

Resonant Multiple Scattering of Light

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Abstract

This educational work presents a new approach towards resonant interaction between classical light and matter. The interaction between light and matter is considered from three different points of view: the light picture (where the material degrees of freedom have been integrated out, and leaving one with scattering theory), the matter picture (where the radiative degrees of freedom have been eliminated and providing one essentially with atomic physics). In addition the polariton approach is discussed, in which the degrees of freedom of light and matter are treated on the same footing. Although the first approach will by far given most of the attention, we will frequently emphasize the equivalence of the three methods. Much of the presented material is selfcontained.

We demonstrate that in the *dynamical* properties of multiple scattering of light the “matter” properties play a dominant role. Several “paradigms of atomic physics” will be discussed from the view point of light scattering theory. We shall introduce the far-reaching analogy between the dielectric “Mie” sphere in classical optics, and the two-level atom in semi-classical atomic physics. This mapping turns out to be much more faithful than the widely used analogy between scattering theory for De Broglie waves and classical waves. In scattering theory the semi-classical two-level atom is equivalent to a point scatterer.

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1. PRINCIPLES

Light propagates along straight lines and with constant intensity [1]. This statement is only true as long as the radiation does not encounter obstacles. Barriers can either absorb part of the light, as for instance sun glasses do, or can change its direction of propagation, as water droplets in a cloud do. The latter phenomenon is called “scattering”. When scattering is efficient for all colors, the object will look white. When absorption is efficient for all colors, it will look black. The origin of light scattering is often caused by refraction, which is - in its simplest form - described by the well-known Snell’s law.

“Multiple scattering” will set in when light propagation is influenced by more than one obstacle. Its occurrence is determined by both absorption and scattering of the individual particles. As a rule of the thumb one might say that multiple scattering of a wave is most likely to occur in the presence of a high concentration of obstacles that each have a large scattering efficiency and hardly absorb any intensity. In a medium where multiple scattering of light fully determines the energy transport, the so-called regime of radiative transfer, the underlying wave character of light seems to be washed out. What remains, appears to be a diffuse intensity pattern with hardly any interesting wave physics involved.

In the past experimentalists working in optics tried to avoid multiple scattering as much as possible. The phenomenon was considered to be dull and in clean optical experiments often a nuisance. A microscope with a milky lens system is useless, leave alone glasses made of opal. It should not come as a surprise that most knowledge of classical multiple light scattering originates from astrophysics. In stellar atmospheres multiple scattering of light is often a major means of energy transport, and determines the emission and absorption line spectrum of a star. The light scattering occurring in the interstellar medium cannot be eliminated as can be done in a terrestrial laboratory and a thorough knowledge of the principles radiative transfer is of vital importance to understand the astrophysical processes [2] [3].

It was not until the late fifties that the first indications arose that multiple scattering is full of rich and fundamental phenomena. However, these advances were not made by addressing multiple scattering of light, but by studying multiple scattering of electrons in the solid state. Electrons scatter from a local variation in potential just like light scatters from a local variation in dielectric constant. The resistivity of metals at low temperatures is to a large extent determined by multiple scattering of electrons from the potentials of impurities. The wave character of the electron plays a crucial role here since it explains why the resistivity is zero in a perfect crystal, and why only deviations from this perfection lead to a finite resistivity. But in order to explain the diffuse motion of an electron caused by scattering off impurities in the crystal, it is not necessary to incorporate the interference of the electronic wave function in this scattering process. Ohm’s famous and classic law stating that the resistance of a conductor is proportional to its length is deeply connected to the “hardly interesting” diffuse propagation of light in the regime of multiple scattering. The new developments in condensed matter focused on the role of *interference* of the electron wave function in multiple scattering. Among the many new concepts that were introduced, “Anderson Localization” is undoubtedly one of the most fascinating. It describes the vanishing of propagation in the regime of very strong multiple scattering due to interference and was suggested as a model for metal-insulator transitions [4].

If electron propagation can be inhibited by interference, why not also the propagation of light? This question was posed only about fifteen years ago. Since then, optimizing multiple scattering

of light has become an active area of research, with localization of light as its major goal. How to characterize the strongly-scattering regime, and how to realize it experimentally were, and still are, two major questions. The initial line of attack was to pack strongly scattering dielectric particles closely and randomly. Indeed, many optimistic theoretical predictions have been put forward for this regime [5] [6] [7] [8]. The scattering of light from one individual dielectric particle can be optimized by tuning the wavelength of light right into a scattering resonance. In order to set a scatterer into resonance its size should be matched to the wavelength of the light. This balancing of length scales together with a suitable shape will turn the particle into a resonator for light. In fact, in search for localization of light, the study of resonant multiple scattering (RMS) has become an interesting and large topic by itself.

In this article we will summarize the many new facts that have recently been discovered by examining the regime of RMS. Perhaps the most important conclusion that can be drawn from all this is the unexpected contrast between the role played by resonances in multiple *electron*-impurity scattering on one hand, and in multiple *light* scattering on the other. Despite the strong resemblance between the physics of classical waves and of De Broglie waves many differences will be shown to exist between the two subjects. It turns out that a much closer connection exists between the theory of resonant scattering of light off finite-size objects and polariton theory. Polaritons are the result of a resonant coupling of light to an object not through a local variation in dielectric constant but through an internal degree of freedom of that object [9]. We speak of the object having an internal resonance, and the polariton amplitude can be considered as a coherent superposition of the light and the internal degree of freedom. We will demonstrate that scattering theory is capable of treating RMS, polariton behavior and the (phenomenological) theory of radiative transfer on the same footing in a unified picture.

In the rest of this introductory section we will first familiarize the reader with the most important issues. Unless stated otherwise, all formula's presented there will be accounted for later on in this paper. Equations pertaining to electrons can easily be discriminated from those pertaining to light as the fundamental parameter determining the time dependence is the energy E for electrons and the frequency ω for classical waves.

An important aspect of the study of the influence of disorder on light propagation is the statistical nature of the disorder. Averaging over disorder is a linear process so that conservation laws will hold both for the non-averaged and the averaged case. The precise structure of the disorder is - almost by definition - beyond the control of the experimentalist. Only some stochastic laws governing probabilities are known. Many experiments effectively involve an average over a large number of realizations. One example would be the measurement of the index of refraction of a gas, in which case averaging over all the possible positions of the atoms in the gas seems a safe procedure. The averaging procedure is usually a simple average over the distribution functions, and only in pathological cases a weighted average is needed. In principle, experiments on quenched solid samples can be envisaged that sample only one realization of the disorder. As no averaging occurs in these type of experiments the optical properties may vary strongly from realization to realization (speckle). Theoreticians find it difficult to deal with a single realization of the disorder. So many of the theoretical concepts are defined as an average over realizations right from the start. To incorporate observed fluctuations on single realizations, theoreticians calculate higher-order moments of the distribution function associated with the observable. For instance fluctuations in the intensity I can be investigated by calculating both the average of $\langle I \rangle$ and the variance $\langle I^2 \rangle - \langle I \rangle^2$.

1.1. Atomic Physics Versus Scattering Theory

When one starts out to describe the interaction between light and matter, the Hamiltonian and the equations of motion following from it, contain both material and radiative degrees of freedom. In most situations it is quite cumbersome to do the bookkeeping in both worlds at the same time. A simplifying picture can be created if one can eliminate or ‘integrate out’ either one of them. The remaining reduced Hamiltonian will be more manageable. Depending on taste and goal (what experiment is being explained?) one can try to integrate out either the material degrees of freedom or the radiative degrees of freedom.

In solid-state textbooks polariton behavior is treated in the full world: keeping track of both matter and light. In this particular case, however, it is easy to integrate out the material degrees of freedom. One winds up with radiation characterized by an index of refraction that is different from the one in vacuum and changes substantially when the frequency approaches that of a material resonance. At any time it is easy to trace back the material degrees of freedom. We will generalize the concept of polariton behavior and define it to apply to any case where the index of refraction of light is substantially changed due to the presence of a collection of resonating scatterers. The generalization being that the resonance of the object is not limited to an internal atomic resonance but could also be caused by an external, geometrical resonance. Later we will introduce a ‘polariton parameter’ that will quantify such polariton behavior. Polariton behavior is not necessarily a quantum effect, as is often suggested implicitly in the literature. In our definition, the material degree of freedom to which the light couples can be either a classical or a quantum-mechanical mode. The only condition for the mode is that its excitation should be accompanied by a simultaneous excitation of the polarizability (and thus of the macroscopic polarization). In fact, many other ‘quantum effects’ acquire a more classical context, simply because they are general features of (resonant) light scattering.

Scattering theory applies as soon as one creates a “light-in light-out” situation. Note that many optical experiments are of this type. Channels “in” and channels “out” are connected mathematically by an S -matrix. When light and matter interact scattering theory can be applied after integrating out the material degrees of freedom and focusing on the light. Material properties that determine this S -matrix are typically the “polarizability” and “conductivity”. If one wishes to describe the light-matter interaction semi-classically, in which case matter is treated quantum mechanically but the radiation classically, these quantum-mechanical material properties serve as input in a *purely classical* scattering theory.

Rather than eliminating material degrees of freedom it is also possible to integrate out light modes and focus on the matter excitations. This is the standard procedure in atomic physics. The mathematical treatment now simplifies to a quantum-mechanical eigenvalue problem of some effective atomic Hamiltonian. The existence of light manifests itself in the radiation shift and the Einstein spontaneous emission coefficients of the atomic eigenstates, as well as in the electric forces between polarizable particles. We emphasize again that in principle both elimination methods should be equivalent. In this paper we prefer the elegant framework of scattering theory to describe many features of (semi-classical) light-matter interaction, and establish the connection with concepts of atomic physics such as polarizability, spontaneous emission, induced dipole-dipole coupling and density of states. Comparison of both methods will lead to a deeper and more complete understanding of the way in which light interacts with matter and *vice versa*. The following “atomic” items can be exposed in a scattering theory for light.

Firstly, we will demonstrate that in classical light scattering a very convenient and well-defined object can be defined: a point scatterer that resonantly scatters light - either elastically or inelastically. One can think of point particles as being much smaller than the wavelength of the incoming light. These point scatterers will enable us to investigate the regime of RMS in great detail. The S -matrix of a point particle turns out to be surprisingly similar to the polarizability of an atom with one excited state. A two-level atom is the paradigm of atomic physics and quantum optics [10].

Subsequently it will be demonstrated that a “dwell time” can be *rigorously* defined in any scattering theory. This dwell time can be *interpreted* as a measure of the time that a wave spends in a specified region during a scattering process. This residence time can be large when the scatterer is on resonance, even much larger than the typical time between collisions, so that a large delay will accumulate in multiple scattering. These properties make the dwell time an important aspect of RMS. The dwell time is the concept closest one can come to the lifetime of an excited level (inverse of the Einstein coefficient for spontaneous emission) in a classical theory of elastic scattering. In the case of light scattering by a two-level atom these times are not only analogous but are in fact identical.

Finally, in atomic physics it is known that the (virtual) exchange of two photons between two neutral particles, when triggered by vacuum fluctuations, gives rise to Van der Waals forces [11] [12] [13]. If the process is set off by an ordinary incoming field the interactions are referred to as induced dipole-dipole coupling. Can these two atomic physics paradigms also emerge in light scattering theory?

A simplistic approximation in multiple-scattering theory is the “independent-scattering approximation (ISA)”. A well-known result of the ISA is the expression for the extinction mean free path $\ell_{\text{ext}} = 1/n\sigma_{\text{ext}}$, in which n is the density and σ_{ext} the extinction cross-section of the scatterers. The extinction mean free path describes the decay of the coherent light beam because energy is used for scattering and absorption. (In this article several types of mean free paths will be examined and we will have to differentiate between them by adding subscripts). Within the ISA only a limited set of all scattering events is included: The condition is that only those scattering events count in which every scatterer is visited by the light at most once. No loops of any form are allowed. The range of validity of the ISA will be treated later. The ISA approximation is very popular, one of the reasons being that it is quite difficult to go beyond. It is frequently used without further comment and without justification.

Extensions of the ISA are called “dependent scattering”. For instance repetitive scattering between two scatterers, a phenomenon that we will call “recurrent scattering”, is not included in the ISA and is one out of many dependent scattering effects. The influence of dependent scattering is large when resonant scattering is involved [14]. Apart from producing corrections to well-known ISA results, the inclusion of dependent scattering supplies the desired connection with induced dipole-dipole coupling raised above. This connection is supported experimentally. Recently, one has directly observed the occurrence of recurrent light scattering in a resonant scattering medium [15].

1.2. Resonant Multiple Scattering (RMS) of Light

One of the purposes of this paper is to elucidate the difference between the “coherent light beam” and the “diffuse intensity”, which unfortunately by convention is referred to as the “incoherent” contribution. Both “coherent” part and “incoherent” part result from multiple scattering. Their

difference has been, and still is, the source of much confusion. The coherent beam is the remnant inside the medium of the source and decays exponentially in space. The diffuse beam refers to the light that comes out of the sample in all directions and its intensity decays only algebraically with distance inside the sample. Depending on the mean-free path relative to the size of the system either one of them can dominate.

The confusion arises from the fact that there is nothing incoherent about the diffuse beam. In the dielectric inhomogeneous systems that we envisage every scattering process to preserve phase. As a result the diffuse beam has the same phase-coherence properties as the coherent beam. In fact, the confusion is initiated by earlier studies on radiation trapping [16]. In elastic light scattering incoming and outgoing light waves have the same frequencies and are phase-locked. In the case of radiation trapping one also deals with resonant multiple light scattering, but now it involves an incoherent sequence of absorption and emission cycles. The absence of phase-locking in fluorescence causes many subtle difference with elastic scattering processes, such as the absence of interference effects.

Some simple theoretical results on the behavior of the coherent beam can be obtained without explicitly resorting to multiple-scattering theory, but can simply be envisaged as the wave propagating in a homogeneous absorbing medium (sometimes known as the effective medium [17] [18] [19]). This is impossible for the diffuse intensity and a thorough understanding of multiple scattering is required in this case. For this reason the diffuse intensity is often considered as the genuine multiple scattering. The ISA approximation applied to the propagation of the diffuse intensity is equivalent to the Boltzmann approximation in transport theory. The energy carried by the diffusive beam comes from the “absorption” suffered by the coherent beam if there are no explicit energy loss mechanisms. The “absorption” of the coherent beam is thus no real absorption and is therefore called extinction. It turns out that the ISA approximation for the coherent beam, together with the well-known Boltzmann approximation for the diffuse intensity describes this energy balance properly. Again, it is very difficult to do better while keeping the balance in tact. In fact all theory on radiative transfer relies on the ISA and the Boltzmann approximation.

The coherent beam is characterized by a complex-valued frequency-dependent index of refraction $m(\omega)$, which can be associated with the index of refraction of an “effective medium”. The real and imaginary part of the refractive index define a velocity, the phase velocity v_p , and a decay length, or extinction mean-free path, ℓ_{ext} according to

$$m(\omega) \equiv \frac{c_0}{v_p} + i \frac{c_0}{2\omega\ell_{\text{ext}}} \equiv \eta + i\kappa. \quad (1.1)$$

The factor of two in the definition for ℓ_{ext} stems from the fact that the coherent intensity, which is the coherent amplitude squared, decays twice as fast as the coherent amplitude and thus with length ℓ_{ext} . The relation between wave vector and frequency of the coherent beam, the dispersion law, $k = \eta(\omega)\omega/c_0$ will give rise to polariton behavior when the frequency of the beam is near a resonance of the scatterers. In the electromagnetic picture polariton behavior is thus a property of the coherent beam. Typical polariton behavior is shown in the dispersion relation presented in Fig. 1, a figure that must be familiar to many readers as it features in many text books.

Rather than limiting the treatment of the polariton to explaining the dispersion curves we will go considerably further in this paper and investigate the dependent-scattering corrections to the polariton and discuss polariton aspects of the diffuse intensity. Generally speaking, multiple-scattered diffuse intensity can be described mathematically by a (frequency-dependent) *diffusion constant*,

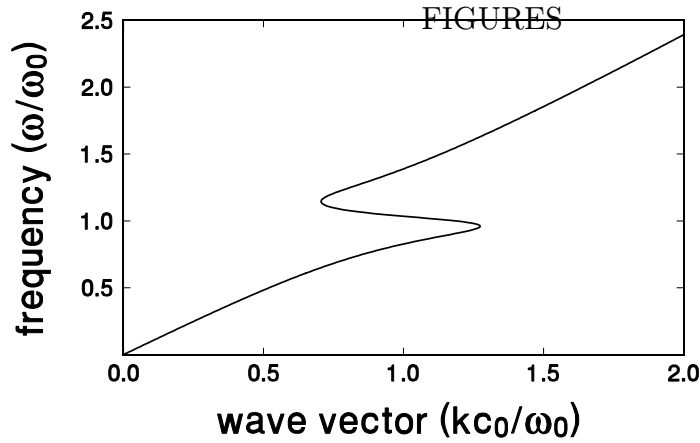


FIG. 1. Typical dispersion relation for a polariton. The following parameters have been used: $\mathcal{S} = 0.25$ and damping $T_2 = 12/\omega_0$, corresponding to a polariton strength of $\mathcal{P} = 1.5$. For definition of these parameters we refer to Section IV.B.2.

$$D(\omega) = \frac{1}{3} v_E(\omega) \ell_{tr}(\omega) . \quad (1.2)$$

consisting of a “transport mean free path” ℓ_{tr} and a “transport speed of light” v_E . The temptation is large to use for ℓ_{tr} the extinction mean free path ℓ_{ext} and for v_E the phase (or possibly group) velocity found for the coherent beam, and to forget about transport theory. It is known for a long time already that anisotropically scattering particles have a transport mean free path different from the decay of the coherent beam, that is $\ell_{tr} \neq \ell_{ext}$. Only recently a theory was developed which showed that the transport velocity v_E in Eq. (1.2) is determined by the energy flow [20]. It was found that neither phase nor group velocity of the coherent beam can be used. Multiple scattered diffuse light does not see the “effective medium” felt by the coherent beam, and its propagation is quite different and characterized by the transport velocity. This speed of propagation of the diffuse light in multiple scattering can be very small when the scatterers are close to a resonance. The large deviation of v_E from v_p is due to the matter, and shows up already in the lowest order of multiple scattering theory (ISA). An important conclusion: In the theory for the light intensity the matter fully reappears although it was integrated out earlier!

A velocity superficially similar to the above transport velocity turns up in the description of (incoherent) radiation trapping mentioned earlier. In that case the light diffusion is like the one of a billiard ball that suffers from temporary trapping at each site before moving the next. Contrary to the transport mechanism associated with the transport velocity the much less interesting radiation trapping involves an inelastic, incoherent transport mechanism where interference plays no role. In yet another context an “energy velocity” showed up in the work of Brillouin [21] which was expanded on later in atomic physics and polariton studies at the end of the sixties, with important contributions by Loudon [22] [23] and Pari and Birman [24]. Since group and phase velocity of a dispersion relation with an S-curve (a polariton) can exceed c_0 , this new velocity was introduced to have a velocity that explicitly did not violate causality. This velocity was implicitly assumed to describe properties of the coherent beam and no link was made then with multiple scattering and long-range diffusion of light. A non-linear variant of the energy velocity occurs in the theory of self-induced transparency [10] [25]. In that case it really describes pulse propagation.

1.3. Multiple Scattering of Electrons

As we will explain in this paper, electrons suffer much less from time delay in RMS than light. The question why electrons and light behave so differently in RMS is fascinating. In electron-impurity scattering the “effective medium” or coherent beam describes the velocity v_E for the diffuse density of electrons satisfactorily. The relation between energy and wave vector of the electron, the dispersion law $E(k)$, is a property of the coherent beam and is given by the underlying crystalline structure and hardly by the impurities. The dispersion relation of the crystal defines an “effective mass” m^* . For most purposes the electron velocity featuring in the diffusion constant can be obtained from the dispersion relation in the following way,

$$v_E(k_F) = \frac{1}{\hbar} \left(\frac{\partial E(k)}{\partial k} \right)_{k=k_F} = \frac{\hbar k_F}{m^*}, \quad (1.3)$$

where k_F is a wave number determined by the Fermi exclusion principle for fermions. A wave vector analogy between light and electrons $k \longleftrightarrow k_F$ can be used. For light the effective medium defines a phase velocity $v_p = \omega/k$ different from c_0 . For electrons we infer that $v_E \sim k_F \sim 1/v_p$ and apparently the electron transport velocity is inversely proportional to its phase velocity. No retardation effects, in contrast to light, are present for electrons.

The absence of microscopic time-delay factors in velocity and diffusion constant for an electron scattering from impurities (in solid state physics better known as “mass-enhancement factors”) may at least be called surprising and must be due to some underlying cancellation mechanism. The appearance of “surprising” cancellation factors in transport theory is well-known by solid-state physicists and applies to other transport quantities, though not all [26] [27] [28]. The cancellation is due to existence of conservation laws in transport theory, and in this particular case due to the conservation of the probability. The fact that such laws are important for the velocity in multiple scattering agrees with our previous observation that the transport velocity of light is to be associated with “energy transport”. An important conclusion that could be drawn by comparing light and electrons is that modifications of conservation laws result in large modifications and thus a deeper understanding of such cancellation theorems.

Other velocities show up in solid-state physics, some of which are not subject to cancellation theorems. We will mention the “drift velocity” \mathbf{v}_d of the electrons in the presence of an applied electric field \mathbf{E} in a conductor. A “linear-response argument” makes this velocity proportional to the applied field so that it is given by the relation,

$$\mathbf{v}_d = \boldsymbol{\mu}_d \cdot \mathbf{E}. \quad (1.4)$$

Here $\boldsymbol{\mu}_d$ is usually called the *drift mobility tensor*. It is instructive to compare this formula to the one defining the *conductivity tensor* $\boldsymbol{\mu}_e$ that describes the induced charge current density \mathbf{J}_e as a result of the driving external field,

$$\mathbf{J}_e = \boldsymbol{\sigma}_e \cdot \mathbf{E} \equiv -en\boldsymbol{\mu}_c \cdot \mathbf{E}. \quad (1.5)$$

(The subscript of $\boldsymbol{\sigma}_e$ is necessary to distinguish it from the cross-sections σ that appear in various places in this article). The electric conductivity is known not to be subject to any time delay factors. Relation (1.5) defines the *conductivity mobility tensor* $\boldsymbol{\mu}_c$. It describes the resulting current

as a result of a driving electric field, and not the velocity. We believe that both mobilities are frequently mixed up in the literature. They certainly do not need to be the same as the relation between any current and velocity in quantum mechanics is given by $\mathbf{J} = \rho \mathbf{p}$, where ρ is the density operator. As ρ is an operator and not simply a constant as in Newtonian mechanics and subject to the conservation laws imposed by quantum mechanics, the two mobilities are differently influenced by mass-enhancement factors. In the language we introduced in the previous subsection one could say that the drift mobility tensor $\boldsymbol{\mu}_d$ is a property of the “coherent beam”, whereas the conductivity mobility tensor $\boldsymbol{\mu}_c$ is a property of the “diffuse beam”.

1.4. Stationary and Dynamic Multiple Scattering

A distinction between stationary and dynamic measurements is important in any discussion on transport of waves. It is physically obvious that time delay can only be measured in dynamic experiments. It seems to be less obvious to decide which transport quantity is dynamic and which one is stationary. To give an example, the DC conductivity and the diffusion constant are often treated on the same footing in multiple scattering of an electron. However, contrary to the wisdom of many textbooks on condensed matter physics, it is *impossible* to obtain the diffusion constant from one stationary measurement.

It is beneficial to treat the velocity v_E and transport mean free path ℓ_{tr} as two individual parameters characterizing the multiple scattering regime, rather than only considering their product in the form of the diffusion constant. A stationary experiment yields information on the transport mean free path only. The transport velocity can only be obtained from a dynamic experiment.

Perhaps the most important stationary transport coefficient for multiple light scattering is the (DC) transmittance T . In the simplest but common case of a disordered slab with thickness L and surface A the transmittance, summed over all channels in and all channels out, rigorous transport theory yields, [3]

$$T = f \times \frac{Ak^2}{2\pi} \times \frac{\ell_{tr}}{L} . \quad (1.6)$$

The factor $Ak^2/2\pi$ is the number of conducting channels per steradian (including two polarization states). A channel is a cavity mode, and in optics often called a coherence area (or a Fresnel zone). It is essentially a discretization of all possible incoming and outgoing angles in such a way that one channel is a coherent source. Here f is a constant factor of order unity that depends on geometry ($f = 4/3$ for the slab geometry).

The stationary transmittance of electrons is deeply connected with the conductance G (inverse resistance) c.q. conductivity σ_e in solid-state physics. For isotropic conductors the conductivity tensor $\boldsymbol{\sigma}_e = \sigma_e \mathbf{I}$. In solid state physics a famous relation between G and T is given by a formula first proposed by R. Landauer in 1957 [29],

$$G \equiv \sigma_e \frac{A}{L} = \frac{e^2}{h} T . \quad (1.7)$$

This relation gave rise to skepticism in the solid-state-physics community since it does not always coincide with Kubo’s original linear-response expression for the conductivity. The applicability of Landauer’s formula beyond simple transport theory, where T is no longer given by relation (1.6), is an active field of research [30]. For our purpose it is most satisfying to see that Eqs. (1.6) and

(1.7) show that the conductivity σ_e does not depend on the dynamic transport velocity, but only on the transport mean free path.

The importance of the transmittance of electrons for the theory of conduction increased considerably after the standard scaling theory of Anderson localization had been developed [31]. This theory scales the size of the random medium, and as such aims to do predictions about the occurrence of localization. In the simplest approach the universal scaling parameter of this theory is the transmission T . In view of this observation, the “vanishing of diffusion” predicted by P.W. Anderson in his famous paper [4] turns out to be a vanishing of the transport mean free path (and thus of the DC conductivity). This is a far-reaching notion: For RMS of light the transport velocity is non-trivial and can also be very small. For light the “vanishing of diffusion” would be an ambiguous statement. The scaling theory is far beyond the scope of this article and will not be discussed any further.

Apart from the transport velocity itself, an important “dynamic” transport quantity is the *density of states* (DOS) per unit volume. This quantity represents the number of microscopic states and can be defined for light and electrons. For light $N(\omega)$ counts the number of states in an infinitesimal frequency range, for electrons $N(E)$ counts the number of states in an infinitesimal energy range. For a large volume V the number of states turns out to be proportional to the volume and it is convenient to introduce the density of states per unit volume. For electrons we use the common notation $\rho(E)$; In this paper we will use the notation W for the density of electromagnetic states per unit volume because it will be shown to coincide with the electromagnetic energy density. In both cases N is the total number of microscopic states per frequency or energy interval. The inverse, $1/N$, can be associated with a frequency (energy) width $\Delta\omega$ (ΔE). Using $\Delta\omega\Delta t \simeq 1$ (for electrons $\Delta E\Delta t \simeq \hbar$) yields that $\Delta t = N$ (for electrons $\hbar N$), demonstrating that N is associated with a time scale of the cavity: a small (large) density of states implies a fast (slow) response.

Among the many situations where the DOS plays a role we just mention the Einstein relation, which takes for electrons the following form [32],

$$\sigma_e(E) = \frac{e^2}{\hbar} \rho(E) D(E) . \quad (1.8)$$

This relation establishes the fundamental connection between electronic conductivity and diffusion, and is one of the most important results of electron transport theory. The left hand side of Eq. (1.8) is not defined for light because Eq. (1.5) has no meaning for light. Fortunately the Landauer formula enables us to rewrite the Einstein relation in terms of transport variables that do also exist for light. For the slab geometry [33] one deduces,

$$T(E) \frac{L}{A} = 2\pi\rho(E) D(E) . \quad (1.9)$$

We will demonstrate that the corresponding result for light transport reads,

$$T(\omega) \frac{L}{A} = 2\pi W(\omega) D(\omega) . \quad (1.10)$$

In the context of our discussion on the difference between dynamic and stationary response these two equations are fascinating since they relate two dynamic transport properties (rhs of Eqs. (1.9) and (1.10)) to one stationary transport quantity (lhs). For the case of light the time-delay in $D(\omega)$ is apparently canceled by $W(\omega)$, because $T(\omega)$ describes the stationary flow of energy, which by definition must be free from delay effects. Examples can be given where delay factors as large as a hundred cancel in the right hand side of relation (1.10).

2. CLASSICAL WAVES VERSUS DE BROGLIE WAVES

Despite the historical fact that Maxwell's electromagnetic wave equations were formulated some thirty years before the first formulation of Schrödinger's equation for De Broglie waves showed up, quantum phenomena have always received more attention. "Quantum" was, and perhaps still is, in fashion. So it is not surprising that modern formulations of classical wave propagation make use of methods that were originally developed for electrons. This is a logical consequence of the close resemblance of the wave equations. Analogies and differences between light and electrons will continue to appear in the rest of this work. An interesting observation one can make is that many "quantum phenomena" were called "quantum" only because they were studied for De Broglie waves but, being essentially interference phenomena, they also exist for classical waves. Recent experimental work on optical systems has indeed shown the existence of many of these [34], such as weak localization and quantized conductances.

