Speed of light in random media

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We give a microscopic derivation of the speed of light relevant for energy transport in media containing randomly distributed scatterers. A comparison is made with the concept of "mass-enhancement" in electron transport theory. The consequences for the Thouless criterion for strong localization are discussed. Finally, quantitative results are obtained for simple scatterers, such as semiclassical oscillators and dielectric spheres. We will also introduce some heuristic approaches and discuss the validity.

I. INTRODUCTION

Recently it was realized that Anderson localization is a phenomenon that can be directly traced down to interference in multiple scattering and should therefore not be limited to the concept of electron propagation, but could apply to any kind of wave. Since then, the field of light localization has become a very active area of research, one reason being that the study of light has the additional advantage that the photons do not mutually interact. The absence of interaction excludes the complication of a Mott transition, caused by correlation.[1] The first observations of weak localization (enhanced backscattering),[2–4] the often assumed precursor of strong localization (vanishing of diffusion), stimulated further research in the hope of ultimately finding the mobility edge.

The similarity between the classical wave equations and the Schrödinger equation substantially simplified the treatment of multiple scattering of classical waves. Many concepts of electron transport theory have been taken over in order to describe the propagation of light in random media, with the usual approach that the Fermi wavelength is replaced by the wavelength of light. The best examples are the formulation of criteria for strong localization of light such as the *Ioffe-Regel criterion*[5] and the *Thouless criterion*.[6] Another crucial example is the application of conservation laws, expressed by means of so-called *Ward identities*. Such identities were developed for electron-impurity scattering and were then simply used to describe the scattering of light from randomly distributed dielectric particles.

Naturally there are some fundamental differences between light waves and electrons. The difference in dispersion laws, parabolic for electrons but linear in the case of light, turns out to be rather insignificant. Another difference is the low-energy behavior of the scattering cross section. In contrast with electrons, light exhibits the well-known E^4 Rayleigh behavior so that localization at low energies is impossible. In general, one expects that the observation of localization is more difficult for classical waves than for electrons. It is anticipated that localization is easiest once the density of states (DOS) is lowest. In fact, this is the primary message of the celebrated Thouless criterion. A suggestion made by John[7] to consider light localization near band edges of optical crystals seems very promising.

The most important difference between the Schrödinger equation and the classical wave equations is the presence of a second-order time derivative in the latter. We will emphasize in this paper that this property has drastic consequences for the microscopic formulation of conservation laws. A second-order derivative with respect to time reflects the presence of more than one scattering channel. This can be illustrated more expliclitly by writing Maxwell's equations as a first-order time-evolution problem with electric and magnetic field taken as scattering channels. It turns out that the classical scattering of light has more resemblance to electron scattering from a two-level system than to ordinary ("single-level") potential scattering.

Although the concept of Anderson localization applies to various kinds of disorder, most experiments deal with topological disorder. To optimize the amount of scattering in such situations and so to minimize the elastic mean free path, it seems beneficial to tune to resonances in the scattering cross section, in combination with a large density of the scatterers. Following this argument, small

diffusion constants[8] were reported and were attributed to small mean free paths and thus to nearness of strong localization. They were also found in our experiments, but an accurate comparison between time-resolved and steady-state measurements[9] demonstrated that the low diffusion constants were caused by an extremely small speed of light. This was recently verified by dynamical speckle experiments.[10] In Ref. 9 we presented a brief treatment of the transport speed of light v_E, entering the diffusion constant via the familiar relation $D = v_E \ell/3$. This approach confirmed the observed smallness of the transport velocity. We demonstrated that no localization concepts were necessary in order to find consistency between different experiments, and a pure Boltzmann approximation was sufficient.

It is the aim of this paper to give a detailed derivation of this so-called transport velocity and to put this quantity in an appropriate physical context. We emphasize that we are dealing with a well-documented quantity, but so far it has only been treated phenomenologically.[11–13] In addition, nobody has ever indicated how to obtain this transport velocity from an experiment. We demonstrate for the first time that this velocity appears as a dynamic vertex correction in the Boltzmann transport equation, and, consequently, in the diffusion constant of light. This last property makes the transport velocity a quantity that is easily inferred from a multiple-scattering experiment.

The existence of a transport velocity different from the phase velocity has serious consequences for the Thouless criterion. In the usual Thouless criterion, the diffusion constant, and thus the transport speed, is an essential ingredient. We point out that, although the diffusion of light is very much affected by dynamic vertex corrections, this would be unacceptable for the Thouless criterion. We will argue the absence of *any* velocity in the Thouless criterion: just like the Ioffe-Regel criterion, any criterion for strong localization should compare *length scales*, not *time scales*. This is consistent with the fact that a decrease of the transport speed by an order of magnitude does not signify the onset of localization. It merely renormalizes the time for a (localized) wave packet to leak away. It turns out that the basic ideas behind the original Thouless criterion can be formulated in terms of length scales. Such a revision brings about the absence of any speed in the prediction of the location of the mobility edge. For electron-impurity scattering, a situation that the Thouless criterion was originally developed for, the modified Thouless criterion agrees with the original one.

II. THE BOLTZMANN EQUATION

In this section we give a microscopic derivation of the transport speed for light waves. Since the basic physics is already contained in the scalar approximation of light, and polarization only causes extra bookkeeping, we consider the scalar wave equation

$$\left(\varepsilon(\mathbf{x})\frac{\partial^2}{\partial t^2} - \nabla\right)\Psi(\mathbf{x},t) = 0\,, \tag{1}$$

in which $\varepsilon = 1/c^2$ is the dielectric constant. The formal solution of the Dyson equation for the configurationally averaged Green function can be written as

$$G(E,p) = \frac{1}{E^2/c_0^2 - p^2 - \Sigma(E,p)}\,, \tag{2}$$

where we have introduced the mass operator $\Sigma(E,p)$. An analogous equation, the Bethe-Salpeter equation, can be written down for the intensity Green function, and defines the irreducible vertex U.[14] By a proper combination of both equations, the scalar correlation function $\langle\Psi(\mathbf{x}_1,t_1)\Psi^*(\mathbf{x}_2,t_2)\rangle$, averaged over disorder, can be shown to satisfy the following generalized Boltzmann equation[15]

$$\left(\frac{-2E\omega}{c_0^2} + 2\mathbf{q}\cdot\mathbf{p} + \Sigma(E^+,\mathbf{p}^+) - \Sigma(E^-,\mathbf{p}^-)\right)\Phi_{\mathbf{p}}(\mathbf{q},\omega|E) = \Delta G(\mathbf{q},\omega|E,\mathbf{p})\left(1 + \sum_{\mathbf{p}'} U_{\mathbf{p}\mathbf{p}'}(\mathbf{q},\omega|E)\Phi_{\mathbf{p}'}(\mathbf{q},\omega|E)\right). \tag{3}$$

