lambda^{(n)}(\boldsymbol{K},\Omega,t)$ be, respectively, the nth eigenvector and eigenvalue of the integral operator of Eq. (49),

$$\int \frac{d^3k_1}{(2\pi)^3}[\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}_1}^{(4)} - \boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega}(t)]\mathbf{\Psi}_{\boldsymbol{k}_1}^{(n)} = \lambda^{(n)}\mathbf{\Psi}_{\boldsymbol{k}}^{(n)}, \tag{52}$$

and $\overline{\mathbf{\Psi}}_{\boldsymbol{k}}^{(n)}(\boldsymbol{K},\Omega,t)$ the eigenvectors of the Hermitian adjoint operator [52]. Then it is straightforward to show [15], with the aid of the completeness relation

$$\sum_n \mathbf{\Psi}_{\boldsymbol{k}}^{(n)} \otimes \overline{\mathbf{\Psi}}_{\boldsymbol{k}'}^{(n)} = \mathbf{1}_{\boldsymbol{k}\boldsymbol{k}'}^{(4)}, \tag{53}$$

that

$$\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega,t) = \sum_n \frac{\mathbf{\Psi}_{\boldsymbol{k}}^{(n)} \otimes \overline{\mathbf{\Psi}}_{\boldsymbol{k}'}^{(n)}}{\lambda^{(n)}} \boldsymbol{f}_{\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega) \tag{54}$$

solves the Bethe-Salpeter equation. When $\boldsymbol{K}=\mathbf{0}$ and $\Omega=t=0$, the quantity $\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)$, defined in Eq. (43), is an eigenvector of Eq. (52) with eigenvalue $\lambda^{(0)}(\mathbf{0},0,0)=0$. To prove this statement we use a special case of one of the Ward identities (see Appendix B1),

$$\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0) = \int \frac{d^3k'}{(2\pi)^3}\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(t=0)\Delta\boldsymbol{G}_{\boldsymbol{k}'}^{\omega}(\mathbf{0},0), \tag{55}$$

and the relation

$$\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0) = \boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0). \tag{56}$$

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 [51]. All other eigenvalues of Eq. (52) are positive, and in the real-space coordinate $\boldsymbol{R}$, they give exponentially decaying contributions to $\mathbf{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(\boldsymbol{K},\Omega,t)$, which are not important at long length scales [15].

The diffusion approximation follows when we calculate $\lambda^{(0)}(\boldsymbol{K},\Omega,t)$ with the help of perturbation theory in the limit $K,\Omega,t\to 0$. First, we have to get the eigenvalue equation in this limit. To proceed we need the following equation, which we derive in Appendix B2:

$$[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)]^{\alpha} \approx [\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha\alpha}\left([\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]_{\alpha} - \frac{\partial[\boldsymbol{G}_0^{-1}]_{\alpha}}{\partial\boldsymbol{k}}\cdot\boldsymbol{K} - \frac{2\omega}{c^2}\Omega\right). \tag{57}$$

As usual, the Greek superscript or subscript α refers, respectively, to the basis ‘‘vectors’’ $\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})$ or $\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}})\otimes\boldsymbol{d}^{\alpha}(\hat{\boldsymbol{k}})$. Equation (57) gives $[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)]^{\alpha}$ to first order in $\boldsymbol{K}$, Ω, and $\mathbf{\Sigma}$. It shows clearly where the wave vector $\boldsymbol{K}$ and the frequency Ω come in. Now, we formulate eigenvalue equation (52) in components, choosing $\boldsymbol{K}=\mathbf{0}$ and $\Omega=0$. Then, we apply the last equation and arrive at

$$\begin{aligned}&\left([\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]_{\alpha} - \frac{\partial[\boldsymbol{G}_0^{-1}]_{\alpha}}{\partial\boldsymbol{k}}\cdot\boldsymbol{K} - \frac{2\omega}{c^2}\Omega\right)\Psi_{\boldsymbol{k}}^{\alpha}\\ &\quad -[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha}\sum_{\beta}\int\frac{d^3k_1}{(2\pi)^3}[\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega}(t)]_{\alpha\beta}\Psi_{\boldsymbol{k}_1}^{\beta}\\ &\quad = \lambda(\boldsymbol{K},\Omega,t)[\Delta\mathbf{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]_{\alpha}\Psi_{\boldsymbol{k}}^{\alpha}\end{aligned} \tag{58}$$

as the eigenvalue equation, which is correct up to first order in $\boldsymbol{K}$, Ω, and $\mathbf{\Sigma}$. In a next step we use the ansatz

$$\Psi_{\boldsymbol{k}}^{\alpha} \propto [\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha}\left[\tilde{\Psi}_0\pi + \sum_{i=1}^{\cdots}\tilde{\Psi}_i\varphi_i^{\alpha}(\hat{\boldsymbol{k}})\right] \tag{59}$$

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

$$[\Delta\boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha} \approx -i\pi(c/\omega)n_{\alpha}(\hat{\boldsymbol{k}})\,\delta\!\left(\frac{\omega}{c}n_{\alpha}(\hat{\boldsymbol{k}}) - k\right). \tag{60}$$

The amplitude $\tilde{\Psi}_0$ represents the zeroth eigenvector, and the second term involves a complete set of real basis functions $\varphi_i(\hat{\boldsymbol{k}})$ on the unit sphere and for the space spanned by $\boldsymbol{e}_1\otimes\boldsymbol{e}_1$ and $\boldsymbol{e}_2\otimes\boldsymbol{e}_2$. We will comment below on how to choose these basis functions. Keeping only dominant terms in the small Ω, K, and t limit, we can write the eigenvalue equation in matrix form as

$$\begin{aligned}&\begin{bmatrix}\dfrac{2\overline{n^3}}{\pi c}[-i\Omega+\mu(\omega,t)] & -i[\mathcal{G}(\boldsymbol{K})]^t\\ -i\mathcal{G}(\boldsymbol{K}) & \mathcal{B}\end{bmatrix}\begin{bmatrix}\tilde{\Psi}_0\\ \vdots\end{bmatrix}\\ &\quad = \lambda(\boldsymbol{K},\Omega,t)\begin{bmatrix}\Sigma & \mathcal{S}_0^t\\ \mathcal{S}_0 & \mathcal{S}_1\end{bmatrix}\begin{bmatrix}\tilde{\Psi}_0\\ \vdots\end{bmatrix}.\end{aligned} \tag{61}$$

The matrix $\mathcal{B}$ basically represents the structure factor $[\boldsymbol{B}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}^{\omega}(0)]_{\alpha\beta}$ in our chosen basis,

$$\begin{aligned}[\mathcal{B}]_{ij} = \pi\sum_{\alpha,\beta}\Bigg\{&\int_{\hat{\boldsymbol{k}}^{\alpha}}\int_{\hat{\boldsymbol{q}}^{\beta}}[\varphi_i^{\alpha}(\hat{\boldsymbol{k}})\varphi_j^{\alpha}(\hat{\boldsymbol{k}}) - \varphi_i^{\alpha}(\hat{\boldsymbol{k}})\varphi_j^{\beta}(\hat{\boldsymbol{q}})]\\ &\times[\boldsymbol{B}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}^{\omega}(0)]_{\alpha\beta}\Bigg\}.\end{aligned} \tag{62}$$

The vector $\mathcal{G}$ with components

$$[\mathcal{G}(\boldsymbol{K})]_i=\pi\sum_\alpha \int_{\hat{\boldsymbol{k}}^\alpha} n_\alpha(\hat{\boldsymbol{k}})\frac{\partial[\boldsymbol{G}_0^{-1}]_\alpha}{\partial\boldsymbol{k}}\bigg|_{\hat{\boldsymbol{k}}}\cdot\boldsymbol{K}\varphi_i^\alpha(\hat{\boldsymbol{k}}) \tag{63}$$

is linear in $\boldsymbol{K}$. Another important term is the coefficient

$$\mu(\omega,t)=\frac{c\pi^3}{2\overline{n^3}}\sum_{\alpha,\beta}\int_{\hat{\boldsymbol{k}}^\alpha}\int_{\hat{\boldsymbol{q}}^\beta}[\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(0)-\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(t)]_{\alpha\beta}, \tag{64}$$

where

$$\overline{n^3}=\frac{1}{8\pi}\sum_\alpha\int d\Omega_{\boldsymbol{k}}\, n_\alpha^3(\hat{\boldsymbol{k}}). \tag{65}$$

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

$$\Sigma=\pi^3\sum_{\alpha,\beta}\int_{\hat{\boldsymbol{k}}^\alpha}\int_{\hat{\boldsymbol{q}}^\beta}[\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(0)]_{\alpha\beta} \tag{66}$$

$$=-\pi\frac{c^2}{\omega^2}\sum_\alpha\int\frac{d^3k}{(2\pi)^3}\times[\Delta\boldsymbol{\Sigma}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)]_\alpha[\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)]^\alpha. \tag{67}$$

The second equation above states that Σ is proportional to the normalization factor of the eigenfunction $\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)$, with $\Delta\boldsymbol{\Sigma}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)$ being the eigenfunction of the Hermitian conjugated problem. The vector $\mathcal{S}_0$ and the matrix $\mathcal{S}_1$ 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 (61), and we want to know how it evolves for small K, Ω, and t. The vector $\mathcal{G}(\boldsymbol{K})$ couples the 00 element with the matrix $\mathcal{B}$. The coupling can be removed by an orthogonal transformation with

$$\mathcal{U}=\begin{bmatrix}1 & i[\mathcal{G}(\boldsymbol{K})]^t\mathcal{B}^{-1}\\ -i\mathcal{B}^{-1}\mathcal{G}(\boldsymbol{K}) & \boldsymbol{1}\end{bmatrix}, \tag{68}$$

which essentially renormalizes the 00 element. Calculating $\lambda^{(0)}$ from the transformed system to leading order in K^2 shows that $\lambda^{(0)}\Sigma$ is equal to the renormalized 00 element, or

$$\lambda^{(0)}=\frac{2\overline{n^3}}{c\Sigma}\left[-i\Omega+\mu(\omega,t)+\frac{\pi c}{2\overline{n^3}}\mathcal{G}(\boldsymbol{K})\cdot\mathcal{B}^{-1}\mathcal{G}(\boldsymbol{K})\right]. \tag{69}$$

Now, we are ready to write down the diffusion approximation of the averaged two-particle Green function from Eq. (54),

$$\boldsymbol{\Phi}^\omega_{\boldsymbol{k}\boldsymbol{k}'}(\boldsymbol{K},\Omega,t)\approx\frac{1}{N}\frac{\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}}(\boldsymbol{0},0)\otimes\Delta\boldsymbol{G}^\omega_{\boldsymbol{k}'}(\boldsymbol{0},0)}{-i\Omega+\mu(\omega,t)+\boldsymbol{K}\cdot\boldsymbol{D}(\omega)\boldsymbol{K}} \tag{70}$$

with

$$\boldsymbol{K}\cdot\boldsymbol{D}(\omega)\boldsymbol{K}=\frac{c}{2\overline{n^3}}\mathcal{G}(\boldsymbol{K})\cdot\mathcal{B}^{-1}\mathcal{G}(\boldsymbol{K}) \tag{71}$$

and $N=-2\overline{n^3}\omega^2/(\pi c^3)$. When $\mu(\omega,t)=0$ the Green function $\boldsymbol{\Phi}^\omega_{\boldsymbol{k}\boldsymbol{k}'}(\boldsymbol{K},\Omega,t)$ possesses a simple diffusion pole with an anisotropic diffusion tensor $\boldsymbol{D}(\omega)$. 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. (71), it is clear that the problem of calculating $\boldsymbol{D}(\omega)$ reduces to the problems of calculating $\mathcal{G}(\boldsymbol{K})$ and the inverse matrix $\mathcal{B}^{-1}$. If we had at our disposal functions $\boldsymbol{\varphi}_i(\hat{\boldsymbol{k}})$, that diagonalize $\mathcal{B}$, our task would be simple:

$$\boldsymbol{K}\cdot\boldsymbol{D}(\omega)\boldsymbol{K}=\frac{c}{2\overline{n^3}}\sum_i\frac{[\mathcal{G}(\boldsymbol{K})]_i^2}{[\mathcal{B}]_{ii}}. \tag{72}$$

