Scattering delay and renormalization of the wave-diffusion constant

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Recent work by van Albada, van Tiggelen, Lagendijk, and Tip reveals the existence of a significant correction to the wave-diffusion constant. We give a simple physical picture for this, along with a derivation for the case of scalar wave and weak disorder. The result of van Albada *et al.* is largely confirmed and its inevitability established using the notion of Wigner scattering time.

In a random medium, a classical wave propagates on a short length scale but diffuses on a length scale much greater than the mean free path.[1-3] The latter regime is characterized by the effective diffusion constant D. If the disorder is weak, D can be calculated using standard techniques[4] and is given by the familiar expression $v_0 l_T/3$, where v_0 is the phase velocity and l_T is the transport mean free path involving the weighted angular average of the differential cross section of a single scatterer

$$l_T=\{n\int\sigma(\theta)[1-\cos(\theta)]\}^{-1},$$

where n is the density of scatterers. In a recent paper van Albada *et al.*[5] point out that this naïve expression for D is, in general, not valid because a sizable correction has so far been overlooked. Using the model of a randomly placed set of identical Mie scatterers of radius R with a volume-filling fraction f, they find the correction at frequency E to be

$$\frac{D}{D_0}=\left[1+\frac{3}{4}\frac{f}{x^2}\sum_{n=1}^{\infty}(2n+1)\left[\frac{d\alpha_n}{dx}+\frac{d\beta_n}{dx}\right]\right]^{-1}, \qquad (1)$$

where D_0 is the "bare" diffusion constant $v_0 l_T/3$, $x=RE/v_0$ is the size parameter, and α_n and β_n are the van de Hulst[6] phase shifts. A derivation of the above result was sketched, and good numerical agreement with experiments was found.

In this paper we present a qualitative physical picture based on the notion of the scattering delay proposed by Wigner.[7] This picture clearly shows why the diffusion-constant correction of the kind proposed by van Albada *et al.* should be expected on general ground; in addition, we present a detailed derivation of the diffusion-constant correction and find qualitative agreement with the results of van Albada *et al.* For simplicity, we restrict ourselves to the discussion of scalar waves; though we expect our results to be qualitatively useful to the case of vector waves, the precise quantitative result remains to be worked out. We also assume the disorder to be weak.

Central to our discussion is the important observation by van Albada *et al.*[5] that a clear distinction must be made between steady-state measurements and dynamical measurements. Typically, the mean free path can be measured under steady-state conditions whereas the diffusion constant must be measured dynamically. The physical picture underlying the diffusion of classical waves in a random medium is the same as that of a classical particle undergoing Brownian motion; both are examples of random walk. If, at $t=0$, a wave packet is introduced at the point $\mathbf{r}=\mathbf{0}$, then to reach a point $|\mathbf{r}|=L\gg l$ requires $N\sim(L/l)^2$ steps and consequently an amount of time τN, where $\tau=l/v_0$ is the time it takes to freely propagate a distance l. The expression of the diffusion constant follows immediately.

At first sight, the above discussion is completely general and watertight. Note, however, that the operational definition of the diffusion constant requires that we keep track of the time evolution of the wave. As a result, we cannot use a monochromatic wave and *must* use a wave packet which involves at least two different frequencies. The use of wave packets brings out the new physics: As pointed out by Wigner,[7] the scattering of a wave packet takes time. Very crudely speaking, we can think of the N scattering events in the following way. Between any two consecutive scatterings the wave propagates a distance l which takes time τ. Each scattering requires an additional Wigner "scattering time" τ_W. Thus the N-step random walk takes $(\tau+\tau_W)N$ to complete. The apparent diffusion constant is then given by $D=D_0\tau/(\tau+\tau_W)$.

Wigner has given a simple derivation of τ_W.[7] Consider an incoming scalar wave packet which consists of two plane waves with slightly different frequencies and wave vectors

$$\Psi_i=\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega t)[1+\exp(i\Delta\mathbf{k}\cdot\mathbf{r}-i\Delta\omega t)],$$

where $\Delta\omega$ and $\Delta\mathbf{k}$ are both small. The outgoing wave is a superposition of the scattered waves and is, in standard notation,

$$\Psi_{\text{out}}=\frac{e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t}}{2ikr}\sum_{l=0}^{\infty}(2l+1)P_l(\cos\theta)(e^{2i\delta_l(\omega)}-1)\times\left[1+\frac{e^{i\Delta\mathbf{k}\cdot\mathbf{r}-i\Delta\omega t}}{1+\Delta k/k}\frac{e^{2i\delta_l(\omega+\Delta\omega)}-1}{e^{2i\delta_l(\omega)}-1}\right], \qquad (2)$$