Here $\Phi_{\mathbf{p}}$ represents $\langle\Psi(\mathbf{x}_1,t_1)\Psi^*(\mathbf{x}_2,t_2)\rangle$ Laplace transformed with respect to time and Fourier transformed with respect to position,

$$\sum_{\mathbf{p}}\int_{-\infty}^{\infty}\frac{dE}{2\pi}\Phi_{\mathbf{p}}(\mathbf{q},\omega|E) \equiv \int d\mathbf{x}\, e^{-i\mathbf{q}\cdot\mathbf{x}}\int_0^{\infty} dt\, e^{i(\omega+i\epsilon)t}\,\langle\Psi(\mathbf{x}_0+\mathbf{x},t_0+t)\Psi^*(\mathbf{x}_0,t_0)\rangle\,. \tag{4}$$

The parameters E and $\mathbf{p}$ are the internal oscillations of the wave packet in time and space, $\Delta G = G(E^+,\mathbf{p}^+) - G(E^-,\mathbf{p}^-)$ is the difference between retarded and advanced Green function, and $E^{\pm} \equiv E \pm i\epsilon \pm \frac{1}{2}\omega$, $\mathbf{p}^{\pm} \equiv \mathbf{p} \pm \frac{1}{2}\mathbf{q}$. The empty-space speed of light is c_0. The sum $\Sigma_{\mathbf{p}}$ stands for the integral $\int d\mathbf{p}/(2\pi)^3$. For the calculation of the diffusion coefficient one requires the solution in the Kubo limit $\mathbf{q},\omega \to 0$. Within the context of light propagation this limit has also been referred to as the "slowly-varying envelope approximation" (SVEA).[16] From now on we shall not explicitly refer to the variables $\mathbf{q}$ and ω, and we will write $\Phi_{\mathbf{p}}(E)$ instead of $\Phi_{\mathbf{p}}(\mathbf{q},\omega|E)$. A careful expansion of the mass operator and the irreducible vertex into first order of ω has to be employed. For simplicity we restrict ourselves to lowest order of the density. With n the density of the scatterers, and $t_{\mathbf{p}\mathbf{p}'}(E)$ the general T matrix, this lowest order in density is

$$\Sigma^{(1)}(E^{\pm},p^{\pm}) = n\,t_{\mathbf{p}(\pm)\mathbf{p}(\pm)}(E^{\pm}) = n\,t_{\mathbf{pp}}(E\pm i\epsilon) \pm \omega n \frac{\partial t_{\mathbf{pp}}(E\pm i\epsilon)}{\partial E} + \cdots ,$$

$$U^{(1)}_{\mathbf{pp}'}(\mathbf{q},\omega|E) = n\,t_{\mathbf{p}(+)\mathbf{p}'(-)}(E^{+})\,t_{\mathbf{p}'(+)\mathbf{p}(-)}(E^{-}) = n\,|t_{\mathbf{pp}'}(E)|^2\left(1+i\omega\frac{\partial\phi_{\mathbf{pp}'}(E)}{\partial E}\right)+\cdots . \tag{5}$$

Here, ϕ denotes the phase shift according to $t_{\mathbf{pp}'} \equiv |t_{\mathbf{pp}'}|\exp(i\phi_{\mathbf{pp}'})$. A similar expansion, involving partial derivatives with respect to p, can be employed in the parameter $\mathbf{q}$. In the low-disorder approximation under consideration, the solution of the Boltzmann equation becomes "on-shell," since it follows from Eq. (3) that $\Phi_{\mathbf{p}}(E) \sim \Delta G$. For $\omega, \mathbf{q} = 0$ and small density,

$$\Delta G(E,\mathbf{p}) = \frac{2i\,\mathrm{Im}\Sigma(E,\mathbf{p})}{(E^2/v_p^2 - p^2)^2 + [\mathrm{Im}\Sigma(E,\mathbf{p})]^2} \rightarrow -2\pi i\Theta(E)\,\delta\left(E^2/v_p^2 - p^2\right) . \tag{6}$$

Here $\Theta(E)$ is the Heaviside function; v_p is the phase velocity, following from Eq. (2),[17]

$$\frac{c_0}{v_p} = \sqrt{1 - \frac{n\,\mathrm{Re}\,t_{\mathbf{pp}}(p)}{p^2}} . \tag{7}$$

The energy flux at energy $E = p\,v_p$ is defined in terms of the yet unspecified transport velocity v_E according to $\mathbf{J}(E) \equiv v_E E \sum_{\mathbf{p}} \hat{\mathbf{p}}\Phi_{\mathbf{p}}(E)$. As a result of the linear dispersion law for light, this expression differs from the (probability) flux in the case of electrons, where it is given by $\mathbf{J}(E) = \sum_{\mathbf{p}}(\mathbf{p}/m)\,\Phi_{\mathbf{p}}(E)$, with m the electron mass.[18] The energy density is identified as $\Xi(E) \equiv E\sum_{\mathbf{p}}\Phi_{\mathbf{p}}(E)$. If we sum both sides of Eq. (3) over $\mathbf{p}$, and apply the optical theorem as well as Eq. (5), we find, in the limit of ω, $\mathbf{q} \rightarrow 0$,

$$\frac{-i\omega}{c_0^2}\Xi(E)\left(1 - n\frac{d}{dp^2}\,\mathrm{Re}\,t_{\mathbf{pp}}(p) + n\int d\Omega \frac{d\sigma}{d\Omega}\frac{d\phi(\Omega)}{dp}\right) + \frac{\mathbf{q}\cdot\mathbf{J}(E)}{v_p\,v_E} = K(E) . \tag{8}$$

We write $d\sigma/d\Omega = |t_{\mathbf{pp}'}|^2/(4\pi)^2$ for the differential cross section in the direction Ω. Obviously, Eq. (8) takes the required form of an equation of continuity (in reciprocal space) only if we let

$$v_E = \frac{c_0^2}{v_p}\left(1 - n\frac{d}{dp^2}\,\mathrm{Re}\,t_{\mathbf{pp}}(p) + n\int d\Omega\frac{d\sigma}{d\Omega}\frac{d\phi(\Omega)}{dp}\right)^{-1} . \tag{9}$$