Unfortunately these functions are difficult to find in anisotropic systems, and we are reduced to seeking usable approximation schemes. One scheme is to choose functions $\boldsymbol{\varphi}_i(\hat{\boldsymbol{k}})$ so that only a few components of $\mathcal{G}(\boldsymbol{K})$, say $[\mathcal{G}(\boldsymbol{K})]_i$ for $i=1,2$, are nonzero. Then, if the off-diagonal components of $[\mathcal{B}]_{ij}$, coupling j for $j>2$ to i for $i=1,2$, are small, $[\mathcal{G}(\boldsymbol{K})]_i[\mathcal{B}^{-1}]_{ij}[\mathcal{G}(\boldsymbol{K})]_j$ can be approximated by using only the $i,j=1,2$ submatrix of $[\mathcal{B}]_{ij}$ to calculate $[\mathcal{B}^{-1}]_{ij}$. We will use this approach in numerical calculations of $\boldsymbol{D}(\omega)$ 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 dielectric tensor originate from density fluctuations only: $\delta\boldsymbol{\varepsilon}\propto\delta\varrho\boldsymbol{1}$. Then the components of the structure factor assume the form

$$[\boldsymbol{B}^\omega_{\boldsymbol{k}^\alpha\boldsymbol{q}^\beta}(0)]_{\alpha\beta}=[\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\cdot\boldsymbol{e}_\beta(\hat{\boldsymbol{q}})]^2B^\omega(\cos\vartheta_s), \tag{73}$$

where $B^\omega(\cos\vartheta_s)$ solely depends on the scattering angle ϑ_s. In isotropic systems, all the polarization vectors perpendicular to $\hat{\boldsymbol{k}}$ are equivalent, and the diffusion constant should not depend on the special choice we make. Therefore, to treat the factor $[\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\cdot\boldsymbol{e}_\beta(\hat{\boldsymbol{q}})]^2$, we can add integrations $\int d\phi_1/(2\pi)\int d\phi_2/(2\pi)$ in the formula for $\mathcal{B}$. The angles ϕ_1 and ϕ_2, respectively, describe rotations of the polarization vectors about $\hat{\boldsymbol{k}}$ and $\hat{\boldsymbol{q}}$. The integration over ϕ_1 and ϕ_2 for a fixed scattering angle ϑ_s is straightforward, and we obtain

$$\int\frac{d\phi_1}{2\pi}\int\frac{d\phi_2}{2\pi}[\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}},\phi_1)\cdot\boldsymbol{e}_\beta(\hat{\boldsymbol{q}},\phi_2)]^2=\frac{1+\cos^2\vartheta_s}{4\varepsilon_0^2}, \tag{74}$$

where the dielectric constant ε_0 comes in through the normalization of the polarization vectors. With this trick we are able to show that $\mathcal{B}$ is essentially diagonalized by spherical harmonics. (Our formulas are written for real basis functions, a generalization to complex ones is straightforward.) As basis functions we choose $\boldsymbol{\varphi}_{1lm}(\hat{\boldsymbol{k}})=[Y_{lm}(\vartheta,\varphi),Y_{lm}(\vartheta,\varphi)]$ and $\boldsymbol{\varphi}_{2lm}(\hat{\boldsymbol{k}})=[Y_{lm}(\vartheta,\varphi),-Y_{lm}(\vartheta,\varphi)]$ with $\cos\vartheta=\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{K}}$. We also use the addition theorem, $P_l(\cos\vartheta_s)\propto\Sigma_m Y^*_{lm}(\vartheta,\varphi)Y_{lm}(\vartheta',\varphi')$ [53], and arrive at $[\mathcal{B}]^{\gamma lm}_{\gamma' l' m'}$

$\propto \delta_{\gamma\gamma'}\delta_{ll'}\delta_{mm'}$. The only nonzero element of $\mathcal{G}$ is $[\mathcal{G}]_{110} = -2\varepsilon_0 K/\sqrt{3}$, and we finally find the formula for the diffusion constant:

$$D = \frac{16}{3}\pi c_0 \left[\int d\cos\vartheta_s (1-\cos\vartheta_s) \right. \\ \left. \times (1+\cos^2\vartheta_s) B^{\omega}(\cos\vartheta_s) \right]^{-1}, \qquad (75)$$

where c_0 stands for the speed of light in the system. Compared to the scalar case we have an additional factor $1+\cos^2\vartheta_s$. $B^{\omega}(\cos\vartheta_s)$ alone describes a scattering process with incoming and outgoing polarization perpendicular to the scattering plane [vertical-vertical (VV) scattering], whereas $\cos^2\vartheta_s B^{\omega}(\cos\vartheta_s)$ belongs to scattering with polarizations in the plane [horizontal-horizontal (HH) scattering] [1]. In a group theoretical language $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}$ transforms under a high-dimensional identity representation of SO(3). The relation $[\mathcal{B}]^{\gamma l m}_{\gamma' l' m'} \propto \delta_{ll'}\delta_{mm'}$ then means that we have decomposed $[\mathcal{B}]^{\gamma l m}_{\gamma' l' m'}$ via the irreducible representations of SO(3). For less symmetric phases, like a nematic liquid crystal, we can at least partially diagonalize $\mathcal{B}$ with the help of group theory. The relevant symmetry group $\mathrm{D}_{\infty h}$ has only one-dimensional representations induced by the functions $\exp(im\varphi)$, where φ is the azimuthal angle around the symmetry axis. Decomposing $\mathcal{B}$ with the help of spherical harmonics gives $[\mathcal{B}]^{\gamma l m}_{\gamma' l' m'} \propto \delta_{mm'}$, where different l and l' now couple to each other (see next section).

The numerator of the Green function in Eq. (70) contains an interesting effect. The second factor $\Delta\boldsymbol{G}^{\omega}_{\boldsymbol{k}'}(\boldsymbol{0},0)$ collects the light sources or the incoming light waves. The first factor determines the energy density $W^{\alpha}(\boldsymbol{R},\boldsymbol{k},T,0)$ of an outgoing light wave independent of the sources. This means that in the diffusion approximation the outgoing light loses all its correlations with the light sources. Integrating $[\Delta\boldsymbol{G}^{\omega}_{\boldsymbol{k}'}(\boldsymbol{0},0)]_{\alpha}$ over the wave number k shows that the ratio for the energy densities $W^{\alpha}(\boldsymbol{R},\hat{\boldsymbol{k}},T,0)$ in polarization states 1 and 2 is $[n_1(\hat{\boldsymbol{k}})/n_2(\hat{\boldsymbol{k}})]^3$. Experiments measure light intensities. In Eq. (18) we learned that only the projection of the Poynting vector $\boldsymbol{S}^{\alpha}$ on $\hat{\boldsymbol{k}}$ is simple: $\boldsymbol{S}^{\alpha}\cdot\hat{\boldsymbol{k}} = (c/n_{\alpha})W^{\alpha}(\boldsymbol{R},\hat{\boldsymbol{k}},T,0)$. Its ratio for the output polarizations 1 and 2 is $[n_1(\hat{\boldsymbol{k}})/n_2(\hat{\boldsymbol{k}})]^2$, which gives the ratio of the output intensities when the Poynting vectors are parallel to $\hat{\boldsymbol{k}}$. This is the case for light traveling along the principal axes of the dielectric tensor. The Green function in Eq. (70) suggests that there is a diffusion equation for each light wave with direction $\hat{\boldsymbol{k}}$ and polarization α. 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 $\boldsymbol{k}$ the Green function in Eq. (70) is equivalent to a diffusion equation for the scalar time-correlation function $W(\boldsymbol{R},T,t)$, introduced in Eq. (26),

$$\left[\frac{\partial}{\partial T} - \nabla\cdot\boldsymbol{D}(\omega)\nabla + \mu(\omega,t)\right] W(\boldsymbol{R},T,t) = \varrho(\boldsymbol{R},T). \qquad (76)$$

This equation is the basis of DWS [14]. Solving it for special boundary conditions and sources $\varrho(\boldsymbol{R},T)$, which depend on experimental arrangements, gives $W(\boldsymbol{R},T,t)$ 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 $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0) - \boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t)]_{\alpha\beta} \ll [\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}$. In this time range we expect to be able to perform a Taylor expansion in t that gives a linear time dependence for $\mu(\omega,t)$:

$$\mu(\omega,t) = \mu_0 t. \qquad (77)$$

The constant μ_0 reflects some averaged dynamical properties of the system. For the diffusion of particles in colloidal suspensions, $\mu_0 = 2D_B\omega^2/(l^*c)$, where D_B is the self-diffusion constant of the particles [49]. 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 $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t)]_{\alpha\beta}$ 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 diplacement is not simply proportional to t [49]. As a result, $\mu(\omega,t)$ does not follow the linear time law of Eq. (77). Experimentalists prefer a different picture for DWS, which they have developed for colloidal suspensions [49]. 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

$$[\mu(\omega,t) - \nabla\cdot\boldsymbol{D}(\omega)\nabla] W(\boldsymbol{R},\ldots,t) = \varrho(\boldsymbol{R},\ldots). \qquad (78)$$

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

$$\left[\frac{\partial}{\partial\tau} - \nabla\cdot\boldsymbol{D}(\omega)\nabla\right] P(\boldsymbol{R},\tau) = \delta(\tau)\varrho(\boldsymbol{R},\ldots) \qquad (79)$$

with

$$W(\boldsymbol{R},\ldots,t) = \int_{-0}^{\infty} P(\boldsymbol{R},\tau)\exp[-\mu(\omega,t)\tau]\,d\tau. \qquad (80)$$

The lower limit -0 means a small negative time τ in order to pick up the δ function. We can interpret $P(\boldsymbol{R},\tau)$ (after an appropriate normalization) as the probability that light, emitted by the source at time $\tau = 0$, arrives at the detector at point $\boldsymbol{R}$ after a time τ. Then the time correlation function $W(\boldsymbol{R},\ldots,t)$ follows after a summation over all light paths where each light path contributes a factor $\exp[-\mu(\omega,t)\tau]$ to the decay of $W(\boldsymbol{R},\ldots,t)$. For isotropic systems τ is directly connected to the path length $s = c\tau/n$ of light and $P(\boldsymbol{R},\tau)$ also represents the path-length distribution. The exact form of $P(\boldsymbol{R},\tau)$ depends on the choice of the light source and the boundary conditions [49], which we do not address here.

