Diffusion of classical waves in random media with microstructure resonances

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The diffusion constant of classical waves propagating in random media with microstructure resonances is obtained. Renormalizations of D that are due to expansions in transferred momentum $\mathbf{q}$ and frequency ω in the Bethe–Salpeter equation are taken into account. The diffusion constant is estimated for scalar waves propagating in the medium with randomly distributed dielectric spheres with the use of the nondiagonal off-shell transition matrix for penetrable scatterers. A detailed comparison of different sources of renormalization is made. Differences between previous calculations based on the on-shell scattering matrix and new results implementing the off-shell scattering matrix are discussed.

1. INTRODUCTION

The resonant effects in the propagation and the multiple scattering of classical waves in random media have recently attracted a lot of attention.[1–11] Usually, a random medium is treated as an unbounded collection of randomly distributed point scatterers.[12–15] This assumption simplifies calculations greatly, allowing one to use the conventional perturbation methods to describe transport properties of waves. In the case of weak disorder when $l/\lambda \gg 1$, where l is the mean free path and λ is the wavelength, one obtains the diffusion equation for the wave intensity with the rate of flow given by the diffusion coefficient $D = cl/3$,[15] where c is the speed of waves in the medium. However, in practice such ideal systems do not exist. All samples are bounded, and all scatterers are of particular shape and size. It is well known that a wave packet scattered from an object of finite size should spend additional time τ_W on the scattering process itself, in contrast to the point scatterers, for which this time is equal to zero. The quantity τ_W is called Wigner or dwelling time in the electron theory.[16] Even if τ_W itself is small, one can obtain sizable corrections to the diffusion constant in a medium with a large number of scatterers, and this effect is even more significant for classical waves because of the resonant behavior of their scattering matrix when scatterers are comparable in size with the wavelength. These resonances can be obtained from the solution of the boundary-value problem for an electromagnetic wave scattered from the dielectric sphere and are known as Mie resonances.[17] Recently, it was suggested by the Amsterdam group[1–3] that these resonances are responsible for the sizable reduction of the transport velocity appearing in the diffusion constant of classical waves, propagating through the random distribution of identical Mie spheres. Thus the conventional expression for D can no longer be applied to describe diffusion in a medium with the scatterers of finite size. The lowering of the diffusion constant as a result of the resonant contribution shows that considerable care is needed in interpreting the low values of D in studies for the search of classical wave localization.[18] To interpret these low values of D properly, one must calculate from first principles all possible sources contributing to its reduction.

The usual way to obtain the diffusion constant is to look for the asymptotic behavior of the average intensity of the wave, $I(\mathbf{q}, \omega)$, where $\mathbf{q}$ and ω are the transferred momentum and frequency, respectively.[4,6,11,15] The function $I(\mathbf{q}, \omega)$ satisfies the Bethe–Salpeter (BS) equation,[19] which is a generalized form of the conventional Boltzmann kinetic equation. Two approaches have been favorable for further studies of the BS equation. One can find D by considering the low-density limit of the BS equation and then expanding in powers of $\mathbf{q}$ and ω, retaining only the lowest-order terms.[1,4,6,9–11] The alternative approach is the coherent potential approximation used by Kroha *et al.*,[7] which leads to numerical computations. Despite the similarities in methods used, different results have been obtained. Barabanenkov and Ozrin[6] and independently Kroha *et al.*[7] have shown that the transport velocity is not lowered, as it was predicted by the Amsterdam group, but rather renormalized in the same way as the phase velocity. However, the Amsterdam group has shown that the conclusion made in Ref. 6, $\nu_E = c_p + O(n^2)$, where n is the density of scatterers, is due to neglect of the off-shell contribution into the photon density of states. The proper treatment of the density of states provides the desirable renormalization of ν_E. The generalization of the well-known coherent potential approximation was developed by Soukoulis *et al.*[8] According to this approach a basic scatterer in the medium is a *coated* dielectric sphere, thus taking into account the short-range order induced by the spherical shape of the scatterers. The effective transport velocity and the diffusion constant were studied numerically, and reasonable agreement with experiment was obtained.

An additional contribution to the renormalization of D was discussed in the paper by Cwilich and Fu.[4] It originates from the coefficients of the $\mathbf{q}$ expansion in the BS equation, whereas the coefficients in the ω expansion are responsible for the renormalization obtained by other authors.[1,6,9] The reason that these terms were

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overlooked in other theories is that the BS equation for both electrons and classical waves can be supplemented by the Ward identity (WI). Substitution of the electronic WI into the BS equation cancels *all* corrections to the diffusion constant. The WI for classical waves derived by Barabanenkov and Ozrin[20] is different from its electronic counterpart, and it preserves terms in the ω expansion in the BS equation. However, for $\omega = 0$ and $\mathbf{q} \neq 0$, both WI's coincide, thus canceling terms in the $\mathbf{q}$ expansion. The authors of Ref. 4 have questioned the applicability of the WI for the case $\omega = 0$ and $\mathbf{q} \neq 0$. They have performed the power-series expansion with respect to the variable $\mathbf{q}$ in the WI and have shown that terms in that expansion are not canceled. They have concluded that the known WI is indeed not valid and that it cannot be used in its present form in the BS equation.