The partial derivatives with respect to E in Eq. (5) are replaced by total derivatives. The fact that the transport velocity is expressed in terms of total derivatives of the on-shell T matrix is a very convenient property. Without the second term in the denominator of Eq. (9), the transport velocity is equal to the *group velocity* defined by $v_g \equiv dE(p)/dp$, in which the dispersion relation of the random medium, $E^2/c_0^2 = p^2 - \Sigma(E,p)$, must be inserted. The group velocity is known to lose its meaning near scattering resonances,[11,13] and may even become negative. Textbooks usually get around this problem by stating that the resonantly enhanced extinction makes a discussion of transport properties irrelevant. Inspection of our formula (9) demonstrates that the true energy velocity takes into account the energy carried by the *elastically* scattered wave. We will show that this collision contribution largely compensates for the anomalous behavior of the group velocity. By means of this term, the considerable delay of the scattered wave near a resonance will cause a large decrease of the transport velocity with respect to the phase velocity, sometimes by an order of magnitude. The group velocity in a random medium has a physical significance only if the attenuated coherent wave is measured.[19] Often a group velocity enters into the diffusion constant,[20,21] but then it does not contain information on impurities and is entirely the result of an effective mass approximation.

By taking the second moment of the transport equation,[18] it is straightforward to prove that the speed appearing in the equation of continuity must also enter the diffusion constant by means of $D = v_E\ell/3$. In fact, this relation can serve to define the transport mean free path ℓ once the diffusion constant and the transport speed have been obtained.[22]

We will now compare our new findings to results obtained for ordinary electron-impurity scattering. It is well known that dynamic vertex corrections do not persist in that case. A rigorous identity,

$$\Sigma(E^{+},\mathbf{p}^{+}) - \Sigma(E^{-},\mathbf{p}^{-}) = \sum_{\mathbf{p}'}\Delta G(\mathbf{q},\omega|E,\mathbf{p})\,U_{\mathbf{pp}'}(\mathbf{q},\omega|E) , \tag{10}$$

usually referred to as the *Ward-Takahashi identity*,[18,23,24] cancels all these corrections, in particular the second and third term in Eq. (9), in the equation of continuity (the prefactor c_0^2/v_p enters as a result of the linear dispersion law for light, and is not present for electrons either). One very fundamental consequence is the absence of the so-called mass-enhancement factor in some crucial transport coefficients such as the dc conductivity.[25] A close examination of the proof of the Ward identity[24] demonstrates its validity for noninteracting, local, and energy-independent potentials. Indeed, it is known that mass-enhancement factors do enter the equation of con-

tinuity once many-particle interactions become important. Examples are Fermi liquids,[1,26] as well as moderately dense gases,[27,28] where these interaction corrections succesfully produce the second virial constant.

Inspection of the scalar wave equation Eq. (1), illustrates that in view of the Schrödinger equation, after insertion of modes $\Psi(\mathbf{x},t) = \tilde{\Psi}(\mathbf{x})\exp(iEt)$, a local but *energy-dependent* "potential" can be identified as $V(\mathbf{x},E) = -[\varepsilon(\mathbf{x})-1]E^2$, so that $V(\mathbf{x},E^+) \neq V(\mathbf{x},E^-)$. An equality here is essential for the Ward identity in Eq. (10) to be valid. This is true only if $\omega = 0$, i.e., as long as *stationary* properties are discussed.

The violation of Eq. (10) for $\omega \neq 0$ has a much deeper origin than indicated so far. The validity for $\omega = 0$ expresses the presence of a conserved quantity, so that it can be considered as a generalized optical theorem. Recalling Eqs. (8) and (4) this conserved quantity is $|\Psi(\mathbf{x},t)|^2$. This is definitely a conserved quantity for (Schrödinger) potential scattering, for which Eq. (3) was developed originally, with the usual probability interpretation. For scalar waves, however, the conserved quantity, associated with energy density, is[29]

$$\mathcal{H} = \tfrac{1}{2}(\partial_t\Psi)^2 + \tfrac{1}{2}c^2(\mathbf{x})(\partial_{\mathbf{x}}\Psi)^2 , \qquad (11)$$

and not $|\Psi|^2$. In the SVEA, both in time and space, and after cycle averaging, it is readily verified that $\mathcal{H}$ coincides with $|\Psi|^2$. This explains why a knowledge of the behavior of the physical quantity $|\Psi(\mathbf{x},t)|^2$ is sufficient for the diffusive, dynamic description of scalar wave propagation, which only needs the solution of the transport equation in the limit where this quantity coincides with the energy density. Nevertheless, the dynamics of $|\Psi|^2$ for scalar waves can be quite different from the one for electrons. This is expressed by the failure of the Ward identity in Eq. (10) for $\omega \neq 0$.

It can be checked that a SVEA in time alone proves $|\Psi|^2$ to be conserved.[30] This is consistent with the validity of the Ward-Takahashi identity for $\omega = 0$, $\mathbf{q} \neq 0$. However, $|\Psi|^2$ then no longer coincides with the energy density. The $\mathbf{q} \neq 0$ Ward identity is violated only when nonlocal potentials are considered. The expansion in the parameter $\mathbf{q}$ in Eq. (3) is not carried out explicitly, since the validity of Eq. (10) for $\mathbf{q} \neq 0$ guarantees all vertex corrections proportional to $\mathbf{q}$ to cancel in the equation of continuity.

A rigorous transport equation should be obtained by considering Maxwell's equations,[31] or the scalar wave equation, as a first-order time-evolution problem. All degrees of freedom (here electric and magnetic field) have to be taken into account, although for the calculation of the dc diffusion constant it suffices to consider Eq. (3). In this respect, (resonant) dielectric scattering of light closely resembles the scattering of a photon from a two-level system or harmonic oscillator. The delay of the scattered wave, expressed by the decrease of the transport speed of light, can be compared to the inverse Einstein spontaneous emission coefficient. We will show in the last section that the energy, temporarily stored in the resonantly excited electron in the Schrödinger case, corresponds to the formation of a standing wave in the dielectric scatterer that leaks away. Away from resonances, such standing waves are absent, and all velocities, including the group velocity, coincide. In the case of potential scattering, there is a "real time" conversion of kinetic and potential energy, and no energy is being stored.

III. THE THOULESS CRITERION FOR LIGHT

Knowledge of the conditions under which strong localization sets in is of extreme importance even if it were only for its experimental significance. One seeks a general, but nevertheless very practical, criterion, hopefully derived from first principles. The Ioffe-Regel (IR) criterion, setting the wavelength equal to the mean free path, $\lambda \approx \ell$, later devised by Mott to $\lambda/2\pi \approx \ell$, was the first attempt to predict the location of the mobility edge in three dimensions. The IR criterion predicts the localization of low-energy electrons and can serve to estimate the desired density of scatterers near scattering resonances.[32,33] Diagrammatic theories[18] as well as nonlinear σ models[34] gave this criterion a microscopic foundation and can be used to generalize the IR criterion to for instance other dimensionalities, or anomalous spectral behavior,[35,21] but a first-principles interpretation is still lacking.