IV. LIGHT DIFFUSION IN NEMATIC LIQUID CRYSTALS

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

$$\boldsymbol{\varepsilon}(\boldsymbol{r},t)=\varepsilon_{\perp}\mathbf{1}+\Delta\varepsilon[\boldsymbol{n}(\boldsymbol{r},t)\otimes\boldsymbol{n}(\boldsymbol{r},t)], \tag{81}$$

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

$$F[\boldsymbol{n}]=\frac{1}{2}\int[K_1(\mathrm{div}\boldsymbol{n})^2+K_2(\boldsymbol{n}\cdot\mathrm{curl}\boldsymbol{n})^2+K_3(\boldsymbol{n}\times\mathrm{curl}\boldsymbol{n})^2 \\ -\Delta\chi(\boldsymbol{n}\cdot\boldsymbol{H})^2]d^3r. \tag{82}$$

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 $\boldsymbol{H}$ is favored. Even in a uniformly aligned sample there exist thermally induced fluctuations of the director:

$$\boldsymbol{n}(\boldsymbol{r},t)=\boldsymbol{n}_0+\delta\boldsymbol{n}(\boldsymbol{r},t). \tag{83}$$

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 uniaxial media. 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

$$\boldsymbol{\varepsilon}_0=\varepsilon_{\perp}\mathbf{1}+\Delta\varepsilon[\boldsymbol{n}_0\otimes\boldsymbol{n}_0]. \tag{84}$$

The equilibrium value $\boldsymbol{n}_0$ 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 (9). They are well described in the literature [47]. 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 $\boldsymbol{e}_2(\hat{\boldsymbol{k}})$ and $\boldsymbol{d}^2(\hat{\boldsymbol{k}})$ are both perpendicular to $\boldsymbol{n}_0$ and $\hat{\boldsymbol{k}}$ 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 $\boldsymbol{n}_0$ parallel to the z axis and write $\hat{\boldsymbol{k}}$ in spherical coordinates:

$$\boldsymbol{n}_0=\begin{bmatrix}0\\0\\1\end{bmatrix} \quad\text{and}\quad \hat{\boldsymbol{k}}=\begin{bmatrix}\sin\vartheta_{\boldsymbol{k}}\cos\varphi_{\boldsymbol{k}}\\ \sin\vartheta_{\boldsymbol{k}}\sin\varphi_{\boldsymbol{k}}\\ \cos\vartheta_{\boldsymbol{k}}\end{bmatrix}. \tag{85}$$

Then the ordinary light ray is represented by

$$n_2=\sqrt{\varepsilon_{\perp}} \quad\text{and}\quad \boldsymbol{e}_2(\hat{\boldsymbol{k}})=\frac{1}{n_2}\begin{bmatrix}-\sin\varphi_{\boldsymbol{k}}\\ \cos\varphi_{\boldsymbol{k}}\\ 0\end{bmatrix}. \tag{86}$$

In the *extraordinary* light mode, the refractive index depends on $\hat{\boldsymbol{k}}$, and the polarization vector $\boldsymbol{e}_1(\hat{\boldsymbol{k}})$ is generally not perpendicular to $\hat{\boldsymbol{k}}$. It follows from $\boldsymbol{e}_1(\hat{\boldsymbol{k}})=\boldsymbol{\varepsilon}_0^{-1}\boldsymbol{d}^1(\hat{\boldsymbol{k}})$ where the polarization vector $\boldsymbol{d}^1(\hat{\boldsymbol{k}})$ is determined by $\boldsymbol{d}^1(\hat{\boldsymbol{k}})\perp\boldsymbol{d}^2(\hat{\boldsymbol{k}})$ and $\boldsymbol{d}^1(\hat{\boldsymbol{k}})\perp\hat{\boldsymbol{k}}$. The refractive index can be calculated from the eigenvalue equation (9) or Fresnel's equation (15):

$$n_1(\hat{\boldsymbol{k}})=n_2\tilde{n}_1(\hat{\boldsymbol{k}}) \quad\text{with}\quad \tilde{n}_1(\hat{\boldsymbol{k}})=\left[\frac{1+\alpha}{1+\alpha\cos^2\vartheta_{\boldsymbol{k}}}\right]^{1/2}, \tag{87}$$

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

$$\alpha=\Delta\varepsilon/\varepsilon_{\perp}. \tag{88}$$

Equation (87) does not represent the most symmetric form for $n_1(\hat{\boldsymbol{k}})$, but we found it useful for our calculations. Finally, we get

$$\boldsymbol{e}_1(\hat{\boldsymbol{k}})=\frac{\tilde{n}_1(\hat{\boldsymbol{k}})}{n_2}\left(\cos\vartheta_{\boldsymbol{k}}\begin{bmatrix}\cos\varphi_{\boldsymbol{k}}\\ \sin\varphi_{\boldsymbol{k}}\\ 0\end{bmatrix}-\frac{\sin\vartheta_{\boldsymbol{k}}}{1+\alpha}\begin{bmatrix}0\\0\\1\end{bmatrix}\right). \tag{89}$$

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

$$C_{\boldsymbol{k}}:=\tilde{n}_1(\hat{\boldsymbol{k}})\cos\vartheta_{\boldsymbol{k}} \quad\text{and}\quad S_{\boldsymbol{k}}:=\tilde{n}_1(\hat{\boldsymbol{k}})\sin\vartheta_{\boldsymbol{k}}, \tag{90}$$

in which the refraction index takes the form

$$\tilde{n}_1(\hat{\boldsymbol{k}})=\sqrt{1+\alpha(1-C_{\boldsymbol{k}}^2)}. \tag{91}$$

The "trigonometric" identity,

$$S_{\boldsymbol{k}}^2+(1+\alpha)C_{\boldsymbol{k}}^2=1+\alpha \tag{92}$$

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

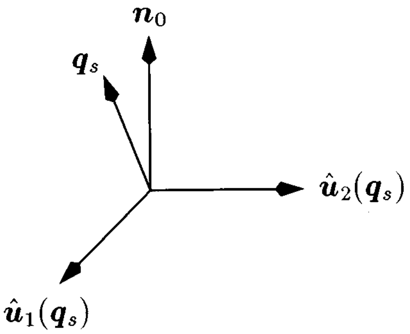

FIG. 4. Basis vectors $\hat{\boldsymbol{u}}_1(\boldsymbol{q}_s)$ and $\hat{\boldsymbol{u}}_2(\boldsymbol{q}_s)$ for given director $\boldsymbol{n}_0$ and wave vector $\boldsymbol{q}_s$.

$$(1+\alpha)dC_{\boldsymbol{k}}=\tilde{n}_1^3(\hat{\boldsymbol{k}})d\cos\vartheta_{\boldsymbol{k}}. \tag{93}$$

The differential $dC_{\boldsymbol{k}}$ absorbs the factor $\tilde{n}_1^3(\hat{\boldsymbol{k}})$, which always comes with our integrals thus making them easier:

$$\int_{\hat{\boldsymbol{k}}^1}\cdots=\int\frac{d\Omega_{\boldsymbol{k}}}{(2\pi)^3}n_1^3(\hat{\boldsymbol{k}})\cdots=n_2^3(1+\alpha)\int\frac{dC_{\boldsymbol{k}}d\varphi_{\boldsymbol{k}}}{(2\pi)^3}\cdots. \tag{94}$$

Alternatively, we can say that we have constructed a complete set of basis functions in the variables $\cos\vartheta_{\boldsymbol{k}}$ and $\varphi_{\boldsymbol{k}}$ with respect to the weight function $\tilde{n}_1(\hat{\boldsymbol{k}})$. In the following we will also use the notation $C_{\boldsymbol{k}}=\cos\vartheta_{\boldsymbol{k}}$ and $S_{\boldsymbol{k}}=\sin\vartheta_{\boldsymbol{k}}$ 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,

$$[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t)]_{\alpha\beta}=\frac{\omega^4}{c^4}\langle\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})\cdot\delta\boldsymbol{\varepsilon}(\boldsymbol{q}_s,t)\boldsymbol{e}_{\beta}(\hat{\boldsymbol{q}})\times[\boldsymbol{e}_{\alpha}(\hat{\boldsymbol{k}})\cdot\delta\boldsymbol{\varepsilon}(\boldsymbol{q}_s,0)\boldsymbol{e}_{\beta}(\hat{\boldsymbol{q}})]^*\rangle, \tag{95}$$

where $\boldsymbol{q}_s=(\omega/c)(n_{\alpha}\hat{\boldsymbol{k}}-n_{\beta}\hat{\boldsymbol{q}})$ stands for the scattering vector. We first need the Fourier component $\delta\boldsymbol{\varepsilon}(\boldsymbol{q}_s,t)$ of the fluctuating part of the dielectric tensor. We insert $\boldsymbol{n}(\boldsymbol{r},t)=\boldsymbol{n}_0+\delta\boldsymbol{n}(\boldsymbol{r},t)$ into $\boldsymbol{\varepsilon}(\boldsymbol{r},t)$ of Eq. (81) and collect the first-order terms in $\delta\boldsymbol{n}(\boldsymbol{r},t)$. After a Fourier transformation we arrive at

$$\delta\boldsymbol{\varepsilon}(\boldsymbol{q}_s,t)=\Delta\varepsilon[\boldsymbol{n}_0\otimes\delta\boldsymbol{n}(\boldsymbol{q}_s,t)+\delta\boldsymbol{n}(\boldsymbol{q}_s,t)\otimes\boldsymbol{n}_0] \tag{96}$$

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

$$\delta\boldsymbol{n}(\boldsymbol{q}_s,t)=\delta n_1(\boldsymbol{q}_s,t)\hat{\boldsymbol{u}}_1(\boldsymbol{q}_s)+\delta n_2(\boldsymbol{q}_s,t)\hat{\boldsymbol{u}}_2(\boldsymbol{q}_s). \tag{97}$$

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

$$\langle\delta\boldsymbol{n}(\boldsymbol{q}_s,t)\otimes\delta\boldsymbol{n}^*(\boldsymbol{q}_s,0)\rangle=\sum_{\delta=1}^{2}\frac{k_BT}{K_{\delta}(\boldsymbol{q}_s)}\exp\left[-\frac{K_{\delta}(\boldsymbol{q}_s)}{\eta_{\delta}(\boldsymbol{q}_s)}t\right]\hat{\boldsymbol{u}}_{\delta}(\boldsymbol{q}_s)\otimes\hat{\boldsymbol{u}}_{\delta}(\boldsymbol{q}_s) \tag{98}$$

with

$$K_{\delta}(\boldsymbol{q}_s)=K_{\delta}q_{\perp}^2+K_3q_{\parallel}^2+\Delta\chi H^2, \tag{99}$$

where $q_{\perp}$ and $q_{\parallel}$ are the components of $\boldsymbol{q}_s$, perpendicular and parallel to $\boldsymbol{n}_0$. The chosen basis for $\delta\boldsymbol{n}(\boldsymbol{q}_s,t)$ provides the normal coordinates, because the correlation function is already diagonal. The free energy of one director mode follows from the Frank-Oseen-Zöcher free energy (82) as $K_{\delta}(\boldsymbol{q}_s)|\delta n_{\delta}(\boldsymbol{q}_s,t)|^2$. For a general vector $\boldsymbol{q}_s$ it is either composed of splay and bend ($\delta=1$) or twist and bend distortions ($\delta=2$). The factor $k_BT/K_{\delta}(\boldsymbol{q}_s)$ 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 [55]. They combine the Navier-Stokes equation for a uniaxial media with dynamical equations for the director. A detailed analysis [55] shows that director modes are diffusive with a relaxation frequency given by the quotient of elastic $[K_{\delta}(\boldsymbol{q}_s)]$ and viscous $[\eta_{\delta}(\boldsymbol{q}_s)]$ forces. This is the origin of the exponential factor in the correlation function. The viscosity $\eta_{\delta}(\boldsymbol{q}_s)$ is a combination of several Leslie viscosities, which we will address in Sec. IV D. We neglect a fast relaxing part that comes from the coupling between the director and the fluid motion. Finally we are able to write down the structure factor

$$[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t)]_{\alpha\beta}=(\Delta\varepsilon)^2k_BT\,\frac{\omega^4}{c^4}\sum_{\delta=1}^{2}\frac{N(\boldsymbol{e}_{\alpha},\boldsymbol{e}_{\beta},\hat{\boldsymbol{u}}_{\delta})}{K_{\delta}(\boldsymbol{q}_s)}\times\exp\left[-\frac{K_{\delta}(\boldsymbol{q}_s)}{\eta_{\delta}(\boldsymbol{q}_s)}t\right] \tag{100}$$

with

$$N(\boldsymbol{e}_{\alpha},\boldsymbol{e}_{\beta},\hat{\boldsymbol{u}}_{\delta})=[(\boldsymbol{n}_0\cdot\boldsymbol{e}_{\beta})(\hat{\boldsymbol{u}}_{\delta}\cdot\boldsymbol{e}_{\alpha})+(\hat{\boldsymbol{u}}_{\delta}\cdot\boldsymbol{e}_{\beta})(\boldsymbol{n}_0\cdot\boldsymbol{e}_{\alpha})]^2 \tag{101}$$