Another aspect of the calculations of the diffusion constant requires additional attention. Resonant corrections to D involve partial derivatives of the scattering t matrix for a single scatterer with respect to the momentum $p = |\mathbf{p}|$ and energy E. The following procedure of numerical evaluation of the corrections to D, based on the fact that the scattering occurs on shell $p = p_0 = E/c_p$, has been adopted in Refs. 1–4. The on-shell approximation has been applied to the scattering matrix $t_{\mathbf{p},\mathbf{p}'}(E)|_{p=E/c} = t(E)$, and then derivatives with respect to E have been taken. As far as derivatives with respect to the momentum are concerned, it was concluded from the dispersion relation $p^2 = E^2/{c_p}^2 + nf(p, E)$ that, in the limit of low densities, $\partial/\partial p \approx c_p \partial/\partial E + O(n)$. These assumptions simplify calculations significantly, since the on-shell scattering matrix is simple enough and well known.[12,14] This approach can, however, lead to incorrect results. The t matrix for point scatterers, for example, is initially independent of momentum, and thus its partial derivatives with respect to momentum should be equal to zero, whereas the above approach leads to a finite result. Moreover, the functional dependence $t(p)$ is completely different from $t(E)$. The off-shell matrix may also include terms proportional to $\mathbf{p}^2 - E^2$, which would be zero if the on-shell approximation is applied first and then derivatives with respect to either p or E are taken. If derivatives are taken before the application of the on-shell approximation, the finite result is obtained. Therefore the off-shell scattering matrix must be used for numerical evaluations.

In the present study we derive the general expression for D with possible sources of renormalization taken into account. This expression is obtained without employing the WI and differs from results obtained by both the Amsterdam group[1–3] and Cwilich and Fu.[4] To make numerical evaluations, we calculate the off-shell t matrix for a penetrable dielectric sphere. A comparison between our correction to the diffusion constant, where the off-shell scattering matrix is used, and the results of the Amsterdam group for scalar Mie scatterers is made. We find that the functional behavior of our correction is different from the results obtained by the Amsterdam group. We also consider the case of acoustic waves in a hydrodynamic medium. We find strong enhancement in the correction to the diffusion constant compared with the results previously obtained by Cwilich and Fu.[4]

2. BASIC EQUATIONS

To calculate the correction to the diffusion coefficient, we use the formalism of Refs. 1 and 4. We consider the wave equation for a scalar monochromatic field $\psi_\Omega(\mathbf{r})$ of frequency Ω:

$$\left[\nabla^2 + \left(\frac{\Omega}{c}\right)^2 \epsilon(\mathbf{r})\right]\psi_\Omega(\mathbf{r}) = 0\,, \tag{1}$$

where c is a speed of wave propagation in the nonabsorbing random medium and $\epsilon(\mathbf{r})$ is the refraction index, which is a random function of $\mathbf{r}$. After the formal substitutions

$$(\Omega/c)^2\langle\epsilon\rangle = E\,, \qquad (\Omega/c)^2[\langle\epsilon\rangle - \epsilon(r)] = V(r, E)\,, \tag{2}$$

where $\langle\cdot\rangle$ stands for averaging over disorder, Eq. (1) formally coincides with the Schrödinger equation for a particle in the energy-dependent potential $V(\mathbf{r}, E)$. We will further use the notation E instead of Ω. The field $\psi(\mathbf{r}, t)$ generated by the point source at $\mathbf{r} = \mathbf{r}'$ is $\psi(\mathbf{r}, t) = G(\mathbf{r}, \mathbf{r}'; t)$, where $G(\mathbf{r}, \mathbf{r}'; t)$ is the Green function of Eq. (1). On account of the condition of macroscopic homogeneity, $|G(\mathbf{r}, \mathbf{r}', t)|^2$ averaged over disorder has translational invariance, i.e., it depends upon $|\mathbf{r} - \mathbf{r}'|$ only, and, furthermore, $\langle|G(\mathbf{r}, \mathbf{r}', t)|^2\rangle$ is a wave intensity $I(\mathbf{r} - \mathbf{r}', t)$ that is due to a source at a point $\mathbf{r}'$. After performing the Fourier transform in the space domain and the Laplace transform in the time domain, we obtain