We will write the equations of motion for light propagation in Schrödinger form which is,

$$i\hbar\partial_t\Psi(\mathbf{r},t) = H\Psi(\mathbf{r},t) . \quad (2.1)$$

The Hamiltonian operator determines the time-evolution and is given by $H = \hbar^2\mathbf{p}^2/2m + V(\mathbf{r})$ for a particle of mass m , momentum \mathbf{p} in a potential $V(\mathbf{r})$. The quantized momentum \mathbf{p} denotes the operator $-i\nabla$.¹

The corresponding eigenvalue equation for energy E is,

$$\left\{ \mathbf{p}^2 + \frac{2m}{\hbar^2} [V(\mathbf{r}) - E] \right\} \Psi_E(\mathbf{r}) = 0 . \quad (2.2)$$

In the rest of this paper we shall use units such that

$$m \equiv \frac{\hbar^2}{2} . \quad (2.3)$$

Maxwell's equations in the presence of a static dielectric substance $\varepsilon(\mathbf{r})$ can be combined to the so-called Helmholtz equation for the electric field $\mathbf{E}(\mathbf{r},t)$. Throughout this paper we will use the Heaviside-Lorentz (Rationalized Gaussian) units as discussed in Table 2 of Jackson [35]. In these units the Helmholtz equation reads,

$$\frac{\varepsilon(\mathbf{r})}{c_0^2} \partial_t^2 \mathbf{E}(\mathbf{r},t) + \nabla \times \nabla \times \mathbf{E}(\mathbf{r},t) = 0 . \quad (2.4)$$

The vector nature of light and the second derivative with respect to time are two differences with the

¹We will use a notation slightly different from conventional quantum mechanics in order to make the analogy with classical waves as clear as possible. In this way we can use the words "momentum" and "wavenumber" next to each other. In addition in formulas when no confusion arises we will not make a notational difference between the momentum operator \mathbf{p} and its eigenvalue \mathbf{p} .

Schrödinger equation. In terms of the quantum-mechanical momentum operator we can represent the double rotation formally as $-\mathbf{p}^2 \Delta_{\mathbf{p}}$. The tensor of rank two defined by

$$(\Delta_{\mathbf{p}})_{ij} \equiv \delta_{ij} - \frac{p_i p_j}{p^2} \equiv (\mathbf{I} - \hat{\mathbf{p}}\hat{\mathbf{p}})_{ij} , \quad (2.5)$$

projects upon the space of transverse fields (normal to \mathbf{p}). The eigenvalue equation for frequency ω can be written as,

$$\left\{ \mathbf{p}^2 \Delta_{\mathbf{p}} + \left(\frac{\omega}{c_0} \right)^2 [1 - \varepsilon(\mathbf{r})] - \left(\frac{\omega}{c_0} \right)^2 \right\} \mathbf{E}_{\omega}(\mathbf{r}) = 0 . \quad (2.6)$$

Comparing both eigenvalue equations leads us to identify the “light energy”

$$E_{\text{light}} = (\omega/c_0)^2 , \quad (2.7)$$

which shows that it is proportional to the square of the frequency, and a “light potential”,

$$V_{\text{light}}(\mathbf{r}) = [1 - \varepsilon(\mathbf{r})] E_{\text{light}} . \quad (2.8)$$

Since for free motion $E \sim \mathbf{p}^2$ and $\omega^2 \sim E_{\text{light}} \sim \mathbf{p}^2$ we find that the dispersion law is parabolic for quantum particles and linear for light. As ω^2 is always positive, the bound states of the Schrödinger equation have been lost in our identification scheme for light in dielectric systems ($\varepsilon(\mathbf{r}) > 0$).

The “energy dependency” of the light potential has far reaching consequences for multiple light scattering and is a crucial property when comparing light with electrons. One of the manifestations of the energy dependence is well known and concerns light scattering at low frequencies. Since the light potential $V_{\text{light}} \sim E_{\text{light}}$ it follows that the strength of the potential is “small” at “small” frequencies, and one may expect the Born approximation for the scattering cross-section σ_{scat} to apply. Then,

$$\sigma_{\text{scat}}(\omega) \sim |V_{\text{light}}|^2 \sim E_{\text{light}}^2 \sim \omega^4 . \quad (2.9)$$

This strong frequency dependence of the scattering cross-section was first predicted by Lord Rayleigh [36] in the previous century and explains why blue sun light is considerably more scattered in the Earth atmosphere than red sun light. In contrast to this long-wavelength behavior the cross-section for electrons goes to a constant - the s-wave limit - and usually far from the first Born approximation. At large energies light scattering can be treated in the (by no means simple) framework of geometrical optics [42].

By construction energy states are stationary solutions of the wave equation. For one (positive) energy the identification scheme between light and quantum waves is one-to-one. The conclusion is that light and quantum waves behave “analogously” in stationary experiments. A dynamical situation is one in which more than one energy eigenfunction features. As soon as one addresses dynamical situations the energy dependence of the interaction will enter in and will cause differences between results obtained for electrons and electromagnetic radiation. The application of conservation laws is an example of an inherently dynamical situation. Indeed, conservation laws in light scattering are notably different from the ones in Schrödinger quantum theory. In general a conservation law is a relation between a density W and a current density \mathbf{J} taking the form,

$$\partial_t W + \nabla \cdot \mathbf{J} = 0 . \quad (2.10)$$

From this formula it is clear that the density W characterizes the dynamic ($\partial_t \neq 0$), and the current

density \mathbf{J} the stationary ($\partial_t = 0$) properties of wave propagation.

For quantum mechanical wave functions in a real-valued potential field it is easily shown from Eq. (2.1) that

$$W = |\Psi|^2 \quad ; \quad \mathbf{J} = \frac{\hbar}{m} \operatorname{Re} \Psi^* \mathbf{p} \Psi , \quad (2.11)$$

interpreted as a probability density and a probability current density. On the other hand for light in a lossless dielectric,

$$W = \frac{1}{2} \varepsilon(\mathbf{r}) |\mathbf{E}|^2 + \frac{1}{2} |\mathbf{B}|^2 \quad ; \quad \mathbf{J} = c_0 \operatorname{Re} \mathbf{E}^* \times \mathbf{B} , \quad (2.12)$$

representing electromagnetic energy density and Poynting vector. Note that $\varepsilon(\mathbf{r})$ and $\mathbf{E}(\mathbf{r})$ depend both on position \mathbf{r} and can be strongly correlated. So the factor $\varepsilon(\mathbf{r})$ in Eq. (2.12) causes a difference between the conserved quantity W in Eqs. (2.11) and (2.12). In section III.B.3 we point out how this difference can be directly related to the energy dependence in the light potential. Here, it suffices to conclude that light and electrons behave differently in dynamic situations.

Contrary to the difference in the densities W , when using our identification scheme, the associated current densities are quite analogous. The major difference is caused by the vector nature of light. In fact, it is the current density that is measured in typical stationary experiments. This suggests one more time that light and electrons should behave “analogously” in stationary situations.

The identification scheme between the two wave equations becomes even clearer by ignoring polarization indices and replacing the electromagnetic field by a scalar field $\Psi(\mathbf{r}, t)$. The Helmholtz equation is then replaced by

$$\frac{\varepsilon(\mathbf{r})}{c_0^2} \partial_t^2 \Psi(\mathbf{r}, t) - \nabla^2 \Psi(\mathbf{r}, t) = 0 . \quad (2.13)$$

The quantity $\varepsilon(\mathbf{r})$ could still be called the dielectric constant, but the identification with a microscopic charge distribution has been lost in a scalar picture. The possible mapping on dynamics of sound will not be discussed here. The associated density and current density for scalar waves are [37],

$$W = \frac{1}{2} \varepsilon(\mathbf{r}) |c_0^{-1} \partial_t \Psi|^2 + \frac{1}{2} |\mathbf{p} \Psi|^2 \quad ; \quad \mathbf{J} = \operatorname{Im} (\partial_t \Psi)^* \mathbf{p} \Psi . \quad (2.14)$$

It is sometimes theoretically convenient to replace light by a scalar field. In that case one can think of $c_0^{-1} \partial_t \Psi$ as the electric field, and $\mathbf{p} \Psi$ as the magnetic field. For most cases discussed in this paper no essential physics will be lost after implementing this simplification. By replacing the double rotation $\nabla \times \nabla \times$ in the Helmholtz equation (2.4) by $-\nabla^2 + \nabla \nabla \cdot$ one can infer one term has gone lost in the scalar picture. This term is called the “logarithmic derivative” since it is, by Maxwell’s first law, proportional to the logarithmic space derivative of the dielectric constant. It is caused by the longitudinal field in Maxwell’s equations, or equivalently, by polarization charges on the boundaries.

3. RESONANT INTERACTION OF LIGHT AND MATTER

In this section we will discuss the interaction between light and matter from two different viewpoints. First we investigate how a material degree of freedom responds to a driving field through its polarizability. The simplest model is to use a polarizable classical or quantum-mechanical oscillator. In the second part we shall discuss how propagation of light is influenced by polarizable matter. We will look for the simplest objects from which light can be scattered according to the restrictions imposed by scattering theory. Our aim is to emphasize the equivalence of two apparently different methods, and to stress the equivalent role of light and matter in a unifying way.

3.1. Field Driven Oscillator

Consider an oscillator that is polarizable. This means that it can be driven by an electric field. The degree of freedom associated with the polarization has some (generalized) space coordinate \mathbf{Q} . The oscillator is coupled to the electromagnetic field through its dipole moment. We assume the dipole moment to be a function of the coordinate \mathbf{Q} and perform a Taylor expansion,

$$\boldsymbol{\mu}(\mathbf{Q}) = \boldsymbol{\mu}(0) + \frac{\partial \boldsymbol{\mu}}{\partial \mathbf{Q}}(0) \cdot \mathbf{Q} + \dots \quad (3.1)$$

If the polarizable oscillator is a harmonically bound bare electron the dipole moment can be simplified according to $(\partial \mu_i / \partial Q_j) = e \delta_{ij}$, in which e is the electronic charge. The interaction of the oscillator with the optical field is given by $V_{\text{int}} = -\boldsymbol{\mu} \cdot \mathbf{E}$, and by using expansion (3.1) we find a contribution proportional to \mathbf{Q} . As minus the gradient of an interaction represents a force, this term gives rise to a total force on the oscillator,

$$\mathbf{F} = \frac{\partial \boldsymbol{\mu}}{\partial \mathbf{Q}} \cdot \mathbf{E}. \quad (3.2)$$

Apparently one can only drive an oscillator by an optical field if its dipole moment changes during vibration. The classical equation of motion reads,

$$\frac{d^2 \mathbf{Q}(t)}{dt^2} + \frac{2}{T_2} \frac{d\mathbf{Q}(t)}{dt} + \omega_0^2 \mathbf{Q}(t) = \frac{q}{m} \mathbf{E}(t). \quad (3.3)$$

The oscillator has been given an eigenfrequency of ω_0 ; For simplicity we adopted $(\partial \mu_i / \partial Q_j) = q \delta_{ij}$. The phenomenological dephasing time T_2 has been introduced that describes the interaction of the oscillator with its environment. Later we will make the connection with scattering theory and find that T_2 should at least include radiation damping. The relative importance of radiative and radiationless decay is characterized by a parameter called the albedo a , $a \leq 1$. A situation with albedo $a = 1$ implies radiation damping only, and sets an upper limit to T_2 . For this reason the often treated lossless polarizable oscillator ($T_2 = \infty$) is unphysical. Radiative decay can be prohibited only in some special cavities.

The solution of equation of motion (3.3) contains two classes of responses. The first type is the transient response and the second is the force-induced reaction. The transient response will have died out after the elapsed time exceeds several damping times and the driven reaction is the only lasting effect.

In principle any electromagnetic field should be real-valued and complex notation should be used with care. If we assume that the interference between rotating and counter-rotating fields does not survive cycle averaging, the simpler notation of complex fields can be used. The treatment of the response to a harmonic driving electric field $\mathbf{E}(t) = \mathbf{E}_0 e^{-i\omega t}$ is then straightforward and can be found in any book on mechanics, electricity, or optics. The result is

$$\mathbf{Q}(t) = \frac{q/m}{\omega_0^2 - \omega^2 - i2\omega/T_2} \cdot \mathbf{E}_0 e^{-i\omega t}. \quad (3.4)$$

The driven displacements give rise to an induced dipole moment \mathbf{p} (no confusion will arise with the momentum operator \mathbf{p} introduced earlier)

$$\mathbf{p}(t) = q \mathbf{Q}(t) \equiv \boldsymbol{\alpha}(\omega) \cdot \mathbf{E}_0 e^{-i\omega t}, \quad (3.5)$$

where the (here diagonal) complex polarizability tensor

$$\boldsymbol{\alpha}(\omega) = \frac{q^2/m}{\omega_0^2 - \omega^2 - i2\omega/T_2} \mathbf{I}, \quad (3.6)$$

has been defined.

Standard arguments based on energy flow, show that the rate at which energy (cycle-averaged) is absorbed from the radiation field by the oscillator from a time-varying electric field with frequency ω is [38],

$$\frac{\partial U_{\text{rad}}}{\partial t} = -\frac{\partial U_{\text{osc}}}{\partial t} = -\ll \mathbf{E}(t) \cdot \frac{d\mathbf{p}}{dt} \gg_{\text{cycles}} = -\frac{\omega}{2} \mathbf{E}_0^* \cdot \text{Im } \boldsymbol{\alpha} \cdot \mathbf{E}_0. \quad (3.7)$$

It is useful to connect the *volume* loss due to extinction to a loss per area, as expressed by an (extinction) cross section. The relation is,

$$\frac{\partial U_{\text{osc}}}{\partial t} = \sigma_{\text{ext}} c_0 \times \left(\frac{1}{2} \mathbf{E}_0^2 \right) \Rightarrow \sigma_{\text{ext}} = \frac{\omega}{c_0} \text{Im } \alpha. \quad (3.8)$$

So far we only considered the ‘matter’ part of the problem. The extinction cross-section informs us about the rate of absorption out of the incoming beam induced by the oscillator. Where is this energy going? Strictly speaking the present ‘matter’ treatment of the oscillator does not allow us to answer that question. Part of the light captured from the incoming beam will be reradiated and requires therefore a treatment of the light propagation. All that one can say is that the reradiation process will be determined by $|\alpha|^2$.

The total cycle-averaged current radiated by an oscillating dipole is known to be $\omega^4 |\alpha|^2 \mathbf{E}_0^2 / 3c_0^5$ [39]. This will later be verified in scattering theory. The scattering cross section can be found from

$$\mathbf{J}_{\text{scat}} = \sigma_{\text{scat}} c_0 \times \left(\frac{1}{2} \mathbf{E}_0^2 \right) \Rightarrow \sigma_{\text{scat}} = \frac{2\omega^4}{3c_0^4} |\alpha|^2. \quad (3.9)$$

The proportionality factor ω^4/c_0^4 can readily be understood when realizing that the polarizability has the dimension of a volume and the cross-section the one of an area. Now we can tell how much of the light taken out of the beam will finally be reradiated and how much is really absorbed. For this purpose one defines the albedo

$$a(\omega) = \frac{\sigma_{\text{scat}}}{\sigma_{\text{ext}}} = \frac{2\omega^3}{3c_0^3} \frac{|\alpha|^2}{\text{Im } \alpha} = \frac{T_2}{T_{\text{rad}}} \leq 1. \quad (3.10)$$

Here $T_{\text{rad}} = 3mc_0^3/q^2\omega^2$ is the radiative decay time of the oscillator. Albedos ranging from 0.99999

(colloidal dielectric spheres and cold atomic gasses [40]) to 0.00001 (molecular oscillators in solids and liquids [41]) are relevant. In condensed matter the albedo can be very small because the polarizable oscillator is strongly coupled (due to small distances) to a large number (due to high density) of non-radiative decay channels. When the only dephasing process is radiation damping the albedo is equal to one and T_2 represents the radiation time, its maximum value. In that case the direction of the light is changed by the oscillator, without loss of energy.

3.2. Single Scattering of Light

The investigation of multiple scattering demands first of all a good understanding of single scattering. Single scattering is the building block for multiple scattering. As has been pointed out in the introduction the onset of multiple scattering is boosted when a number of conditions is met. Firstly, the particles must scatter the light to a very large extent with conservation of energy. In case of energy loss in single scattering, multiple scattering will be exponentially small in the order of scattering. Secondly, the multiple scattering may be enhanced when the single scattering is on a resonance. At a resonance the scattering cross-section reaches its maximum “unitary limit”, where it is proportional to the square of the wavelength. If single scattering is maximal, multiple scattering can be anticipated to be large as well. With modern lasers the frequency of the exciting light can be tuned accurately and chosen to coincide exactly with a resonant frequency.

Because of its simplicity the “Born approximation” for single scattering is popular in solid state physics. It avoids detailed calculations of single scattering and a minor extension suffices to have control over conservation laws. However, resonant scattering can never be addressed in this approach. As a matter of fact, the Born series is known to diverge near a resonance.

In the following subsections we will discuss a very simple model for single scattering of light, and compare it with the exact solution for a dielectric sphere. A number of general properties of single scattering will be addressed which will be of vital importance in multiple scattering: resonances, energy conservation, time delay, and near field. Before doing so we would like to convince the reader that the Mie solution - which already represents an oversimplifying picture of a typical particle in a typical experiment - is too complicated to use in a general multiple-scattering theory.

3.2.1. Mie Scatterers

Generally speaking, single scattering of light from a dielectric object with arbitrary shape is an exercise by itself. In line with a desire for simplicity it seems beneficial to limit oneself to spherical, so-called Mie particles. As the Mie solution is about the only problem that can be solved exactly it is always the first model treated in detail in text books on light scattering [42]. In Fig. 2 we show the scattering cross-section obtained from the Mie solution for a sphere with index of refraction $m = \sqrt{\epsilon} = 2.7$ and a size comparable to the wave length as a function of the frequency. It can be concluded that Mie scattering is full of resonances. The Mie solution is a complicated infinite sum of Bessel functions of the Hertz potentials. Part of the complicated behavior can be appreciated by the large number of resonances in Fig. 2. For two Mie particles already the solution is no longer available in any practical form and one must fall back on a time-consuming numerical integration of Maxwell’s equations [43].

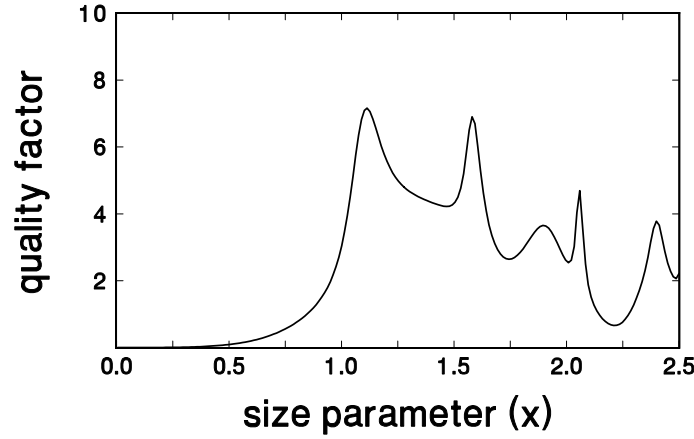


FIG. 2. Dimensionless quality factor of a dielectric sphere, defined as the total cross-section divided by the geometric cross-section, $Q \equiv \sigma/\pi a^2$, as a function of the size parameter $x \equiv \omega a/c_0$. The index of refraction of the sphere is $m = 2.7$.

The Mie solution can only be used for multiple scattering purposes after some tremendous simplifications have been carried out: ISA (see introduction), far-field approximation [17], and diffusion approximation. Even in that case keeping track of all polarization indices is complicated [44] [45].

3.2.2. Point Scatterers

The simplest scatterer one can possibly imagine is a point scatterer. We will now give an educational derivation of the properties of such a scatterer in three dimensions. We will extensively use the bra and ket notation known in quantum mechanics also for classical waves.

In quantum mechanics a point scatterer, labelled by the index i and located at position \mathbf{r}_i , is characterized by the local potential $V_i(\mathbf{r}, \mathbf{r}') = \langle \mathbf{r}' | V_i | \mathbf{r} \rangle = V_0 u \delta(\mathbf{r} - \mathbf{r}_i) \delta(\mathbf{r} - \mathbf{r}')$, where V_0 is a coupling strength with the same dimension as the energy (in our reduced units this is the dimension of an inverse length squared) and u a volume whose role will become clear later. Scattering theory tells us that a plane wave $|\mathbf{k}\rangle$ incident with wave number (momentum) \mathbf{k} and energy $E = \mathbf{k}^2$ on a target is distorted according to,

$$|\Psi_{\mathbf{k}}^+\rangle = [1 + G_0(E) T_i(E)] |\mathbf{k}\rangle. \quad (3.11)$$

In terms of the interacting potential, the T -operator or T -matrix is given by the Born series,

$$T_i(E) = V_i + V_i G_0(E) V_i + V_i G_0(E) V_i G_0(E) V_i + \dots. \quad (3.12)$$

Here $G_0(E) = (E + i\epsilon - H_0)^{-1}$ is the free Green's function (operator). It is the formal representation of a spherical wave emitted by the scatterer. A subtle infinitesimally small ϵ is used so that complex analysis can be used. The fact that ϵ is positive takes care that the spherical wave propagates outwardly and not inwardly. Substitution of Eq. (3.12) into Eq. (3.11) makes clear that the total wave is built up from multiple scattering from the potential combined with free propagation in between.

The Dirac delta-function of the zero-range potential allows us again to find the T -matrix as a simple geometric series when taking matrix elements. Most calculations are easiest when calculating matrix elements in the momentum representation, but they can always be transformed back to real space. We find,

$$\langle \mathbf{p} | T_i(E) | \mathbf{p}' \rangle = t(E) \exp[-i(\mathbf{p} - \mathbf{p}') \cdot \mathbf{r}_i],$$

$$t(E) = \frac{V_0 u}{1 - V_0 u \sum_{\mathbf{p}} G_0(E, \mathbf{p})} , \quad (3.13)$$

in which the diagonal matrix element

$$\langle \mathbf{p} | G_0(E) | \mathbf{p}' \rangle \equiv G_0(E, \mathbf{p}) \delta(\mathbf{p} - \mathbf{p}') = (E^2 - p^2 + i\epsilon)^{-1} \delta(\mathbf{p} - \mathbf{p}') .$$

In real space the T -matrix takes the form

$$T_i(\mathbf{r}_1, \mathbf{r}_2, E) = t(E) \delta(\mathbf{r}_1 - \mathbf{r}_i) \delta(\mathbf{r}_2 - \mathbf{r}_i) , \quad (3.14)$$

which demonstrates clearly the extreme simplification obtained by using point scatterers. Only exactly at the position of the scatterer will a scattering event take place.

Expression (3.12) has been obtained for electrons but may be translated to light scattering. To this end we must replace the free Hamiltonian and the energy by

$$H_0 \rightarrow \mathbf{p}^2 \Delta_{\mathbf{p}} \text{ and } E \rightarrow (\omega/c_0)^2 ,$$

and introduce an energy-dependent potential. Polarization indices can be added to account for the vector character of light. As a result, both the t -matrix and the Green's function become second-rank tensors. In what follows we will evaluate the expression for the t -matrix (3.13) explicitly for electrons and light.

a. Electrons Let us first investigate the point scatterer for De Broglie waves, keeping the interaction of a conduction electron with a defect in the solid state in mind. Since $G_0(E, p) = (E + i\epsilon - p^2)^{-1}$ we rapidly conclude that the momentum summation $\sum_{\mathbf{p}} G_0(E, \mathbf{p})$ in Eq. (3.13) is infinite in three dimensions. We can improve on this by writing²

$$\sum_{\mathbf{p}} G_0(E, \mathbf{p}) = \sum_{\mathbf{p}} \left[\frac{1}{E - p^2} + \frac{1}{p^2} \right] - \sum_{\mathbf{p}} \frac{1}{p^2} , \quad (3.15)$$

where the divergent part has been separated. The first integral is elementary and yields $i\sqrt{E}/4\pi$ if $E \geq 0$. The second term may be regularized by introducing a positive, energy-independent variable Λ that acts as a cut-off. The divergence signifies that a normal point potential does not scatter. We must therefore introduce a physical size a of the target. The cut-off Λ is chosen to be the inverse of the smallest length scale a in our problem: $\Lambda \equiv a^{-1}$. It is consistent to take for the not yet specified volume u , $u \equiv a^3$. With the cut-off procedure we can substitute (3.15) into (3.13) and find

$$t(E) = \left[a^{-1} \left(\frac{1}{V_0 a^2} - 1 \right) + \frac{i}{4\pi} \sqrt{E} \right]^{-1} . \quad (3.16)$$

The variable $a^{-1}(1/V_0 a^2 - 1)$ is a finite quantity which depends only on the scattering potential. We shall denote it by $-1/4\pi f$. We wind up with,

²We are indebted to Dr. Theo Nieuwenhuizen for bringing this simple derivation to our attention. The derivation agrees with the formal boundary value treatment in Ref. [46].

$$t(E) = \frac{-4\pi}{1/f - i\sqrt{E}}. \quad (3.17)$$

This model is known as the Fermi interaction, and was very popular in the early days of nuclear physics. The real-valued parameter f is called the scattering length, and can be positive or negative. The final result we obtained in Eq. (3.17) coincides with mathematical (boundary condition) treatments [46]. The good news is that point scattering exists, the bad news is that this model cannot be used to describe resonant scattering of electrons as there is no resonance at any finite energy E .

b. Light In the case of light most operators become tensors of rank two. The light potential is $V_i(\omega, \mathbf{r}', \mathbf{r}) = -\gamma u (\omega/c_0)^2 \delta(\mathbf{r} - \mathbf{r}_i) \delta(\mathbf{r} - \mathbf{r}')$. Recalling (2.8) we identify γ as the polarizability density of the matter which the scatterer is made of; u is again a volume. With the notation of section II we have,

$$\sum_{\mathbf{p}} \mathbf{G}_0(\omega, \mathbf{p}) = \sum_{\mathbf{p}} \frac{1}{(\omega/c_0)^2 + i\epsilon - \mathbf{p}^2 \Delta_{\mathbf{p}}} = \sum_{\mathbf{p}} \left[\frac{\hat{\mathbf{p}}\hat{\mathbf{p}}}{(\omega/c_0)^2} + \frac{\Delta_{\mathbf{p}}}{(\omega/c_0)^2 + i\epsilon - p^2} \right], \quad (3.18)$$

consisting of a longitudinal part with projector $\hat{\mathbf{p}}\hat{\mathbf{p}}$ and a transverse part with projector $\Delta_{\mathbf{p}}$ orthogonal to it, as defined in Eq. (2.5). The latter describes the travelling wave solutions of Maxwell's equations. The longitudinal part is an electrostatic part and does not propagate. It is divergent but we can regularize with a momentum cut-off such that $\sum_{\mathbf{p}} \hat{\mathbf{p}}\hat{\mathbf{p}} \rightarrow 1/3 u^{-1} \mathbf{I}$. The one-third is recognized as the “depolarization factor” of a sphere and shows up here in the scattering amplitude from the angular integral of the longitudinal projector. The second term on the rhs. of (3.18) is also diverging but can be cut off with a positive parameter Λ as we did for the Fermi pseudo potential, with an additional factor of 2/3 from the angular average of $\Delta_{\mathbf{p}}$. We obtain [47]

$$\mathbf{t}(\omega) = - \left[\left(\frac{1}{\gamma} + \frac{1}{3} \right) \frac{1}{u (\omega/c_0)^2} - \Lambda - \frac{i\omega}{6\pi c_0} \right]^{-1} \mathbf{I}. \quad (3.19)$$

We observe that the longitudinal singularity - responsible for the factor 1/3 - can be absorbed into a modified polarizability. In physical language this implies that the polarizability of the sphere as a whole is different from its local value γu . Indeed, electrostatics [35] (or equivalently the exact Mie solution at low frequencies [42]) shows that the polarizability of a sphere is not γu but rather

$$\gamma_{\text{sph}} u \equiv \alpha_{\text{sph}} = 3 \frac{\varepsilon - 1}{\varepsilon + 2} u, \quad (3.20)$$

coinciding with our cut off procedure. The difference between local and total polarization arises from depolarization charges on the boundary of the sphere. It is a genuine vector effect, absent in a scalar model for light.

The transverse singularity Λ does not have the same frequency dependence as the interaction part $1/(\omega/c_0)^2 \alpha_{\text{sph}}$. Therefore the influence of the cut-off cannot be simply replaced by a frequency-independent scattering length. In more mathematical language this means that the point scatterer for light is not renormalizable in the normal Hilbert space of square-integrable wave-functions. [48]. Introducing the length scale Γ as $4\pi\Gamma \equiv 1/\Lambda$ we shall write the final result in the form,

$$\mathbf{t}(\omega) = \frac{-4\pi\Gamma\omega^2}{\omega_0^2 - \omega^2 - (2/3) i\Gamma\omega^3/c_0} \mathbf{I}, \quad (3.21)$$

in which $\omega_0^2 \equiv 4\pi\Gamma c_0^2/\alpha_{\text{sph}}$. An important conclusion that can be drawn is that the simplest model of dielectric light scattering, contrary to the one of electron scattering, necessarily has a resonance. The t -matrix in Eq. (3.21) cannot be a solution of Schrödinger's equation. The solution has a strong resemblance to the polarizability (3.6) of the radiating dipole with eigenfrequency ω_0 discussed in the previous subsection. The scattering amplitude (3.21) can be reproduced with mathematical boundary-condition treatments [48] [49].