The Thouless criterion, developed in close connection with the scaling theory of localization,[36] can be considered as one of the most important breakthroughs in the description of strong localization. Not only does this theory provide clear and verifiable predictions concerning the appearance of localization in finite media of arbitrary dimension and the value of critical exponents, but it also introduces a fundamental parameter known as the dimensionless conductance: $g = \sigma L^{d-2}$. Here σ is the dc electrical conductivity, and L is a typical size of the d-dimensional random medium. A simple analysis, with application of the Einstein relation connecting diffusion constant, density of states and conductivity, demonstrates the equivalence of this parameter to the Thouless-parameter, defined by

$$\delta(L) = \frac{\Delta E(L)}{\delta E(L)} , \qquad (12)$$

with $\Delta E(L) \sim D/L^2$ the uncertainty in energy due to the finite traversal time of the (diffusive) transport in the medium, and $\delta E(L)$ the average level spacing. It has even been argued that the Thouless parameter $\delta(L)$ is more fundamental than the conductance g, since the latter becomes self-averaging in the presence of absorption.[37] The criterion $\delta(L) = \delta_c \approx 1$ is known as the *Thouless criterion* for localization.

The Thouless parameter in Eq. (12) is proportional to the diffusion constant and, hence, to the transport speed. The enormous decrease of this speed near scattering resonances thus lowers this parameter by an order of magnitude. A straightforward application of the Thouless criterion would locate the mobility edge at much smaller disorders than expected, for instance, on the basis of the Ioffe-Regel criterion. This is physically unacceptable, since the decrease of the transport speed of light

is a renormalization of time scales and does not enhance the correlation of the scattering medium. It merely takes longer to accomplish a certain correlation length, which is determined by the transport mean free path, and not by the diffusion constant as a whole.

To incorporate these ideas we propose a Thouless parameter in terms of *length scales* only:

$$\hat{\delta}(L) = \frac{\Delta p(L)}{\delta p(L)} . \tag{13}$$

Here $\Delta p(L)$ is the uncertainty in momentum and is determined by the path length distribution between two points in real space, a distance L apart. The modified Thouless criterium becomes $\hat{\delta}(L) = \delta_c \approx 1$. We will show that the use of this modified criterion is in agreement with previous work and introduces neither the transport nor the group velocity in a localization criterion.

Before we evaluate Eq. (13) for a specific situation we stress the equivalence of the Thouless parameter $\hat{\delta}(L)$ to the original Thouless parameter $\delta(L)$, if electron-impurity scattering is considered. Since dynamic vertex corrections are absent in the diffusion constant as well as in the density of states per energy interval dE, the velocities entering nominator and denominator of Eq. (12) quantities coincide, and equal the Fermi velocity evaluated at the Fermi surface, without mass-enhancement corrections. Thus Eq. (12) is equivalent to Eq. (13). It seems to us that a Thouless parameter formulated in terms of length scales, as indicated by Eq. (13), is a natural consequence of the scaling theory of localization, notwithstanding the fact that there is a one-to-one correspondence of time and length scales in situations for which the Thouless criterion was developed originally. A dynamical scaling theory was recently presented,[38] but did not yet incorporate our dynamic vertex corrections.

We will now evaluate the revised Thouless criterion (13) for classical waves. For a diffusion process with step length ℓ the path length distribution is given by[4,39,40]

$$P(s) = \frac{3}{(4\pi s\ell)^{3/2}} \exp\left(-\frac{3L^2}{4s\ell}\right) , \tag{14}$$

and has a maximum at $s_m = L^2/2\ell$. The uncertainty in momemtum is thus estimated to be $\Delta p(L) \approx 1/s_m = 2\ell/L^2$. In the presence of localization the transport mean free path ℓ may become scale dependent. The level spacing between momentum states is defined as $\delta p(L) = dp/dN$, in which dN denotes the total number of states with momentum between p and $p+dp$. Since the scalar spectral function $S(E,\mathbf{p})$, given by the imaginary part of the configurationally averaged Green function,[41]

$$S(E,\mathbf{p}) = -\frac{2E}{c_0} \operatorname{Im} G(E,\mathbf{p}) , \tag{15}$$

describes the density of states per unit volume with momentum $\mathbf{p}$ and energy E, it follows that

$$\frac{dn}{dp} \equiv 2 \sum_{\mathbf{p}'} \int_{-\infty}^{\infty} \frac{dE}{2\pi c_0} \delta(p-p') S(E,\mathbf{p}') . \tag{16}$$

The prefactor 2 comes from the spin degeneracy, specific for light. In case of a three-dimensional medium randomly filled with (nonabsorbing) dielectric scatterers, it can be shown (Appendix A), using a sum rule, that

$$\frac{dn}{dp} = \frac{p^2}{\pi^2} \frac{\langle c^2 \rangle}{c_0^2} . \tag{17}$$

Here $\langle c^2 \rangle$ denotes the average-medium value of the square of the speed of light. Equation (17) demonstrates that, apart from the topological factor $\langle c^2 \rangle / c_0^2$, the DOS per momentum interval does not depend on disorder.

For a volume L^3 we find explicitly that $\hat{\delta}(L) = (2/\pi^2)\ell(L)Lp^2\langle c^2 \rangle / c_0^2$, so that indeed group and transport velocity are absent. The absence of these velocities in the prediction of the location of the mobility edge is in agreement with the self-consistent theory of Vollhardt and Wölfle,[18] which has, at least in three dimensions, a stationary ($\omega = 0$) formulation. Their final result can be expressed in terms of a correlation length ξ. The transport mean free path ℓ, the length scale in the diffusion coefficient via $D = v_E \ell/3$, is given by $\ell = \ell_{sc}^2/\xi$, with ℓ_{sc} the scattering mean free path. In the absence of absorption,

$$\frac{1}{\xi} = \frac{1}{\xi_o} + \frac{1}{L} , \tag{18}$$

where ξ_o is the correlation length of the infinite system, $\xi_o = \ell_{sc}^2/(\ell_{sc} - \ell_c)$. The mobility edge is reached once the system is completely correlated, $\xi = L$, so that $\ell = \ell_{sc}^2/L$. Again this is a comparison of length scales. The modified Thouless criterion becomes $\hat{\delta}_c = (2/\pi^2)(p\ell_{sc})_c^2 \langle c^2/c_0^2 \rangle \approx 1$, in agreement with Ref. 42. In a random medium containing perfect metallic scatterers with packing fraction f, the topological factor in Eq. (17) lowers the DOS by a factor $1-f$ so that localization is predicted to become easier.