$$\begin{aligned} I(\mathbf{q}, \omega) &= \int_0^\infty \mathrm{d}t\, \exp[i(\omega + i0)t] \int \mathrm{d}\mathbf{R}'\mathrm{d}\mathbf{R}\mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \\ &\quad \times \exp[i\mathbf{q}(\mathbf{R} - \mathbf{R}') - i\mathbf{p}\mathbf{r} + i\mathbf{p}'\mathbf{r}'] \\ &\quad \times \left\langle G\left(\mathbf{R} + \frac{\mathbf{r}}{2}, \mathbf{R}' + \frac{\mathbf{r}'}{2}; t\right)\right. \\ &\quad \times \left. G\left(\mathbf{R} - \frac{\mathbf{r}}{2}, \mathbf{R}' - \frac{\mathbf{r}'}{2}; t\right)\right\rangle \\ &= \int_{-\infty}^{+\infty} \frac{\mathrm{d}E}{2\pi} \sum_{\mathbf{p},\mathbf{p}'} \langle G^+_{E_+}(\mathbf{p}_+, \mathbf{p}'_+) G^-_{E_-}(\mathbf{p}_-, \mathbf{p}'_-)\rangle \\ &= \int_{-\infty}^{+\infty} \frac{\mathrm{d}E}{2\pi} \sum_{\mathbf{p}} \Phi_{\mathbf{p}}(\mathbf{q}, \omega; E)\,, \end{aligned} \tag{3}$$

where the notations $\mathbf{p}_\pm = \mathbf{p} \pm \mathbf{q}/2$ and $E_\pm = E \pm \omega/2 \pm i0$ were introduced and we defined the advanced (G^+) and the retarded (G^-) Green functions as

$$G^\pm_E(\mathbf{r}, \mathbf{r}') = \int_0^\infty \mathrm{d}t\, \exp[i(E \pm i0)t] G(\mathbf{r}, \mathbf{r}', t)\,. \tag{4}$$

The function

$$P_E(\mathbf{r} - \mathbf{r}', t) = \sum_{\mathbf{p}} \Phi_{\mathbf{p}}(\mathbf{r} - \mathbf{r}', t; E) \tag{5}$$

may be regarded as the Fourier transform of the E component of the averaged intensity excited at $\mathbf{r}'$ at $t = 0$. Therefore, in the weakly scattering regime, $P_E(\mathbf{q}, \omega)$ must exhibit a singular behavior as $\mathbf{q}, \omega \to 0$, from which the diffusion coefficient can be evaluated. The Fourier transform $\Phi_{\mathbf{p}}(\mathbf{q}, \omega; E)$ is related to the disorder-averaged Green functions,

$$\langle G^{\pm}_{E_{\pm}}(\mathbf{p}, \mathbf{p}')\rangle = \delta(\mathbf{p} - \mathbf{p}')[\xi_{\pm}{}^2 - \mathbf{p}^2 - \Sigma^{\pm}(\mathbf{p}, E^{\pm}]^{-1} \equiv G^{\pm}_{E_{\pm}}(\mathbf{p})\delta(\mathbf{p} - \mathbf{p}'), \tag{6}$$

where $\xi_{\pm} = E/c \pm i0$ and $\Sigma^{\pm}$ is the self-energy, defined by the BS equation,

$$\left[2\frac{E\omega}{c^2} + 2\mathbf{q}\cdot\mathbf{p} + \Sigma^{+}(E_{+}, \mathbf{p}_{+}) - \Sigma^{-}(E, \mathbf{p}_{-})\right]\Phi_{\mathbf{p}}(\mathbf{q}, \omega; E) = \Delta G_{\mathbf{p}}(\mathbf{q}, \omega; E)\left[1 + \int \frac{\mathrm{d}^3 p'}{(2\pi)^3} U_{\mathbf{p},\mathbf{p}'}(\mathbf{q}, \omega)\Phi_{\mathbf{p}'}(\mathbf{q}, \omega)\right]. \tag{7}$$

Here $U_{\mathbf{p},\mathbf{p}'}(\mathbf{q}, \omega)$ is the irreducible four-point vertex, and $\Delta G_{\mathbf{p}}(\mathbf{q}, \omega; E) \equiv G^{+}_{E_{+}}(\mathbf{p}_{+}) - G^{-}_{E_{-}}(\mathbf{p}_{-})$. To observe corrections to the diffusion constant, one must take into account the information about such geometrical properties of the individual scatterers as their shape and size. It can be accomplished within the framework of the low-density approximation, according to which Σ and U are expressed through the scattering matrix $t_{\mathbf{p},\mathbf{p}'}$ for a single scatterer: $\Sigma^{\pm}(\mathbf{p}_{\pm}, \omega; E^{\pm}) = nt_{\mathbf{p}_{\pm},\mathbf{p}'_{\pm}}(E^{\pm})$, $U_{\mathbf{p},\mathbf{p}'}(\mathbf{q}, \omega) = nt_{\mathbf{p}_{+},\mathbf{p}'_{+}}(E^{+})t_{\mathbf{p}'_{-},\mathbf{p}_{-}}(E^{-})$. Care must be taken to ensure that the density of scatterers n is small enough to allow the weak-scattering approximation to be valid. Since we are interested in the singular behavior of $\Phi_{\mathbf{p}}(\mathbf{q}, \omega; E)$ when $\mathbf{q}, \omega \to 0$, our next step is to perform an expansion of the self-energy and the vertex in terms of the variables ω and $\mathbf{q}$:

$$\Sigma^{\pm}\left(\mathbf{p} \pm \frac{\mathbf{q}}{2}, E^{\pm} \pm \frac{\omega}{2}\right) = nt_{\mathbf{pp}}(E^{\pm}) \pm n\frac{\omega}{2} \times \frac{\partial t_{\mathbf{pp}}(E^{\pm})}{\partial E} \pm (\mathbf{p}\cdot\mathbf{q})\frac{\partial t_{\mathbf{pp}}(E^{\pm})}{\partial p^2} + O(\omega^2, \omega q, q^2),$$

$$U_{\mathbf{p},\mathbf{p}'}(\mathbf{q}, \omega; E) = n|t_{\mathbf{p},\mathbf{p}'}(E^{+})|^2\left[1 + i\omega\frac{\partial\phi_{\mathbf{p},\mathbf{p}'}(E)}{\partial E}\right] + [\mathbf{q}\cdot(\mathbf{p} + \mathbf{p}')]K(\mathbf{p}, \mathbf{p}') + O(\omega^2, \omega q, q^2), \tag{8}$$

with

$$K(\mathbf{p}, \mathbf{p}') = in\,\mathrm{Im}\left\{\left[\frac{\partial t_{\mathbf{p},\mathbf{p}'}}{\partial p^2} + \frac{1-\mu}{p^2}\frac{\partial t_{\mathbf{p},\mathbf{p}'}}{\partial\mu}\right]t^{*}_{\mathbf{p},\mathbf{p}'}\right\}. \tag{9}$$

In deriving these equations, we have taken into account that in the case of elastic collisions the scattering matrix $t_{\mathbf{p},\mathbf{p}'}$ depends on the magnitude of the momenta, $|\mathbf{p}|^2 = |\mathbf{p}'|^2$, and on the cosine of the scattering angle $\mu = (\mathbf{p}\cdot\mathbf{p}')/p^2$ only. We have also denoted the phase shift of the scattering matrix as $\phi_{\mathbf{p},\mathbf{p}'}(E)$ according to $t_{\mathbf{p},\mathbf{p}'}(E^{+}) = |t_{\mathbf{p},\mathbf{p}'}|\exp(i\phi_{\mathbf{p},\mathbf{p}'})$. After substitution of Eqs. (8) into the BS equation we obtain

$$-\frac{2E\omega}{c^2}\left[1 - n\frac{\partial\mathrm{Re}(t_{\mathbf{pp}})}{\partial\xi^2}\right]\Phi_{p}(\mathbf{q}, \omega) - \frac{in\omega}{c}\Delta G_{\mathbf{p}}(\mathbf{q} = 0, \omega = 0; E)\sum_{\mathbf{p}'}|t_{\mathbf{p},\mathbf{p}'}|^2\frac{\partial\phi_{\mathbf{p},\mathbf{p}'}}{\partial\xi}\Phi_{\mathbf{p}'}(\mathbf{q}, \omega) + (\mathbf{p}\cdot\mathbf{q})\left[1 + n\frac{\partial\mathrm{Re}(t_{\mathbf{pp}})}{\partial p^2}\right]\Phi_{\mathbf{p}}(\mathbf{q}, \omega) - n\Delta G_{\mathbf{p}}(\mathbf{q} = 0, \omega = 0; E)\sum_{\mathbf{p}'}[\mathbf{q}\cdot(\mathbf{p} + \mathbf{p}')] \times K_{\mathbf{p},\mathbf{p}'}(\mathbf{q}, \omega)\Phi_{\mathbf{p}'}(\mathbf{q}, \omega) = \Delta G_{\mathbf{p}}(\mathbf{q}, \omega; E). \tag{10}$$