Adding the two delta-functions as in Eq. (3.14) gives the dipole a position in space, which will make it possible to apply it to multiple scattering. The width of the resonance, equal to $\Gamma\omega_0^2/3c_0$, plays the role of $1/T_2$ when comparing to Eq. (3.6). Further inspection of the solution for the driven atomic oscillator in Eq. (3.6), letting $q = e$, shows that the value for Γ appropriate for the oscillator is $\Gamma \approx e^2/m_e c_0^2 \equiv r_0$, a quantity referred to as the classical electron radius. Since $\Gamma \sim 1/\Lambda$, the appropriate inverse momentum cut-off in Eq. (3.19) thus turns out to be the classical electron radius. This is the only length scale in our problem that can be constructed from *classical* natural constants. For this reason the Bohr radius of the atom disqualifies.

The scattering cross-section defines the scattered flux by the object relative to the incoming flux density (which is per unit of area). To find the scattering cross-section it is convenient - though by no means necessary since the scattered flux does not depend on the distance from the scatterer - to write the scattered wave (3.11) in the far field. It is customary to distinguish between the scattered electric field perpendicular (r) to and within (l) the plane of scattering spanned by incoming and outgoing wave vectors. For a plane light wave with wave vector $\mathbf{k} = \omega\hat{\mathbf{z}}/c_0$ along the z -direction ,

$$\Psi_{\text{in}}(\mathbf{k}, \mathbf{r}) \rightarrow \begin{pmatrix} E_{l,\text{in}} \\ E_{r,\text{in}} \end{pmatrix} e^{ikz} ,$$

the electric field (3.11) far away from the scattering obstacle takes the form

$$\begin{pmatrix} E_l^+ \\ E_r^+ \end{pmatrix} = \begin{pmatrix} E_{l,\text{in}} \\ E_{r,\text{in}} \end{pmatrix} e^{ikz} + \begin{pmatrix} \cos\theta & 0 \\ 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} E_{l,\text{in}} \\ E_{r,\text{in}} \end{pmatrix} t(\omega) \frac{e^{ikr}}{-4\pi r} . \quad (3.22)$$

The differential scattering cross-section, summed over polarization and averaged over the azimuthal angle ϕ of the incident polarization vector (giving a factor of 1/2) then becomes

$$\frac{d\sigma_{\text{scat}}(\omega)}{d\Omega} = \frac{1}{2} \times \frac{1}{4\pi} |t(\omega)|^2 \times (1 + \cos^2\theta) .$$

The total scattering cross-section is found by integrating over $d\theta \sin\theta$,

$$\sigma_{\text{scat}}(\omega) = \frac{|t(\omega)|^2}{6\pi} . \quad (3.23)$$

In Fig. 3 we have plotted the scattering cross-section of a point scatterer as a function of the frequency ω of the incoming field. Compared to the Mie solution for a finite sphere one observes that only one resonant frequency of the Mie sphere has survived.

Two other differences with Mie scattering can be mentioned that cannot be inferred from the plot. In general a Mie sphere scatters the light rather anisotropically. The phase function, which is the angular distribution of the scattered light intensity, of our point scatterer follows from

$$\frac{d\sigma_{\text{scat}}(\omega)}{d\Omega} = \sigma_{\text{scat}}(\omega) \times \Phi(\theta) \Rightarrow \Phi(\theta) = \frac{3}{4} (1 + \cos^2\theta) , \quad (3.24)$$

and is a purely geometrical factor arising from the mutual direction of the polarization vectors. For the Fermi point interaction in quantum mechanics this distribution is isotropic.

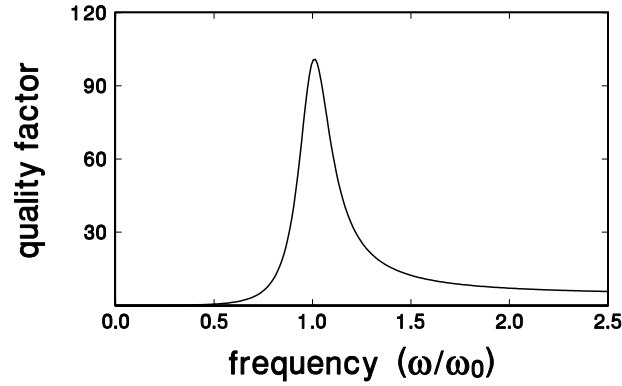


FIG. 3. Dimensionless quality factor of a classical oscillator (equivalent to a point scatterer) with an internal resonance at ω_0 as a function of frequency. The damping of the oscillation is $T_2 = 10/\omega_0$. The quality factor is defined as the total cross-section divided by the “geometric (Thomson) cross-section” of a free electron, $Q \equiv \sigma/\pi r_0^2$, where r_0 is the classical radius of the electron. Due to resonant scattering the cross-section of the harmonically bound oscillator can exceed the one of a free charge by orders of magnitude. Hence Q can exceed unity by orders of magnitude, contrary to the quality factor of the dielectric sphere.

The second difference with a Mie particle concerns the scattered field near the particle. Given an incident electric field $\mathbf{e}_{\text{in}}^{(j)}|\mathbf{k}\rangle$, where $\mathbf{e}_{\text{in}}^{(j)}$ is the polarization vector of the incoming plane wave associated with polarization j , the field radiated by the point particle is the one of a radiating dipole. By using $\mathbf{R} \equiv \mathbf{r} - \mathbf{r}_i$ we can drop the superscript i on the matrix elements of the T -matrix in the momentum representation. According to the Lippmann-Schwinger equation (3.11) the outgoing wave becomes,

$$\langle \mathbf{r} | \mathbf{E}_{\mathbf{k},j}^+ \rangle = \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}) = \{ \mathbf{I} \exp(i\mathbf{k} \cdot \mathbf{r}) + t(\omega) \mathbf{G}_0(\omega, \mathbf{r} - \mathbf{r}_i) \} \cdot \mathbf{e}_{\text{in}}^{(j)}, \quad (3.25)$$

in which $\omega = |\mathbf{k}| c_0$ and,

$$\mathbf{G}_0(\omega, \mathbf{R}) = \frac{e^{ikR}}{-4\pi R} \left\{ [\mathbf{I} - \widehat{\mathbf{R}}\widehat{\mathbf{R}}] - \left(\frac{1}{ikR} + \frac{1}{(kR)^2} \right) [\mathbf{I} - 3\widehat{\mathbf{R}}\widehat{\mathbf{R}}] \right\}.$$

In Mie scattering the near field is much more complex. Anisotropy and near field effects are both very difficult to deal with in multiple scattering, but do not always lead to fundamentally new physics. For that reason the point dipole derived in this section is of high value to investigate the multiple scattering regime.

Having solved the light scattering from a point particle completely, we can make our first attempt to trace back the material degrees of freedom. In general, the local polarization density $\mathbf{P}(\omega, \mathbf{r})$ induced by the eigenfunction (3.25) is given by,

$$\mathbf{P}(\omega, \mathbf{r}) = [\varepsilon(\mathbf{r}) - 1] \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}) \equiv -\frac{V(\omega, \mathbf{r})}{(\omega/c_0)^2} \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}), \quad (3.26)$$

which we have again written in terms of the frequency-dependent potential $V(\omega, \mathbf{r})$. Inserting the electric field scattered by particle i , as given by Eq. (3.11) we obtain,

$$\mathbf{P}_i(\omega, \mathbf{r}) = -\frac{c_0^2}{\omega^2} \langle \mathbf{r} | V_i(\omega) | \mathbf{E}_{\mathbf{k},j}^+ \rangle = -\frac{c_0^2}{\omega^2} \langle \mathbf{r} | \mathbf{T}_i(\omega) | \mathbf{E}_{\mathbf{k},j}^{\text{in}} \rangle = -\frac{c_0^2}{\omega^2} \sum_{\mathbf{p}} e^{i\mathbf{p} \cdot \mathbf{r}} \mathbf{T}_{\mathbf{p}\mathbf{k}}(\omega) \cdot \mathbf{e}_{\text{in}}^{(j)} \quad (3.27)$$

For the point particle we arrive at,

$$\mathbf{P}_i(\omega, \mathbf{r}) = -\delta(\mathbf{r} - \mathbf{r}_i) \frac{\mathbf{t}(\omega)}{(\omega/c_0)^2} \cdot \mathbf{E}_{\mathbf{k},j}^{\text{in}}(\mathbf{r}_i) \quad (3.28)$$

This implies that the polarizability of the scatterer is given by

$$\alpha(\omega) = -\mathbf{t}(\omega) \frac{c_0^2}{\omega^2}. \quad (3.29)$$

This agrees with the material treatment given in the previous section.

c. Optical Theorem Does the point scatterer obtained in Eq. (3.21) scatter light elastically? We could not really obtain this information from our phenomenological model because we needed the equations for the light. But within the framework of our scattering theory we can answer this question. Conservation of energy puts severe constraints on the scattering amplitude by means of the unitarity of the S -matrix. The identity that guarantees conservation of energy for light or probability for electrons is called the Optical Theorem. For light it takes the form [50],

$$-\frac{\text{Im } T_{j\mathbf{k}j\mathbf{k}}(\omega)}{\omega/c_0} = \sum_{j'} \int d\hat{\mathbf{k}}' \frac{|T_{j\mathbf{k}j'\mathbf{k}'}(\omega)|^2}{(4\pi)^2}. \quad (3.30)$$

Here $T_{j\mathbf{k}j'\mathbf{k}'}(\omega)$ is the scattering amplitude for scattering from direction \mathbf{k} and polarization j into direction \mathbf{k}' and polarization j' . Transversality allows two orthogonal polarization states of the light in the far field for each wave vector on the energy shell $|\mathbf{k}| = \omega/c_0$ (three polarization states are nevertheless possible in the near field!). The right hand side of Eq. (3.30) is the total scattering cross-section of the object or, equivalently, the total current associated with the scattered wave. The left hand side is called the extinction cross section, and refers to the current captured by the object.

For the point scatterer derived in Eq. (3.21) the far-field scattering amplitude is

$$T_{j\mathbf{k}j'\mathbf{k}'}(\omega) = t(\omega) \mathbf{e}^{(j)}(\mathbf{k})^* \cdot \mathbf{e}^{(j')}(\mathbf{k}'), \quad (3.31)$$

so that the Optical Theorem simplifies to,

$$-\frac{\text{Im } t(\omega)}{\omega/c_0} = \frac{|t(\omega)|^2}{6\pi}. \quad (3.32)$$

The general form of the t -matrix that obeys this identity is $t(\omega) = -4\pi/[F(\omega) - 2i\omega/3c_0]$, in which $F(\omega)$ is a real-valued, frequency dependent scattering strength, sometimes referred to as the optical potential in view of the Fermi model (3.17). Our point scatterer is of this form when the polarizability is real-valued. In that case the point object scatters the light elastically.

In the previous section we have indicated how to extend the treatment towards cases in which part of the extinction is due to absorption rather than to scattering. In the present treatment this amounts to adding to $F(\omega)$ a negative imaginary part. In that case the lhs. of the Optical Theorem exceeds the rhs. This gives the same inequality as derived in Eq. (3.10).

3.2.3. Time Delay and Near Field

One might suppose that the differential scattering cross-section characterizes a scattering process completely. Indeed for many applications this statement is true. Source and detector are often far

separated from the target and experimentalists are not interested in what is happening near or even inside the particle. All that one measures is the light or the electron that is analyzed in the far field. In the case of De Broglie waves one often goes sometimes one step further by (erroneously) stating that it is even unphysical to pose questions concerning processes occurring in near field.

One might put forward that in multiple scattering the particles are not infinitely far separated from each other and that the near field becomes relevant. However, if the scattering mean free path - the average distance between two collisions - is much larger than the wavelength, a condition usually fulfilled, the far-field approximation suffices, even for multiple scattering. On the basis of this reasoning one would expect that near-field and fields inside the scatterer hardly ever matter.

As soon as one starts asking questions regarding the dynamics of the propagation the situation turns out to become entirely different. The most important dynamical scattering properties are the rigorously defined *delay time* and *dwell time*. The delay time can be interpreted as the duration of the scattering process. The dwell time can be interpreted as the time the wave spends in the potential region (We assume the potential to have finite support. This may be unrealistic for electron scattering but is always true for light). Delay can be anticipated to occur only inside the object since far outside the motion is again free. Hence, both time scales may be anticipated to be roughly equal. Time delay is a characteristic property of resonant scattering. In what follows we shall demonstrate that the field inside the objects indeed plays a crucial role. We will point out that by studying dynamical aspects of light scattering one recover many traces of the material degrees of freedom that have been integrated out earlier.

a. Time Delay of Electrons Most of the knowledge of time delay has been developed in quantum mechanics. The delay time was first introduced by E.P. Wigner in 1946 [51]. If $\Phi(E, \Omega)$ is the quantum mechanical phase shift of the scattered wave (read *t*-matrix) for some channel Ω , the time-scale

$$\tau_\phi(E, \Omega) = \hbar \frac{d\Phi(E, \Omega)}{dE}, \quad (3.33)$$

is called the Wigner phase-delay time for the channel Ω . Wigner demonstrated that this quantum-mechanical definition coincides well with what would classically be called the time delay of an incident particle with energy E . A great contribution of Wigner was his observation that $\tau_\phi(E)$ can be very large if a quantum-mechanical scattering resonance occurs. It is generally a positive quantity, but can also be negative for repulsive interactions. This would correspond to a time advance rather than a delay, but this advance is bound by a lower causality limit related to the extent of the scattering region [52].

Later work by Jauch *et al.* [53] and Martin [54] established a physically more transparent but mathematically equivalent expression for the phase delay time in terms of the exact continuum eigenfunction $\Psi_E^+(\mathbf{r})$,

$$\tau_\phi(E) = \frac{1}{\sigma_{\text{scat}}(E)v} \int d^3\mathbf{r} \left[|\Psi_E^+(\mathbf{r})|^2 - 1 \right], \quad (3.34)$$

$\sigma_{\text{scat}}(E)$ being the scattering cross-section and v the velocity of the incident particle. The delay time has now been averaged over angles, which can be done if we don't care in what direction the scattered wave will finally go. We have divided by the dimensionless number $\sigma_{\text{scat}} k^2 / \pi$ in order to get the delay time per open scattering channel. In words, the space integral of the excess accumulated

probability density around the scatterer is equal to the average time-delay the scattering particle will finally undergo. Since for an electron the local probability density is proportional to the local charge density, the right hand side of Eq. (3.34) can be identified as the total “screened charge” around the scattering potential. Work by Friedel [55] in the sixties demonstrated the vital importance of this notion for electron-impurity scattering in the solid state, namely the existence and modification of the Fermi level E_F in the presence of impurities.

From the point of view of scattering theory Eq. (3.34) is surprising since a dynamic scattering property, the delay time, is related to a stationary eigenfunction. Nevertheless, a prize has to be paid: We need this eigenfunction *everywhere*, not only in the far field. This suggests that “near field” (off-shell) and “dynamics” must have a lot in common. The value of this conclusion cannot be underestimated. It plays a key role in the present paper.

Eq. (3.34) can be understood heuristically. The total excess probability of finding the particle near the scatterer is given by the integral on its right hand side. The rate with which this excess probability is carried away is the total current through a closed surface encompassing the scatterer and given by $\sigma_{\text{scat}} v$. The delay is then typically given by the ratio of excess probability and current. This reproduces the formal scattering result.

We will use Eq. (3.34) to define the “dwell time” in three dimensions as

$$\tau_d(E) \equiv \frac{1}{\sigma_{\text{scat}}(E)v} \int_S d^3\mathbf{r} \left| \Psi_E^+(\mathbf{r}) \right|^2. \quad (3.35)$$

This definition generalizes the one known in one dimension [56] [57]. The difference between Eq. (3.34), describing the phase delay time, and the definition for the dwell time is that in the latter case the integral extends only over the scatterer region S and the contribution of the incident plane wave is no longer subtracted. By construction the dwell time is a property of the quantum mechanical field inside the scattering object only. Quite recently both dwell and delay time have been given renewed attention in the context of one-dimensional quantum tunneling [58].

The Wigner phase-delay time for quantum mechanical scattering in three dimensions, averaged over outgoing channels, can be expressed in terms of the t -matrix. In principle this follows by inserting the eigenfunction defined in Eq. (3.11) into Eq. (3.34). The algebra is tedious but straightforward. One obtains [59],

$$\int d^3\mathbf{r} \left[\left| \Psi_E^+(\mathbf{r}) \right|^2 - 1 \right] = -\frac{d}{dE} \text{Re } T_{\mathbf{k}\mathbf{k}}(E) + 2\sqrt{E} \int d\Omega_{\mathbf{k}'} \frac{d\sigma}{d\Omega} \frac{\partial \Phi(E, \Omega)}{\partial E}. \quad (3.36)$$

Here $T_{\mathbf{k}\mathbf{k}'}(E)$ is the scattering amplitude for scattering from \mathbf{k} to \mathbf{k}' at energy $E = k^2$; $d\sigma/d\Omega = |T_{\mathbf{k}\mathbf{k}'}(E)|^2 / (4\pi)^2$ is the associated differential scattering cross-section. In view of relation (3.33) the first term on the rhs. may look unexpected. It describes the phase-delay in the forward scattering channel. This channel always plays a subtle role in scattering theory since this channel does not give rise to real scattering. In fact this channel is to be associated with the coherent beam mentioned earlier. One of the “beauties” of scattering theory is that energy dependence of the far-field amplitude, the rhs. of Eq. (3.36), provides knowledge of what is going on near the scattering obstacle, the lhs. of Eq. (3.36).

b. Time Delay of Light Most formulae for electrons allow a straightforward mathematical translation for light. Eq. (3.35) for the dwell time becomes for light with frequency ω in a dielectric region S ,

$$\tau_d(\omega) = \frac{W(\omega, S)}{\sigma_{\text{scat}}(\omega)c_0} = \frac{1}{\sigma_{\text{scat}}(\omega)c_0} \int_S d^3\mathbf{r} W(\omega, \mathbf{r}) . \quad (3.37)$$

We recall that units have been chosen such that the energy density of a plane wave in vacuum is normalized to unity. $W(\omega, S)$ is the electromagnetic energy inside S at frequency ω . It can be calculated straightforwardly from the Mie solution [60].

Fig. 4 shows the outcome of a computation for Mie spheres with radius a and index of refraction $m = 2.73$ (corresponding to the value for titanium-dioxide, a material often used in laboratory experiments). Near a resonant frequency both energy density and scattering cross-section become large. Contrary to the cross-section, the accumulated energy density is not subject to a unitary maximum and can be very large. In particular, near the size parameter $x \equiv \omega a/c_0 = 4.59$ we find $W \approx 2500 \frac{4}{3}\pi a^3$ and $\sigma \approx 3.9 \pi a^2$. This results in a dwell time as large as $850 a/c_0$. For particles with a size of 220 nm this corresponds to the time that light needs to propagate 190 μm in vacuum. This is roughly 5 times larger than optically thick samples that are used to study multiple scattering of visible light!

Let us investigate the dwell time $\tau_d(\omega)$. The electromagnetic energy density is given by Eq. (2.12). Let us adopt for simplicity “equipartition” between electric and magnetic energy (this can be demonstrated in a stationary situation). We then obtain,

$$\begin{aligned} W(\omega, S) &= \int_S d^3\mathbf{r} \varepsilon(\mathbf{r}) \left| \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}) \right|^2 = \int_S d^3\mathbf{r} \left| \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}) \right|^2 + \int_S d^3\mathbf{r} [\varepsilon(\mathbf{r}) - 1] \left| \mathbf{E}_{\mathbf{k},j}^+(\mathbf{r}) \right|^2 \\ &\equiv W_{\text{rad}}(\omega, S) + W_{\text{pot}}(\omega, S) . \end{aligned} \quad (3.38)$$

The first term of the right hand side of Eq. (3.38) is analogous to the probability inside S for quantum waves and may here be called the radiation energy. The second term seems to be an extra contribution for light. The dielectric constant defines the local polarization density according to $\mathbf{P}(\mathbf{r}) = [\varepsilon(\mathbf{r}) - 1] \mathbf{E}(\mathbf{r})$ so that this contribution can be identified as the integral of the total potential energy density $\mathbf{E} \cdot \mathbf{P}$ stored in the dielectric object. This partitioning has been effected in Eq. (3.38). In our scattering theory, the potential energy represents the material degrees of freedom. If the dielectric constant of the object is much larger than the vacuum value, the potential energy part dominates completely. As a result, the time that the light spends in the dielectric scatterer is essentially the time it takes to store the potential energy. Note the surprising observation that this argument is apparently not valid for quantum mechanical potential scattering! In section IV.C.2 we will infer that the contribution of longitudinal electric fields in the “radiation energy” (also absent for De Broglie waves) should also be considered as a material contribution. The longitudinal field arises from polarization charges on the boundary of S . The real radiation energy is thus contained in the *transverse*, propagating modes of the Maxwell equations. In the rest of the paper we will frequently relate the dwell time to the matter energy only, and ignore the radiation energy inside the scatterer.

We will find two alternative but equivalent ways to find the dwell time of light. The first one establishes an interesting link with the “energy dependent” potential mentioned in section II. The second method is phenomenological, very educational, and what is most important, extremely useful for numerical calculations. Finally we will obtain explicit results for the point particles introduced in section II.B.2.b.

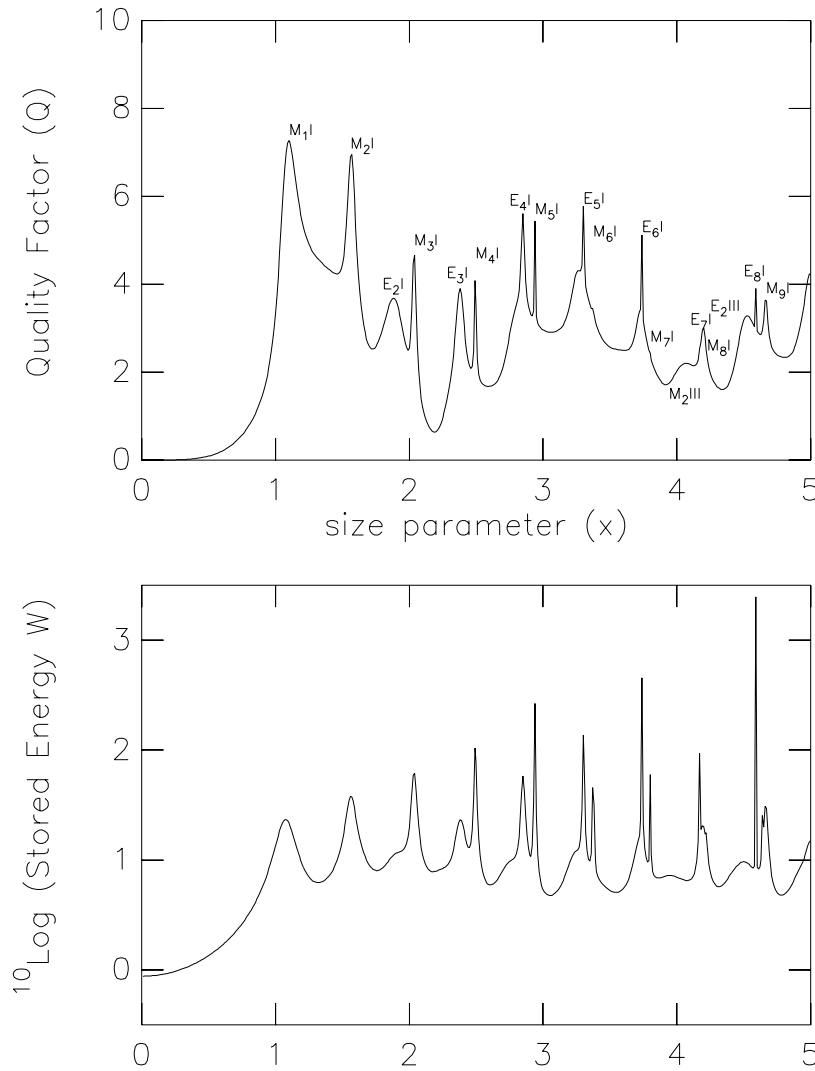


FIG. 4. Two important properties of a Mie sphere with radius a . Top: scattering cross-section σ , relative to its geometrical cross-section πa^2 . Bottom: average electromagnetic energy density W inside the Mie sphere relative to vacuum. Both have been displayed as a function of the size parameter x , defined as the $x = \omega a/c_0$. The index of refraction of the Mie sphere is $m = 2.73$, corresponding to Titanium-dioxide in vacuum. In the graph of the cross-section we identified various geometrical resonances of the Mie sphere. The spectroscopic notation $E_i I$ ($M_i I$) refers to the ground tone of the electric (magnetic) resonance of the i^{th} partial wave. Higher roman number signify overtones as discussed by Stratton [61].

b.1. Relation with Energy-dependent Potential Let us investigate how Eq. (3.36) changes when we want to let it apply to light rather than electrons. In particular, what will change if an energy dependent potential is introduced? If we forget the extra book keeping caused by polarization indices we first notice that the energy dependence in the t -matrix due to energy dependence of the interaction will give rise to extra terms on the right hand side. On the basis of Eq. (3.36) also the left hand side is expected to be subject to an extra contribution due to stored energy. It seems likely that modifications of left and right hand side will compensate so that the outlook of Eq. (3.36) will remain the same. By writing the potential as $V(\mathbf{r}, E) = f(\mathbf{r})V_0(E)$, and using the mathematical identity,

$$\frac{d}{dE} = \left(\frac{\partial}{\partial E} \right)_V + \frac{dV_0}{dE} \left(\frac{\partial}{\partial V_0} \right)_E, \quad (3.39)$$

the extra terms on the rhs. of Eq. (3.36) generated by the energy dependent potential can easily be identified as a derivative “with respect to potential” [62]. The former electronic terms that do not contain derivatives with respect to the potential will cancel as a result of Eq. (3.36). The term remaining on the left is $W_{\omega}^{\text{pot}}(S)$. Recalling that $E \rightarrow \omega^2/c_0^2$ for light this suggests the validity of the relation

$$W_{\text{pot}}(\omega, S) = \left\{ -\frac{\partial}{\partial V_0} \text{Re } T_{\mathbf{k}\mathbf{k}}(\omega) + 2(\omega/c_0) \int d\Omega_{\mathbf{k}'} \frac{d\sigma}{d\Omega} \frac{\partial \Phi(\omega, \Omega)}{\partial V_0} \right\} \times \frac{dV_0}{d(\omega/c_0)^2}, \quad (3.40)$$

where on the right the terms are collected that contain the derivative with respect to the potential. In view of relation (2.8) we can insert $dV_0/d(\omega/c_0)^2 = 1$ for light. Eq. (3.40) can again be derived straightforwardly from scattering theory but we will spare you the details [57] [63]. Polarization indices can easily be added in this equation. In that case the longitudinal field energy will automatically be incorporated in W_{pot} . We first demonstrated that the total excess electromagnetic energy around the scatterer can be obtained from the *on-shell* scattering amplitude). We now have shown that this also holds for the matter contribution to the electromagnetic energy, that is

$$\begin{aligned} \int d^3\mathbf{r} [W(\omega, \mathbf{r}) - 1] &\sim \frac{\partial \Phi(\omega, V_0)}{\partial (\omega/c_0)^2}, \\ \int d^3\mathbf{r} W_{\text{pot}}(\omega, \mathbf{r}) &\sim \frac{\partial \Phi(\omega, V_0)}{\partial V_0}. \end{aligned} \quad (3.41)$$

It may come as a surprise that both near-field properties can be obtained separately from knowledge of far field only.