a geometrical factor. It has two interesting implications. First, there exists no scattering of an ordinary light ray into an ordinary light ray because $\boldsymbol{e}_2\perp\boldsymbol{n}_0$, and therefore $N=0$. Second, forward and backward scattering along $\boldsymbol{n}_0$ is always forbidden. The other terms of $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(t)]_{\alpha\beta}$ are familiar from the previous discussion. The scattering mean free path $l_{\alpha}(\hat{\boldsymbol{k}},\omega)$, defined in Eq. (39), has been already discussed in detail by two groups [56,57]. Its dependence on $\hat{\boldsymbol{k}}$ and polarization was calculated by Val'kov and Romanov [56]. We just stress one point. The structure factor diverges for $H=0$ and $\boldsymbol{q}_s\to\boldsymbol{0}$, and we expect the scattering mean free paths to be zero in an infinite medium. But $\boldsymbol{q}_s=\boldsymbol{0}$ can only occur in scattering events where the extraordinary polarization is preserved. Hence, only the scattering mean free path $l_1(\hat{\boldsymbol{k}},\omega)$ of an extraordinary light ray is zero. In Appendix C we give $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}$ for $\boldsymbol{q}^1\to\boldsymbol{k}^1$ (the notation we use there is explained below). From this form it is obvious that $l_1^{-1}(\hat{\boldsymbol{k}},\omega)$ diverges weakly like $-\ln[\Delta\chi H^2/(K_3n_2^2\omega^2/c^2)]$ for magnetic fields much smaller than $\sqrt{K_3/\Delta\chi}n_2\omega/c$. The explicit form of the structure factor is very complex because it explicitly depends on the direction of the incoming ($\hat{\boldsymbol{k}}$) and outgoing ($\hat{\boldsymbol{q}}$) light through the geometrical factor. We mention some symmetry properties that facilitate its handling. The first two are quite obvious:

$$[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}=[\boldsymbol{B}^{\omega}_{\boldsymbol{q}^{\beta}\boldsymbol{k}^{\alpha}}(0)]_{\beta\alpha} \tag{102}$$

and

$$[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}=[\boldsymbol{B}^{\omega}_{-\boldsymbol{k}^{\alpha}-\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}. \tag{103}$$

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 $\boldsymbol{n}_0$. If we choose

$$\hat{\boldsymbol{k}}=\begin{bmatrix}\sin\vartheta_{\boldsymbol{k}}\cos\varphi_{\boldsymbol{k}}\\ \sin\vartheta_{\boldsymbol{k}}\sin\varphi_{\boldsymbol{k}}\\ \cos\vartheta_{\boldsymbol{k}}\end{bmatrix} \quad \text{and} \quad \hat{\boldsymbol{q}}=\begin{bmatrix}\sin\vartheta_{\boldsymbol{q}}\cos\varphi_{\boldsymbol{q}}\\ \sin\vartheta_{\boldsymbol{q}}\sin\varphi_{\boldsymbol{q}}\\ \cos\vartheta_{\boldsymbol{q}}\end{bmatrix}, \tag{104}$$

then $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}$ depends only on the relative azimuthal angle $\varphi=\varphi_{\boldsymbol{q}}-\varphi_{\boldsymbol{k}}$. The existence of mirror planes, containing $\boldsymbol{n}_0$, implies that the structure factor should be invariant under $\varphi\rightarrow-\varphi$. In Appendix C we give its explicit form in terms of $C_{\boldsymbol{k}}$, $S_{\boldsymbol{k}}$, $C_{\boldsymbol{q}}$, $S_{\boldsymbol{q}}$, and φ. We also introduce a scaled structure factor $[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}$ through

$$[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}=(\Delta\varepsilon)^2\frac{\omega^2}{c^2}\frac{k_BT}{K_3n_2^6}[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}, \tag{105}$$

which depends on scaled parameters: the relative dielectric anisotropy $\alpha=\Delta\varepsilon/\varepsilon_{\perp}$, the Frank elastic constants

$$\overline{K_1}=K_1/K_3 \quad \text{and} \quad \overline{K_2}=K_2/K_3, \tag{106}$$

and the magnetic field

$$h=H/H_0 \quad \text{with} \quad H_0=n_2\frac{\omega}{c}\sqrt{\frac{K_3}{\Delta\chi}}. \tag{107}$$

If we introduce the magnetic coherence length

$$\xi_3=\sqrt{\frac{K_3}{\Delta\chi H^2}}, \tag{108}$$

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

$$h=\frac{\lambda}{2\pi\xi_3}, \tag{109}$$

where $\lambda=n_2\omega/c$. Thus, $h=1$ 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 5CB 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 [58]. The bend elastic constant is $K_3=5.3\times10^{-7}$dyn and is, as usual for conventional thermotropic nematic liquid crystals, larger than the splay and twist constants: $\overline{K_1}=0.79$ and $\overline{K_2}=0.43$. For green light ($\omega/c=1.15\times10^5$ cm^{-1}) the dielectric constant is $\varepsilon_{\perp}=2.381$ and the anisotropy is $\alpha=0.228$ corresponding to the refractive indices $n_1(\vartheta=90°)=1.710$ and $n_2=1.543$. Finally, the magnetic anisotropy is $\Delta\chi=0.95\times10^{-7}$, from which we obtain a characteristic magnetic field of $H_0=3.6\times10^5$ G.

C. Light diffusion – results

In this subsection we discuss the two essential components $D_{\parallel}$ and $D_{\perp}$ of the diffusion tensor $\boldsymbol{D}(\omega)$, which describe the diffusion of light, respectively, parallel and perpendicular to the director $\boldsymbol{n}_0$. Before we can apply our general formulas from Eqs. (62), (63), and (71) we need to be more specific about our basis functions $\varphi_i(\hat{\boldsymbol{k}})$, and we need the derivatives of the components $[\boldsymbol{G}_0^{-1}]_{\alpha}$, which we gave in Eq. (21). The calculations for the latter are straightforward and the results for the extraordinary and ordinary light mode are, respectively,

$$\widetilde{n}_1(\hat{\boldsymbol{k}})\frac{\partial[\boldsymbol{G}_0^{-1}]_1}{\partial\boldsymbol{k}}\bigg|_{\hat{\boldsymbol{k}}}\cdot\boldsymbol{K}=-\frac{2}{\varepsilon_{\perp}}\left(\frac{\sqrt{1-C_{\boldsymbol{k}}^2}\cos\varphi_{\boldsymbol{k}}}{\sqrt{1+\alpha}}K_{\perp}+C_{\boldsymbol{k}}K_{\parallel}\right) \tag{110}$$

and

$$\frac{\partial[\boldsymbol{G}_0^{-1}]_2}{\partial\boldsymbol{k}}\bigg|_{\hat{\boldsymbol{k}}}\cdot\boldsymbol{K}=-\frac{2}{\varepsilon_{\perp}}(\sqrt{1-C_{\boldsymbol{k}}^2}\cos\varphi_{\boldsymbol{k}}K_{\perp}+C_{\boldsymbol{k}}K_{\parallel}). \tag{111}$$

In the above, we chose

$$\boldsymbol{K}=[K_{\perp},0,K_{\parallel}], \tag{112}$$

wrote $\hat{\boldsymbol{k}}$ in spherical coordinates as in Eq. (104) and then switched to the new C coordinate. The right-hand side of Eq. (111) for the ordinary mode is the isotropic result $\hat{\boldsymbol{k}}\cdot\boldsymbol{K}$, which is modified in the extraordinary case. As our basis functions, we choose

$$\varphi_i^{\alpha}(\hat{\boldsymbol{k}})\rightarrow\varphi_{\gamma lm}^{\alpha}(\hat{\boldsymbol{k}})=\delta_{\gamma}^{\alpha}\varphi_{lm}(C_{\boldsymbol{k}},\varphi_{\boldsymbol{k}}), \tag{113}$$

where $\varphi_{lm}(C_{\boldsymbol{k}},\varphi_{\boldsymbol{k}})$ stands for a real combination of the spherical harmonics $Y_{lm}(C_{\boldsymbol{k}},\varphi_{\boldsymbol{k}})$ and $Y_{l-m}(C_{\boldsymbol{k}},\varphi_{\boldsymbol{k}})$ in the new coordinate $C_{\boldsymbol{k}}$ instead of $\cos\vartheta_{\boldsymbol{k}}$. It will soon become clear why the coordinate $C_{\boldsymbol{k}}$ is so helpful. The index γ stands for the basis of the tensor space, which we identify here with our basis $\boldsymbol{e}_{\alpha}\otimes\boldsymbol{e}_{\alpha}$. 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 $[\boldsymbol{G}_0^{-1}]_{\alpha}$. Furthermore, we only need functions $\varphi_{lm}(C_{\boldsymbol{k}},\varphi_{\boldsymbol{k}})$ containing $\cos m\varphi_{\boldsymbol{k}}$ ($m\geqslant0$); $\sin m\varphi_{\boldsymbol{k}}$ is not necessary because of the symmetry of $[\boldsymbol{B}^{\omega}_{\boldsymbol{k}^{\alpha}\boldsymbol{q}^{\beta}}(0)]_{\alpha\beta}$. With this choice of basis functions the matrix $\mathcal{B}$ is decomposed into submatrices for each m, $[\mathcal{B}]^{\gamma lm}_{\gamma' l' m'}\propto\delta_{mm'}$, because a term $\cos m\varphi_{\boldsymbol{k}}\cos n\varphi_{\boldsymbol{q}}$ ($n\neq m$) is not compatible with the rotational symmetry of the structure factor. Then, the diffusion constants $D_{\parallel}$ and $D_{\perp}$ are, respectively, related to $m=0$ and $m=1$:

$$D_{\parallel}K_{\parallel}^{2}=\frac{c}{2\overline{n^{3}}}\sum_{\gamma\gamma' l l'}[\mathcal{G}(\boldsymbol{K})]_{\gamma l 0}[\mathcal{B}^{-1}]^{\gamma l 0}_{\gamma' l' 0}[\mathcal{G}(\boldsymbol{K})]_{\gamma' l' 0}, \tag{114}$$

$$D_{\perp}K_{\perp}^{2}=\frac{c}{2\overline{n^{3}}}\sum_{\gamma\gamma' l l'}[\mathcal{G}(\boldsymbol{K})]_{\gamma l 1}[\mathcal{B}^{-1}]^{\gamma l 1}_{\gamma' l' 1}[\mathcal{G}(\boldsymbol{K})]_{\gamma' l' 1} \tag{115}$$

with

$$\overline{n^{3}}=n_{2}^{3}(1+\alpha/2)=(\varepsilon_{\perp})^{3/2}(1+\alpha/2). \tag{116}$$

Only elements of $\mathcal{G}(\boldsymbol{K})$ with $l=1$ are nonzero, $[\mathcal{G}(\boldsymbol{K})]_{\gamma 1 m}\neq 0$, since the right-hand sides of Eqs. (110) and (111) depend only on $l=1$ basis functions. This raises the question of how important higher-l contributions are. For our calculations we, therefore, choose $l=1$ and $l=3$ functions:

$$\varphi_{10}(C,\varphi)=\sqrt{6}\pi C, \tag{117a}$$

$$\varphi_{11}(C,\varphi)=\sqrt{6}\pi\sqrt{1-C^{2}}\cos\varphi, \tag{117b}$$

$$\varphi_{30}(C,\varphi)=\sqrt{\tfrac{7}{2}}\pi C(5C^{2}-3), \tag{117c}$$

$$\varphi_{31}(C,\varphi)=\frac{\sqrt{21}}{2}\pi\sqrt{1-C^{2}}(1-5C^{2})\cos\varphi \tag{117d}$$

with the normalization

$$\int\frac{dC\,d\varphi}{(2\pi)^{3}}\varphi_{lm}(C,\varphi)\varphi_{l'm'}(C,\varphi)=\delta_{ll'}\delta_{mm'}. \tag{118}$$