In order to find the diffusion coefficient from this equation, we find it useful to introduce, besides the function $P_E(\mathbf{q}, \omega)$ defined by Eq. (5), the correlation current $J_E(\mathbf{q}, \omega) = \sum_{\mathbf{p}}(\mathbf{p}\cdot\mathbf{q})\Phi_{\mathbf{p}}(\mathbf{q}, \omega; E)$. The quantities P_E and J_E satisfy a system of equations that we can obtain in two steps: first, by integrating Eq. (10) over all outgoing momenta and, second, by multiplying Eq. (10) by $\mathbf{p}\cdot\mathbf{q}$ and performing the same integration. This yields two relations for P_E and J_E:

$$-\frac{2\xi\omega}{c}P_E\left\{1 - n\frac{\partial\mathrm{Re}[t_{\mathbf{pp}}(E)]}{\partial\xi^2}\bigg|_{p=\xi} + in\sum_{\mathbf{p}'}\Delta G_{\mathbf{p}'}|t_{\mathbf{p},\mathbf{p}'}|^2 \times \frac{\partial\phi_{\mathbf{p},\mathbf{p}'}(E)}{\partial\xi^2}\bigg|_{p=\xi}\right\} + 2J_E\left\{1 + n\frac{\partial\mathrm{Re}[t_{\mathbf{pp}}(E)]}{\partial p^2}\bigg|_{p=\xi} + \frac{inE}{4\pi c_p}\langle(1+\mu)K(\mathbf{p}, \mathbf{p}')\rangle_{\mu}\right\} = -\frac{iE}{2\pi c_p}, \tag{11}$$

$$\frac{1}{3}q^2\xi^2 P_E\left\{1 + n\frac{\partial\mathrm{Re}[t_{\mathbf{pp}}(E)]}{\partial p^2}\bigg|_{p=\xi} + \frac{iE}{4\pi c_p}\langle(1+\mu)K(\mathbf{p}, \mathbf{p}')\rangle_{\mu}\right\} = -inJ_E\left[\frac{E}{4\pi c_p}\langle\mu|t_{\mathbf{p},\mathbf{p}'}|^2\rangle_{\mu} + \mathrm{Im}(t_{\mathbf{pp}})\right], \tag{12}$$

where $\langle\cdot\rangle_{\mu}$ denotes the angular averaging over all outgoing momenta and the phase velocity c_p is defined as $c_p \equiv c/[1 - nc^2\,\mathrm{Re}(t_{pp})/E^2]^{1/2}$. In deriving Eqs. (11) and (12), we have used the fact that, first, in the low-density approximation the density of photon states is sharply peaked at $|\mathbf{p}| = E/c_p$ and, second, that the imaginary part of the scattering matrix is related to the differential cross section of scattering by the optical theorem:

$$-\mathrm{Im}(t_{\mathbf{pp}}) = \frac{E}{4\pi c_p}\langle|t_{\mathbf{p},\mathbf{p}'}|^2\rangle_{\mu}. \tag{13}$$

Solving Eqs. (11) and (12) for P_E and taking into account that it has a diffusive pole $P_E \propto (-i\omega + q^2 D)^{-1}$, we find the diffusion constant D:

$$D = D_0\left(\frac{c}{c_p}\right)^2 \times \frac{1 + 2n\left.\frac{\partial \mathrm{Re}(t_{\mathbf{pp}})}{\partial p^2}\right|_{p=\xi} + \frac{inE}{2\pi c_p}\langle(1+\mu)K(\mathbf{p}, \mathbf{p}')\rangle_\mu}{1 - n\left.\frac{\partial \mathrm{Re}(t_{\mathbf{pp}})}{\partial \xi^2}\right|_{p=\xi} + in\sum_{\mathbf{p}} \Delta G_{\mathbf{p}'}|t_{\mathbf{p},\mathbf{p}'}|^2 \left.\frac{\partial \phi_{\mathbf{p},\mathbf{p}'}}{\partial \xi^2}\right|_{p=\xi}}, \tag{14}$$

where $D_0 = 1/3c_p l_T$ is the classical diffusive constant with the transport mean free path l_T defined as

$$l_T = \frac{1}{n\langle(1-\mu)|t_{\mathbf{p},\mathbf{p}'}|^2\rangle_\mu}\cdot \tag{15}$$

Since Eq. (14) is valid for low densities, we rewrite it, retaining terms of the first order in n only:

$$D(E) = D_0\{1 + f[a(E) + \Delta(E)]\},$$

$$\Delta(E) = \frac{2}{V}\left.\frac{\partial \mathrm{Re}[t_{\mathbf{p},\mathbf{p}}(E)]}{\partial p^2}\right|_{p=\xi} + \frac{iE}{2\pi c_p V}\langle(1+\mu)K(\mathbf{p}, \mathbf{p}')\rangle_\mu,$$

$$a(E) = \frac{1}{V}\left.\frac{\partial \mathrm{Re}[t_{\mathbf{p},\mathbf{p}}(E)]}{\partial \xi^2}\right|_{p=\xi} - \frac{i}{V}\sum_{\mathbf{p}'} \Delta G_{\mathbf{p}'}|t_{\mathbf{p},\mathbf{p}'}|^2 \left.\frac{\partial \phi_{\mathbf{p},\mathbf{p}'}(E)}{\partial \xi^2}\right|_{p=\xi}, \tag{16}$$

where $f = Vn$ is the volume-filling fraction and V is a characteristic volume of a scatterer. The terms $\Delta(E)$ and $a(E)$ originate from the nominator and the denominator of Eq. (14), respectively.