Relation (3.40) establishes the desired connection between the concept of energy-dependent potential and “stored energy”. Energy can be stored *if and only if* some degree of freedom has been integrated out. Apparently, this is automatically the case when light scatters from dielectric media. An explicit connection between such a degree of freedom will be established later for the point particle defined in Eq. (3.21) where Eq. (3.40) will allow us to calculate the potential energy easily. In the fifties H. Feshbach demonstrated elegantly that integrating out a closed internal channel gives rise to a potential that depends on energy [37]. Quite conveniently he referred to it as an “optical potential” even when no light is involved at all. Optical potentials are common in nuclear physics when intermediate particle states, with finite lifetime, are integrated out.

b.2. Relation with Absorption Using elementary arguments from geometrical optics one can arrive at almost an exact solution for the dwell time. To this end consider Fig. 5. A plane wave is incident on a relatively big particle with index of refraction $m = \eta + i\kappa$. In the particle this wave can be represented by

$$\Psi(s, t) = \exp[ikms - i\omega t],$$

where s is some hypothetical coordinate along the path of the wave. The amplitude decays exponentially when the imaginary part of the index $\kappa > 0$. The amplitude of the wave that finally comes out is

$$|\Psi_{\text{out}}|^2 = \exp[-2\kappa kL].$$

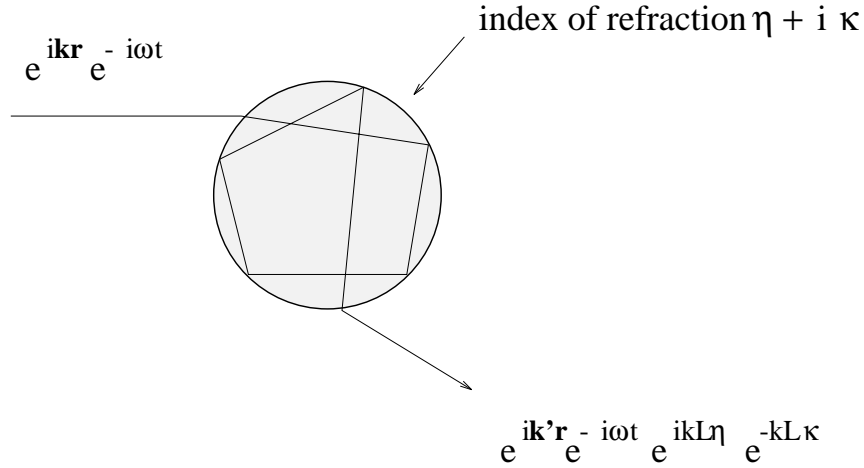


FIG. 5. The absorption picture in geometrical optics. It illustrates how an incident ray is scattered by a sphere. This picture allows us to estimate the traversed path length of the wave in the scattering object from the amount of absorption that it has suffered. Despite the simplistic derivation, this estimate turns out to give the exact solution for the dwell time of a scalar scatterer, and an accurate expression for vector waves.

Here L is the path length of the wave in the particle. Since the speed of light inside is given by c_0/η , the residence time becomes $\tau = \eta L/c_0$. Furthermore $|\Psi_{\text{out}}|^2$ can be identified as the albedo a of the scattering particle, that is the ratio of scattering and extinction. The albedo represents the deviation from the optical theorem (3.30) due to dissipation of energy inside the particle. This yields for the dwell time of waves in a particle without absorption,

$$\tau_d^{\text{abs}}(\omega) = \lim_{\kappa \rightarrow 0} \frac{1 - a(\omega)}{2\omega\kappa} m. \quad (3.42)$$

For scalar waves it can be proven rigorously that τ_d^{abs} is equal to the dwell time. Its formal proof uses the Cauchy-Riemann equations for an analytic function [63] [64]. These equations tell that differentiating the real part of an analytic function with respect to the imaginary part of its argument is the same as differentiating the imaginary part of the function with respect to the real part of its argument. Scattering theory shows the t -matrix to be analytic in the potential. In this way the equivalence of relations (3.42) and (3.37) can be established.

In Fig. 6 we show absorption time, dwell time and Wigner delay time for a vector Mie sphere with index of refraction $m = 3.0$. Numerically they all seem to be rather close. The fact that dwell and absorption time are only approximately equal in the numerical computations above is a genuine vector effect and due to the longitudinal fields inside the particles. The absorption argument is an argument for travelling waves and does not address the longitudinally stored energy.

b.3. Application to Point Scatterers In this subsection we find explicit results for delay properties of the point scatterer we developed in section III.B.2.b. Let us start by inserting the expression of the t -matrix into result (3.40) we obtained for the potential energy. For vector scattering we simply add a summation over two orthogonal polarization channels in the scattering term. The result is,

$$W_{\text{pot}}(\omega) = \frac{c_0^4}{\alpha_{\text{sph}}} \cdot \frac{(4\pi\Gamma)^2}{(\omega_0^2 - \omega^2)^2 + (2\Gamma\omega^3/3c_0)^2}. \quad (3.43)$$

Here α_{sph} is the polarizability of the point scatterer given by Eq. (3.20). Recalling the expression for the total scattering cross-section (3.23), we find for the dwell time (which we calculate here only from the potential energy),

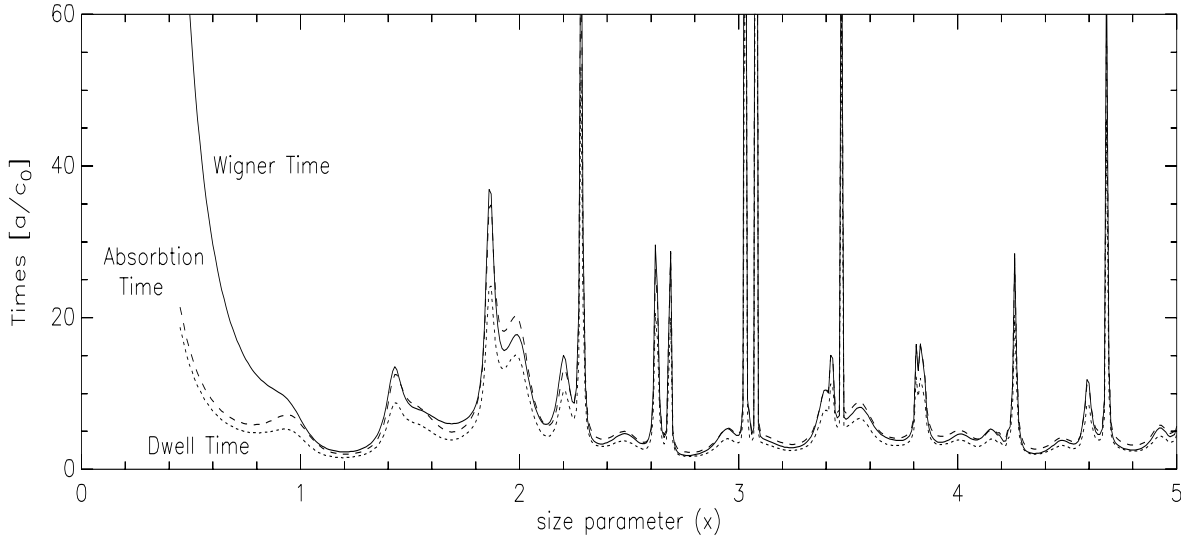


FIG. 6. Dwell time, Wigner time and absorption time obtained calculated for a Mie sphere with index of refraction 3.0. In the resonant scattering regime they are all approximately equal. In the Rayleigh regime at low frequencies they deviate from each other, but they all diverge towards infinity, and no longer seem to be reliable characteristic time scales.

$$\tau_d(\omega) = \left(\frac{2}{3} \frac{\alpha_{\text{sph}}}{4\pi c_0^3} \omega^4 \right)^{-1}. \quad (3.44)$$

For a very sharp resonance ($\Gamma\omega_0/c_0 \ll 1$) the dwell time is roughly constant over the resonance. This is a specific property of point particles and not true for scatterers with finite size such as Mie spheres. The results of section III.B.2.b. show that $\omega_0^2 = 4\pi\Gamma c_0^2/\alpha_{\text{sph}}$. This implies that the dwell time near the resonance in this model is simply the inverse frequency width of the resonance.

An interesting connection with atomic physics occurs by doing some formula manipulations. Let $W_0(\omega, j)$ being the density of states of the radiation per unit volume per polarization direction j at frequency ω in vacuum, and found by summing the eigenvalues,

$$W_0(\omega, j) = \frac{2\omega}{c_0^2} \sum_{\mathbf{p}} \delta\left((\omega/c_0)^2 - \mathbf{p}^2 \Delta_{\mathbf{p}}\right)_{jj} = \frac{\omega^2}{3\pi^2 c_0^3}. \quad (3.45)$$

This expression will be given more attention later in section IV.C of this paper. The factor 2ω originates from $d\omega^2 = 2\omega d\omega$. The inverse dwell time is expressed as

$$\frac{1}{\tau_d} = \frac{\pi}{2} \alpha_{\text{sph}} \omega^2 W_0(\omega, j). \quad (3.46)$$

This is a purely classical expression. It reminds us that the dwell time is determined by the polarizability α_{sph} of the matter and the number of states available for the leaving waves.

We can now insert the quantum mechanical expressions for the atomic polarizability in terms of the atomic dipole matrix element \mathbf{d} : $\alpha_{\text{sph}}/4\pi = 2|\mathbf{d}|^2/\hbar\omega$ [65], and the expectation value of the electric field (along the direction i of the dipole moment) triggered by vacuum quantum fluctuations,

$$\langle \mathbf{E}_j^2(\omega) \rangle = \frac{1}{2} \hbar\omega \times W_0(\omega, j). \quad (3.47)$$

This gives

$$\frac{1}{\tau_d} = \frac{2\pi}{\hbar^2} |\mathbf{d}|^2 \langle \mathbf{E}_j^2 \rangle . \quad (3.48)$$

This is readily recognized as the Fermi Golden Rule for the Einstein spontaneous emission coefficient. We conclude that the dwell time and inverse spontaneous coefficient are mathematically identical for this case. Physically, the spontaneous emission coefficient A can only be addressed in a matter picture, with the excited state as initial state, and not an incoming laser beam. In the former case the emitted light has a spread in frequency proportional to A due to uncertainty. In our light picture there is only one frequency because the scattering process is purely elastic with conservation of phase.

Quantum mechanics and \hbar play a spurious role in Eq. (3.48) since the derivation is classical. The presence of \hbar reminds us here that the matter has a quantum description and that the perturbing light originates from vacuum fluctuations. Recent work in quantum optics concentrates on the modification of spontaneous emission in materials other than vacuum [66] [67] [68]. The change in density of states in Eq. (3.46) then plays a leading role. This conclusion could have been drawn using strictly classical considerations.

4. MULTIPLE SCATTERING OF LIGHT

The single scattering properties discussed in the previous section will now be used to investigate multiple scattering. The medium that we envisage is a medium with typical size L containing a large number of identical particles, located randomly. The degree of multiple scattering that occurs in such a medium may be expected to be determined by the size of the medium relative to the extinction mean free path of the waves,

$$\tau \equiv \frac{L}{\ell_{\text{ext}}} = Ln\sigma_{\text{ext}} . \quad (4.1)$$

The second equality in relation (4.1) is based on the independent scattering approximation (ISA). In optics and astrophysics the parameter τ is called the optical thickness. If $\tau \gg 1$ multiple scattering will be large, when $\tau \ll 1$ multiple scattering will be small. On the basis of Eq. (4.1) one infers that multiple scattering will be prominent when the density of the scattering particles is large and when the scattering of the individual particles is strong. This conclusion seems to be physically sound, but objections can be raised. First of all, one can imagine that for particles with large but predominantly forward scattering, multiple scattering will not be very efficient. However, anisotropy in scattering does not seem to play any role in the definition in (4.1). An additional complication, that might show up for a very large density of scatterers, is the importance of “beyond-ISA” events, like repeated scattering from one particle. In that case optimizing individual scattering may not necessarily optimize collective scattering.

This section on multiple scattering is divided into six parts. First we pay attention to the averaging procedure needed to describe random media. Next, we treat the average amplitude (or field) under near-resonant scattering conditions and establish the link with polaritons. We will introduce the density of states (DOS) and the spectral function for light. We will have to extend the quantum-mechanical notion of these concepts. In the fourth part we deal with the transport of the average intensity of light with a frequency near scattering resonances of the medium. We shall derive the equation of radiative transfer, and point out how the average amplitude and average intensity are

related. We shall prove the validity of an Einstein relation. In a separate section we briefly discuss extensions of standard radiative transfer theory.

4.1. Averaging

Generally speaking, physicists immediately apply averaging procedures as soon as “disorder” is introduced in their model [69] [70]. When the scattered intensity is averaged over some random variable, a transport theory emerges. When workers start out with a transport theory, they often do not realize that this theory was obtained after performing an average over the disorder. Standard transport theory focuses both on the average field amplitude $\langle\langle\Psi\rangle\rangle$ and on the average intensity $\langle\langle\Psi\Psi^*\rangle\rangle$. With these first two moments of the field Ψ it is in principle impossible to reconstruct the whole distribution of Ψ , and information will be lost. The big question is: do these two lowest moments in practice suffice to describe the experiment? This question can be divided into two parts:

- Does the experiment always deal with averaging and is it the same average as performed in theory?
- Are higher moments expressible in lower moments and is their influence limited to a numerical correction to known phenomena, or do these higher moments really provide new physical insight?

Observation of speckle is an example where at least $\langle\langle\Psi\Psi\Psi^*\Psi^*\rangle\rangle$ is necessary to interpret the observations.

In some special circumstances the whole distribution function of some observable Φ , not necessarily the field amplitude Ψ , is determined by the lowest moment $\langle\langle\Phi\rangle\rangle$. The Poisson distribution is a well known example. Sometimes the distribution function has only one value for which it is non-zero. The quantity Φ is then called “self-averaging” and the random process has in fact become deterministic. Knowledge of the average then provides knowledge about “almost every” individual realization of the random system. This property implies that the average value applies to *every single* realization of the system except for a few special ones (with measure zero), among which is the realization of purely crystalline structure. Propagation of waves in one dimensional random systems (meaning that two out of three dimensions are translationally invariant and only the third is random) embodies such a special case for which self-averaging can be demonstrated mathematically [71] [72]. For a one-dimensional system one can prove that all eigenfunctions decay exponentially in space for “almost any” energy or frequency for “almost any” realization of the disorder. The decay length is a function of energy or frequency only, and not of the precise realization of the system. In three dimensions, proofs of self-averaging are rare and in most cases quantities are not self-averaging. One quantity for which self averaging has been demonstrated in three dimensions is the integrated density of states of Schrödinger Hamiltonians [73].

For electron propagation in disordered metals the averaging is usually brought about in a natural way by thermal fluctuations. Thermal fluctuations destroy the phase of the electronic wave function and thereby kill all interference at length scales typically on the order of a micrometer. The conductivity of copper is $6 \times 10^7 (\Omega m)^{-1}$ at room temperature and a normal copper wire has a resistance of, let us say, 100 Ω . There are no large fluctuations of this resistance induced by quantum interferences. Under these circumstances knowledge of the second moment $\langle\langle\Psi\Psi^*\rangle\rangle$ alone suffices.

In the eighties a new branch of condensed matter physics developed: mesoscopic physics. New technological developments made it possible to manufacture very tiny samples. In such mesoscopic samples the phase of the electronic wave function is not destroyed [74]. Yet, propagation of the electron can be both in the ballistic ($L \ll \ell$) and in the opaque ($L \gg \ell$) regime.

In opaque mesoscopic systems the movement of *one* single defect can give rise to typical variations of the conductance of order $e^2/h \simeq (25.8 \text{ k}\Omega)^{-1}$. These fluctuations are known as the Universal Conductance Fluctuations (UCF). They do not disappear when the medium is enlarged as long as its size remains smaller than phase-destroying lengths caused mainly by electron-phonon coupling. To explain conductivity measurements in mesoscopic systems one needs the whole distribution of the conductance. In general correlations between physical processes cause a distribution to deviate from a Gaussian or Poissonian statistics. These distributions functions apply when assumptions about independence and decorrelation are made. For example, assumptions of this type lead to a Gaussian distribution for the complex optical field amplitude in transmission and a Poisson distribution for the intensity [75]. Deviations from these distributions functions are caused by correlations e.g. originating from interference. For multiple light scattering such deviations are a topic of current interest; They have indeed been predicted theoretically [76] and observed experimentally [77].

In experiments on optical structures with quenched disorder the averaging is not automatically performed by thermal fluctuations. This lack of automatic averaging has both advantages and disadvantages. The advantage of studying mesoscopic physics of light rather than of electrons is that the former is experimentally easier and cheaper. To study electronic systems one needs milliKelvin temperatures and nanometer samples. Interesting fluctuations in light intensity, referred to as speckles, come almost for free! The drawback is that the magnitude of these optical intensity fluctuations are of the same order as the intensity itself. Consequently, optical experimentalists must average over some parameter - either over time or frequency, or over position (rotation or translation) - of the sample in order to get rid of the large fluctuations. In the accompanying theory one averages over all possible locations of the scattering objects. The question - actually the first one posed earlier this section - whether experimental and theoretical averaging procedures are equivalent is an unsolved one.

4.2. Scattering Theory for the Average Amplitude

Given the polarizability $\alpha(\omega)$ of one individual oscillator, or dielectric particle, in vacuum at frequency ω , the polarization density \mathbf{P} of an ensemble of oscillators can be found by simply multiplying the single-oscillator polarization \mathbf{p} with the number density n of the oscillators. In this way the standard “textbook result” for the complex dielectric constant is obtained,

$$\varepsilon(\omega) \equiv 1 + \frac{P}{E} = 1 + n\alpha(\omega) . \quad (4.2)$$

Strong objections can be raised against such a phenomenological procedure. Insertion of this dielectric constant into the macroscopic Maxwell’s equations shows that dynamic electric fields propagate, and that their frequencies and wavelengths are related through the (complex) dispersion law

$$k^2 = \varepsilon(\omega) \frac{\omega^2}{c_0^2} , \quad (4.3)$$

referred to as the “Maxwell relation”. Yet, retardation effects between the different oscillators - inherently connected to propagation - have been ignored completely in the simplistic derivation of relation (4.2). In principle this standard derivation can only be justified for electrostatics ($\omega = 0$).

For the same reason the treatment of “local-field” concepts in some text books is hard to defend at optical frequencies. In these approaches an oscillator “feels” a so-called local electric field, caused by nearby oscillators, that is different from the macroscopic field, according to

$$\mathbf{E}_{\text{loc}} = \mathbf{E} + \frac{\mathbf{P}}{3} . \quad (4.4)$$

This local field polarizes the oscillator, $\mathbf{p} = \alpha \mathbf{E}_{\text{loc}}$, and inclusion of this local field leads to a polarization density of $\mathbf{P} = n\alpha \mathbf{E}_{\text{loc}}$. Using Eq. (4.4) to eliminate \mathbf{P} we find

$$\mathbf{E}_{\text{loc}} = \frac{1}{1 - \frac{1}{3}n\alpha} \mathbf{E} \equiv \varphi_{\text{loc}} \mathbf{E} . \quad (4.5)$$

The local field factor φ_{loc} describes the enhancement of the local field with respect to the macroscopic field and is expressed in Eq. (4.5) in terms of microscopic polarizabilities. If we want to have φ_{loc} expressed in terms of the, experimentally available, macroscopic dielectric constant this can be done as Eq. (4.5) implies the following relation between the dielectric constant and the polarizability,

$$\varepsilon = 1 + \frac{n\alpha}{1 - \frac{1}{3}n\alpha} , \quad (4.6)$$

so that

$$\varphi_{\text{loc}} = \frac{\varepsilon + 2}{3} . \quad (4.7)$$

This expression is familiar in linear and even nonlinear optics [38] to relate microscopic polarizabilities to macroscopic fields. It works very good for gasses and surprisingly well for solids [78]. Relation (4.6) can be inverted into,

$$n\alpha = 3 \frac{\varepsilon - 1}{\varepsilon + 2} . \quad (4.8)$$

One can see that local-field corrections are at least second-order in the density and are thereby one out of many other “beyond-ISA” modifications of the dielectric constant to be discussed later.

More advanced considerations [39] [79] (among which the original one given by Lorentz [80] !) do take into account retardation effects. More or less surprisingly and after considerable effort, it turns out that under mild conditions the static outcome indeed applies to finite frequency and Eqs. (4.4), (4.5), (4.6), (4.7), and (4.8) remain valid. One just replaces α by $\alpha(\omega)$ and ε by $\varepsilon(\omega)$. The explicit consideration of retardation is not necessary. The dynamic version of Eq. (4.8) is known as the Lorentz-Lorenz formula. We will investigate the validity of the Lorentz-Lorenz formula using the modern formulation of multiple scattering theory. The fact that the oscillators can be replaced by an effective medium with a dielectric constant given by the Lorentz-Lorenz formula is known as the Ewald-Oseen extinction theorem. This is a theorem for the coherent wave.

4.2.1. Dyson’s Equation

We will first shortly illustrate the Green’s function formalism for an electron in a (random) potential and then show how this framework will have to be modified to let it apply to light. The

quantum-mechanical eigenfunctions of the Hamilton operator H are given by $H\psi_i = E_i\psi_i$. In many applications it is more convenient to have the inverse wave operator in which the eigenvalues of the Hamiltonian show up as poles, and not the Hamiltonian. For this purpose one defines the (amplitude) Green's function operator $G(z)$ [81],

$$G(z) \equiv \frac{1}{z - H} . \quad (4.9)$$

One can take matrix elements of this operator. Both the \mathbf{r} and the \mathbf{p} -representation are relevant. The matrix element $G(z, \mathbf{r}, \mathbf{r}') \equiv \langle \mathbf{r} | G(z) | \mathbf{r}' \rangle$ describes the propagation of the electron from \mathbf{r} to \mathbf{r}' . The average propagation is determined by the average of the Green's function over all possible realizations. This averaging restores translational symmetry, so that momentum is a conserved quantity. In real space this implies $\langle\langle G(z, \mathbf{r}, \mathbf{r}') \rangle\rangle = \langle\langle G(z, \mathbf{r} - \mathbf{r}', 0) \rangle\rangle \equiv G(z, \mathbf{r} - \mathbf{r}')$. It is convenient to take matrix-elements in the \mathbf{p} -representation,

$$\langle\langle \langle \mathbf{p} | G(z) | \mathbf{p}' \rangle \rangle \equiv G(z, \mathbf{p}) \delta(\mathbf{p} - \mathbf{p}') . \quad (4.10)$$

We define a self energy (sometimes called mass operator due to its role in quantum field theory) $\Sigma(z, \mathbf{p})$ according to,

$$G(z, \mathbf{p}) \equiv \frac{1}{z - \mathbf{p}^2 - \Sigma(z, \mathbf{p})} . \quad (4.11)$$

Definition (4.11) is known as Dyson's equation. All information has been hidden in the new object $\Sigma(z, \mathbf{p})$, the self-energy. For free motion the self-energy obviously vanishes. As the Dyson equation is exact, the exact calculation of $\Sigma(z, \mathbf{p})$ remains as difficult as the exact calculation of $G(z, \mathbf{p})$. However, it is often much easier to obtain reliable approximations for $\Sigma(z, \mathbf{p})$ than for $G(z, \mathbf{p})$. Diagrammatic perturbation theory can be developed and it turns out that the self energy is determined by only a limited set of diagrams, that are characterized by a special "connected" structure [70] [82].

Let us now find the analogous quantities for the propagation of light. The vector Green's function has to work on three-dimensional vector fields and is therefore a tensor operator of rank two. We still call it the "vector" Green's function,

$$\mathbf{G}(\omega) = \frac{1}{\boldsymbol{\varepsilon}(\mathbf{x}) (\omega/c_0)^2 - \mathcal{F}(\mathbf{p})} , \quad (4.12)$$

where we regard $\boldsymbol{\varepsilon}(\mathbf{x})$ as the operator with real-space matrix element $\langle \mathbf{r} | \boldsymbol{\varepsilon}(\mathbf{x}) | \mathbf{r}' \rangle = \boldsymbol{\varepsilon}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}')$, as is done in quantum mechanics. We have defined the operator $\mathcal{F}(\mathbf{p})$

$$\mathcal{F}(\mathbf{p}) = \mathbf{p}^2 - \mathbf{p}\mathbf{p} . \quad (4.13)$$

The Green's function (4.12) is intimately related to Huygens' principle (stating that the propagation of light is a subsequent sum of spherical waves) and it is referred to as the amplitude Green's function. However, it is not the Green's function that one would expect to show up here when comparing with the electron case. To emphasize this point we introduce the frequency eigenfunctions of the Helmholtz equation $\{|\mathbf{E}_i\rangle\}$,

$$\begin{aligned} \varepsilon(\mathbf{x})^{-1} \cdot \mathcal{F}(\mathbf{p}) \cdot |\mathbf{E}_i\rangle &= (\omega_i/c_0)^2 |\mathbf{E}_i\rangle \\ \text{or : } \varepsilon(\mathbf{x})^{-1/2} \cdot \mathcal{F}(\mathbf{p}) \cdot \varepsilon(\mathbf{x})^{-1/2} \cdot \{\varepsilon(\mathbf{x})^{1/2} \cdot |\mathbf{E}_i\rangle\} &= (\omega_i/c_0)^2 \{\varepsilon(\mathbf{x})^{1/2} \cdot |\mathbf{E}_i\rangle\} \end{aligned} \quad (4.14)$$

We arranged the operators $\varepsilon(\mathbf{x})$ such that we obtain an Hermitean Green's operator. This vector eigenvalue equation calls for another vector Green's function namely,

$$\begin{aligned} \widetilde{\mathbf{G}}(\omega) &\equiv \frac{1}{(\omega/c_0)^2 - \varepsilon(\mathbf{x})^{-1/2} \cdot \mathcal{F}(\mathbf{p}) \cdot \varepsilon(\mathbf{x})^{-1/2}} = \varepsilon(\mathbf{x})^{1/2} \cdot \frac{1}{\varepsilon(\mathbf{x}) (\omega/c_0)^2 - \mathcal{F}(\mathbf{p})} \cdot \varepsilon(\mathbf{x})^{1/2} \\ &= \varepsilon(\mathbf{x})^{1/2} \cdot \mathbf{G}(\omega) \cdot \varepsilon(\mathbf{x})^{1/2}, \end{aligned} \quad (4.15)$$

By construction, a number of mathematical properties holding for the electronic $G(E)$ also hold for $\widetilde{\mathbf{G}}(\omega)$. However, in most physical applications the average amplitude Green's function $\mathbf{G}(\omega)$ shows up because this one describes the electrical field propagating from one place to another. The difference between the two is the sandwiching by $\varepsilon(\mathbf{x})^{1/2}$. One should realize that $\varepsilon(\mathbf{x})$ is in fact a random variable. Consequently the average of $\widetilde{\mathbf{G}}$ is more complicated than the one of \mathbf{G} . This section is devoted to the average of the amplitude Green's function \mathbf{G} .

The formulation of a Dyson equation for the Green's function \mathbf{G} is equivalent to the electron case, only changing scalar operators into tensors of rank two. As with electrons we define a (vector) self energy $\Sigma(\omega, \mathbf{p})$ according to,

$$\mathbf{G}(\omega, \mathbf{p}) \equiv \frac{1}{(\omega/c_0)^2 - \Sigma(\omega, \mathbf{p}) - \mathbf{p}^2 \Delta_{\mathbf{p}}}. \quad (4.16)$$

We have used $\langle \mathbf{p} | \mathcal{F} | \mathbf{p}' \rangle = \mathbf{p}^2 \Delta_{\mathbf{p}} \delta(\mathbf{p} - \mathbf{p}')$ as in definition (2.4).

The vector self-energy is intimately related to the dielectric function $\varepsilon(\omega, \mathbf{p})$. The definition of the dielectric function of the medium is,

$$\varepsilon(\omega, \mathbf{p}) \equiv 1 - \frac{\Sigma(\omega, \mathbf{p})}{(\omega/c_0)^2}. \quad (4.17)$$

In isotropic media all tensor operators can be decomposed into a fully transverse and a fully longitudinal component: in the \mathbf{p} -representation we divide the identity operator into $\mathbf{I} = \hat{\mathbf{p}}\hat{\mathbf{p}} + \Delta_{\mathbf{p}}$. We can pursue this decomposition for the dielectric function,

$$\varepsilon(\omega, \mathbf{p}) = \varepsilon_{\parallel}(\omega, \mathbf{p}) \hat{\mathbf{p}}\hat{\mathbf{p}} + \varepsilon_{\perp}(\omega, \mathbf{p}) \Delta_{\mathbf{p}}, \quad (4.18)$$

so that for the Green's function,

$$\mathbf{G}(\omega, \mathbf{p}) = \frac{\hat{\mathbf{p}}\hat{\mathbf{p}}}{\varepsilon_{\parallel}(\omega, \mathbf{p}) (\omega/c_0)^2} + \frac{\Delta_{\mathbf{p}}}{\varepsilon_{\perp}(\omega, \mathbf{p}) (\omega/c_0)^2 - \mathbf{p}^2}. \quad (4.19)$$

The (complex) poles of the Green's function $\mathbf{G}(\omega, \mathbf{p})$ determine the excitations of the system. The real part of the poles determine the frequency of the excitations and the imaginary part represents their damping. For transverse excitations we obtain the complex dispersion law $\mathbf{K}^2 = \varepsilon_{\perp}(\omega, \mathbf{K}) (\omega/c_0)^2$, which is similar to Eq. (4.3). Longitudinal excitations, if any, satisfy $\varepsilon_{\parallel}(\omega, \mathbf{K}) = 0$. If the dielectric function is p -independent, both longitudinal and transverse dielectric constant are equal and written as the dielectric constant $\varepsilon(\omega)$. Momentum dependence of $\varepsilon(\omega, \mathbf{p})$ - usually called spatial dispersion [83] [84] - can be induced by cluster scattering or by spatial correlations of the scatterers, both being beyond the independent scattering approximation (ISA). In the ISA spatial dispersion can be due to the finiteness of the scatterers. For point scatterers or atoms spatial dispersion is absent in the ISA.