The nonzero components of $\mathcal{G}(\boldsymbol{K})$ turn out to be

$$[\mathcal{G}(\boldsymbol{K})]_{110}=-2\varepsilon_{\perp}(1+\alpha)K_{\parallel}/\sqrt{6}, \tag{119a}$$

$$[\mathcal{G}(\boldsymbol{K})]_{210}=-2\varepsilon_{\perp}K_{\parallel}/\sqrt{6}, \tag{119b}$$

$$[\mathcal{G}(\boldsymbol{K})]_{111}=-2\varepsilon_{\perp}\sqrt{1+\alpha}K_{\perp}/\sqrt{6}, \tag{119c}$$

$$[\mathcal{G}(\boldsymbol{K})]_{211}=-2\varepsilon_{\perp}K_{\perp}/\sqrt{6}. \tag{119d}$$

The integrations to obtain the matrix elements of $\mathcal{B}$ require more effort. We were able to calculate the integrals over φ analytically with the help of

$$\int d\varphi_{\boldsymbol{k}}d\varphi_{\boldsymbol{q}}f(\varphi_{\boldsymbol{q}}-\varphi_{\boldsymbol{k}})=2\pi\int d\varphi f(\varphi), \tag{120a}$$

$$\int d\varphi_{\boldsymbol{k}}d\varphi_{\boldsymbol{q}}f(\varphi)\cos\varphi_{\boldsymbol{k}}\cos\varphi_{\boldsymbol{q}}=\pi\int d\varphi f(\varphi)\cos\varphi, \tag{120b}$$

$$\int d\varphi_{\boldsymbol{k}}d\varphi_{\boldsymbol{q}}f(\varphi)\cos^{2}\varphi_{\boldsymbol{k}}=\pi\int d\varphi f(\varphi). \tag{120c}$$

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,

$$l_{0}^{*}=9\pi\frac{c_{\perp}^{2}}{\omega^{2}k_{B}T}\frac{K_{3}}{\alpha^{2}} \tag{121}$$

and write the diffusion constant $D_{\parallel}$ and $D_{\perp}$ in the form

$$D_{\parallel}=c_{\perp}l_{0}^{*}\tilde{D}_{\parallel}/3,\quad D_{\perp}=c_{\perp}l_{0}^{*}\tilde{D}_{\perp}/3, \tag{122}$$

reminiscent of isotropic systems. The numerical factors $\tilde{D}_{\parallel}$ and $\tilde{D}_{\perp}$ only depend on the scaled parameters α, 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. [42]). The factor 9π 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 $N(\boldsymbol{e}_{\alpha},\boldsymbol{e}_{\beta},\hat{\boldsymbol{u}}_{\delta})$ of Eq. (101). 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 Eqs. (114) and (115) 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 $N(\boldsymbol{e}_{\alpha},\boldsymbol{e}_{\beta},\hat{\boldsymbol{u}}_{\delta})$ 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 [41,42].

In the following we will explore the dependence of the diffusion constants on the scaled parameters K_1, K_2, α, and h. In Figs. 5 and 6 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. 6. 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. 7 how the diffusion constants $\tilde{D}_{\parallel}$ and $\tilde{D}_{\perp}$ and the relative anisotropy $(D_{\parallel}-D_{\perp})/D_{\perp}$ 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 5×10^{4} G, which corresponds to a magnetic coherence length ξ_3 of approximately 1 μm, the changes in $\tilde{D}_{\parallel}$ and $\tilde{D}_{\perp}$ are small. The values for $H=0$ read $\tilde{D}_{\parallel}=0.95$ and

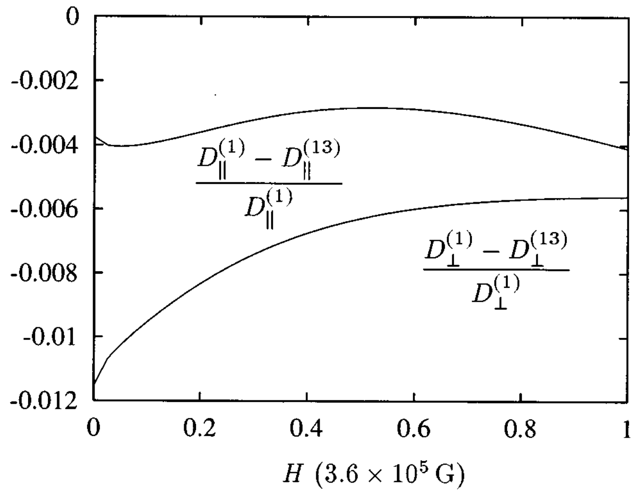

FIG. 5. Field dependence of the relative changes of diffusion constants after $l=3$ spherical harmonics (superscript 13) are included in addition to $l=1$ spherical harmonics (superscript 1). Parameters are $K_1=K_2=1$ and $\alpha=0$. The magnetic field is given relative to the characteristic field $H_0=3.6\times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

$\tilde{D}_\perp = 0.65$ with a ratio $\tilde{D}_\parallel/\tilde{D}_\perp = 1.45$ in good agreement with experiments [41,42]. The reason why we plot $\tilde{D}_\parallel$, $\tilde{D}_\perp$, and $(D_\parallel - D_\perp)/D_\perp$ 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 $\boldsymbol{q}_s$. 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 $1-\cos\vartheta_s$ in formula (75) cancels the singularity from $\boldsymbol{q}_s^2 \propto 1-\cos\vartheta_s$.

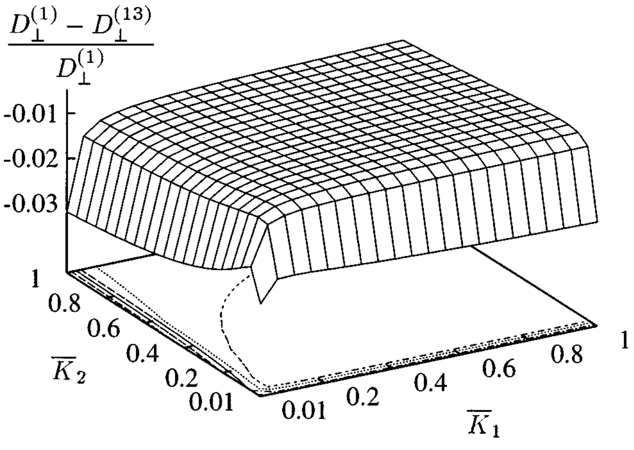

FIG. 6. Relative difference as a function of $\overline{K_1}$ and $\overline{K_2}$ at $\alpha=0$, $h=0.1$ between $D_\perp^{(1)}$ calculated with $l=1$ spherical harmonics only and $D_\perp^{(13)}$ calculated with both $l=1$ and $l=3$.

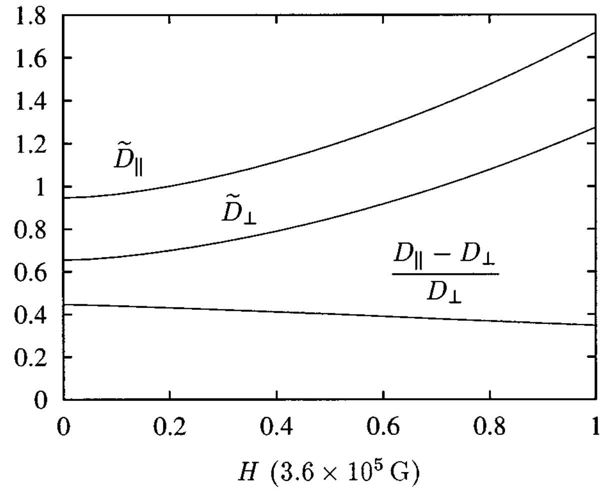

FIG. 7. Field dependence of the diffusion constants $\tilde{D}_\parallel$ and $\tilde{D}_\perp$ and the relative anisotropy $(D_\parallel - D_\perp)/D_\perp$ for the nematic compound 5CB ($K_1=0.79$, $K_2=0.43$, $\alpha=0.228$, and $H_0=3.6\times 10^5$ G).

In Fig. 8 we explore the anisotropy in the diffusion depending on the anisotropy α in the dielectric constants. As already discussed, when $\alpha=0$ there is a small nonzero value of $(D_\parallel - D_\perp)/D_\perp$. This grows with α 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 α is negative.

Finally, we discuss the dependence of the diffusion on the elastic constants K_1 and K_2. In Fig. 9 we show that $\tilde{D}_\perp$ 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 $\tilde{D}_\perp = 0.07$. The contour lines reveal an asymmetry between the splay (K_1) and the twist (K_2) distortions. $\tilde{D}_\perp$ decreases more strongly with K_2. The diffusion constant $\tilde{D}_\parallel$ shows a similar behavior. Figure 10 gives the anisotropy $(D_\parallel - D_\perp)/D_\perp$ for the same range. It grows with decreasing elastic constants showing that $\tilde{D}_\perp$ is more affected by splay and twist distortions than $\tilde{D}_\parallel$. The asymmetry between 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

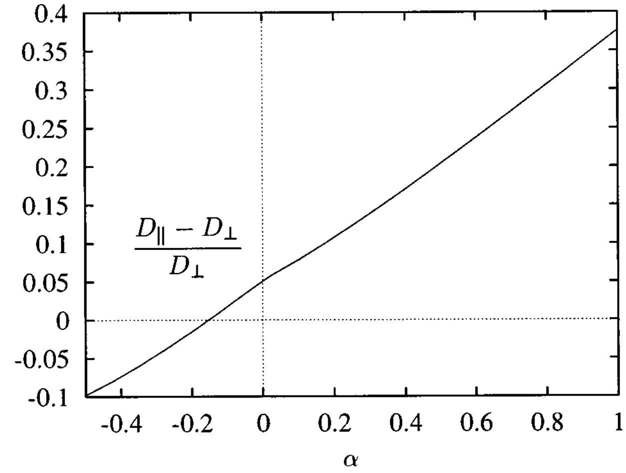

FIG. 8. Relative anisotropy $(D_\parallel - D_\perp)/D_\perp$ depending on the dielectric anisotropy α for $K_1=K_2=1$ and $h=0.01$.

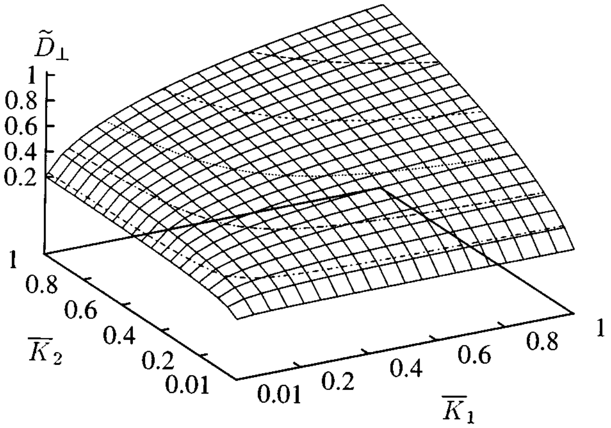

FIG. 9. Diffusion constant $\tilde D_\perp$ depending on $\bar K_1$ and $\bar K_2$. Parameters are $\alpha=0$ and $h=0.1$.

this range to $\bar K_1=\bar K_2=10$ and observe that the anisotropy $(D_\parallel-D_\perp)/D_\perp$ 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 $\bar K_1>0.6$ and $\bar K_2>1.4$. In a smectic-*A* phase (Sm-*A*) twist and bend deformations are expelled by the layered structure [59], hence $\bar K_1\ll 1$. 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 Sm-*A*-nematic phase transition, where the layered structure softens (Sm-*A*) 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 $\bar K_1=0.85$ and $\bar K_2=0.07$ with an absolute value of $K_3=4.7\times10^{-7}$ dyn, 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 α and scatters light only weakly. However, it should be possible to find systems that are more favorable regarding α.