IV. NUMERICAL EVALUATION

The speed derived in Eq. (9) can be worked out for some special cases. We notice that this expression, although derived for scalar waves, allows for a straightforward inclusion of polarization once different polarization channels are sufficiently decoupled. In that case we must sum the third term in the denominator over all possible polarization states of the scattered wave and evaluate the second term, since it represents the coherent wave, for equal polarization of the incoming and outgoing wave.

(1) First of all let us consider a very elementary scattering situation for which phenomenological formulations of the transport velocity already exist in literature. This involves the semiclassical treatment of light scattering from an harmonic oscillator. The outcome of Refs. 12 and 13 can be written in terms of the following on-shell T matrix:

$$t_{\mathbf{pp}'}(E = pc_0) = \frac{-4\pi p^2 r_e}{p_o^2 - p^2 - \frac{2}{3} i p^3 r_e} \mathbf{g}_{\text{in}} \cdot \mathbf{g}_{\text{out}} . \tag{19}$$

Here $\mathbf{g}_{\rm in}$ and $\mathbf{g}_{\rm out}$ are the normalized polarization vectors of incoming and outgoing waves, and r_e is the classical electron radius. Furthermore, $p = p'$. It can readily be verified that

$$\sum_{\mathbf{g}_{\rm out}} \int \frac{d\Omega}{4\pi} (\mathbf{g}_{\rm in}\cdot\mathbf{g}_{\rm out})^2 = \frac{2}{3}. \tag{20}$$

We introduce the phase angle α according to $t_{\mathbf{pp}'}(p) = -(6\pi/p)\sin\alpha\,\exp(i\alpha)F(\Omega)$, with $\tan\alpha = \frac{2}{3}p^3 r_e/(p_0^2 - p^2)$. Upon direct differentiation,

$$v_E = \frac{c_0^2}{v_p}\left(1 + \frac{3\pi n}{p^2}\frac{d\alpha}{dp} + \frac{3\pi n}{2p^3}\sin 2\alpha\right)^{-1}. \tag{21}$$

Neglecting higher orders in density, yields the final result

$$\frac{v_E}{c_0} = \left(1 + \frac{2\pi n r_e (p_o^2 + p^2)}{(p_o^2 - p^2)^2 + (\frac{2}{3}p^3 r_e)^2}\right)^{-1}. \tag{22}$$

This speed is seen to be always less than unity, in sharp contrast to v_g, which equals, in the same approximation,

$$\frac{v_g}{c_0} = \left(1 + 2\pi n r_e \frac{(p_o^2 - p^2)^2(p_o^2 + p^2) + (\frac{2}{3}p^3 r_e)^2(3p^2 - 5p_o^2)}{\left[(p_o^2 - p^2)^2 + (\frac{2}{3}p^3 r_e)^2\right]^2}\right)^{-1}, \tag{23}$$

which becomes anomalous near the resonance at $p = p_o$. Inspection of Eq. (22) proves that the transport velocity drops sharply in the neighborhood of a resonance. This drop is essentially determined by the product of density, total cross section, and lifetime of the resonance. We have plotted the velocities in Fig. 1. Sufficiently far from the resonance, all velocities coincide.

The result obtained by Loudon,[12] as well as Brillouin,[11] is in agreement with Eq. (22). This is not self-evident because their model differs from ours. The T matrix in Eq. (19) is the result of a coupling of the radiation field to and a subsequent integration over internal degrees of freedom (exited states), which then become internal resonances. On the other hand, Eq. (9) was derived without inclusion of any internal degree of freedom. Nevertheless, from the appearance of the on-shell T matrix alone one cannot distinguish between both kinds of resonances. This is consistent with the fact that an application of our formula to their T matrix yields the same result as an explicit treatment of these internal degrees of freedom.

(2) Since explicit internal degrees of freedom are absent in dielectric scattering, a resonance in that situation must be a shape resonance, in which case the wave is trapped inside the dielectric, and a standing wave is built up. We evaluate Eq. (9) for spherical dielectric scatterers with radius r_m and real index of refraction m, known as Mie scatterers. The on-shell T matrix, in terms of the copolarized and cross-polarized channels, is given by[43]

$$\mathbf{t}_{\mathbf{pp}'}(E = pc_0) = -\frac{4\pi i}{p}\begin{pmatrix} S_2^*(\theta)\cos\varphi & 0 \\ 0 & S_1^*(\theta)\sin\varphi \end{pmatrix}, \tag{24}$$

where,

$$S_1(\theta) = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\left[a_n(x)\pi_n(\cos\theta) + b_n(x)\tau_n(\cos\theta)\right], \tag{25}$$

$$S_2(\theta) = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\left[b_n(x)\pi_n(\cos\theta) + a_n(x)\tau_n(\cos\theta)\right].$$

Here $x = pr_m$ is the size parameter. The phase shift ϕ_i of a particular channel is given by the phase of the corresponding $S_i(\Theta)$. Using the orthogonality relations[44]

$$\langle\pi_l\pi_n + \tau_l\tau_n\rangle = \delta_{nl}\frac{l^2(l+1)^2}{2l+1}, \quad \langle\pi_l\tau_n + \tau_l\pi_n\rangle = 0, \tag{26}$$

it follows that the third term in Eq. (9), the collision contribution, can be worked out:

$$\frac{4\pi}{p^2}\sum_i \int \frac{d\Theta,\varphi)}{4\pi}|S_i(\Theta)|^2\frac{\partial\phi_i(x,\Theta)}{\partial x}\cos^2\varphi = \frac{2\pi}{p^2}\sum_{n=1}^{\infty}\left(|a_n|^2\frac{d\alpha_n}{dx} + |b_n|^2\frac{d\beta_n}{dx}\right).$$

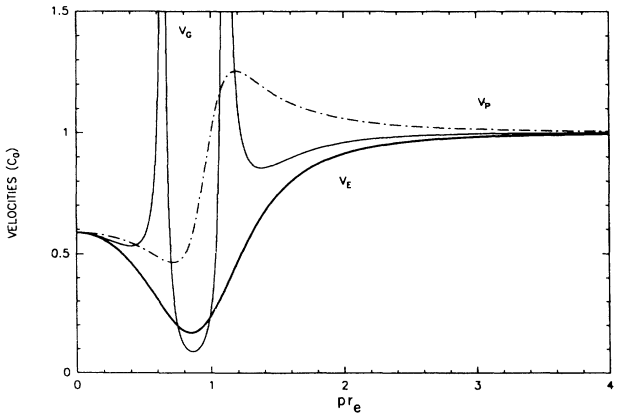

FIG. 1. Transport velocity (bold solid), group velocity (solid), and phase velocity (dashed dotted) for the semiclassical model discussed in the text. We have used $2\pi n r_e^3 = 0.7$ and $p_0 r_e = 1$. The two anomalous singularities of the group velocity are characteristic of an S-shaped dispersion law. Velocities are given in units of c_o.