The ISA includes all multiple scattering up to infinite order, but ignores any form of recurrent scattering between different particles. The ISA is widely used, often without any justification. The precise nature of the ISA can be readily appreciated when examining the approximation that has to be made to the self energy in order to produce the ISA. The ISA self-energy is just given by the product of the scattering contribution of one particle and the density of scatterers,

$$\Sigma^{(1)}(\omega, \mathbf{p}) = n \mathbf{T}_{\mathbf{pp}}(\omega) . \quad (4.20)$$

This formula shows nicely that scattering from one particle has become a “building block” for the average amplitude. Since recurrent scattering from clusters is going to be higher order in particle density, one expects that the ISA is the first term in a density expansion (we skip questions concerning convergence and analyticity of such an expansion). Inserting (4.20) into (4.17) gives the ISA expression for the dielectric function.

$$\varepsilon(\omega, \mathbf{p}) \longrightarrow \mathbf{I} - \frac{n \mathbf{T}_{\mathbf{pp}}(\omega)}{(\omega/c_0)^2} . \quad (4.21)$$

For isotropic scatterers is $\mathbf{T}_{\mathbf{pp}}(\omega) = T_{\mathbf{pp}}(\omega) \mathbf{I}$ so that the dielectric function satisfies $\varepsilon_{\parallel}(\omega, \mathbf{p}) = \varepsilon_{\perp}(\omega, \mathbf{p})$. For isotropic point particles we have a second simplification namely $\mathbf{T}_{\mathbf{pp}}(\omega) = t(\omega) \mathbf{I}$. For the polarizability of the medium this implies

$$\alpha(\omega) = \frac{1 - \varepsilon(\omega)}{(\omega/c_0)^2} = -\frac{nt(\omega)}{(\omega/c_0)^2} . \quad (4.22)$$

(See also the discussion following Eq. (3.28)). Hence we recover relation (4.2), at least for point particles. This proves the validity of relation (4.2) even in the case of retardation effects. The local-field correction is higher-order in density and strictly speaking beyond ISA.

The wave number following from $K^2 = \varepsilon(\omega) (\omega/c_0)^2$ has in general a real and imaginary part. The optical theorem (3.30) shows that the imaginary part must be positive. This is found in many other cases and is a manifestation of the dissipation-fluctuation theorem [85]. Hence the imaginary part of K gives rise to a decay of the propagating modes. We stress here already a feature - which cannot be addressed by considering the average field only - that this decay cannot be due to (only) absorption but is due to scattering out of the forward direction. Decay and propagation are characterized by the complex index of refraction $m(\omega) \equiv \sqrt{\varepsilon(\omega)} = K c_0 / \omega \equiv (k + ik'') c_0 / \omega$. Phase velocity and scattering mean free path can be defined as already announced in relation (1.1). In real space the average electric field of a transverse excitation takes the form,

$$\langle\langle \mathbf{E} \rangle\rangle = \mathbf{e}^{(1,2)} \exp(-i\omega t) \exp\left(i \frac{\omega}{v_p} r - \frac{r}{2\ell_{\text{ext}}}\right) . \quad (4.23)$$

being the solution of the wave equation with complex index of refraction $m(\omega)$.

4.2.2. Polaritons

More insight into the consequences of the dispersion relation (1.1) can be obtained when we use the t -matrix of a point dipole. We have shown that point dipoles necessarily have a frequency resonance, and that this resonance will influence the dielectric function and the index of refraction. The

resulting resonant behavior of the index of refraction gives rise to a rather complicated dispersion law, an S-curve, that is called polariton behavior. Polaritons come in many forms. An important characteristic property is how much of the incoming flux the point dipole will scatter, and how much it absorbs. We have introduced earlier the concept of albedo a to characterize the relative importance of scattering and absorption. To describe the point scatterer we can use both the phenomenological dielectric constant with a variable T_2 in Eq. (3.3) and the fundamental microscopic t -matrix (with $a = 1$). As explained earlier in this paper polaritons exist with albedo's ranging from close to one down to 10^{-6} .

We will illustrate polariton behavior by using the conventional notation as much as possible. Up to now we have described $\varepsilon(\omega)$ arising from only one type of point scatterer. In general there may be several types of scatterers in a medium, with resonances at different frequencies. If one remains with the frequency of the incoming light in the neighborhood of one particular resonance, well separated from the others, we can adapt our formalism without difficulty to include this situation. If only one type of resonance is present the dielectric function at frequencies higher than this resonance will be 1. However in a real situation there could still be resonances at much higher frequencies from which there is still a static contribution. We deal with this by generalizing this into $\varepsilon(\omega = \infty) \equiv \varepsilon_\infty \geq 1$. It is conventional to define the dimensionless quantity \mathcal{S} ,

$$\mathcal{S} \equiv \varepsilon(\omega = 0) - \varepsilon_\infty \equiv \varepsilon_{\text{low}} - \varepsilon_\infty. \quad (4.24)$$

For point dipoles we identify

$$\mathcal{S} = 4\pi n \frac{\Gamma}{(\omega_0/c_0)^2}, \quad (4.25)$$

so that the dielectric constant is now given by

$$\varepsilon(\omega) = \varepsilon_\infty + \mathcal{S} \left(\frac{\omega_0^2}{\omega_0^2 - \omega^2 - i\Gamma\omega^3/c_0} \right). \quad (4.26)$$

We expect the largest influence on the dielectric constant to occur near the resonance frequency. The term between brackets is then of order $c_0/\Gamma\omega_0$. So $\mathcal{S}c_0/\Gamma\omega_0$ is a measure for the “swing” of the dielectric constant near the resonance. We would like to define some kind of polariton parameter that measures the coupling between the light and the matter. We expect this parameter to depend on the density of the scatterers and on the sharpness of the resonance. It is clear that $\mathcal{S}c_0/\Gamma\omega_0$ is ideal. So it makes sense to define a polariton parameter according to

$$\mathcal{P} \equiv \frac{\mathcal{S}c_0}{\Gamma\omega_0} = \frac{4\pi n}{(\omega_0/c_0)^3}. \quad (4.27)$$

In terms of the resonance width $\Delta\omega/\omega_0 = 1/\omega_0 T_2$ the polariton parameter would be

$$\mathcal{P} \equiv \frac{1}{2} \mathcal{S} \omega_0 T_2. \quad (4.28)$$

In terms of the extinction mean free path \mathcal{P} can be expressed as,

$$\mathcal{P} = (k\ell_{\text{ext}})^{-1}. \quad (4.29)$$

This expression establishes the link with multiple scattering theory, in which $1/k\ell_{\text{ext}}$ shows up as an expansion parameter [86].

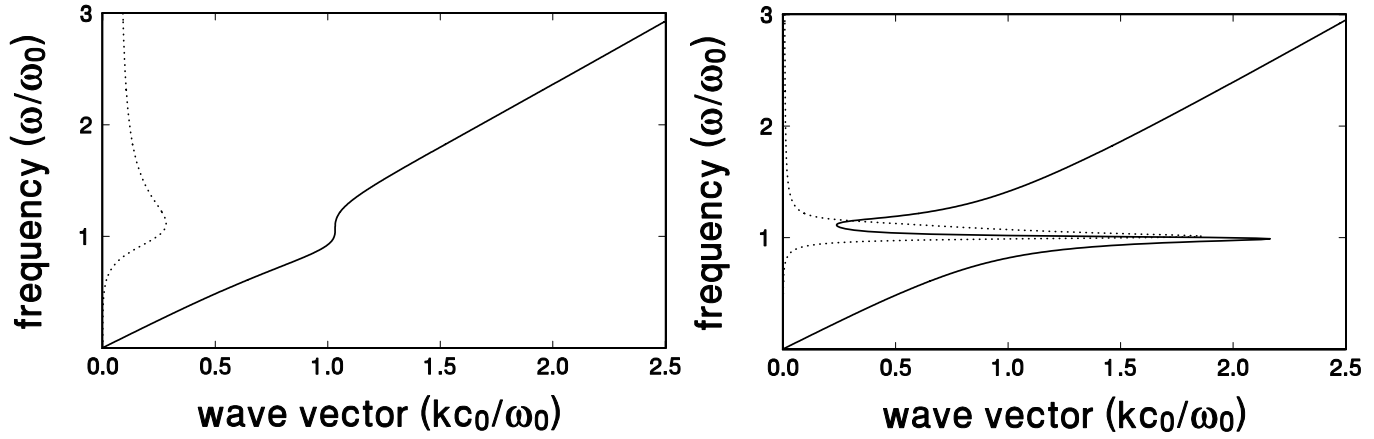


FIG. 7. Dispersion law (solid line) for a polariton with only small ‘polariton character’. The aptiodotted line represents the imaginary part of the wave vector. The following parameters have been used: $\mathcal{S} = 0.25$ and damping $T_2 = 4/\omega_0$. In this case the polariton parameter is $\mathcal{P} = 0.5$. Since this is less than the critical value $\mathcal{P}_C = 2$ no spectral (pseudo) gap is present.

FIG. 8. Dispersion relation (solid line) for a polariton with strong ‘polariton character’. The dotted line represents the imaginary part of the wave vector. The following parameters have been used: $\mathcal{S} = 0.25$, and damping $T_2 = 50/\omega_0$, so that $\mathcal{P} = 6.25$. Note the spectral gap that opens up beyond the resonant frequency.

In Fig. 7 we show real and imaginary part of the wave vector K calculated for a medium with resonant point scatterers under various conditions. In Fig. 7 we have used $\mathcal{S} = 0.25$ and $\omega_0 T_2 = 4$ meaning that $\mathcal{P} = 0.5$. This describes a rather broad resonance with only moderate polariton character. Close to the resonant frequency the dispersion is curved and the polariton acquires some matter character, although we are still looking at it by means of the light. If we increase the polariton character the dispersion law takes a more complicated S-like form as we already saw in Fig. 1, where we used $\mathcal{S} = 0.25$ and $\omega_0 T_2 = 12$, corresponding to $\mathcal{P} = 1.5$. In the neighborhood of the S-curve the dispersion becomes ‘anomalous’. This terminology refers to the negative slope of the dispersion law. In that region of the frequency the polariton is called ‘material-like’. In the regime where the dispersion law is normally increasing, the excitation is referred to as ‘photon-like’.

If we increase the polariton character even more by making $\mathcal{S} = 0.25$ and $\omega_0 T_2 = 50$, corresponding to $\mathcal{P} = 6.25$ (Fig. 8) the dielectric function becomes even more complicated. For a range of frequencies the real part of the wave vector is smaller than the imaginary part: the wave is overdamped or evanescent. This is the region where the real part of the dielectric constant is negative. If one would neglect the imaginary part this would give rise to an imaginary wave vector as $k \sim \sqrt{\epsilon}$ and no propagation would be possible. One calls this a frequency gap or stop band. It is not a real gap as the imaginary value cannot be neglected. Nevertheless, in the region for which the real part of the wave vector is smaller than the imaginary part, propagation will be seriously hampered. The critical parameter for which this occurs is $\mathcal{P} = 2$ or $kl_{\text{ext}} = 0.5$.

In atomic physics one often says that the sharpness of the resonance increases when the losses decrease. In that terminology Fig. 8 represents an ‘almost lossless’ case since the resonance is very sharp. However, from the point of view of scattering any value of the damping parameter can represent a ‘lossless’ situation. This is the case if the imaginary part of the wave vector is induced by *elastic* scattering. For the energy balance one should be willing to count the energy of the scattered waves as well.

The group velocity v_g can be defined as the local slope of the dispersion relation,

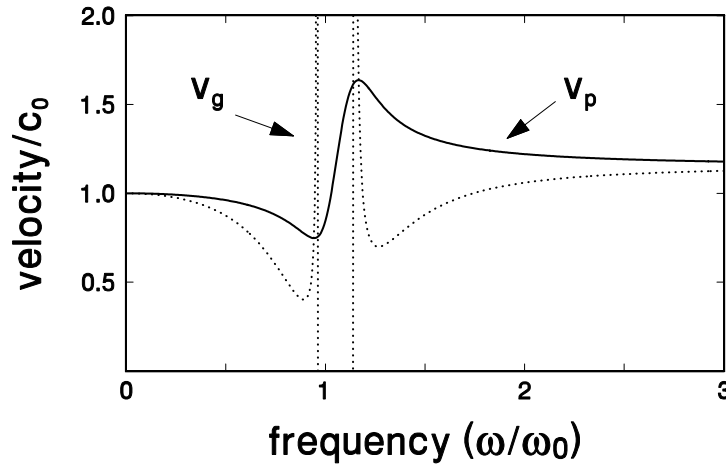


FIG. 9. Phase velocity v_p (solid line) and group velocity v_g (dotted) of electromagnetic waves propagating through a collection of point scatterers having an internal resonance with frequency ω_0 as a function of frequency. The following parameters have been used: $S = 0.25$ and the damping $T_2 = 12/\omega_0$ ($P = 1.5$).

$$v_g(\omega) \equiv \frac{\partial \omega}{\partial k} . \quad (4.30)$$

We emphasize that this is merely a formal definition. One still has to prove the physical relevance of this velocity. Textbooks [87] prove the group velocity to determine transport of energy by considering the propagation of a pulse inside a dispersive medium. But this notion only applies when the dispersion law is real-valued, which is not the case for polaritons. Especially for frequencies near the resonance frequency ω_0 the imaginary part becomes very large and the group velocity loses its connection with energy transport.

The physics behind the imaginary part has already been explained. It signifies extinction. As is illustrated in Eq. (4.23) the amplitude decays exponentially in space, since its energy will be used for other purposes, either elastic scattering, absorption or inelastic processes. Over distances small compared to the extinction decay length one might neglect extinction at all, and take the real part as in Eq. (4.30) and say that this group velocity describes the speed of energy propagation. Thus at the very best, the group velocity describes the speed of propagation of the coherent beam over one extinction mean free path.

In Fig. 9 we show the group velocity for a polariton with the same parameters as in Fig. 1. We observe that the group velocity is diverging near resonance, and may even become negative. The group velocity is said to lose its meaning near the resonant frequency. In the next section we will show that its role will be taken over by a transport velocity for which this apparently acausal behavior has been removed by adding the scattering channels.

4.3. Density of States

An important concept in multiple scattering theory is the density of states (DOS). We will first introduce this concept for electrons in a (random) potential. For a quantum particle with Hamiltonian H the expression

$$\int_{-\infty}^E dE' \sum_i \delta(E' - E_i) = \int_{-\infty}^E dE' \text{Tr} \delta(E' - H) , \quad (4.31)$$

counts the number of states with energy less than E . The integrand $\text{Tr} \delta(E - H)$ is the density of states $N(E)$. As always in quantum mechanics the trace of an operator, $\text{Tr} A = \sum_i \langle i | A | i \rangle$, is independent of the choice of a complete set of functions. As a result,

$$N(E) = \text{Tr} \delta(E - H) . \quad (4.32)$$

Additional spectral information can be obtained by projecting on a specific representation. It is profitable to consider both the \mathbf{r} and \mathbf{p} -representation. The local density of states gives the number of states in an energy range dE at position \mathbf{r} ,

$$\rho(E, \mathbf{r}) \equiv \langle \mathbf{r} | \delta(E - H) | \mathbf{r} \rangle = -\frac{1}{\pi} \text{Im} G(E, \mathbf{r}, \mathbf{r}) . \quad (4.33)$$

We will now consider an infinite, disordered system and add the averaging procedure. Averaging induces translational symmetry. In that case the local density of states is equal to the density of states per unit volume so that the local density of states does not render any more insight than the total density of states. Consequently, some other additional characteristic of the modes may be looked for. Since momentum is conserved in a translationally symmetric system, this seems to be a good candidate. A spectral function $S(E, \mathbf{p})$ can be introduced which gives upon integration over momenta the density of states per unit volume,

$$\sum_{\mathbf{p}} S(E, \mathbf{p}) = \langle \langle \rho(E) \rangle \rangle . \quad (4.34)$$

The spectral function represents the number of modes (per unit volume) with energy E and wavenumber \mathbf{p} ,

$$S(E, \mathbf{p}) (2\pi)^3 \delta(\mathbf{p} - \mathbf{p}') \equiv \langle \langle \langle \mathbf{p} | \delta(E - H) | \mathbf{p}' \rangle \rangle \rangle . \quad (4.35)$$

For an electron obeying Schrödinger's equation the spectral density $S(E, \mathbf{p})$ can be obtained from the Dyson Green's function $G(E, \mathbf{p})$. Using again the operator identity $1/(x + i\epsilon) = P(1/x) - i\pi\delta(x)$ we see that $\text{Im} G(E + i\epsilon) = -i\pi\delta(E - H)$, so that

$$S(E, \mathbf{p}) = -\frac{1}{\pi} \text{Im} G(E, \mathbf{p}) . \quad (4.36)$$

Apparently all spectral information can be obtained from $G(E, \mathbf{p})$.

We would like use the same formalism for light and translate all these expressions for the density of states to light, using the analogies discussed in section II. The results for the electrons act as a guide, but the differences will be large. This has to do with the energy dependence of the "light potential". We have indicated already in section III.B.1 that in fact two Green's functions exist. The fact that the equations for light are not scalar but vector equations is an extra complication, but turns out to be of less principal nature.

We consider some finite inhomogeneous dielectric medium with volume V . By definition, to find the density of states, we first consider the eigenvalues of the wave equation in a frequency range $d\omega^2$,

$$N(\omega^2) = \sum_i \delta(\omega^2 - \omega_i^2) = \text{Tr} \delta(\omega^2 - c_0^2 \epsilon(\mathbf{x})^{-1/2} \cdot \mathcal{F}(\mathbf{p}) \cdot \epsilon(\mathbf{x})^{-1/2}) \quad (4.37)$$

$$= -\frac{1}{\pi} \text{Tr} \text{Im} \widetilde{\mathbf{G}}(\omega) c_0^{-2} . \quad (4.38)$$

The local density of states of electromagnetic waves can be defined as

$$W(\omega^2, \mathbf{r}) = \langle \mathbf{r} | \delta(\omega^2 - c_0^2 \boldsymbol{\varepsilon}(\mathbf{x})^{-1/2} \cdot \mathcal{F}(\mathbf{p}) \cdot \boldsymbol{\varepsilon}(\mathbf{x})^{-1/2}) | \mathbf{r} \rangle = -\frac{1}{\pi} \text{Im } \widetilde{\mathbf{G}}(\omega, \mathbf{r}, \mathbf{r}) c_0^{-2}. \quad (4.39)$$

In terms of $\widetilde{\mathbf{G}}$ the expression for the local density of states is similar to Eq. (4.33) found for electrons. However, we would like to make the connection between the (local) density of states and the more physical Green's function \mathbf{G} for the electric field. Let us first generalize the Helmholtz equation as a kind of Schrödinger equation with an energy-dependent potential taken at fixed frequency ω ,

$$\{-\mathcal{F}(\mathbf{p}) + [1 - \boldsymbol{\varepsilon}(\mathbf{x})] (\omega/c_0)^2\} |\mathbf{E}_i(\omega)\rangle = (\omega_i/c_0)^2 |\mathbf{E}_i(\omega)\rangle \quad (4.40)$$

For *any fixed* ω the light potential is a fixed operator and standard Schrödinger operator theory guarantees the existence of a complete set of solutions $\mathbf{E}_i(\omega)$ of this equation. Using this set we can work out the trace occurring in Eq. (4.37) as,

$$\begin{aligned} N(\omega^2) &= \sum_i \langle \mathbf{E}_i(\omega) | \delta(\omega^2 - c_0^2 \boldsymbol{\varepsilon}(\mathbf{x})^{-1/2} \cdot \mathcal{F}(\mathbf{p}) \cdot \boldsymbol{\varepsilon}(\mathbf{x})^{-1/2}) | \mathbf{E}_i(\omega) \rangle, \\ &= c_0^{-2} \text{Tr} \delta(\boldsymbol{\varepsilon}(\mathbf{x}) (\omega/c_0)^2 - \mathcal{F}(\mathbf{p})) + \sum_i \langle \mathbf{E}_i(\omega_i) | \boldsymbol{\varepsilon}(\mathbf{x}) - 1 | \mathbf{E}_i(\omega_i) \rangle \delta(\omega^2 - \omega_i^2) \end{aligned} \quad (4.41)$$

The decomposition carried out in the second equality is not only mathematical but also physical in the sense that it seems to separate the propagating response from the material response. We come back to this point later.

In terms of the Green's function \mathbf{G} the first term in Eq. (4.41) is seen to be similar to the electron case since

$$-\frac{1}{\pi} \text{Im } \mathbf{G}(\omega) = -\frac{1}{\pi} \text{Im} \frac{1}{\boldsymbol{\varepsilon}(\mathbf{x}) (\omega/c_0)^2 + i\epsilon - \mathcal{F}} = \delta(\boldsymbol{\varepsilon}(\mathbf{x}) (\omega/c_0)^2 - \mathcal{F}). \quad (4.42)$$

The second term in $N(\omega^2)$ strongly resembles the extra term in Eq. (3.38) that was shown earlier to arise from stored energy inside the dielectric objects. This term is absent for electrons. Quite conveniently we observe that for this term only the solution of the normal Helmholtz equation - that is $\omega^2 = \omega_i^2$ - is required.

The trace in the first term of Eq. (4.41) is best evaluated by using the complete set $|\mathbf{r}\rangle$. As usual, the summation over the eigenfunction index i at frequency ω_i goes over into the volume times the integral over wave vectors and a summation over polarization indices j ,

$$\sum_i \rightarrow V \sum_{\mathbf{p}, j} = \frac{V}{(2\pi)^3} \int_0^\infty dp p^2 \int d\hat{\mathbf{p}} \sum_j. \quad (4.43)$$

Writing $d\omega^2 = 2\omega d\omega$ we get for the density of states per frequency range $d\omega$, per polarization channel j ,

$$N(\omega, j) = -\frac{2\omega}{\pi c_0^2} \int d^3\mathbf{r} \text{Im } \mathbf{G}_{jj}(\omega, \mathbf{r}, \mathbf{r}) + \frac{\omega^2}{2\pi^2} \int \frac{d\hat{\mathbf{k}}}{4\pi} \int d^3\mathbf{r} [\boldsymbol{\varepsilon}(\mathbf{r}) - 1] |\mathbf{E}_{\mathbf{k}, j}^+(\mathbf{r})|^2. \quad (4.44)$$

In the second term on the rhs. of Eq. (4.44) the volume factor V has been put into the field to give it the same normalization as a plane wave. This transforms the eigenfunction $\mathbf{E}_i(\omega_i)$ into a Lippmann-Schwinger distorted plane wave $\mathbf{E}_{\mathbf{k}, j}^+(\mathbf{r})$ given in (3.11).

Up to now we considered a finite volume and did not average the density of states over the disorder and the results so far apply to any realization. Let us now calculate the average density of states, by averaging over all possible positions of the scatterers, and take the thermodynamic limit in which both the number of particles and the total volume go to infinity at constant number density n of the particles. In that case the average local density of states becomes independent of position and is simply the density of states per unit volume per polarization channel. This yields,

$$\begin{aligned} \frac{1}{V} \langle\langle N(\omega, j) \rangle\rangle &\equiv \langle\langle W(\omega, j) \rangle\rangle \\ &= -\frac{2\omega}{\pi c_0^2} \text{Im} \mathbf{G}_{jj}(\omega, \mathbf{r} = 0) + \frac{\omega^2}{2\pi^2} n \int \frac{d\hat{\mathbf{k}}}{4\pi} \int_S d^3\mathbf{r} [\boldsymbol{\varepsilon}(\mathbf{r}) - 1] |\mathbf{E}_{\mathbf{k},j}^+(\mathbf{r})|^2. \end{aligned} \quad (4.45)$$

S denotes the volume of only one scatterer, but $\mathbf{E}_{\mathbf{k},j}^+(\mathbf{r})$ is still the exact distorted plane wave in the medium. By symmetry $\langle\langle W(\omega, j) \rangle\rangle$ does not depend on the polarization channel j . We use the symbol W for the density of states per unit volume of the light since it obviously coincides with the electromagnetic energy density for which this symbol was used earlier. In leading order of the particle density $\mathbf{E}_{\mathbf{k},j}^+(\mathbf{r})$ may be replaced by the eigenfunction for one scatterer in vacuum; $\mathbf{G}(\omega, \mathbf{r})$ is the averaged Green's function in real space. In view of Eq. (3.38) the two terms in Eq. (4.45) can be interpreted physically as the contributions from radiation and material degrees of freedom. Indeed the second term is absent in vacuum. For later purposes it is convenient to write the radiation part as an integral over wave numbers. From Eq. (4.19) we can infer that the radiation part can be further separated into a transverse and longitudinal part. This yields the final expression,

$$\langle\langle W(\omega, j) \rangle\rangle = \langle\langle W(\omega, j) \rangle\rangle_{\text{rad}} + \langle\langle W(\omega, j) \rangle\rangle_{\text{long}} + \langle\langle W(\omega, j) \rangle\rangle_{\text{mat}}, \quad (4.46)$$

where

$$\begin{aligned} \langle\langle W_{\text{rad}}(\omega, j) \rangle\rangle &= -\frac{2\omega}{\pi c_0^2} \frac{2}{3} \sum_{\mathbf{p}} \text{Im} \left[\varepsilon_{\parallel}(\omega, p) (\omega/c_0)^2 + i\epsilon - \mathbf{p}^2 \right]^{-1}, \\ \langle\langle W_{\text{long}}(\omega, j) \rangle\rangle &= -\frac{2}{\pi\omega} \frac{1}{3} \sum_{\mathbf{p}} \text{Im} \varepsilon_{\perp}(\omega, p)^{-1}, \\ \langle\langle W_{\text{mat}}(\omega, j) \rangle\rangle &= \frac{\omega^2}{2\pi^2} n \int \frac{d\hat{\mathbf{k}}}{4\pi} \int_S d^3\mathbf{r} [\boldsymbol{\varepsilon}(\mathbf{r}) - 1] |\mathbf{E}_{\mathbf{k},j}^+(\mathbf{r})|^2. \end{aligned} \quad (4.47)$$

The numerical factors $1/3$ and $2/3$ arise from the angular momentum integral. The dielectric function is real-valued in vacuum and so the longitudinal density of states is also absent in vacuum. From that point of view the longitudinal contribution can also be considered as a material contribution.

It is instructive to evaluate the various terms explicitly for an ensemble of identical point particles in the independent scattering approximation (ISA) introduced earlier. The t -matrix is given by Eq. (3.21) and the average dielectric constant induced by these particles in Eq. (4.21). The longitudinal part is divergent but can be regularized with the same cut-off as in section III.B.2.b. The expressions can be put into the following form, using ISA,

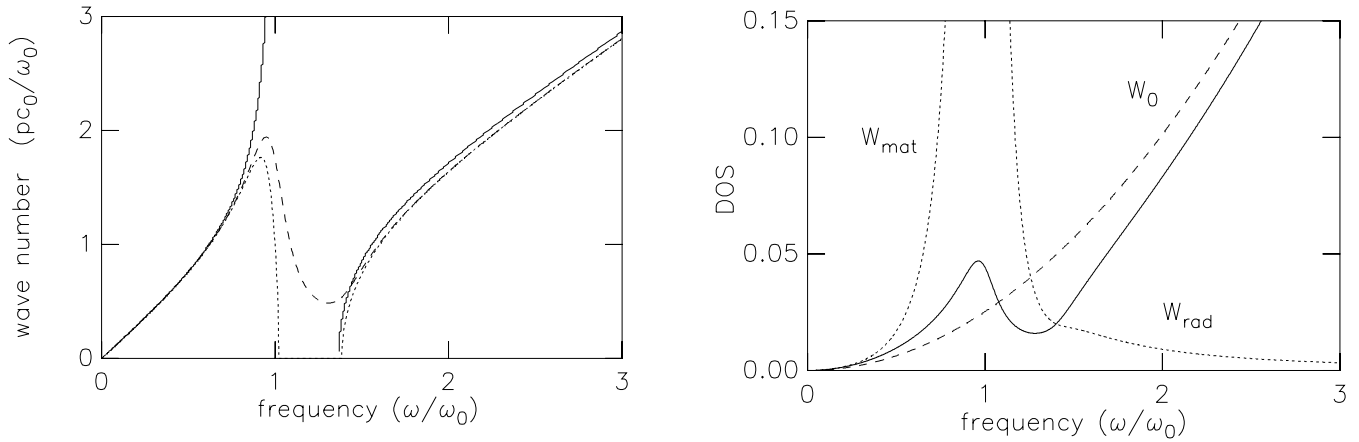


FIG. 10. Polariton behavior and spectral function $S(\omega, p)$. Top figure: Dispersion laws $\omega(p)$ defined in three different ways for a polariton with $\mathcal{P} = 10$ and $T_2 = 10/\omega_0$. We adopted $\varepsilon_\infty = 1$. Bold: maxima of $S(\omega, p)$ for fixed momentum p ; Finely dashed: maxima of $S(\omega, p)$ for fixed frequency ω ; Broadly dashed: real part of the pole of the Dyson Green's function. Far away from the resonant frequency they all coincide. In the regime of resonant scattering they all differ, and the one to be used depends on the experiment. The first in fact resembles the dispersion law of a “lossless” polariton. Like the second dispersion law it has no solution in a small spectral region beyond the resonant frequency. The third dispersion law is strongly anomalous in this region, and its slope is usually associated with the group velocity. Bottom figure: Density of states (DOS) per unit volume (energy density) for the same polariton. In bold is displayed the radiation contribution; Finely dashed we show the the energy density of the matter only. The free density of states W_0 is shown broadly dashed. It is seen that on the left hand side of the resonance the radiation energy density is increased, and on the right hand side - the spectral polariton gap - it is suppressed.

$$\begin{aligned}
 \langle\langle W_{\text{rad}}(\omega, j) \rangle\rangle &= v_p^{-1} W_0(\omega, j), \\
 \langle\langle W_{\text{long}}(\omega, j) \rangle\rangle &= \frac{\gamma}{\gamma + 3} W_0(\omega, j) \times nW_{\text{pot}}(\omega), \\
 \langle\langle W_{\text{mat}}(\omega, j) \rangle\rangle &= \frac{3}{\gamma + 3} W_0(\omega, j) \times nW_{\text{pot}}(\omega),
 \end{aligned} \tag{4.48}$$

where $W_0(\omega, j) = \omega^2/3\pi^2c_0^3$ is the radiation density per polarization direction in vacuum; We recall that $\gamma = \varepsilon - 1$ is the polarizability density of the matter inside the scatterer. The potential energy W_{pot} of one particle is given in Eq. (3.43). Longitudinal and material part have been evaluated in leading order of the number density. Together they constitute the potential energy density nW_{pot} stored in the medium. From this example it is clear that the density of states per unit volume for light is in fact the electromagnetic energy density in the medium. Far away from resonant scattering (in particular in the static limit of small frequencies) the total energy is the one in vacuum times the index of refraction cubed: $(1 + n\alpha_{\text{sph}})^3$.