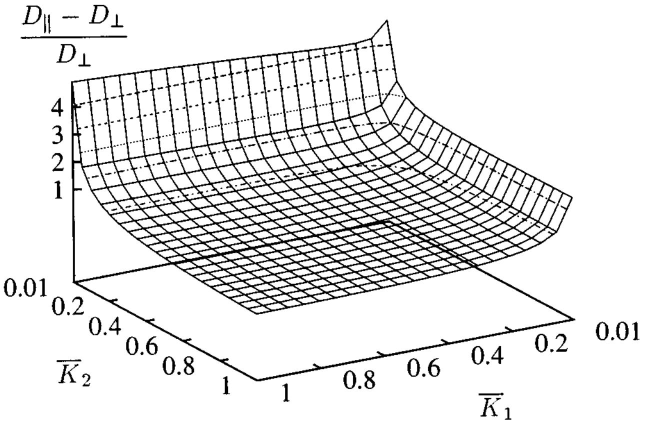

FIG. 10. Relative anisotropy $(D_\parallel-D_\perp)/D_\perp$ depending on $\bar K_1$ and $\bar K_2$, which range from 0.01 to 1. Parameters are $\alpha=0$ and $h=0.1$.

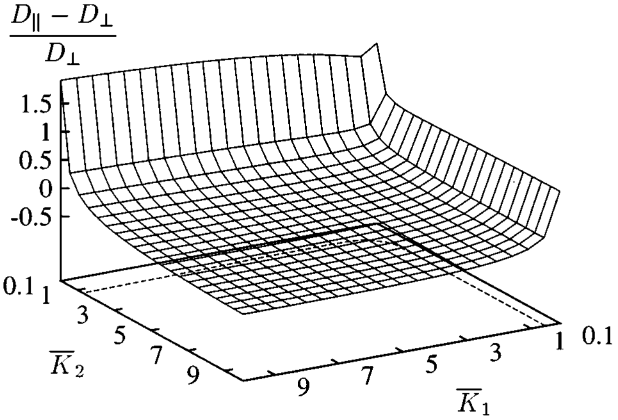

FIG. 11. Relative anisotropy $(D_\parallel-D_\perp)/D_\perp$ depending on $\bar K_1$ and $\bar K_2$, which range from 0.1 to 10. Parameters are $\alpha=0$ and $h=0.1$.

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

$$\begin{aligned}\frac{D_\parallel-D_\perp}{D_\perp}\approx{}&0.06-0.1(\bar K_1-1)-0.3(\bar K_2-1)\\&+0.3\alpha+0.1(\bar K_1-1)^2+0.2(\bar K_2-1)^2\\&-0.08(\bar K_1-1)(\bar K_2-1),\end{aligned}\tag{123}$$

where the coefficients are material independent. Second order terms in α and couplings to $\bar K_1$ and $\bar K_2$ 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. (64). Director modes are purely diffusive, as described by Eq. (100) for the structure factor, and possess a viscosity $\eta_\delta(\boldsymbol{q}_s)$, which we specify here [54]:

$$\eta_1(\boldsymbol{q}_s)=\gamma-\frac{(\mu_3 q_\perp^2-\mu_2 q_\parallel^2)^2}{\eta_b q_\perp^4+\eta_c q_\parallel^4+\eta_m q_\perp^2 q_\parallel^2},\tag{124a}$$

$$\eta_2(\boldsymbol{q}_s)=\gamma-\frac{\mu_2^2 q_\parallel^2}{\eta_a q_\perp^2+\eta_b q_\parallel^2}\tag{124b}$$

with

$$\eta_a=\mu_4/2,\tag{125a}$$

$$\eta_b=(-\mu_2+\mu_4+\mu_5)/2,\tag{125b}$$

$$\eta_c=(\mu_3+\mu_4+\mu_6)/2,\tag{125c}$$

$$\eta_m=\mu_1+\eta_b+\eta_c.\tag{125d}$$

The Leslie viscosities μ_i govern the viscous flow of the fluid and couple it to the director motion. The Miesowicz viscosities η_a, η_b, and η_c can be measured in pure flow experiments. The rotational viscosity γ characterizes viscous forces due to rotations of the director. With the explicit formula (100) for the structure factor and small enough times to expand the exponential time factor, $\mu(\omega,t)$ becomes proportional to time t,

$$\mu(\omega,t)\approx(\Delta\varepsilon)^2 k_B T \, \frac{\pi^3 \ \omega^4}{2\overline{n}^3 c^3} \sum_{\alpha,\beta,\delta} \int_{\hat{\boldsymbol{k}}^{\alpha}} \int_{\hat{\boldsymbol{q}}^{\beta}} \frac{N(\boldsymbol{e}_\alpha,\boldsymbol{e}_\beta,\hat{\boldsymbol{u}}_\delta)}{\eta_\delta(\boldsymbol{q}_s)} t. \tag{126}$$

If we collect all the prefactors, the dynamic absorption coefficient can be written as

$$\mu(\omega,t)=\mu_0 t \quad \text{with} \quad \mu_0=\alpha^2 \frac{2k_B T\omega^4}{9\pi \ c_\perp^3} \frac{\tilde{\mu}}{\gamma}. \tag{127}$$

Here the numerical factor $\tilde{\mu}$ is a dimensionless angular average involving the geometrical factor and the viscosities of the director modes. It depends on the Leslie viscosities relative to γ and the dielectric anisotropy α, and it is equal 1 if $\eta_\delta(\boldsymbol{q}_s)=\gamma$ and $\alpha=0$. When $\eta_\delta(\boldsymbol{q}_s)=\gamma$, $\tilde{\mu}$ can be evaluated analytically even for $\alpha\neq 0$, and we find $\tilde{\mu}=(1+\alpha/4)/(1+\alpha/2)$. For the compound 5CB, $\gamma/\tilde{\mu}=0.60\pm 0.20$ P was experimentally determined by Kao *et al.* using DWS and the last formula [41,42]. This value is in good agreement with the rotational viscosity $\gamma=0.81$ P of 5CB [58] and shows the validity of the theory. Of course γ is larger than the measured $\gamma/\tilde{\mu}$ since $\eta_\delta(\boldsymbol{q}_s)$ is smaller than γ [see Eqs. (124a) and (124b)] so that $\tilde{\mu}$ exceeds 1. However, the values of the Leslie viscosities are such that $\tilde{\mu}$ is of order 1 in usual thermotropic nematics. Furthermore, the Leslie-Erickson theory seems to describe the director modes properly down to 4×10^{-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 $\eta_\delta(\boldsymbol{q}_s)$ for different values of $\boldsymbol{q}_s$ differ by factors of 10^2-10^3 are polymer nematic liquid crystals [62] 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 (126) 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 [49] $\mu_0=2D_B\omega^2/(l^* c)$, 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

$$\begin{aligned}&\boldsymbol{\Phi}(\boldsymbol{x}_1,\boldsymbol{y}_1,\tau_1,\overline{\tau_1};\boldsymbol{x}_2,\boldsymbol{y}_2,\tau_2,\overline{\tau_2})\\ &\quad=\langle \boldsymbol{G}^R(\boldsymbol{x}_1,\boldsymbol{x}_2;\tau_1,\tau_2)\otimes\boldsymbol{G}^A(\boldsymbol{y}_2,\boldsymbol{y}_1;\overline{\tau_2},\overline{\tau_1})\rangle^{(N)}.\end{aligned} \tag{A1}$$

After integration over electric field sources $\boldsymbol{W}_0(2)$ at points $(\boldsymbol{x}_2,\tau_2)$ and $(\boldsymbol{y}_2,\overline{\tau_2})$ it provides us with the full autocorrelation function $\boldsymbol{W}(1)$ of the electric light field at $(\boldsymbol{x}_1,\tau_1)$ and $(\boldsymbol{y}_1,\overline{\tau_1})$:

$$\boldsymbol{W}(1)=\int d2\boldsymbol{\Phi}(1,2)\boldsymbol{W}_0(2). \tag{A2}$$

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]

$$\boldsymbol{\Phi}(1,4)=\boldsymbol{f}(1,4)+\int d2d3\boldsymbol{f}(1,2)\boldsymbol{U}(2,3)\boldsymbol{\Phi}(3,4). \tag{A3}$$

The quantity $\boldsymbol{f}(1,2)$ stands for the tensor product of the averaged one-particle Green functions:

$$\boldsymbol{f}(1,2)=[\langle \boldsymbol{G}^R\rangle(\boldsymbol{x}_1-\boldsymbol{x}_2;\tau_1-\tau_2)\otimes\langle \boldsymbol{G}^A\rangle(\boldsymbol{y}_2-\boldsymbol{y}_1;\overline{\tau_2}-\overline{\tau_1})]^{(N)}. \tag{A4}$$

It propagates two electric field modes from 2 to 1 between their scattering events. The irreducible vertex function $\boldsymbol{U}$ 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'' $(\boldsymbol{R}_i,T_i)$ and relative $(\boldsymbol{r}_i,t_i)$ coordinates:

$$\begin{aligned}\boldsymbol{x}_i&=\boldsymbol{R}_i+\boldsymbol{r}_i/2, \quad \boldsymbol{y}_i=\boldsymbol{R}_i-\boldsymbol{r}_i/2,\\ \tau_i&=T_i+t_i/2, \quad \overline{\tau_i}=T_i-t_i/2.\end{aligned} \tag{A5}$$

It is straightforward to show that the Jacobian determinant for this coordinate transformation is 1, and we have

$$di=d^3R_i d^3r_i dTdt. \tag{A6}$$

All of our quantities only depend on differences in the center-of-''mass'' coordinates $\boldsymbol{R}_i$ and T_i because of the homogeneity in space time. To discuss the Bethe-Salpeter equation we perform a Fourier transformation,

$$\begin{aligned}&\int d^3(R_i-R_j)d^3(T_i-T_j)d^3r_i d^3r_j d^3t_i d^3t_j\cdots\\ &\quad\times\exp\{-i[\boldsymbol{K}\cdot(\boldsymbol{R}_i-\boldsymbol{R}_j)+\boldsymbol{k}_i\cdot\boldsymbol{r}_i-\boldsymbol{k}_j\cdot\boldsymbol{r}_j]\}\\ &\quad\times\exp\{i[\Omega(T_i-T_j)+\omega_i t_i-\omega_j t_j]\},\end{aligned} \tag{A7}$$

which transforms our quantities as follows:

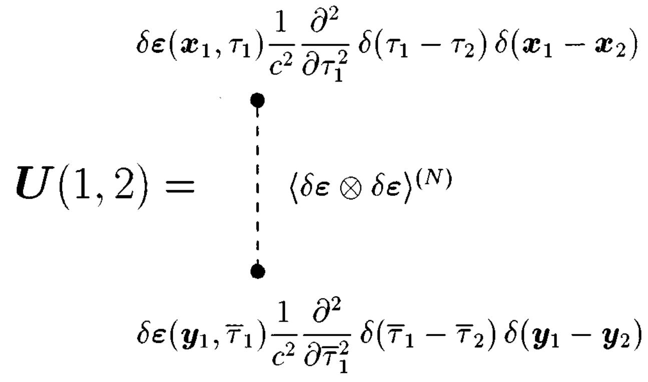

FIG. 12. The irreducible vertex function $\boldsymbol{U}(1,2)$ in the weak-scattering approximation. The second pair of points in space time is tied to the structure factor via delta functions.