The phases $\alpha_n(x)$ and $\beta_n(x)$ of the different partial waves are defined according to

$$a_n(x) \equiv \tfrac{1}{2}(1 - e^{-2i\alpha_n(x)}) \ , \ b_n(x) \equiv \tfrac{1}{2}(1 - e^{-2i\beta_n(x)}) \ . \tag{27}$$

In the forward direction $\Theta = 0$ it can be shown that $S_1 = S_2$. Direct differentation yields

$$-\frac{1}{2p^2}\frac{\partial(p\,\mathrm{Re}t_{pp})}{\partial p} = \frac{\pi}{p^2}\sum_{n=1}^{\infty}(2n+1)\left(\cos 2\alpha_n \frac{d\alpha_n}{dx} + \cos 2\beta_n \frac{d\beta_n}{dx}\right) .$$

Insertion of both parts into the expression for v_E gives the final answer,

$$\frac{v_E}{c_0} = \frac{c_0}{v_p}\left[1 + \frac{3}{4}\frac{f}{x^2}\sum_{n=1}^{\infty}(2n+1)\left(\frac{d\alpha_n}{dx} + \frac{d\beta_n}{dx}\right) - \frac{1}{2}f\,C(x)\right]^{-1} . \tag{28}$$

We have defined the packing fraction $f = \frac{4}{3}\pi n r_m^3$ and

$$C(x) \equiv \frac{3}{2x^3}\sum_{n=1}^{\infty}(2n+1)\,[\mathrm{Im}\,a_n + \mathrm{Im}\,b_n] \ . \tag{29}$$

In terms of this parameter, the phase velocity is given by $v_p = c_0/\sqrt{1+fC}$. Figure 2 shows the numerical evaluation of Eq. (28) for Mie scatterers with an index of refraction $m = 2.73$ and packing $f = 36\%$, relevant for our experiments.[9,10] It can be seen that the transport velocity sharply drops near scattering resonances, and differs considerably from the phase velocity. The reason is that the spectrum is dominated by multipole resonances and their overtones. Only sufficiently far away from such resonances can we expect all velocities to coincide. In fact, this holds for the Rayleigh limit $x \to 0$. Indeed, we find from the electric dipole contribution

$$v_E = v_p = v_g = c_0\left(1 + 3f\frac{m^2-1}{m^2+2}\right)^{-1/2} \quad (x \to 0) . \tag{30}$$

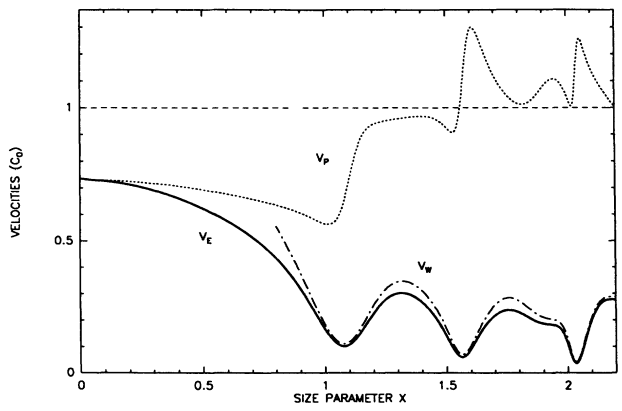

FIG. 2. Transport velocity (bold solid) and phase velocity (dashed) for $m = 2.73$ dielectric spheres, with packing fraction $f = 36\,\%$. The dashed dotted line is the heuristic speed v_W obtained from energy-density arguments. The size parameter $x \approx 1.1$ corresponds to the first magnetic dipole resonance. The size parameters in our experiments are distributed near this resonance. There $v_E \approx v_W \approx v_a \approx 0.13\,c_0$.

V. HEURISTIC APPROACHES

In this section we indicate the relation between the transport speed, and the formation of standing waves inside the scatterer, responsible for the considerable delay of the scattering near resonances. To this end let us reconsider the semiclassical model of Eq. (19). We have argued before that, in this case, the delay expressed by the decrease of the transport velocity can be understood in terms of *spontaneous emission*. The linear scattering of light (from a harmonic oscillator) can be viewed quantum mechanically as a single-photon excitation followed by a subsequent spontaneous decay. The delay of the scattering process is expressed by the inverse Einstein spontaneous emission coefficient A, and should be taken into account in the transport of photons. The average time between two scattering events is the *scattering* mean free time $\tau = 1/n\sigma c_0$. We obtain for the transport velocity

$$\frac{v_E}{v_p} \approx \frac{\tau}{\tau + 1/A} . \tag{31}$$

The uncertainty relation relates the energy-width of the cross-section to the Einstein coefficient according to (see Eq. 4.91 of Ref. 12) $A = \frac{2}{3}p^2 r_e c_0$. Equation (31) can be proven to coincide exactly with the microscopic outcome Eq. (22).

A similar procedure can be followed for the Mie scatterer.[45] The time Δt_W needed for the incoming plane wave with flux $S = c_0\Xi_o$ to "charge" the volume V of the dielectric to energy $\int dV\Xi \equiv W\,V\,\Xi_o$ is $\Delta t_W = V\Xi_o(W-1)/\sigma S c_0$. Here $\Xi_o\,,\Xi$ denote the EM energy density of the vacuum and scatterer in a steady-state situation. Again using $\tau = 1/n\sigma c_0$ we estimate

$$\frac{v_W}{v_p} = \frac{1}{1 + \Delta t_W/\tau} = \frac{1}{1 + f(W-1)} . \tag{32}$$

This velocity is expected to give a good estimate provided the EM energy is well confined within the sphere, which is the case if $m \gg 1$, $x \gg 1$. A large value for W reflects the formation of a standing wave inside the scatterer.