The term $a(E)$ coincides exactly with the correction to D obtained by Barabanenkov and Ozrin.[20] Since the Amsterdam group has considered the transport velocity ν_E rather than the diffusion constant, comparison between their results and Eqs. (16) is straightforward if one assumes the applicability of the expression $D = \frac{1}{3}\nu_E l_T$ for the diffusion constant. Then the term $a(E)$ is equal to the correction following from the results of Refs. 1–3. The term $\Delta(E)$ is the result of the expansion in the variable $\mathbf{q}$ and is missing in Refs. 1–3 and 6. It was first discussed by Cwilich and Fu.[4] Thus Eqs. (16) represent the most general expression for the renormalization of the diffusion constant in the low-density approximation. For the purpose of numerical comparison between Eqs. (16) and previously obtained results the exact expression for the nondiagonal off-shell t matrix must be known. The details of the calculation of $t_{\mathbf{p},\mathbf{p}'}$ for the case of scalar waves are given in the next section.

3. CALCULATION OF THE OFF-SHELL t MATRIX

In order to calculate a nondiagonal off-shell transition or t matrix for a single scatterer, we will utilize the general formalism developed in Refs. 23 and 24. We introduce the retarded and advanced Green functions $\Im_E^{\pm}(\mathbf{r}, \mathbf{r}')$ for a problem of scattering from a single scatterer that satisfy the differential equations

$$(\nabla^2 + \xi_\pm{}^2)\Im_{E,\mathrm{outside}}^{\pm}(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \tag{17}$$

outside the single scatterer and

$$(\nabla^2 + M^2\xi_\pm{}^2)\Im_{E,\mathrm{inside}}^{\pm}(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \tag{18}$$

inside the scatterer of average index of refraction M. Equations (17) and (18) should be supplemented by boundary conditions that for permeable scatterers have the following form:

$$\Im_{E,\mathrm{outside|surface}}^{\pm} = \Im_{E,\mathrm{inside|surface}}^{\pm},$$
$$\left.\frac{\partial \Im_{E,\mathrm{outside}}^{\pm}}{\partial \mathbf{r}}\right|_{\mathrm{surface}} = \left.\frac{\partial \Im_{E,\mathrm{inside}}^{\pm}}{\partial \mathbf{r}}\right|_{\mathrm{surface}}. \tag{19}$$

The equation that defines a transition matrix for a single scatterer can be written in a wave-number representation as

$$\Im_E^{\pm}(\mathbf{p}, \mathbf{p}') = \Im_{E,0}^{\pm}(\mathbf{p}, \mathbf{p}') + \Im_{E,0}^{\pm}(\mathbf{p}, \mathbf{p}')t_{\mathbf{p},\mathbf{p}'}^{\pm}(E)\Im_{E,0}^{\pm}(\mathbf{p}, \mathbf{p}'), \tag{20}$$

with the matrix elements of the free-space Green operator $\Im_{E,0}$ given in the wave-number representation by

$$\Im_{E,0}^{\pm}(\mathbf{p}, \mathbf{p}') = (\xi_\pm{}^2 - p^2)^{-1}\delta(\mathbf{p} - \mathbf{p}'). \tag{21}$$

Then the desired matrix elements are found to be

$$t_{\mathbf{p},\mathbf{p}'}(E^\pm) = [\Im_{E,0}^{\pm}(\mathbf{p})]^{-1}\Im_E^{\pm}(\mathbf{p}, \mathbf{p}')[\Im_{E,0}^{\pm}(\mathbf{p}')]^{-1} - \delta(\mathbf{p} - \mathbf{p}')[\Im_{E,0}^{\pm}(\mathbf{p})]^{-1}. \tag{22}$$

In order to calculate the fill Green function $\Im_E^{\pm}(\mathbf{p}, \mathbf{p}')$, we find it convenient to solve for $\Im_E^{\pm}(\mathbf{r}, \mathbf{p}')$, which satisfies the differential equations

$$(\nabla^2 + \xi_\pm{}^2)\Im_{E,\mathrm{outside}}^{\pm}(\mathbf{r}, \mathbf{p}') = \exp(i\mathbf{p}'\mathbf{r}), \tag{23}$$
$$(\nabla^2 + M^2\xi_\pm{}^2)\Im_{E,\mathrm{inside}}^{\pm}(\mathbf{r}, \mathbf{p}') = \exp(i\mathbf{p}'\mathbf{r}) \tag{24}$$

with the boundary conditions given by Eqs. (19) and then to perform the Fourier transform with respect to the remaining space variable. For a dielectric sphere of index of refraction M and radius a one obtains, after lengthy but straightforward calculations,