where the phase shifts δ_l are, in general, functions of ω. The motion of the center of the outgoing wave packet can be determined by looking at $|\Psi|^2$, and the lth partial wave moves according to $(\Delta\mathbf{k})\cdot\mathbf{r}-(\Delta\omega)(t-d\delta_l/d\omega)=\text{const}$. The center of the wave packet is seen to move at the group velocity, but the origin of time is shifted by the derivative $d\delta_l/d\omega$, which is identified, following Wigner, to be the "scattering time" of the lth partial wave. If we define the effective scattering time τ_W by the average of $d\delta_l/d\omega$ over all the partial waves we obtain

$$\tau_W = \frac{\sum_{l=0}^{\infty}(2l+1)(d\delta_l/d\omega)\sin^2\delta_l}{\sum_{l=0}^{\infty}(2l+1)\sin^2\delta_l} = \frac{4\pi}{\sigma k^2}\sum_{l=0}^{\infty}(2l+1)(d\delta_l/d\omega)\sin^2\delta_l \;, \tag{3}$$

where σ is the total cross section. Using $f=4\pi na^3/3$ and $\tau=(n\sigma v_0)^{-1}$, we have

$$\frac{D_0}{D} = 1 + \frac{3f}{x^2}\sum_{l=0}^{\infty}(2l+1)\sin^2\delta_l\frac{d\delta_l(x)}{dx} \;, \tag{4}$$

which strongly resembles Eq. (1), the result of van Albada *et al.*

Our derivation is of course nothing more than a hand-waving argument; the choice for the weighting factor $(2l+1)\sin^2\delta_l$ is at best suggestive. Still, we believe it captures the essence of the diffusion-constant correction as discovered by van Albada *et al.* and gives physical meaning to the detailed formal derivation given in Ref. 5 and below. The underlying physical reason for the diffusion-constant correction is that, in addition to the time of flight between scatterings, there is generally a "scattering delay" or "dwelling time" during the scattering. This changes the time it takes for the wave to undergo a given number of multiple scatterings and consequently modifies the diffusion constant. The diffusion constant may decrease or increase, depending on whether the delay time is positive or negative, though if it is negative the phrase scattering "delay" is of course a misnomer.

We now calculate the correction explicitly, using the formalism of Ref. 4. The natural object to study is the Fourier transform of the *two-fields* Green's function $G^2(\mathbf{r},t;\mathbf{r}',0)$, and to express it in terms of the disorder-averaged *one-field* Green's function

$$\langle G_E(\mathbf{p},\mathbf{p}')\rangle = \delta(\mathbf{p}-\mathbf{p}')[(E/v_0)^2 - p^2 - \Sigma(E,\mathbf{p})]^{-1} \equiv G_E(\mathbf{p}) \;,$$

where Σ is the self energy. Following closely the derivation in Ref. 4, a generalized Boltzmann equation can be obtained for

$$\mathcal{C}_{\mathbf{p}}(\mathbf{q},\omega) \equiv \int d^3\mathbf{p}'/(2\pi)^3 \langle G^+_{E_+}(\mathbf{p}_+,\mathbf{p}'_+)G^-_{E_-}(\mathbf{p}'_-,\mathbf{p}_-)\rangle \;,$$

$$\left[-2\frac{E\omega}{v_0^2} + 2\mathbf{q}\cdot\mathbf{p} + \Sigma^+(E_+,\mathbf{p}_+) - \Sigma^-(E_-,\mathbf{p}_-)\right]\mathcal{C}_{\mathbf{p}}(\mathbf{q},\omega) = \Delta G(E,\mathbf{p},\mathbf{q},\omega)\left[1 + \int\frac{d^3p'}{(2\pi)^3}U_{\mathbf{pp}'}(\mathbf{q},\omega)\mathcal{C}_{\mathbf{p}'}(\mathbf{q},\omega)\right] \;, \tag{5}$$

with the notations $E_\pm = E\pm\omega/2$ and $\mathbf{p}_\pm = \mathbf{p}\pm\mathbf{q}/2$. Here $U_{\mathbf{pp}'}(\mathbf{q},\omega)$ is the irreducible four-point function, connecting the lower and upper parts of the "bubble," and $\Delta G(E,\mathbf{p},\mathbf{q},\omega) \equiv G^+_{E_+}(\mathbf{p}_+) - G^-_{E_-}(\mathbf{p}_-)$.