$$\begin{aligned}\boldsymbol{\Phi}(1,4)&\rightarrow\boldsymbol{\Phi}_{\boldsymbol{k}_1\boldsymbol{k}_4}^{\omega_1\omega_4}(\boldsymbol{K},\Omega),\\ \boldsymbol{U}(2,3)&\rightarrow\boldsymbol{U}_{\boldsymbol{k}_2\boldsymbol{k}_3}^{\omega_2\omega_3}(\boldsymbol{K},\Omega),\\ \boldsymbol{f}(1,2)&\rightarrow\boldsymbol{f}_{\boldsymbol{k}_1}^{\omega_1}(\boldsymbol{K},\Omega)\,\delta_{\omega_1\omega_2}\mathbf{1}_{\boldsymbol{k}_1\boldsymbol{k}_2}^{(4)},\end{aligned}\tag{A8}$$

where $\delta_{\omega_1\omega_2}=2\pi\delta(\omega_1-\omega_2)$, and $\mathbf{1}_{\boldsymbol{k}_1\boldsymbol{k}_2}^{(4)}$, defined in Eq. (51), contains $\delta(\boldsymbol{k}_1-\boldsymbol{k}_2)$. The δ functions appear since $\boldsymbol{f}(1,2)$ only depends on the differences of the relative coordinates. In Eq. (50) we give the explicit form of $\boldsymbol{f}_{\boldsymbol{k}_1}^{\omega_1}(\boldsymbol{K},\Omega)$. The Fourier transformed Bethe-Salpeter equation finally takes the form

$$\begin{aligned}\int\frac{d^3k_1}{(2\pi)^3}\frac{d\omega_1}{2\pi}&[\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}_1}^{(4)}\delta_{\omega\omega_1}\\ &-\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)\boldsymbol{U}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega\omega_1}(\boldsymbol{K},\Omega)]\boldsymbol{\Phi}_{\boldsymbol{k}_1\boldsymbol{k}'}^{\omega_1\omega'}(\boldsymbol{K},\Omega)\\ &=\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}'}^{(4)}\delta_{\omega\omega'}\end{aligned}\tag{A9}$$

and the autocorrelation function $\boldsymbol{W}(\boldsymbol{K},\boldsymbol{k},\Omega,\omega)$ for light with frequency ω and wave vector $\boldsymbol{k}$ follows from

$$\boldsymbol{W}(\boldsymbol{K},\boldsymbol{k},\Omega,\omega)=\int\frac{d^3k'}{(2\pi)^3}\frac{d\omega'}{2\pi}\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega\omega'}(\boldsymbol{K},\Omega)\boldsymbol{W}_0(\boldsymbol{K},\boldsymbol{k}',\Omega,\omega').\tag{A10}$$

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 [50]

$$\begin{aligned}\boldsymbol{U}(1,2)\approx&\langle\delta\boldsymbol{\varepsilon}(\boldsymbol{x}_1,\tau_1)\otimes\delta\boldsymbol{\varepsilon}(\boldsymbol{y}_1,\overline{\tau_1})\rangle^{(N)}\frac{1}{c^4}\frac{\partial^2}{\partial\tau_1^2}\frac{\partial^2}{\partial\overline{\tau_1}^2}\\ &\times\delta(\boldsymbol{x}_1-\boldsymbol{x}_2)\,\delta(\boldsymbol{y}_1-\boldsymbol{y}_2)\,\delta(\tau_1-\tau_2)\,\delta(\overline{\tau_1}-\overline{\tau_2}).\end{aligned}\tag{A11}$$

It only considers scattering events of the two electric field modes, which are tied together through the structure factor. Figure 12 gives a graphic representation of $\boldsymbol{U}(1,2)$. In center-of-‘‘mass’’ and relative coordinates we obtain

$$\begin{aligned}\boldsymbol{U}(1,2)=&\langle\delta\boldsymbol{\varepsilon}(\boldsymbol{r}_1,t_1)\otimes\delta\boldsymbol{\varepsilon}(\mathbf{0},0)\rangle^{(N)}\frac{1}{c^4}\frac{\partial^4}{\partial t_1^4}\\ &\times\delta\left(\boldsymbol{R}_1-\boldsymbol{R}_2+\frac{\boldsymbol{r}_1-\boldsymbol{r}_2}{2}\right)\delta\left(\boldsymbol{R}_1-\boldsymbol{R}_2-\frac{\boldsymbol{r}_1-\boldsymbol{r}_2}{2}\right)\\ &\times\delta\left(T_1-T_2+\frac{t_1-t_2}{2}\right)\delta\left(T_1-T_2-\frac{t_1-t_2}{2}\right),\end{aligned}\tag{A12}$$

where we neglect the partial derivative $\partial/\partial T$, which probes time variations on much longer time scales than the time period of light. Then, in Fourier space, we obtain

$$\boldsymbol{U}_{\boldsymbol{k}_1\boldsymbol{k}_2}^{\omega_1\omega_2}(\boldsymbol{K},\Omega)=\frac{\omega_2^4}{c^4}\langle\delta\boldsymbol{\varepsilon}\otimes\delta\boldsymbol{\varepsilon}\rangle^{(N)}(\boldsymbol{k}_1-\boldsymbol{k}_2,\omega_1-\omega_2),\tag{A13}$$

where the time derivative of a δ 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, $\boldsymbol{U}_{\boldsymbol{k}_1\boldsymbol{k}_2}^{\omega_1\omega_2}(\boldsymbol{K},\Omega)$ is strongly peaked around $\omega_1=\omega_2$. The Bethe-Salpeter equation (A9) then implies the same behavior for $\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega\omega'}(\boldsymbol{K},\Omega)$, i.e., $\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega\omega'}(\boldsymbol{K},\Omega)\propto p(\omega-\omega')g(\omega')$, where $p(\omega-\omega')$ stands for the strongly peaked part around $\omega=\omega'$ and $g(\omega')$ for the remaining smooth function in ω'. The Green function $\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega\omega'}(\boldsymbol{K},\Omega)$ picks up a source term of frequency ω' and produces an autocorrelation function with frequencies ω centered narrowly around ω'. In the time domain this corresponds to $\exp(i\omega' t)$ times a slowly varying factor in t. It is therefore appropriate to introduce the Green function

$$\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega'}(\boldsymbol{K},\Omega,t)=\int d^3(\omega-\omega')\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega\omega'}e^{-i(\omega-\omega')t},\tag{A14}$$

which gives this factor for light sources of frequency ω'. If we multiply Eq. (A9) by $\exp[-i(\omega-\omega')t]$, rewrite the argument of $\langle\delta\boldsymbol{\varepsilon}\otimes\delta\boldsymbol{\varepsilon}\rangle^{(N)}$ as $\omega-\omega_1=\omega-\omega'-(\omega_1-\omega')$, and integrate over t, we finally arrive at the Bethe-Salpeter equation of Sec. III B for $\boldsymbol{\Phi}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega'}(\boldsymbol{K},\Omega,t)$:

$$\begin{aligned}\int\frac{d^3k_1}{(2\pi)^3}&[\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}_1}^{(4)}-\boldsymbol{f}_{\boldsymbol{k}}^{\omega'}(\boldsymbol{K},\Omega)\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega'}(t)]\boldsymbol{\Phi}_{\boldsymbol{k}_1\boldsymbol{k}'}^{\omega'}(\boldsymbol{K},\Omega,t)\\ &=\boldsymbol{f}_{\boldsymbol{k}}^{\omega'}(\boldsymbol{K},\Omega)\mathbf{1}_{\boldsymbol{k}\boldsymbol{k}'}^{(4)}\end{aligned}\tag{A15}$$

with

$$\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega'}(t)=\frac{(\omega')^4}{c^4}\langle\delta\boldsymbol{\varepsilon}\otimes\delta\boldsymbol{\varepsilon}\rangle^{(N)}(\boldsymbol{k}-\boldsymbol{k}_1,t).\tag{A16}$$

In deriving the last equation we replaced the argument ω in $\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$ in Eq. (A9) by ω' and the factor ω_1^4 in $\boldsymbol{U}_{\boldsymbol{k}\boldsymbol{k}_1}^{\omega\omega_1}(\boldsymbol{K},\Omega)$ by $(\omega')^4$.

APPENDIX B: TWO IDENTITIES

1. Ward identity

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

$$\Delta \boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},0)=\int \frac{d^3k'}{(2\pi)^3}\boldsymbol{B}_{\boldsymbol{k}\boldsymbol{k}'}^{\omega}(t=0)\Delta \boldsymbol{G}_{\boldsymbol{k}'}^{\omega}(\boldsymbol{K},0), \tag{B1}$$

which can be proven by a variable transformation of the integrand. For the general case see Vollhardt and Wölfle [51].

2. A useful identity

In this subsection we derive a useful relation between $\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$, $\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$, and $\Delta \boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$. We start with the definition

$$\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)=\langle \boldsymbol{G}^R\rangle(\boldsymbol{k}_+,\omega_+)-\langle \boldsymbol{G}^A\rangle(\boldsymbol{k}_-,\omega_-) \tag{B2}$$

and insert the unit tensor $\mathbf{1}$ in the appropriate representation:

$$\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)=\langle \boldsymbol{G}^R\rangle(\boldsymbol{k}_+,\omega_+)([\langle \boldsymbol{G}^A\rangle(\boldsymbol{k}_-,\omega_-)]^{-1} -[\langle \boldsymbol{G}^R\rangle(\boldsymbol{k}_+,\omega_+)]^{-1})\langle \boldsymbol{G}^A\rangle(\boldsymbol{k}_-,\omega_-). \tag{B3}$$

Then we introduce $\langle \boldsymbol{G}^{R/A}\rangle(\boldsymbol{k}_\pm,\omega_\pm)$ from Eq. (33), use definitions (44) and (50) for $\Delta\boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$ and $\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)$, and arrive at the final equation:

$$\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)=\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)[\Delta \boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)-\{\boldsymbol{G}_0^{-1}(\boldsymbol{k}_+,\omega_+) -\boldsymbol{G}_0^{-1}(\boldsymbol{k}_-,\omega_-)\}]. \tag{B4}$$

In the case $K,\Omega\rightarrow 0$, we get to first order in $\boldsymbol{K}$ and Ω:

$$\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)\approx \boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)\left[\Delta \boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)-\frac{\partial \boldsymbol{G}_0^{-1}}{\partial \boldsymbol{k}}\boldsymbol{K}-\frac{\partial \boldsymbol{G}_0^{-1}}{\partial \omega}\Omega\right], \tag{B5}$$

where we neglect first-order terms from $\boldsymbol{f}_{\boldsymbol{k}}^{\omega}$ and $\Delta\boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}$ because the components of $\Delta\boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}$ are already much smaller than one. The last equation shows that the leading order in $\boldsymbol{K}$ and Ω comes solely from $\boldsymbol{G}_0(\boldsymbol{k},\omega)$.

In Sec. III A we calculated the Green functions $\langle \boldsymbol{G}^{R/A}\rangle(\boldsymbol{k},\omega)$ to first order in $\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)$. They were diagonal and only involved the diagonal elements of $\boldsymbol{\Sigma}^{R/A}(\boldsymbol{k},\omega)$. Let us look at Eq. (B5) 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'' $\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\alpha$ or $\boldsymbol{d}^\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{d}^\alpha$. For $\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)$ we find

$$\begin{aligned}\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)&=[\langle \boldsymbol{G}^R\rangle(\boldsymbol{k},\omega)\otimes\langle \boldsymbol{G}^A\rangle(\boldsymbol{k},\omega)]^{(N)}\\ &\approx\sum_{\alpha,\beta}[\langle \boldsymbol{G}^R\rangle(\boldsymbol{k},\omega)]^{\alpha}[\langle \boldsymbol{G}^A\rangle(\boldsymbol{k},\omega)]^{\beta}\\ &\quad\times\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\beta(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{e}_\beta(\hat{\boldsymbol{k}}).\end{aligned} \tag{B6}$$

Terms such as $\boldsymbol{e}_1\otimes\boldsymbol{e}_2\otimes\boldsymbol{e}_1\otimes\boldsymbol{e}_2$ appear because the superscript (N) tells us to interchange the second and third basis vectors in $\langle \boldsymbol{G}^R\rangle\otimes\langle \boldsymbol{G}^A\rangle$. They only couple nondiagonal components to each other. Since nondiagonal elements are beyond our approximation we do not have to consider them. Then we are able to write

$$[\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha\beta}=\begin{cases}[\langle \boldsymbol{G}^R\rangle(\boldsymbol{k},\omega)]^{\alpha}[\langle \boldsymbol{G}^A\rangle(\boldsymbol{k},\omega)]^{\alpha}, & \alpha=\beta,\\ 0, \quad \alpha\neq\beta.\end{cases} \tag{B7}$$