The number of states available for the travelling waves is changed by the disorder by means of the phase velocity. This is a very important conclusion. Any process that depends on the number of available travelling waves will be changed by this factor. In the lower part of Fig. 10 we show $\langle\langle W_{\text{rad}}(\omega, j) \rangle\rangle$ for a polariton with a very sharp resonance. In the (pseudo) gap that opens up on

the right hand side ($\omega > \omega_0$) of the resonant frequency the number of travelling waves is inhibited since the phase velocity is nearly infinite. On the left hand side ($\omega < \omega_0$) the radiation density of states is increased. Such a behavior may be anticipated since no states are destroyed or created in the medium. The increment of a scattering parameter on one side of the resonance, and the associated decrement on the other is a typical feature of resonant multiple scattering. Theoretical indications exist that this characteristic behavior persists for high densities of the scatterers.

One of the factors that depends strongly on the number of travelling waves is the Einstein spontaneous coefficient of an atom. It can be argued that the density-of-states factor in Eq. (3.48) for the spontaneous emission rate will in general be replaced by the one of the travelling waves, and *not* the total one. It is well known that spontaneous emission can be seriously inhibited for atoms with their resonant frequency in a region with small density of states, e.g. due to the polariton behavior discussed above [88]. Contrary to what is generally believed it is not the total density of states that determines the Einstein spontaneous emission coefficient, but rather the (local) *radiation* density of states W_{rad} . This notion means that the suppression of spontaneous emission is larger than is expected on the basis of the *total* density of states.

4.3.1. Spectral Function

As we discussed before shortly the local density of states does not render any more insight than the total density of states if the system is translationally symmetric. A spectral function $S(\omega, j, \mathbf{p})$ can be introduced which gives upon integration over momenta the density of states per unit volume per polarization channel,

$$\sum_{\mathbf{p}, j} S(\omega, \mathbf{p}, j) = \langle \langle W(\omega) \rangle \rangle . \quad (4.49)$$

The spectral function counts the number of modes (per unit volume) with frequency ω , polarization j and wavenumber \mathbf{p} . For the travelling-wave part - the genuine radiation part - we can just copy what was found in Eq. (4.36) for electrons,

$$S_{\text{rad}}(\omega, j, \mathbf{p}) = \frac{2\omega}{\pi c_0^2} \frac{1}{(\omega^2/c_0^2 + \text{Re}\Sigma_{\perp}(\omega, \mathbf{p}) - \mathbf{p}^2)^2 + (\text{Im}\Sigma_{\perp})^2} (\Delta_{\mathbf{p}})_{jj} ,$$

which translates in more physical quantities to,

$$S_{\text{rad}}(\omega, j, \mathbf{p}) = \frac{2\omega}{\pi c_0^2} \frac{1}{(k^2 - 1/4\ell_{\text{ext}}^2 - \mathbf{p}^2)^2 + (k/\ell_{\text{ext}})^2} (\Delta_{\mathbf{p}})_{jj} . \quad (4.50)$$

We used the definition of wave vector $k(\omega)$ and extinction mean free path $\ell_{\text{ext}}(\omega)$ to convert the Dyson self-energy into physical quantities. The form of this spectral distribution function depends heavily on the value of $k\ell_{\text{ext}}$.

When $k\ell_{\text{ext}} \gg \frac{1}{2}$ this distribution has most of its weight near the “momentum shell” defined by $p = k(\omega)$. To be more precise, the maxima of the spectral function in the $\omega - p$ plane are, for a given value of ω , located on the curve

$$p_{\text{max}}^2(\omega) = k^2 - \frac{1}{4\ell_{\text{ext}}^2} . \quad (4.51)$$

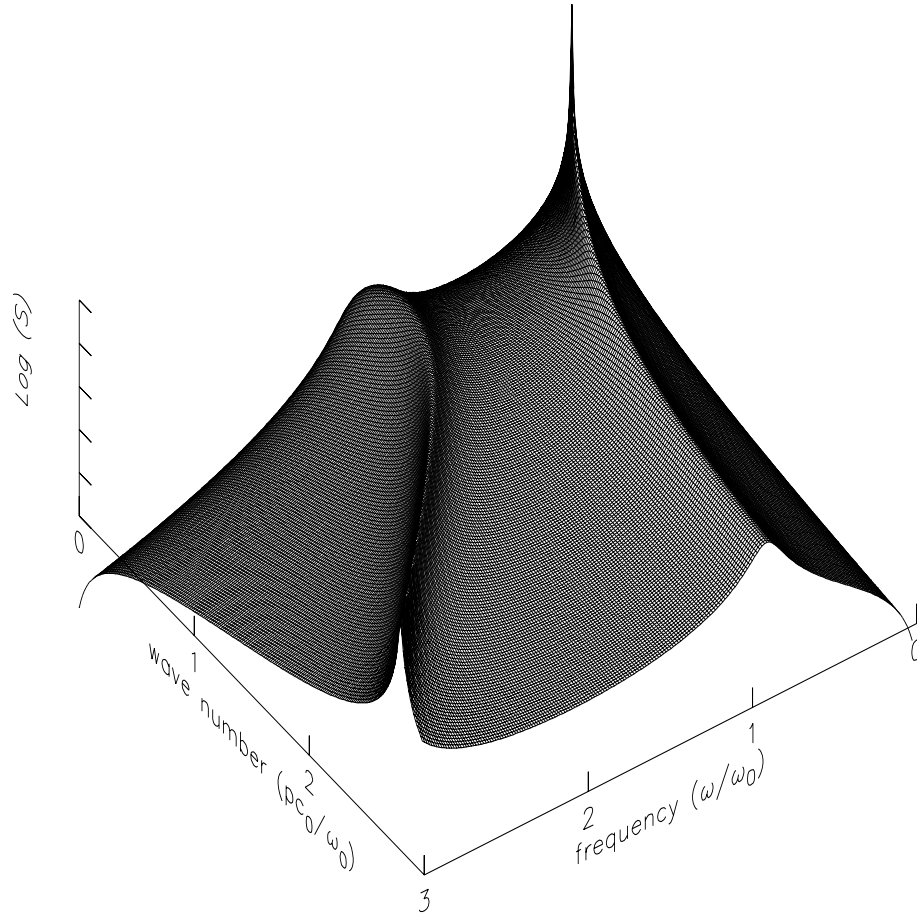


FIG. 11. Three dimensional visualisation of the total spectral function (including the material states) in the $\omega - p$ plane for the same polariton as in Fig. 10 (polariton parameter $\mathcal{P} = 10$). Notice the two branches where the maximum of $S(\omega, p)$ occurs, and the (pseudo-) gap in between.

The maximum is seen to be slightly different from the momentum shell due to shift of the mean free path. On the other hand for a fixed value of p , the maxima are two separate branches resembling a polariton without damping. This is called the *Raman dispersion curve* of the polariton, since it determines the peaks in spontaneous Raman scattering [89] (if the polariton is Raman active).

On the other hand when $kl_{\text{ext}} < \frac{1}{2}$ the shell has completely disappeared and the maximum of the spectral distribution occurs at $p = 0$: the waves are evanescent. In the upper part of Fig. 10 of we compare the dispersion law $\omega(k)$ to the relation given by Eq. (4.51) and the Raman curve. It should be remembered that the two latter curves only visualize a *maximum* of the spectral function. A better understanding can be obtained from the three-dimensional plot 11. Exactly in the evanescent region the maxima of the spectral function do not coincide with the dispersion law.

These results imply that the frequency regime characterized by

$$kl_{\text{ext}} \leq \frac{1}{2}, \quad (4.52)$$

is no longer one in which one can think in terms of plane-wave propagation and can therefore be called the strongly scattering regime. A similar line of reasoning was put forward in 1960 by A.F. Ioffe and A.R. Regel [90] in their discussion of electron propagation in strongly doped

semiconductors. The criterion (4.52) is therefore known as the Ioffe-Regel criterion for the strongly scattering regime. It is frequently misused as a criterion for Anderson localization. We will comment on this in section IV.F.4.

The mean free path that goes into the criterion (4.52) is the extinction mean free path, and counts everything, including inelastic scattering. For atomic oscillators the inelastic component in the light scattering is sometimes very large, and the strongly scattering regime can easily be realized.

Sofar we concentrated on the spectral function of the transverse propagating modes only. From the expression of the density of states for the longitudinal solutions of the Maxwell equations, the associated spectral function can be written down at once. For the matter part more work has to be done. Nevertheless, the *total* spectral function, counting all possible states, can be directly obtained from Eq. (4.41),

$$\begin{aligned} \langle\langle W(\omega) \rangle\rangle &= 2\omega \langle\langle \text{Tr} \delta [\omega^2 - c_0^2 \epsilon(\mathbf{x})^{-1} \cdot \mathbf{p}^2 \Delta_{\mathbf{p}}] \rangle\rangle \\ &= \langle\langle \text{Tr} [\epsilon(\mathbf{x}) - c_0^2 \omega^{-2} \mathbf{p}^2 \Delta_{\mathbf{p}}] \delta [\epsilon(\mathbf{x}) \omega^2 - c_0^2 \mathbf{p}^2 \Delta_{\mathbf{p}}] \rangle\rangle \\ &\quad + \text{Tr} c_0^2 \omega^{-2} \mathbf{p}^2 \Delta_{\mathbf{p}} \cdot 2\omega \langle\langle \delta [\epsilon(\mathbf{x}) (\omega/c_0)^2 - \mathbf{p}^2 \Delta_{\mathbf{p}}] \rangle\rangle. \end{aligned} \quad (4.53)$$

We have made use of the cyclic property of the trace. The first term in this formula vanishes. The trace can be replaced again by a momentum integral. The average of the delta-function is in momentum space is the spectral function for the transverse electromagnetic waves. We arrive at a very simple formula, namely [59]

$$S(\omega, \mathbf{p}, j) = \frac{p^2 c_0^2}{\omega^2} \times S_{\text{rad}}(\omega, j, \mathbf{p}). \quad (4.54)$$

This formula demonstrates nicely that near the dispersion law the states are only travelling waves. For $p^2 \gg (\omega/c_0)^2$ one is also including the non propagating states of the matter. In the regime $p^2 \ll (\omega/c_0)^2$ only very few evanescent states are found. If there are no propagating states with frequency ω and wave vector \mathbf{p} , there are no states at all with these parameters.

The relation for the total spectral function looks deceptively simple, despite the fact that it contains *all* information of the excitations. We would like to emphasize that a full knowledge of $S(\omega, \mathbf{p}, j)$ for all arguments is never available. Furthermore, whereas the strongly simplifying shell approximation for the spectral function (replacing it by a delta-function, with an appropriate weight) is reliable for electrons, for light it would completely disregard the states of the matter that do not propagate.

4.4. Average Intensity from Scattering Theory

The most important element in physics is symmetry and the associated conservation laws. If a quantity is conserved globally, the removal of a local imbalance will be slow (non exponential in time) as transport over all of space is needed to get rid of the asymmetry. If there is no conservation law operating we encounter the rather uninteresting situation that a local disturbance is removed quickly (that is exponential in time). Whenever a quantity is conserved, the transport of its density will be slow. Transport over long distances can then be modelled with a random walk picture and a diffusion law. Sometimes the conservation law is not obeyed rigorously and a small amount of local absorption is possible. If the absorption is small it can be dealt with as a perturbation. If the

infringement of the conservation law is serious the whole concept of a random walk and diffusion will break down.

Let us focus now on light scattering in disordered materials. We have seen in the previous section that the coherent light beam loses its energy after one mean free path. Nevertheless, the scattering objects satisfy the Optical Theorem so that conservation of energy is guaranteed during scattering. It is physically obvious that the energy of the coherent beam is used for scattering in other directions. This scattering can never be contained in the coherent beam, because this one propagates in a translationally symmetric medium in which the direction of propagation is necessarily conserved. Once scattered in some direction the wave serves as a new coherent beam and again decays in space. This picture strongly suggests that a random walk model for the light indeed applies. The step length is given by the mean free path of the light, and not by the average distance between the scattering centers, which is often much smaller.

A number of objections can be raised against this simple picture. First of all, in the random walk picture all different paths the wave can take are completely unaware of each other. All interference has been neglected, and we are in fact following the motion of a soccer ball shot into a wood (physically equal to the classical Lorentz gas). Secondly, it is unclear what happens if the scattering objects come close together. The wave is no longer scattering from one particle but rather from clusters of particles. Within this microscopic cluster, the random walk model fails completely. A strict derivation of the laws that govern the transport of energy on all scales is thus demanding.

4.4.1. Microscopic Transport Equation

In this section we derive a rigorous transport equation starting from the wave equation. A vector treatment for the various Stokes parameters has been given by Papanicolaou and Burridge [91]. A scalar treatment will be given in order not to drown in polarization indices. The average radiation density at time t and position \mathbf{r} is proportional to the absolute square of the field amplitude:

$$I(\mathbf{r}, t) \sim \langle \langle |\Psi(\mathbf{r}, t)|^2 \rangle \rangle . \quad (4.55)$$

We will represent the scalar field by $\Psi(\mathbf{r}, t)$ and sometimes think of it as the electric field. Given some perturbation we want to follow the average intensity in space and time.

Let us first focus on the time and forget about the averaging. If the perturbation occurs at time $t = 0$ the intensity must be zero for $t < 0$. This asks for a Laplace transformation,

$$I(\Omega) = \int_0^\infty dt |\Psi(t)|^2 e^{i(\Omega + i\epsilon)t} . \quad (4.56)$$

An infinitesimally small positive ϵ makes sure that this integral converges in critical cases. It is convenient to disentangle the perturbation into frequency components, and write

$$\Psi(t) = \int_{-\infty}^\infty \frac{d\omega}{2\pi} \Psi(\omega) e^{i\omega t} . \quad (4.57)$$

Inserting into Eq. (4.56) gives,

$$I(\Omega) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Psi(\omega + \Omega/2 + i\epsilon) \Psi^*(\omega - \Omega/2 - i\epsilon) . \quad (4.58)$$

Use has been made of the fact that $\Psi(\omega)$ can be analytically continued in the lower sheet $\text{Im } \omega < 0$ since $\Psi(t < 0) = 0$. The frequency function under the integral sign determines the time aspects of the propagation.

Let us next consider the spatial part, and add the averaging over realizations. We will drop the frequency indices and add them later. The propagation from \mathbf{r} to \mathbf{r}' is determined by the Green's function $G(\mathbf{r}, \mathbf{r}')$. Knowledge of the average of the square of the Green's function,

$$L(\mathbf{r}, \mathbf{r}') = \langle \langle G(\mathbf{r}, \mathbf{r}') G^*(\mathbf{r}, \mathbf{r}') \rangle \rangle , \quad (4.59)$$

suffices to find the average disturbance at any position \mathbf{r}' in the medium given a source at another specified position \mathbf{r} . In general this function can depend on four positions, but for our purposes we only need two. As was the case for the average amplitude the derivation is far more elegant in momentum representation. We write,

$$G(\mathbf{r}, \mathbf{r}') = \sum_{\mathbf{p}_1, \mathbf{p}_2} G(\mathbf{p}_1, \mathbf{p}_2) e^{i(-\mathbf{p}_1 \cdot \mathbf{r} + \mathbf{p}_2 \cdot \mathbf{r}')} . \quad (4.60)$$

Averaging restores translational symmetry so that $L(\mathbf{r}, \mathbf{r}') = L(\mathbf{r} - \mathbf{r}')$. This implies momentum conservation,

$$\langle \langle G(\mathbf{p}_1, \mathbf{p}_2) G^*(\mathbf{p}_3, \mathbf{p}_4) \rangle \rangle \equiv \Phi_{\mathbf{p}\mathbf{p}'}(\mathbf{q}) \times (2\pi)^3 \delta(-\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) . \quad (4.61)$$

For later convenience we defined new momentum variables \mathbf{p} , \mathbf{p}' and \mathbf{q} according to $\mathbf{p}_1 = \mathbf{p} + \mathbf{q}/2$, $\mathbf{p}_2 = \mathbf{p}' + \mathbf{q}/2$, $\mathbf{p}_3 = \mathbf{p} - \mathbf{q}/2$. By momentum conservation $\mathbf{p}_4 = \mathbf{p}' - \mathbf{q}/2$. Inserting this into Eq. (4.59) gives us

$$L(\mathbf{r}) = \sum_{\mathbf{q}} \left\{ \sum_{\mathbf{p}\mathbf{p}'} \Phi_{\mathbf{p}\mathbf{p}'}(\mathbf{q}) \right\} e^{-i\mathbf{q} \cdot \mathbf{r}} . \quad (4.62)$$

Combining the results found in Eqs. (4.58) and (4.62) the object that determines the exact microscopic space-time behavior of a disturbance is $\Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q})$ and is defined by

$$\begin{aligned} \langle \langle G(\omega + \Omega/2 + i\epsilon, \mathbf{p}_1, \mathbf{p}_2) G(\omega - \Omega/2 - i\epsilon, \mathbf{p}_3, \mathbf{p}_4) \rangle \rangle \\ \equiv \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) \times (2\pi)^3 \delta(-\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) . \end{aligned} \quad (4.63)$$

The frequency Ω and the momentum \mathbf{q} are identified as the conjugate variables of the carrier wave that supports the disturbance. The frequency ω and the momentum \mathbf{p} denote genuine internal oscillations of the wave packet in space and time. In most cases these parameters are orders of magnitude larger than the slow variables Ω and \mathbf{q} . From now on we shall discriminate between slow and fast variables by referring to the fast variables by means of subscripts (Fig. 12). The momentum \mathbf{p}' is associated with spatial characteristics of the source and is automatically summed over when the initial disturbance originates from a point. The transport quantity is then,

$$\Phi_{\omega \mathbf{p}}(\Omega, \mathbf{q}) = \sum_{\mathbf{p}'} \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) . \quad (4.64)$$

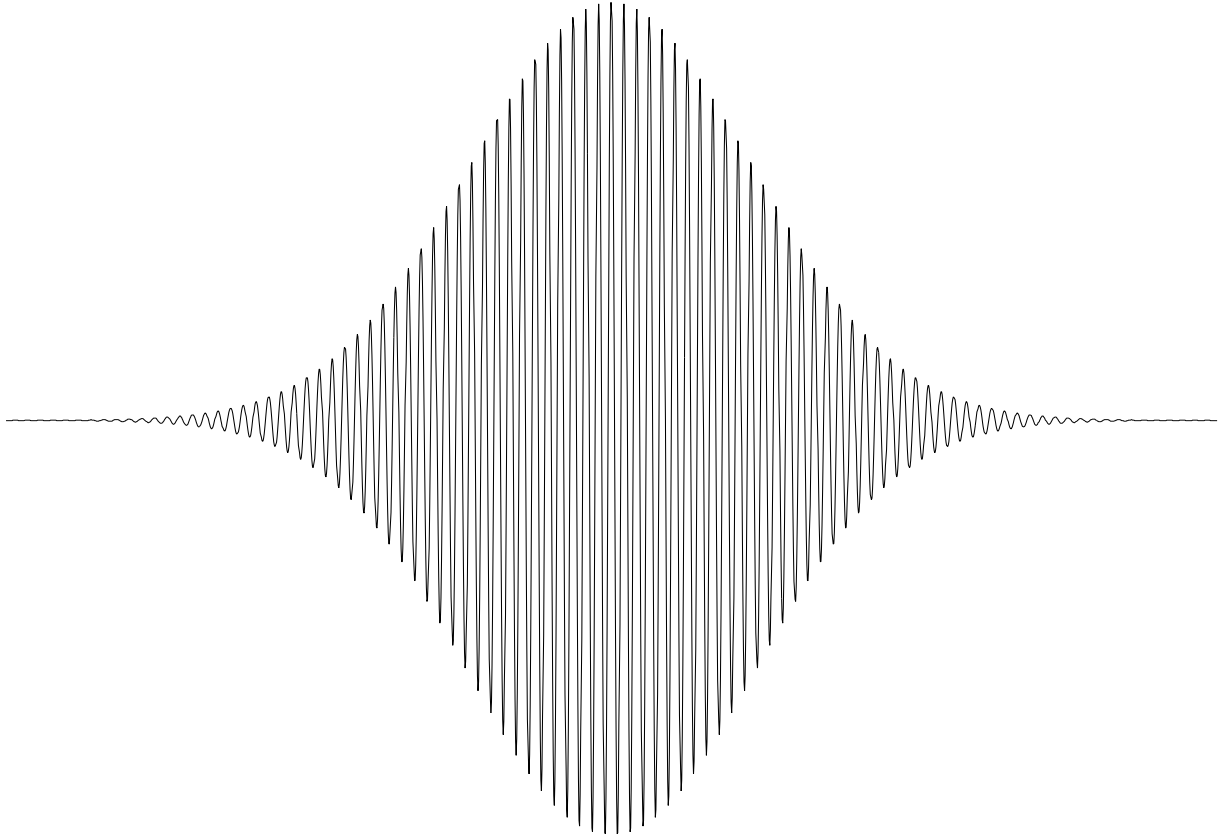


FIG. 12. A typical wave packet (either in space or time) that we envisage in our transport equation. Internal oscillations occur at frequency ω (in time) and wave vector \mathbf{p} (in space); The envelope is described by the frequency Ω (in time) and the wave vector \mathbf{q} (in space). These two variables are much smaller than the ones characterizing the internal oscillations. This is the “Slowly Varying Envelope Approximation”. The wave packet is denoted mathematically by $\Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q})$. Apart from numerical constants this is the exact microscopic definition of the “specific intensity” featuring in radiative transport theory.

Having identified the appropriate transport quantity, we are left with the determination of its properties. As was the case with the self energy Σ in the coherent beam, a definition of a building block for the intensity will give us deeper insight. The *Irreducible Vertex* U is defined by the (operator) relation,

$$\langle\langle G(\omega) G^*(\omega') \rangle\rangle = G(\omega) G^*(\omega') + G(\omega) G^*(\omega') U(\omega, \omega') \langle\langle G(\omega) G^*(\omega') \rangle\rangle, \quad (4.65)$$

$G(\omega)$ denotes the averaged Dyson Green’s function that was already obtained earlier for which we do not show the averaging brackets explicitly. The equation shown here is widely known as the Bethe-Salpeter equation. In a translationally invariant medium full use can be made of momentum conservation, and the Bethe-Salpeter equation can be transformed into one for the object we need, namely $\Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q})$. We will spare the reader some algebraic details (see e.g. [92] [93] [94]). What comes out is,

$$\left[\frac{i\omega\Omega}{c_0^2} - i\mathbf{p} \cdot \mathbf{q} + \Delta\Sigma_{\omega\mathbf{p}}(\Omega, \mathbf{q}) \right] \Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q}) = \Delta G_{\omega\mathbf{p}}(\Omega, \mathbf{q}) \left[1 + \sum_{\mathbf{p}'} U_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) \Phi_{\omega\mathbf{p}'}(\Omega, \mathbf{q}) \right]. \quad (4.66)$$

In this equation we introduced

$$\Delta\Sigma_{\omega\mathbf{p}}(\Omega, \mathbf{q}) \equiv \frac{1}{2i} \{ \Sigma(\omega + \Omega/2 + i\epsilon, \mathbf{p} + \mathbf{q}/2) - \Sigma(\omega - \Omega/2 - i\epsilon, \mathbf{p} - \mathbf{q}/2) \} ,$$

in terms of the Dyson self energies defined earlier. Exactly the same definition holds for $\Delta G_{\omega\mathbf{p}}(\Omega, \mathbf{q})$ in terms of the Dyson Green's functions. The convention of the matrix element $U_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q})$ is exactly the same as for $\Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q})$.

The physical interpretation of Eq. (4.66) is the following. The Ω -term on the left hand side will act as a time derivative in time coordinates. The second term is a typical hydrodynamic flow term, and contains in coordinate space a gradient. The $\Delta\Sigma$ -term comes from the average amplitude and was already shown to represent loss of energy (extinction). The second term on the right hand side contains crucial new information. It represents the scattered intensity from all directions \mathbf{p}' into the direction \mathbf{p} . Not all absolute values of \mathbf{p}' count since the ΔG -factor makes sure that only momenta near the dispersion relation come in, as expected physically. The object $U_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q})$ is the rigorous microscopic “building block” for scattering processes in the medium, $\Delta\Sigma_{\omega\mathbf{p}}(\Omega, \mathbf{q})$ the one for extinction. The first term on the right hand side is a source term that shows up from the initial value problem. The physical interpretation of Eq. (4.66) is therefore

$$[\partial_t + \mathbf{v} \cdot \nabla + \text{losses}] I_{\mathbf{v}}(\mathbf{r}, t) = \text{Source} + \text{scattering} . \quad (4.67)$$

This looks very much like a transport equation for a particle; \mathbf{v} is the velocity associated with the particle, which maps onto the momentum \mathbf{p} of the wave. This balance equation can be recognized as a generalized Boltzmann equation. It is the most important equation of transport theory. Generalized, because all microscopic time and space correlations are still included.

As always with exact microscopic equations they cannot be solved exactly and one has to rely on approximations. In lowest order of the scatterer density we anticipate the building block to be determined by the exact single scattering solution. Concerning the loss term this was already shown to be true in Eq. (4.20). A similar formula holds for the single-scattering contribution to the Irreducible Vertex. We have

$$\begin{aligned} \Sigma^{(1)}(\omega, \mathbf{p}) &= n T_{\mathbf{p}\mathbf{p}}(\omega) , \\ U_{\omega, \mathbf{p}\mathbf{p}'}^{(1)}(\Omega, \mathbf{q}) &= n T_{\mathbf{p}^+\mathbf{p}^+} \left(\omega + \frac{\Omega}{2} + i\epsilon \right) T_{\mathbf{p}'^-\mathbf{p}^-} \left(\omega - \frac{\Omega}{2} - i\epsilon \right) . \end{aligned} \quad (4.68)$$

The replacement of U by $U^{(1)}$ is widely known as the *Boltzmann approximation*. If the single particle properties are known completely, the transport equation can in principle be solved. We emphasize that this equation is derived for an infinite medium. In all applications one uses this equation supported by boundary conditions to find the transport in a finite medium, with considerable success. This is not a good approach for optically thin media.

Two important physical quantities to be determined are the (average) local energy density of the radiation and the local current density with which it propagates. The radiation density was already found in Eq. (4.46) for light in a stationary ($\Omega = 0$) situation. For scalar waves the local, time-dependent radiation density is proportional to $|\Psi(\mathbf{r}, t)|^2$ given in Eq. (4.55). Expressed in Fourier and Laplace variables it becomes,

$$W_{\omega}^{\text{rad}}(\Omega, \mathbf{q}) \equiv (\omega/c_0)^2 \sum_{\mathbf{p}} \Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q}). \quad (4.69)$$

We added the frequency factor in order to be consistent with Eq. (2.14) in section II. Similarly the current density at frequency ω can be identified as,

$$\mathbf{J}_{\omega}(\mathbf{q}) = \omega \sum_{\mathbf{p}} \mathbf{p} \Phi_{\omega\mathbf{p}}(\mathbf{q}). \quad (4.70)$$

4.4.2. Steady State: $\Omega = 0$

A steady state is characterized by a stationary flow of particles or energy. This is not the same as equilibrium. In equilibrium the flow should be zero because the flow of two different mechanisms compensate (think of a copper wire in thermal equilibrium, where the diffusion flux of electrons is compensated by the drift induced by an external electric field). A steady state is only achievable in a *finite* medium. In an infinite medium a small perturbation will spread without end.