At this point, the usual calculation of D (Refs. 4, 8, and 9) proceeds by invoking the Ward identity (WI)

$$\Sigma^+(E_+,\mathbf{p}_+) - \Sigma^-(E_-,\mathbf{p}_-) = \int\frac{d^3p'}{(2\pi)^3}U_{pp'}(\mathbf{q},\omega)\Delta G(E,\mathbf{p}',\mathbf{q},\omega) \;, \tag{6}$$

though its validity in our context has been a point of contention.[5] The WI was proved for the problem of electrons by Vollhardt and Wölfle[10] using a term-by-term diagrammatic analysis. It is therefore important to ask whether such a perturbative analysis is applicable for our purpose. We believe the answer is no. To observe the correction to the diffusion constant, care must be taken to retain information about the properties of the scatterers such as their shape and size. This is explicitly taken into account by writing Σ and U in terms of the scattering matrix for a single scatterer (see below). Generally speaking, one cannot expect to recover the diffusion-constant correction without using information about the individual scatterers.

The usual perturbative analysis almost invariably takes the following approach. One writes down the wave equation $[\nabla^2+\omega^2/v^2(\mathbf{r})]u=0$, where $v(\mathbf{r})$ is the local wave velocity, assumed to be random. The zeroth order problem is defined by a certain average velocity v_0 defined by $\langle 1/v^2(\mathbf{r})\rangle = 1/v_0^2$, so that $1/v^2(\mathbf{r}) \equiv (1/v_0^2)[1+\varepsilon(\mathbf{r})]$, with $\langle\varepsilon(\mathbf{r})\rangle=0$. One now carries out a perturbation theory in ε, *assuming that its statistical properties are completely known.* It is at this stage that the crucial information about the individual scatterers is lost. The true distribution of ε for randomly distributed spherical scatterers must obey, among other things, an infinite set of hard sphere conditions; although it is, *in principle,* possible to choose ε in such a way as to represent faithfully the wave-velocity distribution, in practice this is never done. Instead, one often assumes ε to be a Gaussian white noise. This greatly simplifies the analytical calculation and is, as a rule, good enough for many problems of classical wave localization and diffusion, as long as the physics does not depend critically on the properties of individual scatterers. The problem of the diffusion-constant correction is an exception.

In the analysis of Zhang and Sheng,[4] Σ is *not* calculated perturbatively from some microscopic random potential; instead, it is determined by appealing to the effective medium theory and demanding that the real part of the self-energy vanish at certain frequency E. Once this is done, the effective wave velocity v_0 is fixed, and the self-energy is completely determined to be $n\mathbf{t}_{\mathbf{p},\mathbf{p}}$. Since it is no longer determined order by order from U, there is no *a priori* reason why Σ and U should be related by the WI. A direct calculation along the lines indicated below shows that the WI is indeed not valid, except at $\mathbf{q}=0$, $\omega=0$ where it reduces to the optical theorem, a consequence of unitarity, which remains valid and is sufficient to ensure energy conservation. The nonvalidity of the

WI for $\mathbf{q}$, $\omega \neq 0$ leads directly to the diffusion constant correction.

To solve Eq. (5), we introduce the correlation function integrated over all incoming and scattering momenta $\mathcal{S}(\mathbf{q},\omega)=\int d^3p/(2\pi)^3 \mathcal{C}_{\mathbf{p}}(\mathbf{q},\omega)$, and the corresponding correlation current $\mathcal{T}(\mathbf{q},\omega)=\int d^3p/(2\pi)^3(\mathbf{p}\cdot\mathbf{q})\mathcal{C}_{\mathbf{p}}(\mathbf{q},\omega)$. In the Kubo limit, the integrated energy density $\mathcal{S}$ has a diffusive pole, from which the diffusion constant can be evaluated.

We will assume from here on that n is small such that the transport mean free path l_T is large compared to v_0/E, the average wavelength of the wave. Only under this condition does a diffusive process take place. To the lowest order in n, the vertex function U is $U_{\mathbf{p},\mathbf{p}'}(\mathbf{q},\omega)=n\,\mathbf{t}_{\mathbf{p}_+,\mathbf{p}'_+}\mathbf{t}^*_{\mathbf{p}'_-,\mathbf{p}_-}$. Recalling that the scattering matrix, which describes an elastic collision in an isotropic space, depends only on the modulus of the wave vectors $|\mathbf{p}|^2=|\mathbf{p}'|^2$ and on the cosine of the scattering angle $\mathbf{p}\cdot\mathbf{p}'/p^2\equiv\mu$, we expand U to the lowest order in $\mathbf{q}$