The component 0 refers to terms such as $\boldsymbol{e}_1\otimes\boldsymbol{e}_1\otimes\boldsymbol{e}_2\otimes\boldsymbol{e}_2$. The derivation of $\boldsymbol{G}_0^{-1}$ with respect to ω just gives

$$\frac{\partial[\boldsymbol{G}_0^{-1}]_\alpha}{\partial\omega}\Omega=\frac{2\omega}{c^2}\Omega. \tag{B8}$$

The derivation with respect to $\boldsymbol{k}$ contains two contributions:

$$\frac{\partial \boldsymbol{G}_0^{-1}}{\partial \boldsymbol{k}}=\sum_\alpha\left[\frac{\partial[\boldsymbol{G}_0^{-1}]_\alpha}{\partial \boldsymbol{k}}\boldsymbol{d}_\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{d}_\alpha(\hat{\boldsymbol{k}}) +[\boldsymbol{G}_0^{-1}]_\alpha\frac{\partial}{\partial \boldsymbol{k}}[\boldsymbol{d}^\alpha(\hat{\boldsymbol{k}})\otimes\boldsymbol{d}^\alpha(\hat{\boldsymbol{k}})]\right]. \tag{B9}$$

The second one only produces nondiagonal elements that we do not have to consider. This statement seems to be obvious because a small rotation of $\boldsymbol{k}$ rotates $\boldsymbol{d}_\alpha(\hat{\boldsymbol{k}})$. But our basis vectors are more general and we have to look at it more carefully. We have to show that $\boldsymbol{e}_i(\hat{\boldsymbol{k}})\cdot(\partial/\partial k_j)\boldsymbol{d}^i(\hat{\boldsymbol{k}})$ is zero. With $\boldsymbol{d}^i=\boldsymbol{\varepsilon}_0\boldsymbol{e}_i$ and $\boldsymbol{\varepsilon}_0$ being symmetric and independent of $\boldsymbol{k}$ we can write

$$\boldsymbol{e}_i(\hat{\boldsymbol{k}})\cdot\frac{\partial}{\partial k_j}\boldsymbol{d}^i(\hat{\boldsymbol{k}})=\boldsymbol{d}^i(\hat{\boldsymbol{k}})\cdot\frac{\partial}{\partial k_j}\boldsymbol{e}_i(\hat{\boldsymbol{k}}). \tag{B10}$$

From the biorthogonality relation it is clear that

$$\boldsymbol{d}^i(\hat{\boldsymbol{k}})\cdot\frac{\partial}{\partial k_j}\boldsymbol{e}_i(\hat{\boldsymbol{k}})=-\boldsymbol{e}_i(\hat{\boldsymbol{k}})\cdot\frac{\partial}{\partial k_j}\boldsymbol{d}^i(\hat{\boldsymbol{k}}), \tag{B11}$$

which verifies the statement. We are now able to write down Eq. (B5) within our approximation:

$$[\Delta \boldsymbol{G}_{\boldsymbol{k}}^{\omega}(\boldsymbol{K},\Omega)]^{\alpha}\approx[\boldsymbol{f}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]^{\alpha\alpha}\left([\Delta\boldsymbol{\Sigma}_{\boldsymbol{k}}^{\omega}(\mathbf{0},0)]_\alpha -\frac{\partial[\boldsymbol{G}_0^{-1}]_\alpha}{\partial \boldsymbol{k}}\cdot\boldsymbol{K}-\frac{2\omega}{c^2}\Omega\right). \tag{B12}$$

APPENDIX C: STRUCTURE FACTOR

We give the two important structure factors in scaled form and in the coordinates $C_{\boldsymbol{k}}$, $S_{\boldsymbol{k}}$, $C_{\boldsymbol{q}}$, $S_{\boldsymbol{q}}$, and φ. From the notation it is clear whether they belong to an extraordinary or ordinary light ray. The parameters are α, $\overline{K}_1$, $\overline{K}_2$, and h:

$$[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^2}(0)]_{12}=\frac{1}{(1+\alpha)^2}\,\frac{S_{\boldsymbol{k}}^2}{Q_\perp^2}\left[\frac{S_{\boldsymbol{k}}^2\sin^2\varphi}{\overline{K}_1Q_\perp^2+Q_\parallel^2+h^2}+\frac{(S_{\boldsymbol{k}}\cos\varphi-S_{\boldsymbol{q}})^2}{\overline{K}_2Q_\perp^2+Q_\parallel^2+h^2}\right], \tag{C1a}$$

$$[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}=\frac{1}{(1+\alpha)^2}\,\frac{1}{Q_\perp^2}\left[\frac{\cos^2\varphi N_1+2\cos\varphi N_2+N_3}{\overline{K}_1Q_\perp^2+Q_\parallel^2+h^2}+\frac{\sin^2\varphi N_4}{\overline{K}_2Q_\perp^2+Q_\parallel^2+h^2}\right] \tag{C1b}$$

with

$$Q_\parallel^2=(C_{\boldsymbol{k}}-C_{\boldsymbol{q}})^2,\quad Q_\perp^2=S_{\boldsymbol{k}}^2-2S_{\boldsymbol{k}}S_{\boldsymbol{q}}\cos\varphi+S_{\boldsymbol{q}}^2 \tag{C2}$$

and

$$\begin{gathered}N_1=(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}-S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})^2,\quad N_2=S_{\boldsymbol{k}}S_{\boldsymbol{q}}(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}-S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})(C_{\boldsymbol{k}}-C_{\boldsymbol{q}})\\ N_3=S_{\boldsymbol{k}}^2S_{\boldsymbol{q}}^2(C_{\boldsymbol{k}}-C_{\boldsymbol{q}})^2,\quad N_4=(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}+S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})^2.\end{gathered} \tag{C3}$$

The integration over φ gives

$$\int [\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^2}(0)]_{12}d\varphi=\frac{\pi}{2}\,\frac{S_{\boldsymbol{k}}^2}{S_{\boldsymbol{q}}^2}\left[\frac{1}{\overline{K}_2}-\frac{1}{\overline{K}_1}+\frac{1}{C(h)}\left(\frac{I^{-1}(\overline{K}_1)}{\overline{K}_1}-\frac{I^{-1}(\overline{K}_2)}{\overline{K}_2}\right)+4S_{\boldsymbol{q}}^2I(\overline{K}_2)\right], \tag{C4a}$$

$$\begin{aligned}\int \cos\varphi[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^2}(0)]_{12}d\varphi=\frac{\pi}{4}\,\frac{S_{\boldsymbol{k}}}{S_{\boldsymbol{q}}^3}\Bigg[&\frac{C(h)+2\overline{K}_2(S_{\boldsymbol{k}}^2-S_{\boldsymbol{q}}^2)}{\overline{K}_2^2}-\frac{C(h)+2\overline{K}_1(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_1^2}+\frac{1}{C(h)}\left(\frac{C(h)+\overline{K}_1(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_1^2I(\overline{K}_1)}\right.\\&\left.-\frac{C(h)+\overline{K}_2(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_2^2I(\overline{K}_2)}\right)+\frac{4S_{\boldsymbol{q}}^2}{\overline{K}_2}[C(h)+\overline{K}_2(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)]I(\overline{K}_2)\Bigg],\end{aligned} \tag{C4b}$$

$$\begin{aligned}\int [\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}d\varphi=\frac{\pi}{2}\Bigg[&\left(\frac{S_{\boldsymbol{k}}^2}{S_{\boldsymbol{q}}^2}C_{\boldsymbol{q}}^2+\frac{S_{\boldsymbol{q}}^2}{S_{\boldsymbol{k}}^2}C_{\boldsymbol{k}}^2\right)\left(\frac{1}{\overline{K}_1}-\frac{1}{\overline{K}_2}\right)-2C_{\boldsymbol{k}}C_{\boldsymbol{q}}\left(\frac{1}{\overline{K}_1}+\frac{1}{\overline{K}_2}\right)+\frac{1}{C(h)}\left(\frac{S_{\boldsymbol{k}}}{S_{\boldsymbol{q}}}C_{\boldsymbol{q}}+\frac{S_{\boldsymbol{q}}}{S_{\boldsymbol{k}}}C_{\boldsymbol{k}}\right)^2\left(\frac{I^{-1}(\overline{K}_2)}{\overline{K}_2}-\frac{I^{-1}(\overline{K}_1)}{\overline{K}_1}\right)\\&+\frac{4}{\overline{K}_1}I(\overline{K}_1)[\overline{K}_1(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}+S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})(C_{\boldsymbol{k}}+C_{\boldsymbol{q}})+C(h)C_{\boldsymbol{k}}C_{\boldsymbol{q}}]\Bigg]\end{aligned} \tag{C4c}$$

$$\begin{aligned}\int \cos\varphi[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}d\varphi=\frac{\pi}{4}\,\frac{1}{S_{\boldsymbol{k}}^3S_{\boldsymbol{q}}^3}\Bigg[&\frac{1}{\overline{K}_1^2}[(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}-S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})^2C(h)+2\overline{K}_1(S_{\boldsymbol{k}}^4C_{\boldsymbol{q}}^2-S_{\boldsymbol{q}}^4C_{\boldsymbol{k}}^2)(S_{\boldsymbol{k}}^2-S_{\boldsymbol{q}}^2)]-\frac{1}{\overline{K}_2^2}(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}+S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})^2[C(h)\\&+2\overline{K}_2(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)]+\frac{1}{C(h)}(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}+S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})^2\left(\frac{C(h)+\overline{K}_2(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_2^2I(\overline{K}_2)}-\frac{C(h)+\overline{K}_1(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_1^2I(\overline{K}_1)}\right)\\&+4S_{\boldsymbol{k}}^2S_{\boldsymbol{q}}^2I(\overline{K}_1)\frac{C(h)+\overline{K}_1(S_{\boldsymbol{k}}^2+S_{\boldsymbol{q}}^2)}{\overline{K}_1^2}[\overline{K}_1(S_{\boldsymbol{k}}^2C_{\boldsymbol{q}}+S_{\boldsymbol{q}}^2C_{\boldsymbol{k}})(C_{\boldsymbol{k}}+C_{\boldsymbol{q}})+C(h)C_{\boldsymbol{k}}C_{\boldsymbol{q}}]\Bigg]\end{aligned} \tag{C4d}$$

with

$$C(h)=(C_{\boldsymbol{k}}-C_{\boldsymbol{q}})^2+h^2\quad\text{and}\quad I(\overline{K}_i)=\{[\overline{K}_i(S_{\boldsymbol{k}}-S_{\boldsymbol{q}})^2+C(h)][\overline{K}_i(S_{\boldsymbol{k}}+S_{\boldsymbol{q}})^2+C(h)]\}^{-1/2}. \tag{C5}$$

The structure factor $[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}$ diverges for $h=0$ and $\boldsymbol{q}^1\to\boldsymbol{k}^1$. We give $[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}$ for small φ and $\delta C=C_{\boldsymbol{k}}-C_{\boldsymbol{q}}$:

$$\begin{aligned}[\widetilde{\boldsymbol{B}}^{\omega}_{\boldsymbol{k}^1\boldsymbol{q}^1}(0)]_{11}=\frac{1}{(1+\alpha)^2}\,\frac{4C_{\boldsymbol{k}}^2}{(1+\alpha)^2C_{\boldsymbol{k}}^2\delta C^2/S_{\boldsymbol{k}}^2+S_{\boldsymbol{k}}^2\varphi^2}\Bigg[&\frac{(1+\alpha)^2C_{\boldsymbol{k}}^2\delta C^2}{[\overline{K}_1(1+\alpha)^2C_{\boldsymbol{k}}^2/S_{\boldsymbol{k}}^2+1]\delta C^2+\overline{K}_1S_{\boldsymbol{k}}^2\varphi^2+h^2}\\&+\frac{S_{\boldsymbol{k}}^2C_{\boldsymbol{k}}^2\varphi^2}{[\overline{K}_2(1+\alpha)^2C_{\boldsymbol{k}}^2/S_{\boldsymbol{k}}^2+1]\delta C^2+\overline{K}_2S_{\boldsymbol{k}}^2\varphi^2+h^2}\Bigg].\end{aligned} \tag{C6}$$

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