The stationary case is the typical case encountered in astrophysics. Sources are usually time independent on the time scales that light needs to travel through a stellar atmosphere or interstellar cloud. Accordingly, this situation is extremely well documented in standard astrophysical references [2] [3]. In laboratory experiments the steady state is reached when a scattering medium is being exposed to a stationary light source. In many cases this is a laser with a frequency ω in the optical regime. The rapid optical oscillations of the laser light are not considered time dependent since any detector will average them out automatically.

Mathematically, the steady state is characterized by $\Omega = 0$. We are now following the motion of $|\Psi_{\omega}(\mathbf{r})|^2$ in space. Only one frequency, namely ω , enters the problem. For that reason we will no longer use the variable Ω explicitly.

Let us first find a rigorous result that we get almost for free. If we adopt scatterers (no matter how difficult) that scatter light purely elastically, no energy will be lost in the medium. In a stationary situation this implies that the divergence of the current density (4.70) should vanish, or be equal to possible local sources. If we integrate left and right hand side of our transport equation over the momentum \mathbf{p} all losses are to be compensated by the gain. This provides us with,

$$\Delta \Sigma_{\omega\mathbf{p}}(\mathbf{q}) = \sum_{\mathbf{p}'} \Delta G_{\omega\mathbf{p}'}(\mathbf{q}) U_{\omega,\mathbf{p}'\mathbf{p}}(\mathbf{q}). \quad (4.71)$$

This is an extremely important result. It tells us that conservation of energy gives rise to an identity between the average amplitude and the average intensity. In field theory such an equation is called a Ward Identity.

We will proceed with the low-density approximation (4.68). In general one has to be very careful in approximating exact equations because the approximation might break the symmetry and destroy the associated conservation law. The only way to make the approximation (4.68) consistent with the exact identity (4.71) is to let

$$\Delta G_{\omega\mathbf{p}}(\mathbf{q}) \rightarrow \Delta G_{\omega\mathbf{p}}^0(\mathbf{q}). \quad (4.72)$$

The index zero indicates vacuum. The momentum \mathbf{q} is the conjugate variable of macroscopic length scales, which are assumed larger than the physical size of the scattering objects. We may therefore

neglect it *consistently* in both $\Delta\Sigma$, ΔG and U . Physically we have now imposed that the scatterers see each other in their far field. This approximation does not break the symmetry, but simplifies the transport enormously. It can be inferred that $\Delta G^0(\mathbf{q}=0) = -\pi\delta((\omega/c_0)^2 - p^2)$, so that momentum comes on the momentum shell in vacuum. The Ward identity Eq. (4.71) has now become equal to the Optical Theorem (3.30) derived earlier for single scattering. One often prefers to choose the momentum shell in the medium defined by the phase velocity in order to appreciate the impact of the effective medium on the current (all modifications in second order of the particle density caused in this way are not rigorous). A *specific intensity* can be introduced according to,

$$\frac{\omega^2}{c_0^2} \Phi_{\omega\mathbf{p}}(\mathbf{q}) \equiv \frac{16\pi^3 v_p}{\omega c_0} \delta\left(\frac{\omega^2}{v_p^2} - p^2\right) I_{\omega,(\omega/v_p)\hat{\mathbf{p}}}(\mathbf{q}). \quad (4.73)$$

Following the convention in astrophysical literature, we chose the front factor such that the radiation density (4.69) equals the angular integral of the specific intensity, divided by c_0 :

$$W_{\omega}^{\text{rad}}(\Omega, \mathbf{q}) = \frac{1}{c_0} \int d^2\hat{\mathbf{p}} I_{\omega,(\omega/v_p)\hat{\mathbf{p}}}(\mathbf{q}). \quad (4.74)$$

By \mathbf{k} we shall always denote a wave vector on the momentum shell $|\mathbf{k}| = \omega/v_p$. The generalized Boltzmann equation simplifies to a closed equation for the specific intensity,

$$\left[-i\frac{c_0}{v_p} \hat{\mathbf{k}} \cdot \mathbf{q} - n \frac{\text{Im } T_{\mathbf{k}\mathbf{k}}(\omega)}{\omega/c_0} \right] I_{\omega\mathbf{k}}(\mathbf{q}) = \text{constant} + n \int d^2\hat{\mathbf{k}}' \frac{|T_{\mathbf{k}\mathbf{k}'}(\omega)|^2}{(4\pi)^2} I_{\omega\mathbf{k}'}(\mathbf{q}). \quad (4.75)$$

For practical purposes this equation must be transformed back to real space. The loss term can be identified as $n\sigma_{\text{scat}}$ (σ_{scat} being the total scattering cross-section) and equals the inverse mean free path. The gain term contains the differential cross section for scattering from \mathbf{k}' to \mathbf{k} . Writing this in terms of the phase function,

$$n \frac{d\sigma}{d\Omega}(\omega, \mathbf{k}' \rightarrow \mathbf{k}) = \frac{1}{\ell} \times \Phi_{\omega}(\mathbf{k}, \mathbf{k}'),$$

and scaling the distance with the mean free path $\boldsymbol{\tau} = \mathbf{r}\ell$, we can write

$$\left[\frac{c_0}{v_p} \hat{\mathbf{k}} \cdot \boldsymbol{\nabla}_{\boldsymbol{\tau}} + 1 \right] I_{\omega\mathbf{k}}(\boldsymbol{\tau}) = \text{source} + \int d^2\hat{\mathbf{k}}' \Phi_{\omega}(\mathbf{k}, \mathbf{k}') I_{\omega\mathbf{k}'}(\boldsymbol{\tau}). \quad (4.76)$$

This equation is the Equation of Radiative Transfer, first derived phenomenologically by astrophysicists. It can straightforwardly be extended for vector scattering [2]. The source is often replaced by boundary conditions for the specific intensity in some direction. In case absorption or inelastic scattering occur, an additional loss term enters. The factor c_0/v_p is the index of refraction of the effective medium.

In the stationary situation the measurable quantity turns out to be the specific intensity in some direction. In section II it was mentioned that in comparing light with electrons the currents are very similar. For that reason the Equation of Radiative Transfer must hold for electron-impurity scattering as well. Only minor modifications will come in if the underlying structure is crystalline and not vacuum. Indeed, Eq. (4.76) is frequently used to find the Ohmic resistance of a doped metal.

a. Diffusion Approximation: $\mathbf{q} \rightarrow \mathbf{0}$, $\Omega = \mathbf{0}$ Even for simple geometries and phase functions the exact solution of Eq. (4.76) must be obtained numerically. We will now discuss an approximate solution which works very well in practise, and provides closed-form formulas even for complicated geometries and phase functions: the diffusion approximation [95].

The derivation is simplest in momentum space. As mentioned before the variable \mathbf{q} is a small variable as it represents the wave vector of a macroscopic perturbation from homogeneity. Small inhomogeneities give rise to currents so that the local specific intensity is no longer isotropic. We can account for such anisotropy by expanding the specific intensity as

$$I_{\omega\mathbf{k}}(\mathbf{q}) = \frac{c_0}{4\pi} \left[W_{\omega}^{\text{rad}}(\mathbf{q}) + 3\frac{v_p}{c_0^2} \hat{\mathbf{k}} \cdot \mathbf{J}_{\omega}(\mathbf{q}) + \dots \right]. \quad (4.77)$$

Higher multipoles can be added to improve accuracy. The diffusion approximation consists of considering only the isotropic term and the dipole term. In the diffusion approximation radiation density and current density have become the only two characteristics of the intensity pattern and the rest has been ignored. Their relation can now be found from the first two \mathbf{k} -moments of Eq. (4.75):

$$\begin{aligned} -i\mathbf{q} \cdot \mathbf{J}_{\omega}(\mathbf{q}) &= \text{constant}, \\ \frac{1}{3} \frac{c_0^2}{v_p} i\mathbf{q} W_{\omega}^{\text{rad}}(\mathbf{q}) &= \ell^{-1} \int d^2\hat{\mathbf{k}}' \Phi_{\omega}(\mathbf{k}, \mathbf{k}') [1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}'] \mathbf{J}_{\omega}(\mathbf{q}). \end{aligned} \quad (4.78)$$

In coordinate space this can be written as,

$$\begin{aligned} \nabla \cdot \mathbf{J}_{\omega}(\mathbf{r}) &\sim \delta(\mathbf{r}) = \text{source}, \\ \mathbf{J}_{\omega}(\mathbf{r}) &= -\frac{1}{3} \frac{c_0^2}{v_p} \ell_{\text{tr}} \nabla W_{\omega}^{\text{rad}}(\mathbf{r}). \end{aligned} \quad (4.79)$$

They can be combined to one equation,

$$-\frac{1}{3} \frac{c_0^2}{v_p} \ell_{\text{tr}} \nabla^2 W_{\omega}^{\text{rad}}(\mathbf{r}) \sim \delta(\mathbf{r}) = \text{source}. \quad (4.80)$$

The *transport mean free path* has been defined as,

$$\ell_{\text{tr}} \equiv \frac{\ell}{\int d^2\hat{\mathbf{k}}' \Phi_{\omega}(\mathbf{k}, \mathbf{k}') [1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}']} = \frac{\ell}{1 - \langle \cos \theta \rangle}. \quad (4.81)$$

The length scale in diffusion is not equal to the scattering mean free path if the particles scatter anisotropically. This is the first difference we encounter between average amplitude and average intensity. Strictly speaking, the transport mean free path is not a length scale like the scattering mean free path since nothing decays exponentially with this length. Equation (4.80) resembles the electrostatic potential of a point charge. Hence the radiation energy $W_{\omega}^{\text{rad}}(\mathbf{r})$ decays as $1/r$ Coulomb law from the source. It may seem a lot work to derive this simple result, but we succeeded in giving the diffusion phenomenon a microscopic base.

The diffusion approximation works surprisingly well experimentally [96], and turns out to be valid far beyond the regime for which it was originally derived. Despite the name “diffusion approximation”, no diffusion constant showed up so far. We will deal with this now.

4.4.3. Dynamic Aspects: $\Omega \neq 0$

The dynamics of light propagation comes in when a finite value for Ω is adopted. Let us first mention a *wrong* result that is frequently encountered in literature. In a phenomenological treatment time dependence of the specific intensity can be incorporated by adding a time derivative in the Equation of Radiative Transfer, which then reads,

$$\left[-\tau_{\text{mf}} \partial_t + \frac{c_0}{v_p} \hat{\mathbf{k}} \cdot \nabla_{\boldsymbol{\tau}} + 1 \right] I_{\omega \mathbf{k}}(\mathbf{r}, t) = \text{source} + \int d^2 \hat{\mathbf{k}}' \Phi_{\omega}(\mathbf{k}, \mathbf{k}') I_{\omega \mathbf{k}'}(\mathbf{r}; t) . \quad (4.82)$$

The time $\tau_{\text{mf}} = \ell/v_p$ is the mean free time between the collisions. This equation turns out to have no microscopic base! See here the risk of phenomenological derivations. It has turned out experimentally that for resonant multiple scattering of light this equation is simply wrong. The explanation is that it does not take into account the delay that a wave undergoes during scattering. In resonant light scattering this time is known to be considerably larger than the average time between two collisions. The error in Eq. (4.82) is due to the difference between light and electrons: Eq. (4.82) is correct for electrons.

a. Transport Velocity Let us first see how the equation of continuity looks like for the light transport in the random medium. To this end we have to go back to our exact transport equation (4.66) which we can integrate over all momenta \mathbf{p} . Doing this yields,

$$\begin{aligned} i\Omega W_{\omega}^{\text{rad}}(\Omega, \mathbf{q}) - i\mathbf{q} \cdot \mathbf{J}_{\omega}(\Omega, \mathbf{q}) + \omega \sum_{\mathbf{p}} \left\{ \Delta \Sigma_{\omega \mathbf{p}}(\Omega, \mathbf{q}) - \sum_{\mathbf{p}'} \Delta G_{\omega \mathbf{p}'}(\Omega, \mathbf{q}) U_{\omega, \mathbf{p}' \mathbf{p}}(\Omega, \mathbf{q}) \right\} \Phi_{\omega \mathbf{p}}(\Omega, \mathbf{q}) \\ = \omega \sum_{\mathbf{p}} \Delta G_{\omega \mathbf{p}}(\Omega, \mathbf{q}) . \end{aligned} \quad (4.83)$$

The structure of this equation is nearly the one of a continuity equation. From the exact expression (2.14) derived in the section II it is obvious that W_{ω}^{rad} is not the proper conserved quantity. In the previous chapter we called $|\Psi(\mathbf{r}, t)|^2$ the radiation contribution to the total energy density. From this it follows that the complicated third term in Eq. (4.83) must represent the potential energy of the matter. The conservation law (4.71) found earlier shows that this term must at least be proportional to Ω , in order to correspond to a time derivative. Thus in principle we obtain,

$$\partial_t W_{\omega}^{\text{rad}} + \nabla \cdot \mathbf{J}_{\omega} + \partial_t W_{\omega}^{\text{pot}} = \text{source} . \quad (4.84)$$

Two basic conclusions can be drawn immediately. First for De Broglie waves we already know that the potential energy is not allowed to show up in the equation of continuity: $|\Psi(\mathbf{r}, t)|^2$ is the exact conserved quantity. This simple notion implies that

$$\text{quantum} \Rightarrow \Delta \Sigma_{\omega \mathbf{p}}(\Omega, \mathbf{q}) - \sum_{\mathbf{p}'} \Delta G_{\omega \mathbf{p}'}(\Omega, \mathbf{q}) U_{\omega, \mathbf{p}' \mathbf{p}}(\Omega, \mathbf{q}) = 0 . \quad (4.85)$$

Secondly, the violation of this identity in case of finite Ω for scalar waves should represent the potential energy W_{ω}^{pot} stored in the dielectric scatterers. Thus Eq. (4.85) is *wrong* for light when $\Omega \neq 0$. From the results of section III.B.3 a violation of Eq. (4.85) can indeed be anticipated for classical waves since extra terms will be generated by the presence of the frequency dependent potential (2.8). As has been shown such frequency dependence describes internal degrees of freedom.

The internal degree of freedom shows up elegantly and automatically in the energy balance. We shall denote the deviation of relation (4.85) by $i\Omega\omega \times \delta_\omega$. Thus,

$$\text{classical waves} \Rightarrow \Delta\Sigma_{\omega\mathbf{p}}(\Omega, \mathbf{q}) - \sum_{\mathbf{p}'} \Delta G_{\omega\mathbf{p}'}(\Omega, \mathbf{q}) U_{\omega, \mathbf{p}'\mathbf{p}}(\Omega, \mathbf{q}) \equiv i \frac{\Omega\omega}{c_0^2} \times \delta_\omega(\Omega, \mathbf{q}) \neq 0. \quad (4.86)$$

We calculate δ_ω making the same low-density approximation given by Eqs. (4.68) and (4.72). We adopt the same simplification as in the steady-state case, that is ISA and Boltzmann approximation. This results in,

$$\delta_\omega^{(1)} = n \left[-\frac{\partial \text{Re } t_{\mathbf{k}\mathbf{k}}(\omega)}{\partial (\omega/c_0)^2} + \mathcal{R}_\omega + c_0 \int d\Omega_{\mathbf{k}'} \frac{d\sigma}{d\Omega}(\mathbf{k}' \rightarrow \mathbf{k}) \frac{\partial \Phi(\omega, \mathbf{k}, \mathbf{k}')}{\partial \omega} \right] + \mathcal{O}(\Omega). \quad (4.87)$$

We abbreviated,

$$\mathcal{R}_\omega = -n \sum_{\mathbf{p}} \frac{\partial \text{Re } G_0(\omega, p)}{\partial (\omega/c_0)^2} \cdot \frac{|t_{\mathbf{k}\mathbf{p}}(\omega)|^2}{(4\pi)^2}.$$

Next we can apply the fact that $\delta_\omega = 0$ for quantum wave functions. As a result only terms caused by the “frequency dependent potential” come in. The term \mathcal{R}_ω is in its full extent subject to the quantum cancellation, since it does not suffer from a frequency dependence of the light potential. Using Eq. (3.39), we get the desired result that

$$\delta_\omega^{(1)} = n W_{\text{pot}}(\omega),$$

where the potential energy of one particle (normalized to the outside) is given by Eq. (3.40). The equation of continuity takes therefore exactly the form as was announced in Eq. (4.84). We find that the stored energy is at any time proportional to the radiation energy. Therefore the radiation satisfies the continuity equation,

$$\partial_t [1 + \delta_\omega^{(1)}] W_\omega^{\text{rad}} + \nabla \cdot \mathbf{J}_\omega = \text{source}. \quad (4.88)$$

The matter has now been integrated out and a modified equation of continuity for the radiation remains.

In general an equation of continuity may serve as definition of a velocity, keeping in mind that a current is a density times a velocity. The *transport velocity* v_E can be defined as,

$$v_E = \frac{J_\omega}{W_\omega^{\text{tot}}} = \frac{1}{1 + \delta_\omega^{(1)}} \cdot \frac{J_\omega}{W_\omega^{\text{rad}}}. \quad (4.89)$$

Since $J_\omega/W_\omega^{\text{rad}} = c_0^2/v_p$,

$$v_E(\omega) = \frac{c_0^2}{v_p} \cdot \frac{1}{1 + \delta_\omega^{(1)}} = \frac{c_0^2}{v_p} \cdot \frac{1}{1 + n W_\omega^{\text{pot}}}. \quad (4.90)$$

This transport velocity is the one that determines energy transport. As such, it must be subject to Einstein causality and always be less than the velocity in vacuum. In Fig. 13 we show the transport velocity of light in a medium with resonant point scatterers (we used the same parameters as in

Fig. 9). In the previous chapter it was noticed that group and phase velocity do not satisfy causality and can therefore never be the correct velocity of light in the multiple scattering regime.

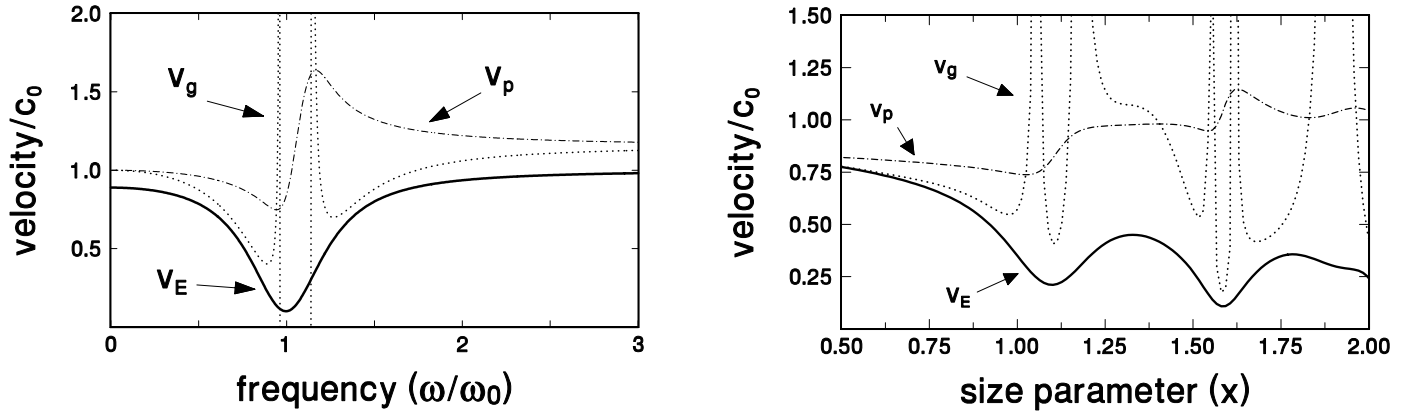


FIG. 13. Phase velocity v_p (dash-dotted line), group velocity v_g (dotted, for clarity the absolute value has been taken) and transport velocity v_E (solid line) of electromagnetic waves propagating through a collection of point scatterers having an internal resonance with frequency ω_0 as a function of frequency ω . The following parameters have been used: $\mathcal{S} = 0.25$ and the damping $T_2 = 12/\omega_0$ (polariton parameter $\mathcal{P} = 1.5$). All parameters are the same as in Fig. 9, in which the transport velocity was not presented.

FIG. 14. Phase velocity v_p (dash-dotted), group velocity v_g (dotted, as in the previous figure the absolute value has been taken), and transport velocity v_E (solid line) of vector (light) waves propagating through a collection of finite size scatterers at a volume fraction of $f = 0.2$ and index of refraction of the scatterers $m = 2.7$. On the horizontal axis the size parameter $x \equiv \omega a/c_0$. Note the large reduction of v_E from c_0 .

As can be seen from Fig. 9 the transport velocity is smooth, causal, and in fact very small near the resonance. In atomic systems the velocity can be orders of magnitude smaller than c_0 . For Mie scatterers we have calculated W_ω^{pot} already in section III.B.1. In Fig. 14 we show the transport velocity for Mie spheres with index of refraction $m = 2.73$ (Titanium-dioxide in vacuum). The velocity is seen to be small near every resonance. This has been confirmed by experiments [20].

One might pose the question in which special circumstance transport and phase velocity coincide, and in which special circumstance group and transport velocity coincide. The second question can be answered by inspection of Eq. (4.87). This equation contains three terms. Recalling Eqs. (4.21) and (4.30) the first term can be recognized as the contribution of the group velocity of the coherent wave. The second term \mathcal{R}_ω is more difficult to identify, but insertion of a point scatterer model (for which the t -matrix is independent of momentum) reveals that it is zero. It is therefore not likely that this term is going to be a crucial one. The third term is a collisional contribution which could only be found by considering the average intensity, and not only the coherent beam. It counts the delay a scattered wave will undergo. Apparently it largely compensates for the anomalous behavior of the group velocity. Group and transport velocity thus coincide in the absence of scattering. This is true in a homogeneous, dispersive dielectric medium.

The difference between transport and phase velocity is due to microscopic resonances. These are absent at low frequencies. It can easily be shown that at low frequencies (the Rayleigh regime) the Boltzmann approximation gives $v_E = v_p$, no matter how complicated the dielectric scatterer. Note that we need the collisional contribution - a rather technical element - to arrive at this elementary conclusion. Without this term we would have obtained $v_E = c_0^2/v_p$ for the Rayleigh regime. In fact it is also possible to find a model for which $v_E = c_0^2/v_p$. This happens when the dielectric constant is infinite as is true for perfect metallic scatterers. An erroneous conclusion that one always arrives

at $v_E = v_p$ for scalar waves was published by Barabanenkov and Ozrin [97]. The error resulted from the replacement of the spectral function $S(E, p)$ by a Dirac delta distribution [98]. We mentioned in section III.C.1 that this simplification - successfully used for electron-impurity scattering - strongly underestimates the role of the non-propagating material states.

Using the results of the previous chapter, very transparent expressions can be obtained for the transport velocity. In terms of the dwell time the transport velocity becomes,

$$v_E \approx \frac{c_0}{1 + \tau_d/\tau_{mf}} . \quad (4.91)$$

In our approach v_E emerges in a full transport theory, starting from the wave equations. As such it will appear in all dynamic transport properties like the diffusion constant, for any possible scatterer. Expression (4.91) looks simple and could have been obtained heuristically. We want to emphasize that such an heuristic reasoning would lead to an erroneous conclusion for De Broglie waves.

Eq. (4.91) applied to point scatterers coincides with expressions obtained by others for semi-classical oscillators and two-level atoms. Brillouin [21] and Loudon [23] derived an “energy velocity” by considering the energy balance. Whether the energy travels ballistically or diffuse is not addressed in their work. Holstein [16] found the transport velocity in a phenomenological way in the context of radiation trapping, where it figures in the diffusion constant of *incoherent* light.

b. Diffusion Approximation: $\mathbf{q} \rightarrow 0$, $\Omega \rightarrow 0$ We replaced earlier the Equation of Radiative Transfer by a diffusion equation. In the following we investigate how this equation will change in dynamic situations. The frequency Ω describes the macroscopic time dependence of a perturbation. As such it can be expected to be small compared to for instance the optical frequency ω . Thus the diffusion approximation will now apply when both Ω and \mathbf{q} are small. In response theory this is called the “Kubo limit”. Note that the range of the two limits is very important, and cannot be interchanged.

Given the results for $\Omega = 0$ and for v_E it is not difficult to infer that the diffusion approximation as defined already in Eq. (4.77) generalizes in dynamic situations to,

$$\begin{aligned} i\Omega [1 + \delta_\omega] W_\omega^{\text{rad}}(\Omega, \mathbf{q}) - i\mathbf{q} \cdot \mathbf{J}_\omega(\Omega, \mathbf{q}) &= \text{constant} , \\ -i\frac{\Omega}{c_0} [1 + \kappa_\omega] \mathbf{J}_\omega(\Omega, \mathbf{q}) + \frac{1}{3} \frac{c_0^2}{v_p} i\mathbf{q} W_\omega^{\text{rad}}(\Omega, \mathbf{q}) &= \frac{1}{\ell_{\text{tr}}} \mathbf{J}_\omega(\Omega, \mathbf{q}) . \end{aligned} \quad (4.92)$$

The quantity κ_ω that shows up here has a similar expression as δ_ω , except for an extra cosine weighting in the scattering term. In the diffusion approximation it is neglected, since it goes together with an Ω^2 -term in the final expression for the radiation density. Combination yields,

$$W_\omega^{\text{rad}}(\Omega, \mathbf{q}) = \frac{1}{-i\Omega/v_E + \frac{1}{3}\ell_{\text{tr}}\mathbf{q}^2} . \quad (4.93)$$

Translating to space and time variables the equation takes the form,

$$\left[\frac{1}{v_E} \partial_t - \frac{1}{3} \ell_{\text{tr}} \nabla^2 \right] W_\omega(\mathbf{r}, t) = \text{source} . \quad (4.94)$$

This is recognized as a diffusion equation. We identify the *diffusion constant*

$$D = \frac{1}{3} v_E \ell_{\text{tr}} . \quad (4.95)$$

This relation between mean free path and diffusion coefficient is very general and can be found, besides the field of radiative transport theory, in many other areas of statistical physics where diffusion occurs. For electromagnetic wave diffusion the velocity entering the diffusion constant of the radiation is the transport velocity, and not the group or phase velocity. We want to emphasize that velocity and mean free path show up separately in the diffusion equation, and not as the diffusion constant as a whole. In a stationary situation, when $\Omega = 0$, the transport velocity has no physical meaning.

Although derived for an infinite medium, the diffusion equation can be solved straightforwardly for any geometry once boundary conditions are added. As such both D and ℓ_{tr} are quantities that can be determined *independently* from the experiment. This makes the velocity v_E an observable quantity, albeit indirect.

How small Ω and \mathbf{q} have to be for the diffusion approximation to become valid, can easily be estimated. The relevant length scale is the (transport) mean free path ℓ_{tr} and the relevant time scale is the transport mean free time $\tau_{\text{tr}} = \ell_{\text{tr}}/v_E$. So the diffusion approximation is expected to apply when

$$q\ell_{\text{tr}} \ll 1 \quad ; \quad \Omega\tau_{\text{tr}} \ll 1. \quad (4.96)$$

In a *finite* system with thickness L the stationary $\Omega = 0$ theory applies when

$$\Omega < \frac{D(\omega)}{L^2}. \quad (4.97)$$

This is the regime of transient diffusion.

4.5. Einstein Relation For Light Conductance

The Equation of Radiative Transfer offers insight into the various direction channels that go in and come out. The DC light conductance can be calculated as the “all-channel-in all-channel-out transmission coefficient. We shall calculate it for a slab with length L , and surface A within the diffusion approximation discussed earlier [95]. A surface A gives rise to a number Ak^2/π of conducting channels. If this number is large enough all these channels can be considered to contribute independently to the transmittance.