$$U_{\mathbf{p},\mathbf{p}'}(\mathbf{q},\omega)=n\,|\mathbf{t}_{\mathbf{p},\mathbf{p}'}|^2+[\mathbf{q}(\mathbf{p}+\mathbf{p}')]U^{(1)}_{\mathbf{p},\mathbf{p}'}+O(q^2)\ , \tag{7}$$

with

$$U^{(1)}\equiv in\ \mathrm{Im}\left[\left[\frac{\partial}{\partial p^2}\mathbf{t}_{\mathbf{p},\mathbf{p}'}+\frac{1-\mu}{p^2}\frac{\partial}{\partial\mu}\mathbf{t}_{\mathbf{p},\mathbf{p}'}\right]\mathbf{t}^*_{\mathbf{p}',\mathbf{p}}\right]\ . \tag{8}$$

We obtain two relations between $\mathcal{S}$ and $\mathcal{T}$ by multiplying the Boltzmann equation by 1 and $(\mathbf{p}\cdot\mathbf{q})$ respectively, and then integrating over all outgoing momenta, after expanding U, G, and Σ in ω and $\mathbf{q}$; keeping terms to order ω and q^2, we have

$$\mathcal{S}\left[-2\frac{E\omega}{v_0^2}\right]+2\mathcal{T}\left[1+\frac{\partial}{\partial p^2}\mathrm{Re}\Sigma^+ +\frac{i}{4\pi}\left[\frac{E}{v_0}\right]\langle U^{(1)}(1+\mu)\rangle\right]=-\frac{i}{2\pi}\left[\frac{E}{v_0}\right]\ , \tag{9}$$

$$\tfrac{2}{3}q^2\left[\frac{E}{v_0}\right]^2\mathcal{S}\left[1+\frac{\partial}{\partial p^2}\mathrm{Re}\Sigma^+ +\frac{i}{4\pi}\left[\frac{E}{v_0}\right]\langle U^{(1)}(1+\mu)\rangle\right]=-\mathcal{T}\left[\frac{ni}{2\pi}\left[\frac{E}{v_0}\right]\langle|\mathbf{t}_{\mathbf{p},\mathbf{p}'}|^2\mu\rangle+2i\,\mathrm{Im}\Sigma^+\right]\ . \tag{10}$$

To obtain the relations (9) and (10), several nonsingular terms, which do not contribute to the leading divergence (the diffusive pole) have been left out, and only contributions to the relevant order in n in each case have been kept. The details of these calculations will be published elsewhere.[11] All the expressions are evaluated *on shell* (at $|\mathbf{p}|=E/v_0$) due to the delta-functionlike structure[12] of ΔG and $\mathcal{C}_{\mathbf{p}}$, and the angular averages $\langle\cdots\rangle$ are over all relative outgoing directions.

Solving for the energy density, we find $\mathcal{S}(\mathbf{q},\omega)=(v_0/4\pi)(-i\omega+Dq^2)^{-1}$ from which we read off the diffusion constant

$$D=\frac{1}{3}E\frac{[1+(\partial/\partial p^2)\mathrm{Re}\Sigma^+ +(i/4\pi)(E/v_0)\langle U^{(1)}(1+\mu)\rangle]^2}{-\mathrm{Im}\Sigma^+ -(n/4\pi)(E/v_0)\langle|\mathbf{t}_{\mathbf{p},\mathbf{p}'}|^2\mu\rangle}\ . \tag{11}$$

The imaginary part of the self-energy $\Sigma^+(\mathbf{p})=n\,\mathbf{t}_{\mathbf{p},\mathbf{p}}$ is related to the differential cross section of scattering $|\mathbf{t}_{\mathbf{p},\mathbf{p}'}/4\pi|^2$ by the optical theorem, and in this way we obtain the leading correction to the diffusion constant

$$D=D_0\left[1+2n\frac{\partial}{\partial p^2}\mathrm{Re}\mathbf{t}_{\mathbf{p},\mathbf{p}}+\frac{i}{2\pi}\left[\frac{E}{v_0}\right]\langle U^{(1)}(1+\mu)\rangle\right]\ , \tag{12}$$

with $D_0=v_0l_T/3$, the classical diffusion constant, and $U^{(1)}$ given in Eq. (8).