$$\begin{aligned} t_{\mathbf{p},\mathbf{p}'}(E^\pm) &= G_{E,0}{}^{-1}(\mathbf{p}) \times \frac{4\pi a^2 j_1(|\mathbf{p} - \mathbf{p}'|)(1 - M^2)\xi_\pm{}^2}{|\mathbf{p} - \mathbf{p}'|(M^2\xi_\pm{}^2 - p'^2)} + G_{E,0}{}^{-1}(\mathbf{p}) \\ &\times \frac{4\pi a^2(M^2 - 1)\xi_\pm{}^2}{(M^2\xi_\pm{}^2 - p'^2)(M^2\xi_\pm{}^2 - p^2)}\sum_l (2l+1) \\ &\times \frac{j_l'(pa)h_l^{(\pm)}(\xi_\pm a) - j_l(pa)h_l^{(\pm)\prime}(\xi_\pm a)}{Mj_l'(M\xi_\pm a)h_l^{(\pm)}(\xi_\pm a) - j_l(M\xi_\pm a)h_l^{(\pm)\prime}(\xi_\pm a)} \\ &\times P_l(\cos\theta)[M\xi_\pm j_{l+1}(M\xi_\pm a)j_l(pa) \\ &- pj_l(M\xi_\pm a)j_{l+1}(pa)] + \frac{4\pi a^2(M^2 - 1)\xi_\pm{}^2}{(M^2\xi_\pm{}^2 - p'^2)} \\ &\times \sum_l \frac{Mj_l(pa)j_l'(M\xi_\pm a) - j_l'(pa)j_l(M\xi_\pm a)}{Mj_l'(M\xi_\pm a)h_l^{(\pm)}(\xi_\pm a) - j_l(M\xi_\pm a)h_l^{(\pm)\prime}(\xi_\pm a)} \\ &\times (2l+1)P_l(\cos\theta)[\xi_\pm h_{l+1}^{(\pm)}(\xi_\pm a)j_l(pa) \\ &- ph_l^{(\pm)}(\xi_\pm a)j_{l+1}(pa)], \end{aligned} \tag{25}$$

where $j_l(x)$ are spherical Bessel functions of lth order, $h_l^{(\pm)}(x)$ are spherical Neumann functions of the first $[h_l^{(+)}]$ and the second $[h_l^{(-)}]$ kind, $P_l(\mu)$ is the Legendre polynomial of lth order, and $j_l'(x) = \mathrm{d}j_l(x)/\mathrm{d}x$. It is worthwhile to mention that when the on-shell limit is used in Eq. (25) the well-known result[22] for scalar Mie scatterers is reproduced.

4. DIFFUSION CONSTANT FOR SCALAR WAVES

After performing the angular averaging in Eqs. (16), we obtain with the help of Eq. (25) expressions for Δ and a for the case of scalar waves:

$$\begin{aligned}\Delta(x) \equiv & -4 - \frac{3}{x^2}\Bigg(\sum_{l=0}^{\infty} 4(l+1)^2 \operatorname{Im}[b_{l+1}^*(x) b_l(x)] \\ & - \sum_{l=0}^{\infty}(2l+1)\Bigg\{x \operatorname{Re}[b_l(x)] \times \Bigg\langle P_l(\mu)(1+\mu) \\ & \times \frac{j_1[2x(\sin\theta)/2]}{(\sin\theta)/2}\Bigg\rangle_\mu + \operatorname{Im}[B_l(x)] \\ & + \operatorname{Im}[B_l^*(x) b_l(x)]\Bigg\} + \sum_{l=0}^{\infty}(l+1)\operatorname{Im}[B_{l+1}(x) b_l^*(x) \\ & + B_l(x) b_{l+1}^*(x)]\Bigg), \\ a(x) = & -1 - \frac{3}{2x^2}\Bigg(\sum_{l=0}^{\infty}(2l+1)\operatorname{Im}[A_l(x)] + \frac{3}{x^2} \\ & \times \sum_{l=0}^{\infty}(2l+1)\{\operatorname{Re}[b_l(x)]\operatorname{Im}[A_l(x)] \\ & - \operatorname{Re}[A_l(x)]\operatorname{Im}[b_l(x)] \\ & + \frac{3}{x}\sum_{l=0}^{\infty}(2l+1)\operatorname{Re}[b_l(x)] \\ & \times \Bigg\langle P_l(\cos\theta)\,\frac{j_1[2x(\sin\theta)/2]}{(\sin\theta)/2}\Bigg\rangle_\mu\Bigg),\end{aligned} \tag{26}$$

where R is the radius of the scatterer and $x = pR$ is the size parameter. The coefficients A_l and B_l are defined as

$$\begin{aligned}A_l(x) = & -\frac{4b_l(x)}{(M^2-1)x} + \frac{\partial b_l(x,y)}{\partial y}\bigg|_{y=x} + ib_l(x)[xh_l^{(-)}(x)j_l(x) \\ & + xh_{l+1}^{(-)}(x)j_{l+1}(x) \\ & - (l+1)h_{l+1}^{(-)}(x)j_l(x) - lh_l^{(-)}(x)j_{l+1}(x)], \\ B_l(x) = & \frac{4b_l(x)}{(M^2-1)x} + \frac{\partial b_l(x,y)}{\partial x}\bigg|_{y=x} + ib_l(x)[xh_l^{(-)}(x)j_l(x) \\ & + xh_{l+1}^{(-)}(x)j_{l+1}(x) \\ & - (l+1)h_l^{(-)}(x)j_{l+1}(x) - lh_{l+1}^{(-)}(x)j_l(x)].\end{aligned} \tag{27}$$