To find the transmission coefficient of one channel we must deal with the boundary conditions. The boundary at which the incident field comes in must be subject to the condition that the flux coming in is $\mathbf{J}_\omega^{\text{in}}$, along the z -axis; The other boundary has the property that nothing is coming in. Within the diffusion approximation the specific intensity is given by Eq. (4.77). For the flux going to the left J_ω^+ and the flux going to the right J_ω^- we obtain,

$$\begin{aligned} J_\omega^+(z) &= 2\pi \frac{c_0^2}{v_p} \int_0^{+1} d\cos\theta I_\omega(\theta, z) \cos\theta = \frac{1}{4} \frac{c_0^2}{v_p} W_\omega^{\text{rad}}(z) + \frac{1}{2} J_\omega(z) , \\ J_\omega^-(z) &= 2\pi \frac{c_0^2}{v_p} \int_0^{-1} d\cos\theta I_\omega(\theta, z) \cos\theta = \frac{1}{4} \frac{c_0^2}{v_p} W_\omega^{\text{rad}}(z) - \frac{1}{2} J_\omega(z) . \end{aligned} \quad (4.98)$$

The total current along the z -axis is $J_\omega = J_\omega^+ - J_\omega^-$. By Eq. (4.79) it equals,

$$J_\omega(z) = - \left(\frac{1}{3} \frac{c_0^2}{v_p} \ell_{\text{tr}} \right) \partial_z W_\omega^{\text{rad}}(z) . \quad (4.99)$$

The boundary conditions become, in the diffusion approximation,

$$J_{\omega}^{+}(z=0) = J_{\omega}^{\text{in}} \quad ; \quad J_{\omega}^{-}(z=L) = 0 \quad , \quad (4.100)$$

and are called the radiative boundary conditions. The solution of the diffusion equation supplied by these boundary conditions is,

$$W_{\omega}^{\text{rad}}(z) = \frac{J_{\omega}^{\text{in}}}{v_p} \cdot 4 \frac{L + \frac{2}{3}\ell_{\text{tr}}}{L + \frac{4}{3}\ell_{\text{tr}}} \left(1 - \frac{z}{L + \frac{2}{3}\ell_{\text{tr}}} \right) . \quad (4.101)$$

Multiplying by the number of channels we obtain for the transmittance,

$$\begin{aligned} T(\omega) &\equiv \frac{Ak^2}{2\pi} \cdot \frac{J_{\omega}(z=L)}{J_{\omega}^{\text{in}}} = \frac{Ak^2}{2\pi} \cdot \frac{-1}{J_{\omega}^{\text{in}}} \left(\frac{1}{3} \frac{c_0^2}{v_p} \ell_{\text{tr}} \right) \partial_z W_{\omega}^{\text{rad}} \\ &= 2\pi W_{\text{rad}}(\omega) \cdot \frac{1}{3} \frac{c_0^2}{v_p} \ell_{\text{tr}} \cdot \frac{A}{L + \frac{4}{3}\ell_{\text{tr}}} . \end{aligned} \quad (4.102)$$

Here $W_{\text{rad}}(\omega)$ is the radiation density of states per unit volume given in Eq. (4.48). This formula can be *rigorously* derived from the Equation of Radiative Transfer (see the book of Van de Hulst [3]). The transmittance is inversely proportional to the length of the slab, and proportional to its surface. In solid state physics this result is known as Ohms law for the DC conductance. The DC conductivity for light at optical frequency ω can now be defined as for electrons (that is Eq. (1.7) with $e, \hbar = 1$),

$$2\pi\sigma_{\text{light}}(\omega) \equiv T(\omega) \frac{L}{A} . \quad (4.103)$$

We can rearrange Eq. (4.102) into a different form by multiplying numerator and denominator by the transport velocity v_E . In terms of the transport velocity, formula (4.48) for the total electromagnetic energy density can be rewritten as

$$W(\omega) = W_{\text{trans}}(1 + \delta_{\omega}) = \frac{c_0^2}{v_E v_p} W_{\text{rad}} , \quad (4.104)$$

so that the light conductivity takes the form,

$$\sigma_{\text{light}}(\omega) = W(\omega) D(\omega) . \quad (4.105)$$

For the DC conductivity of electrons this result would be called the Einstein relation. It is one of the most important results of electron transport theory and is here derived for light. The Einstein relation is apparently true for light only when the *total* energy density is used, and not only the one of the travelling waves.

The interesting feature of the Einstein relation for light is that the diffusion constant D and the energy density W are both dynamic transport parameters. On the other hand the conductivity is a stationary quantity. By virtue of the Einstein relation it is tempting to translate observations made on the DC conductivity into statements regarding the diffusion constant. Not only does this ignore the role of the density-of-states factor, it also suggests that the diffusion constant and all characteristics associated with it are DC properties of wave propagation. In a stationary experiment one can measure the mean free path, the conductance or transmission, but never the diffusion constant.

4.6. Restrictions of Boltzmann Theory

So far we considered transport of light using the scattering of one particle as a building block. We showed that this provides a mathematically consistent framework. If the scatterers are closely packed this may no longer be a good physical description of the random medium. There are a number of modifications of normal Boltzmann theory that one can think of when the particle density becomes large.

- Light scatters no longer from one particle, but rather from clusters of particles. For light, the scattering from clusters is referred to as *dependent scattering*. Dependent, because the scattering from an object depends on other the objects in its local environment. To describe this feature one has to be very careful not to “double count” the propagation of a wave from one particle to the other, and do the book keeping properly. In addition, one must include scattering from clusters without violating energy conservation. Without energy conservation the diffusion constant loses its meaning since even macroscopic disturbances will disappear exponentially in time. Dependent scattering can be severe when the particles scatter the light strongly, that is for resonant scattering [14].
- Concerning the averaging over all possible locations of the scattering objects one has to keep track of excluded volumes and correlations. This implies a deviation from Poissonian statistics of the location for the scattering centers. Realizations of the random system where two or more dielectric particles are put on top of each other are to be excluded.
- *Interference* of two different light paths in the random medium has been fully neglected in the Boltzmann approximation. The occurrence of interference is an important difference with radiation trapping [16].
- Scatterers see each other in their *near* radiation field. The far $1/r$ field described by the scattering cross-section will no longer suffice to describe multiple scattering.

The following hand waving argument may be applied to estimate the importance of recurrent scattering in the coherent beam. We will allow absorption by introducing explicitly the albedo a . A leading correction to Boltzmann theory is likely to be the recurrent scattering of a wave from a cluster of two particles. For point particles and scalar light its contribution to the self energy reads,

$$\begin{aligned}
 & \langle \mathbf{p}_1 | \Sigma(\omega, 1 - 2 - 1) | \mathbf{p}_2 \rangle \\
 &= n^2 \int d^3 \mathbf{r}_1 \int d^3 \mathbf{r}_2 t_1(\omega) G_0(\omega, \mathbf{r}_{12}) t_2(\omega) G_0(\omega, \mathbf{r}_{21}) t_1(\omega) e^{i \mathbf{r}_1 \cdot (\mathbf{p}_2 - \mathbf{p}_1)} \\
 &= i \frac{n^2 t(\omega)^3}{8\pi\omega/c_0} \times (2\pi)^3 \delta(\mathbf{p}_1 - \mathbf{p}_2).
 \end{aligned} \tag{4.106}$$

Comparing this to the independent-scattering approximation $nt(\omega) \times (2\pi)^3 \delta(\mathbf{p}_1 - \mathbf{p}_2)$ brings us to the conclusion that the modification is of order one when $n|t(\omega)|^2/8\pi \geq \omega/c_0$. We can identify $|t|^2/4\pi$ as the scattering cross section for scalar waves. It can be transformed into the extinction cross-section using the albedo a , which is at most unity in the presence of purely elastic scattering. We find that dependent scattering becomes dominant when,

$$k\ell_{\text{ext}} \leq 2a. \tag{4.107}$$

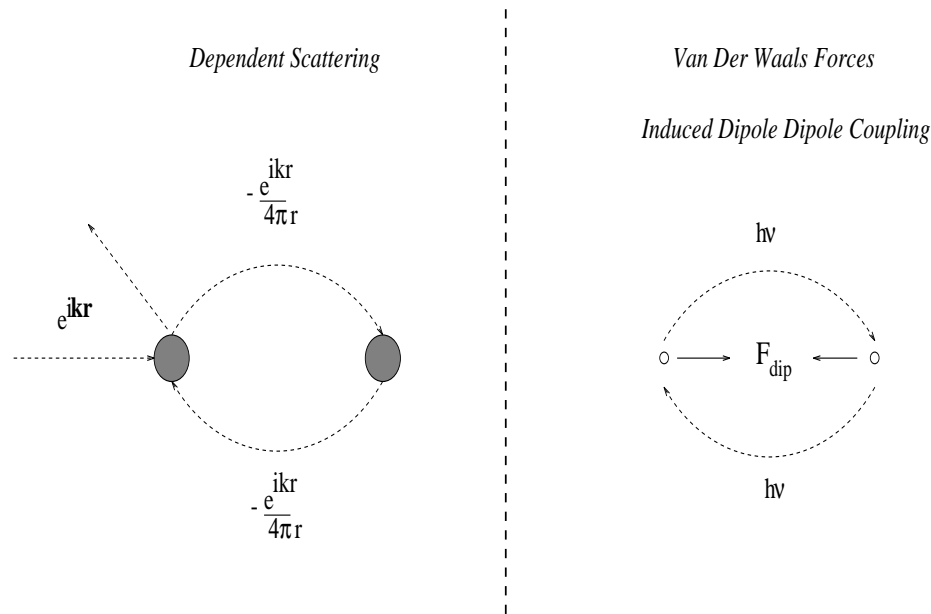


FIG. 15. The Van der Waals interaction and the Induced Dipole-Dipole interaction known in atomic physics compared to dependent scattering modifications in transport theory. For the first one has integrated out the light, for the second one has integrated out the matter.

The role of dependent scattering is thus determined by a criterion similar to the criterion (4.52) for the strongly scattering regime. However, the albedo factor makes sure that strong absorption or inelastic scattering suppresses recurrent scattering considerably.

Correlations and dependent scattering are often included by means of an “effective medium which is self-consistently obtained using a non-linear equation [18] [19]. Very often, a modified Boltzmann picture shows up and one can really speak of engineering with Boltzmann theory. We want to expose here some *genuine* new effects that show up when one goes beyond Boltzmann theory. These should not be seen as corrections to an old picture but really demand a new picture. We will show how dipole-dipole forces between two particles expose themselves as a dependent scattering phenomenon in a multiple light scattering theory. We discuss briefly Coherent Backscattering resulting from interference in multiple light scattering. We shall explain why the inclusion of interference is necessary to restore reciprocity. We point out how the Lorentz-Lorenz formula (4.8) naturally arises from the Dyson equation with pair correlation. Finally we briefly sketch the possible dramatic consequences of interference: Anderson localization.

4.6.1. Induced Dipole-Dipole Coupling & Dependent Scattering

It is known from atomic physics that Van-Der-Waals forces between two neutral polarizable atoms at distance r originate from vacuum fluctuations of the quantized electromagnetic field. A fluctuation of the electric field polarizes one atom, which starts radiating and polarize its neighbor. The radiation that the first atom receives back from the second can be put into a binding energy of the two atoms, which upon integration over the total fluctuation spectrum yields the $1/r^6$ (non-retarded) Van Der Waals force and $1/r^7$ (retarded) Casimir-Polder force [11] [12] [13] (see Fig. 15).

From this picture it appears that the Van Der Waals force is basically due to multiple light scattering from two point scatterers, since we pointed out earlier that a point scatterer can be

viewed as a simple atom. We expect that a lot of quantum physics of the Van Der Waals interaction is already contained in the classical solution for light scattering from two dielectric point scatterers separated by a distance r . To be more precisely, it must be contained in the material part of our solution $W_{\text{pot}}(\omega)$ of the two particle cluster.

The calculation of $W_{\text{pot}}(\omega)$ in (3.43) for one point particle in a monochromatic radiation field with frequency ω can be straightforwardly extended to two point particles separated by a distance r . The detailed derivation will not be given here. For the total potential energy relative to vacuum of a pair of point scatterers we obtain the expression

$$W_{\text{pot}}^{\text{pair}}(\omega, \mathbf{r}) = 2W_{\text{pot}}^0(\omega) - \frac{4\pi}{\alpha_{\text{sph}}(\omega/c_0)^5} \text{Im Tr}_{\text{pol}} \frac{t^3(\omega) \mathbf{G}^2(\omega, \mathbf{r})}{\mathbf{I} - t^2(\omega) \mathbf{G}^2(\omega, \mathbf{r})}. \quad (4.108)$$

The trace is over the three polarization vectors. The first term contains the contribution of two independent scatterers given in Eq. (3.43). The second term is the dipole-dipole binding energy. This term *without* the denominator (multiplied by the vacuum expectation value of the square of the electric field) is the one found in textbooks and describes one recurrent scattering of a “virtual” photon between the two particles. The denominator describes higher orders of recurrent scattering, but is important only at separations comparable to the wavelength. If \mathbf{r} is not large compared to the wavelength there will be three orthogonal polarization directions contributing to the trace and not two.

Let us make the link with transport theory. In section III.D.2 we demonstrated that the transport velocity of light in a random medium is determined by the total stored energy in the medium. The binding energy in this random medium due to induced dipole-dipole coupling can easily be found from Eq. (4.108) and equals

$$W_{\text{pot}}^{\text{dd}} = -n^2 \frac{6\pi}{\alpha_{\text{sph}}(\omega/c_0)^5} \text{Im} \int d^3\mathbf{r} \frac{t^3(\omega) \mathbf{G}^2(\omega, \mathbf{r})}{\mathbf{I} - t^2(\omega) \mathbf{G}^2(\omega, \mathbf{r})}. \quad (4.109)$$

Note that summing all orders of recurrent scattering automatically makes sure that this integral converges at small distances, and no new divergencies are encountered for two point particles. It can be estimated that the dipole-dipole energy becomes significant when $n(\omega_0/c_0)^3$ becomes of order unity.

The dipole-dipole binding energy must give rise to the first dependent-scattering modification of the transport velocity in multiple scattering. Although derived here in a different way, this conclusion can be ascertained by considering all recurrent scattering from two particles in the transport equation [99]. Incorporating cluster scattering from two particles in a transport theory for light thus includes the induced dipole-dipole forces between the particles. In the latter case the light has been integrated out.

4.6.2. Reciprocity & Coherent Backscattering

Contrary to what is frequently believed, the Equation of Radiative Transfer, and thus the Boltzmann approximation for multiple scattering, does not obey reciprocity. In transport theory reciprocity is often defined by interchanging the location of detector and source (both assumed dealing with the intensity, not the complex field), and saying that the same signal should be measured. Recalling the intensity Green’s function (4.59)

$$L(\mathbf{x}, \mathbf{y}) = \langle \langle G(\mathbf{x}, \mathbf{y}) G^*(\mathbf{x}, \mathbf{y}) \rangle \rangle .$$

Reciprocity between source and detector requires $L(\mathbf{x}, \mathbf{y}) = L(\mathbf{y}, \mathbf{x})$. However, this is necessary but not sufficient. In principle four positions enter this vertex,

$$L(\mathbf{x}, \mathbf{y}, \mathbf{x}', \mathbf{y}') = \langle \langle G(\mathbf{x}, \mathbf{y}) G^*(\mathbf{x}', \mathbf{y}') \rangle \rangle ,$$

and reciprocity of the underlying wave equation requires that,

$$L(\mathbf{x}, \mathbf{y}, \mathbf{x}', \mathbf{y}') = L(\mathbf{y}, \mathbf{x}, \mathbf{y}', \mathbf{x}') = L(\mathbf{y}, \mathbf{x}, \mathbf{x}', \mathbf{y}') . \quad (4.110)$$

In vector scattering L becomes a tensor of rank four with additional symmetry [44]. Expressed in Fourier and Laplace variables the operator L manifests itself as $\Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q})$ defined in Eq. (4.63). Reciprocity for this object amounts to [92],

$$\Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) = \Phi_{\omega, -\mathbf{p}', -\mathbf{p}}(\Omega, -\mathbf{q}) = \Phi_{\omega, \frac{\mathbf{p}-\mathbf{p}'+\mathbf{q}}{2}, \frac{\mathbf{p}'-\mathbf{p}+\mathbf{q}}{2}}(\Omega, \mathbf{p} + \mathbf{p}') . \quad (4.111)$$

In the first equality of (4.110) and (4.111) the wave and its complex conjugate have both been turned around, which corresponds to interchanging detector and source. Only this identity is satisfied by the Equation of Radiative Transfer.

The second equality is not obeyed by ordinary Boltzmann theory. It corresponds to only turning around the path propagated by the wave itself, but keeping the track fixed of its complex conjugate. Obviously we are now speaking about an interference contribution, which is only possible for waves, not for particles. In the Boltzmann approximation the wave and its complex conjugate travel the same path. Apparently, inclusion of interference is necessary to restore reciprocity.

Reciprocity can be repaired by including the interference of every path with its time-reversed counterpart, and so extend Boltzmann theory. The physical consequences of the very existence of reciprocity in the underlying wave equations are enormous. At *exact backscattering* from a random sample it can be easily be shown that the interference of two counter propagating is constructive (Fig. 16) and, by the second reciprocity equality, *exactly* equal to the value obtained from the Equation of Radiative Transfer. At backscattering, Boltzmann theory is thus a factor of two wrong [100].

This factor of two does not persist away from the exact backscattering direction, since the interference will dephase. What remains is a sharp peak at backscattering, called Coherent Backscattering. The angular width of this peak equals the wavelength divided by the transport mean free path :

$$\Delta\theta \approx \frac{\lambda}{2\pi\ell_{\text{tr}}} = \frac{1}{k\ell_{\text{tr}}} . \quad (4.112)$$

The Coherent Backscattering effect has been observed experimentally in light scattering [101] [102] [103] [104] [105] (Fig. 17). It is the first *direct* experimental evidence that Boltzmann theory and the associated Equation of Radiative Transfer are incomplete for light, and that interference persists in multiple scattering.

4.6.3. Excluded Volume & Lorentz Cavity

Atoms have a much smaller size than their scattering length. So contrary to random dielectric samples, condensed matter can contain hundreds of atoms per optical volume λ^3 . The role of dependent scattering may be expected to be enormous, but the role of pair correlation may be expected to be negligible. In what follows we will argue exactly the opposite.

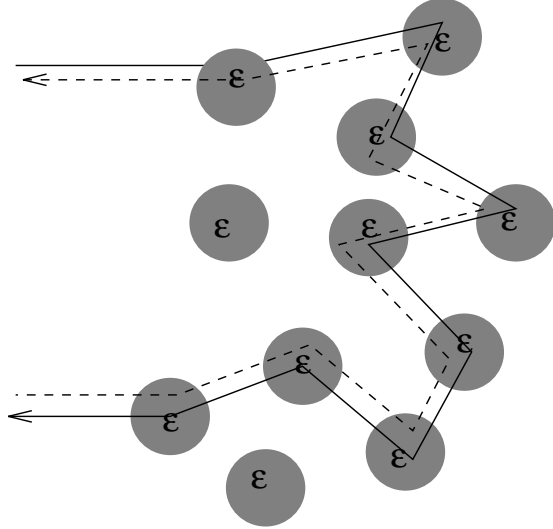


FIG. 16. The principle of Coherent Backscattering. It consists of the interference of two waves in opposite direction, along the same path. At backscattering the interference is always constructive, and by reciprocity arguments equal to the conventional background given by the theory of radiative transport. At angle $\theta > 1/k\ell$ the signal dephases rapidly.

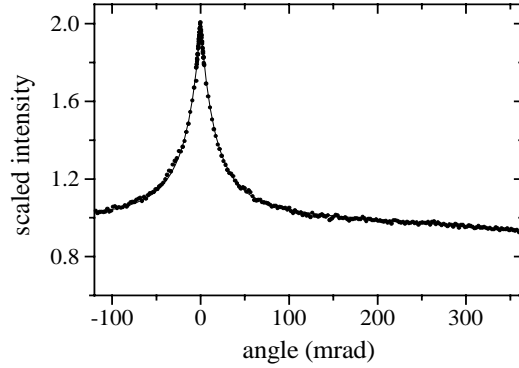


FIG. 17. A measurement of coherent backscattering from a random medium, done with circularly polarized light with vacuum wavelength $\lambda = 632.8$ nm. The backscattered intensity is plotted against the scattering angle where zero corresponds to exact backscattering. The sample is $BaSO_4$ with a transport mean free path of about $2.1 \mu\text{m}$ (Taken from Ref. [15]).

The Lorentz cavity was originally introduced by H.A. Lorentz in 1880 to give an atom an excluded volume [80]. Lorentz showed that propagation of light is influenced by the polarizability of the atoms. The presence of the atoms changes the effective medium dielectric constant which in turn determines the speed of light. The introduction of an *infinitesimally small* cavity around each atom does not give the “independent scattering result given in Eq. (4.21), but automatically yields the Lorentz-Lorenz formula (4.8). Apparently, an infinitesimally small pair correlation provides a severe modification in the dielectric constant.

Let us verify this statement in Dyson’s equation. To this end, we first find the Helmholtz Green’s function in real space by Fourier transformation ($k = \omega/c_0$),

$$\begin{aligned} \mathbf{G}_0(\omega, \mathbf{r}) &= \sum_{\mathbf{p}} \left[\frac{\hat{\mathbf{p}}\hat{\mathbf{p}}}{(\omega/c_0)^2} + \frac{\Delta_{\mathbf{p}}}{(\omega/c_0)^2 + i\epsilon - p^2} \right] e^{i\mathbf{p}\cdot\mathbf{r}} \\ &= -\frac{e^{ikr}}{4\pi r} [P(kr) \Delta_{\mathbf{r}} + Q(kr) \hat{\mathbf{r}}\hat{\mathbf{r}}] + \frac{\mathbf{I}}{3(\omega/c_0)^2} \delta(\mathbf{r}) . \end{aligned} \quad (4.113)$$

The delta-function is a very subtle feature. Its presence can best be verified by the observation $\mathbf{G}_0(\omega, \mathbf{p} = 0) = c_0^2/\omega^2$ must equal the space integral of $\mathbf{G}_0(\omega, \mathbf{r})$. Outside a point source it will never play a role and is for that reason often ignored and perhaps even forgotten. In that case

the functions P and Q determine the electric field vector perpendicular and along the axis $\hat{\mathbf{r}}$ of propagation,

$$\begin{aligned} P(y) &= 1 - \frac{1}{iy} - \frac{1}{y^2}, \\ Q(y) &= 2 \left(\frac{1}{iy} + \frac{1}{y^2} \right). \end{aligned} \quad (4.114)$$

Correlation in the Dyson self energy can be included by the notion that any contribution must be connected. The connected structure can either be due to recurrent scattering from one scatterer (dependent scattering) or to correlation between two or more particles (as may perhaps be familiar from linked cluster expansions in the statistical mechanics of the equation of state). Pair correlation between two particles 1 and 2 is described by a pair correlation function $h(\mathbf{r}_{12})$. In the simple case of exclude volume,

$$h(\mathbf{r}) = \begin{cases} -1 & r < a \\ 0 & r > a \end{cases}. \quad (4.115)$$

In the following we introduce a Lorentz cavity with size a around each atom and finally take the limit $a \rightarrow 0$. The pair correlation contribution of two identical point particles to the self energy is given by [70],

$$\langle \mathbf{p}_1 | \Sigma_{\text{corr}}^{(2)}(\omega) | \mathbf{p}_2 \rangle = n^2 \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_2 t(\omega) h(\mathbf{r}_{12}) \mathbf{G}_0(\omega, \mathbf{r}_{12}) t(\omega) \exp(-i\mathbf{p}_1 \cdot \mathbf{r}_1 + i\mathbf{p}_2 \cdot \mathbf{r}_2).$$

A factor $(2\pi)^3 \delta(\mathbf{p}_1 - \mathbf{p}_2)$ can be split off by translational symmetry. For a very small cavity only the delta function in the vacuum Helmholtz Green's function (4.113) contributes. This gives,

$$\Sigma_{\text{corr}}^{(2)}(\omega, \mathbf{p}) = -\frac{1}{3} \frac{n^2 t(\omega)^2}{(\omega/c_0)^2} = (\omega/c_0)^2 \left\{ -\frac{1}{3} n^2 \alpha(\omega)^2 \right\}.$$

We recall that the relation between polarizability $\alpha(\omega)$ and t -matrix is given in Eq. (3.28). Correlations between three and more particles can be acknowledged similarly: For three particles we require that the joint distribution vanishes if the three particles are in the same Lorentz cavity. In this way we find a geometric series. We obtain for the dielectric function,

$$\varepsilon(\omega, \mathbf{p}) \rightarrow 1 + n\alpha(\omega) + \frac{1}{3} n^2 \alpha(\omega)^2 + \left(\frac{1}{3}\right)^2 n^3 \alpha(\omega)^3 + \dots = 1 + \frac{n\alpha(\omega)}{1 - \frac{1}{3} n\alpha(\omega)}. \quad (4.116)$$

This is the Lorentz-Lorenz formula. It follows rigorously from multiple scattering theory when all dependent scattering is neglected and excluded volume is incorporated by means of the Lorentz cavity. It is a genuine vector effect, and is absent for scalar waves. We want to make two comments to this result.

Firstly, many textbooks³ introduce a macroscopic artificial Lorentz cavity containing many atoms. Next, they calculate how the electrostatic field inside is influenced by surface charges, thereby

³An exception is the book *Principles of Optics* by Born and Wolf [39], where an exact microscopic derivation can be found. This is heavy reading.

introducing a local field. This treatment is correct for $\omega = 0$, that is in electrostatics. It will never be able to find the Lorentz-Lorenz formula for propagating waves since one needs $\omega \neq 0$ to address travelling waves. As such the original microscopic argument of Lorentz is often destroyed by an oversimplified picture.

Secondly, one might pose the question when the neglect of dependent scattering can be justified. If the atoms scatter light inelastically ($a < 1$), as is often the case, recurrent scattering will be suppressed considerably as has been pointed in Eq. (4.107). In that case the Lorentz-Lorenz formula seems all-right. If on the other hand the objects scatter light purely elastically ($a = 1$), the Lorentz-Lorenz formula is nothing more than one among other effective medium approximations. This means that in the regime where the Lorentz-Lorenz formula gives results different from independent scattering calculations, various other corrections can be shown to be of the same order of magnitude [106].

4.6.4. Anderson Localization of Light

The phenomenon of Anderson localization was first introduced in 1958 by P.W. Anderson in his famous paper [4]. After this publication an explosion of literature appeared on the subject. The message was that the quantum nature of the electron may cause it to be localized in space when disorder is present. In that case diffusion and conductivity will vanish and the substance turns into an isolator [107].

It is an interesting question whether this phenomenon can be sought in a transport theory going beyond the Boltzmann approximation. Normal Boltzmann theory is known to give long range diffusion. We want to discuss localization briefly as a phenomenon that nicely illustrates our preceding discussions about the difference between amplitude and intensity. In addition, we want to rephrase the ideas behind Anderson localization in such a form that it can be *unambiguously* applied to RMS of light.

Localization was originally defined as the vanishing of the DC conductivity, because that was the quantity measured in the laboratory [31]. Let us therefore recall that the “light conductance” was defined in section IV.E as the all-channel transmittance $T(\omega)$. For a slab with thickness L and surface A it is obtained from,

$$T(\omega) \frac{L}{A} = W(\omega) \times D(\omega) \sim W(\omega) \times v_E(\omega) \times \ell_{tr}(\omega) . \quad (4.117)$$

It is the product of the density of states and the diffusion constant. The diffusion coefficient factorizes in turn into a transport velocity and a transport mean free path. All four quantities, T , v_E , W , and ℓ_{tr} can in principle be measured but for light $T(\omega)$ is the most difficult one to measure (for electrons the simplest because they are located on the Fermi surface so that all channels are automatically occupied). Since any of these factors can be “small” this raises the question which of them contains the real localization.

We have seen that in RMS of light the velocity can be very small. However, in section III.E we noticed that the suppression of the transport velocity is compensated by the large material component of the density of states. With this cancellation included, the DC conductance takes the form,

$$T(\omega) \frac{L}{A} \sim W_{rad}(\omega) \times \frac{1}{v_p} \ell_{tr}(\omega) \sim \left(\frac{W_{rad}(\omega)}{W_0(\omega)} \right)^2 \times \ell_{tr} . \quad (4.118)$$

This formula shows explicitly that the suppression of the transport velocity should not be categorized as localization, although it suppresses the diffusion constant. Indeed, small diffusion constants reported in optical experiments could be attributed to small velocities and not to localization [20] [108]. It further leaves us with two possible mechanisms for light localization: a vanishing of the transport mean free path, and a vanishing of the radiation density. The difference between both cases becomes very clear by realizing that the transport mean free path is a property of the average *intensity*. The density of states of the travelling waves, the radiation density, concerns the average *amplitude*. The amplitude is only subject to *superposition*, but in the intensity the wave and its complex conjugate are allowed to *interfere*. Theoretically it turns out that in disordered media only interference is capable of generating a genuine vanishing of Eq. (4.118) in an infinite medium:

$$\ell_{\text{tr}}(\omega) = 0 . \quad (4.119)$$

A transport theory of localization was developed by W. Götze [109], P. Wölffe and D. Vollhardt [92] [110] at the end of the seventies. It emphasizes the role of interference in transport phenomena, and puts forward the weak localization effect discussed in section IV.F.2 as a possible basic mechanism. The criterion for localization found in this theory for an infinite random medium (in three dimensions with purely elastic isotropic scattering) turns out to be,

$$\ell_{\text{tr}} = 0 \quad \text{when} \quad k\ell_{\text{scat}} \preceq 1 . \quad (4.120)$$

The second mechanism to obtain a small transmission remains nevertheless interesting to pay attention to. If we recall the results of section III.C.1,

$$\frac{W_{\text{rad}}}{W_0} \ll 1 \quad \text{when} \quad k\ell_{\text{ext}} \preceq \frac{1}{2} \quad (4.121)$$

This criterion is remarkably close to the one of localization. It is for this reason that both criteria are frequently confused, and that criterion (4.52) is frequently misused as a criterion for localization. The difference becomes immediately clear by applying both criteria to a system with absorption. In that case one can still satisfy $k\ell_{\text{ext}} \preceq \frac{1}{2}$, but no longer the localization criterion (4.119).

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