We now express the scattering matrix in terms of the phase shifts and perform the angular average to obtain, finally,

$$\frac{D}{D_0}=1+3fF(x)\ , \tag{13}$$

with

$$\begin{aligned} F(x)&\equiv-\frac{1}{x^3}(S_1+xS_2)\ ,\\ S_1&=\sum_{l=0}^{\infty}4(l+1)^2\sin\delta_l\sin\delta_{l+1}\sin(\delta_{l+1}-\delta_l)\\ &\quad-\tfrac{1}{2}\sum_{l=0}^{\infty}(2l+1)\sin 2\delta_l\ ,\\ S_2&=\sum_{l=0}^{\infty}\frac{d\delta_l(x)}{dx}[(l+1)\cos^2(\delta_l-\delta_{l+1})\\ &\quad+l\cos^2(\delta_l-\delta_{l-1})]\ , \end{aligned} \tag{14}$$

where δ_{-1} is undefined and not needed. These expressions are completely general for scalar waves in the limit of small n, and for small phase shifts formally agree with Eq. (1), the expression found by van Albada *et al.* for the vector wave case.

We now discuss some simple applications of our general result. Because our result applies directly to scalar waves, we shall consider acoustic waves in a hydrodynamic medium. Consider a fluid of density ρ_1 and compressibility ξ_1 in which scatterers of radius R, density ρ_2, and compressibility ξ_2 are introduced and fixed in random positions. Using the size parameter $x = RE/v_1$ where $v_1 = (\xi_1/\rho_1)^{1/2}$ is the wave velocity *outside* the scatterer and E the frequency of the incipient wave, we find the lth phase shift to be given by

$$\tan\delta_l = \frac{z j_l'(y) j_l(x) - j_l(y) j_l'(x)}{z j_l'(y) n_l(x) - j_l(y) n_l'(x)} , \tag{15}$$

where j_l and n_l are the lth order spherical Bessel and spherical Neumann functions, respectively, $j'(x) \equiv dj(x)/dx$, $y \equiv Mx$, where $M = v_1/v_2$ is the index of refraction, v_2 is the wave velocity inside the sphere, and $z = v_1\rho_1/v_2\rho_2$ is the ratio of impedances. Examples of $F(x)$ for selected values of M and z are given in Fig. 1. We see that the classical diffusion constant can be either enhanced or reduced, and that even small changes in the parameters or the wave frequency can bring about this sign change. In general, if $M > 1$, i.e., if the wave velocity is smaller inside the scatterers as it is in a typical optical experiment, we have $F(x) < 0$ for small x, corresponding to a reduction in D. We also see in some cases a rich structure with a multitude of spikes at frequencies close to internal resonances of the scatterers.

The dual limitations of weak disorder and scalar wave preclude a direct comparison with the experiments based on the strong scattering of electromagnetic waves. We have used the lowest-order expressions for $\Sigma(\mathbf{p})$ and U in our calculations [see the discussion preceding Eq. (9)], which is consistent with the other assumptions of our theory but which implies that, by construction, our result is reliable only in the limit of $|D/D_0 - 1| \ll 1$. To see a significant correction, it is necessary to calculate or at least estimate the higher-order corrections; this has not been done. The scalar wave assumption is less restrictive, and, in principle, it is possible to work out the vector wave version of our theory, which can then be directly applied to optical experiments. In summary, we have given a simple physical picture for the diffusion constant correction proposed by van Albada *et al.* This correction is of a dynamical origin; it can be interpreted in terms of a modified energy transport velocity and can be detected in time-domain experiments. This correction modifies the relation between the bare mean free path calculated from the cross section $l = 1/n\sigma$ and the diffusion constant. Similar corrections may affect the renormalized transmission, reflection, and other aspects of wave propagation in a random medium.

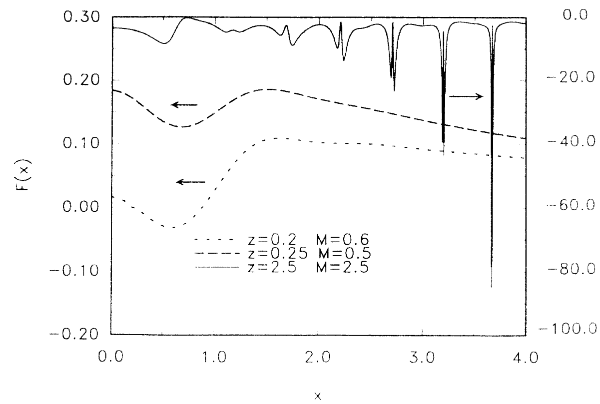

FIG. 1. The diffusion constant correction factor $F(x)$, Eq. (13).

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[12]This delta-functionlike structure makes the expansion in **q** somewhat subtle; the details will appear in Ref. 11.