The delay that occurs in resonant scattering can also be estimated in the following way.[46] Imagine a dielectric scatterer in which little absorption is introduced, by making the index of refraction slightly complex, with imaginary part $m_i > 0$. The albedo a is defined as the ratio of scattering and extinction. Since $1/m_i p$ is the absorption length, the albedo can be expected to decay according to

$a = \exp(-2L_a m_i p)$. Here L_a represents the *path length* of the wave in the dielectric barrier Fig. 3. The time that the wave spends in this barrier can be estimated by $L_a m/c_0$. Writing $a \equiv 1 - Q_{\rm abs}/Q_e$, with $Q_{\rm abs}$, Q_e the (Mie) quality factors of absorption and extinction and r_m the Mie radius, it follows for this time

$$\Delta t_a = \lim_{m_i \to 0} \frac{1}{2} \frac{m}{x} \frac{Q_{\rm abs}}{m_i} \frac{r_m}{Q_e c_0} . \tag{33}$$

In analogy with Eq. (31), a transport velocity can be proposed

$$\frac{v_a}{v_p} = \frac{\tau}{\tau + \Delta t_a} . \tag{34}$$

The energy of electromagnetic waves within a dielectric sphere was discussed by Bott *et al.*[47] These authors calculated W for the general case on the basis of the Mie coefficients c_n and d_n that characterize the field in the sphere. They subsequently showed that in the limit of weak absorption

$$W \approx \lim_{m_i \to 0} \frac{3}{8} \frac{m}{x} \frac{Q_{\rm abs}}{m_i} + \mathcal{O}\left(\frac{1}{m}\right) . \tag{35}$$

A combination of Eqs. (32), (33), (34), and (35) establishes the approximate equivalence of v_a and v_W in this limit, provided $W \gg 1$, that is when a standing wave picture is valid.

We are left with the problem of relating the *exact* expression to these results. We show in Appendix B that for $m \gg 1$, $mx \gg 1$,

$$W - 1 \approx \frac{3}{4} \frac{1}{x^2} \sum_{n=1}^{\infty} (2n+1) \left(\frac{d\alpha_n}{dx} + \frac{d\beta_n}{dx} \right) . \tag{36}$$

A combination of Eqs. (36), (32), and (28) demonstrates the equivalence of v_W and v_E, again when x, $mx \gg 1$. Our numerical calculations clearly demonstrated that this agreement is valid even beyond this regime as can be inferred from Fig. 2.

An estimate of the path length inside the scatterer can also be obtained from our energy velocity using $L_E = c_0 \Delta t_E / m$, with Δt_E defined in terms of the difference between phase and transport velocity according to

$$\frac{v_E}{v_p} \equiv \frac{\tau}{\tau + \Delta t_E} = \frac{1}{1 + f \Delta t_E / f\tau} , \tag{37}$$

for small f; τ is again the scattering mean free time. In Fig. 3 we have plotted the path lengths of the wave in the dielectric (nonabsorbing) sphere, calculated from our microscopic expression as well as from absorption. The agreement is very good. Table I shows some numerical results for $m = 2.73$ spheres, near different scattering resonances. The parameter x^∞ is the size parameter at which a "free vibration" of the sphere is expected in the limit $m \to \infty$.[48] The coincidence of the time scales Δt_W, Δt_a and Δt_E is roughly 10%. For larger index m we found even better agreement.

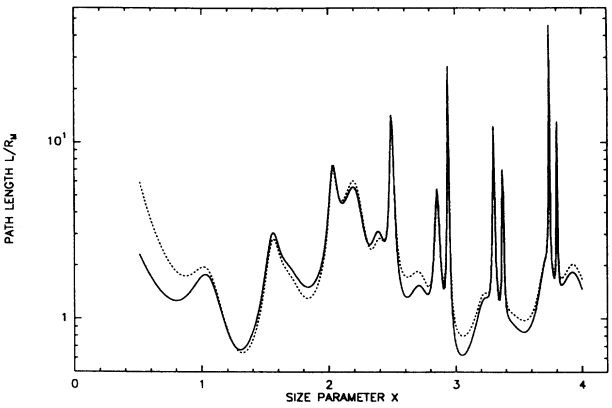

FIG. 3. Path length of the light wave in the dielectric sphere ($m = 2.73$) inferred from absorption (dashed line) as well as from microscopic transport velocity (solid line).

VI. CONCLUSIONS

Using a microscopic theory as a starting point we have obtained a general formula for the energy transport velocity in random media, and established its appearence in

TABLE I. Numerical evaluation of the time scales Δt_W, Δt_a, associated with the transport velocity obtained from energy density, and absorption arguments, as well as Δt_E derived from microscopic theory; τ is the scattering mean free time. We considered resonant size parameters x of a $m = 2.73$ Mie sphere. The ground tone of the magnetic (electric) multipole resonance of order n is indicated by MnI (EnI), the overtones are given by higher roman numbers; $E(n-1)$ coincides approximately with Mn. The columns on the right give two independent estimates of the path length of the wave in the dielectric barrier, as discussed in the text.

x	x^∞	Resonance	$\Delta t_W/f\tau$	$\Delta t_a/f\tau$	$\Delta t_E/f\tau$	L_a/r_m	L_E/r_m
1.100	1.15	*M1I*	20.82	23.21	22.06	1.56	1.48
1.566	1.65	*M2I*	37.12	40.48	43.83	2.83	3.07
2.036	2.11	*M3I*	63.32	69.05	74.00	7.02	7.53
2.380	2.30	*M1II*	22.38	22.26	24.88	2.78	3.11
2.494	2.60	*M4I*	126.31	137.30	145.01	14.43	15.24
2.850	2.83	*M2II*	57.25	55.89	62.61	4.86	5.44
2.939	3.00	*M5I*	293.23	316.76	332.25	27.24	28.57

the diffusion constant. The large decrease of this speed near resonances is translated into a path length of the wave in the dielectric barrier, and agrees reasonably with a path length inferred from absorption. We indicated that the Thouless criterion for localization, when reformulated in terms of length scales, is not altered by our findings. Our general expression for the energy velocity can easily be evaluated for absorbing scatterers. This is beyond the scope of this paper.

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APPENDIX A

This appendix deals with a useful sum rule valid for the spectral function, defined in terms of the averaged amplitude Green function in Eq. (15). The mass-operator $\Sigma(z)$ is defined according to

$$G(z) = \frac{1}{z^2 - p^2 - \Sigma(z)} = \left\langle \frac{1}{z^2\varepsilon - p^2} \right\rangle . \tag{A1}$$

Here ε is the dielectric constant of the random medium, $< >$ denotes ensemble averaging in the thermodynamic limit, and z is a complex energy. We set $c_0 = 1$. By letting $z \to \infty$ it follows that

$$\lim_{z\to\infty} \left(1 - \frac{\Sigma(z)}{z^2}\right) = \langle \varepsilon^{-1} \rangle^{-1} . \tag{A2}$$

This is still an operator identity but can easily be formulated for the matrix element $\Sigma(z,p)$, using the diagonality in momentum space. For a vacuum filled with dielectric scatterers with packing fraction f and speed of light c_{sc} we then obtain for the right-hand side of Eq. (A2) $\langle c^2 \rangle = (1-f) + f c_{sc}^2$. It can be inferred from Eq. (A1) that the matrix element $G(z,p)$ must be analytic in both sheets $\mathrm{Im}\, z >< 0$, since p and ε are real valued. The scalar wave equation has *two* physical sheets, separated by a branch cut (the spectrum) located along the whole real axis. A consequence is the following sum rule:

$$\int_{-\infty}^{\infty} \frac{dE}{2\pi} S(E,p) = \langle c^2 \rangle . \tag{A3}$$

The proof is straightforward. First we write $S(E,p) = iE\,[G(E^+,p) - G(E^-,p)]$, with $E^{\pm} \equiv E \pm i\epsilon$. Since $G(E,p)$ asymptotically decays as $\langle c^2 \rangle / E^2$, both terms cannot be handled separately. If we substract the asymptotical limit of Eq. (A2) from both terms, with

$$f(z) \equiv \frac{z}{z^2 - p^2 - \Sigma(z,p)} - \frac{\langle c^2 \rangle}{z} ,$$

we arrive at

$$\int_{-\infty}^{\infty} \frac{dE}{2\pi i} [f(E^-) - f(E^+)] = \int_{\Gamma^-} \frac{dz}{2\pi i} f(z) - \int_{\Gamma^+} \frac{dz}{2\pi i} f(z) - \oint_{z=0} \frac{dz}{2\pi i} f(z) .$$

$\Gamma^{\pm}$ denotes a very large closed half circle in the upper and lower sheet, respectively. Because $f(z)$ is analytic in both sheets, the first two integrals vanish. The third Cauchy integral is easily shown to be equal to $-\langle c^2 \rangle$.

APPENDIX B

In this appendix analytical results are obtained for Mie scatterers. First we consider the scattering from ideal reflectors $|m| = \infty$, and evaluate the geometrical limit $x \to \infty$. We shall prove

$$\lim_{x\to\infty} \frac{3}{4}\frac{1}{x^2} \sum_{n=1}^{\infty} (2n+1) \left(\frac{d\alpha_n}{dx} + \frac{d\beta_n}{dx} \right) = -1 . \tag{B1}$$

To this end we note that the "Van de Hulst coefficients" defined in Eq. (27) take the form

$$a_n(x) = \frac{\psi_n'(x)}{\xi_n'(x)} , \quad b_n(x) = \frac{\psi_n(x)}{\xi_n(x)} , \tag{B2}$$

where ψ_n and ξ_n are Ricatti-Bessel functions of order n. Since $\psi_n \to \sin(x - n\pi/2)$ and $\xi_n \to -\cos(x - n\pi/2)$, if $x \gg n$, it follows that in the same limit $\alpha_n' = \beta_n' \to -1$, and the causality limit is exactly reached. Partial waves with $x > n$ correspond geometrically to rays reaching the sphere (the "localization principle").[43] A closer inspection of the phase angles demonstrates[43] that $\alpha_n, \beta_n \to -xf(\tau) \pm K$, where $f(\tau) = \sin\tau - \tau\cos\tau$, if $\cos\tau \equiv (n+\frac{1}{2})/x \leq 1$. It can be checked that $\alpha_n' + \beta_n' = -2\sin\tau$. The sum is rewritten as

$$\frac{3}{2} \left(\sum_{n+\frac{1}{2}=\frac{3}{2}}^{[x]} + \sum_{n+\frac{1}{2}=[x]}^{\infty} \right) \frac{\Delta(n+\frac{1}{2})}{x} \frac{(n+\frac{1}{2})}{x} \left(\frac{d\alpha_n}{dx} + \frac{d\beta_n}{dx} \right) .$$

The second sum vanishes as $x \to \infty$. The first term approaches the integral

$$-3 \int_0^1 d\cos\tau \cos\tau \sin\tau = -1 ,$$

which proves the statement. A similar treatment proves that $C(x)$, defined in Eq. (29), goes to zero, so that the

phase velocity approaches c_0.

In the next part we demonstrate analytically the equivalence of microscopic and heuristic velocities, at least in the limit where the heuristic approach is believed to hold, that is $m \gg 1$, $mx \gg 1$. In this regime the phase velocity is of order c_0. It follows by inspection that

$$\frac{da_n}{dx} = \frac{da_n^\infty}{dx} + m\left(\frac{\partial a_n(x,y)}{\partial y}\right)_x + \mathcal{O}\left(\frac{1}{m}\right).$$

A same approximation holds for b_n. For clarity, we have written a_n^∞ for the coefficient of the ideal reflector as given by Eq. (B2). Since $d/dm_i = -ix\partial/\partial y$, we arrive at

$$\frac{da_n}{dx} = \frac{da_n^\infty}{dx} + \frac{im}{x}\frac{da_n}{dm_i} + \mathcal{O}\left(\frac{1}{m}\right).$$

We can apply Bott's result Eq. (35) to obtain

$$\begin{aligned} W &\approx \frac{3}{8}\frac{m}{x}\lim_{m_i\to 0}\frac{Q_{\rm abs}}{m_i} = \frac{3}{4}\frac{m}{x^3}\lim_{m_i\to 0}\sum_{n=1}^{\infty}(2n+1)\left(\mathrm{Re}\,\frac{da_n}{dm_i} - \frac{d(a_n a_n^*)}{dm_i} + \mathrm{Re}\,\frac{db_n}{dm_i} - \frac{d(b_n b_n^*)}{dm_i}\right) \\ &\approx \frac{3}{4x^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Im}\left(\frac{da_n}{dx}(1-2a_n^*)\right) - \frac{3}{4x^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Im}\left(\frac{da_n^\infty}{dx}(1-2a_n^{\infty *})\right) \\ &\quad + \frac{3}{4x^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Im}\left((a_n^\infty - a_n)\frac{da_n^{\infty *}}{dx}\right) + (\cdots), \end{aligned} \tag{B3}$$

where $(\cdots)$ represents the three previous terms with q_n replaced by b_n. Inserting (27), applying the large x limit to approximate the second term as well as the corresponding term with b_n^∞, and neglecting the difference between a_n and a_n^∞, we find

$$W - 1 \approx \frac{3}{4}\frac{1}{x^2}\sum_{n=1}^{\infty}(2n+1)\left(\frac{d\alpha_n}{dx} + \frac{d\beta_n}{dx}\right). \tag{B4}$$

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