The function $b_l(x, y)$ is given by

$$b_l(x, y) = \frac{Mj_l'(My)j_l(x) - j_l(My)j_l'(x)}{Mj_l'(My)h_l^{(\pm)}(y) - j_l(My)h_l^{(\pm)\prime}(y)} \tag{28}$$

and can be recognized as a van de Hulst coefficient for the TE mode of the vector Mie sphere[22] when $y = x$. Below we attempt a numerical comparison between Eqs. (26)–(28) and the previously obtained results[1–3] for the scattering by scalar Mie spheres. We would like to mention that our results are also valid for acoustic waves

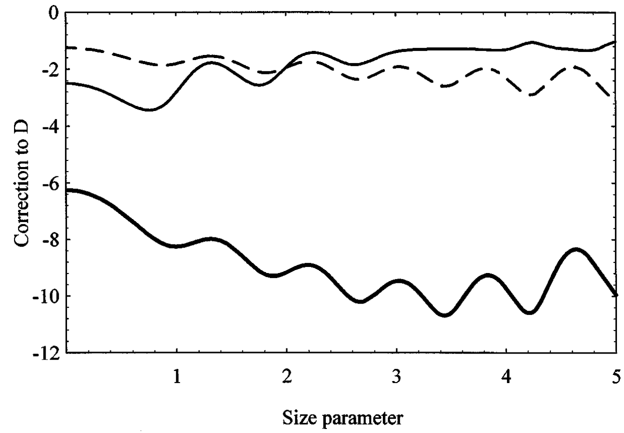

Fig. 1. Comparison among corrections to the diffusion constant as a function of size parameter x for $M = 1.5$: thick curve, correction calculated in this paper; dashed curve, correction $a(x)$ obtained by the Amsterdam group[1–3]; thin curve, $\Delta(x)$, correction obtained by Cwilich and Fu.[4]

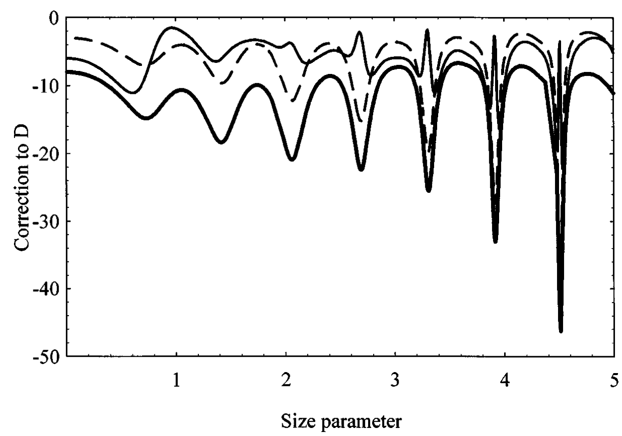

Fig. 2. Same comparison as in Fig. 1 but for $M = 2$. In this case, in contrast to Fig. 1, all three corrections show a definite structure with spikes at frequencies close to the internal resonances of the scatterers.

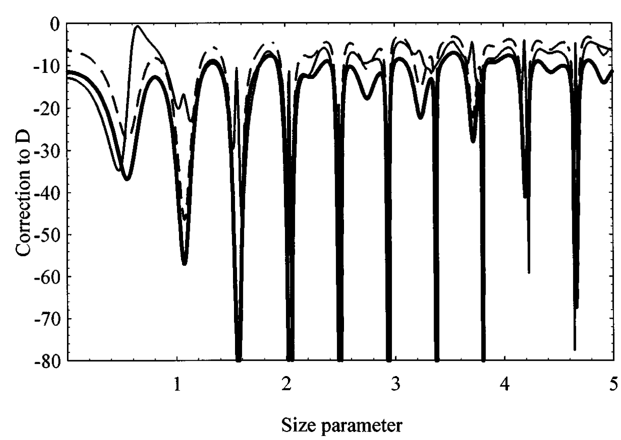

Fig. 3. Same comparison as in Fig. 1 but for $M = 2.73$. In order to provide a good resolution of the functional behavior of all corrections, we have omitted the principal Mie resonances that are in the form of narrow spikes with an amplitude as large as 700.

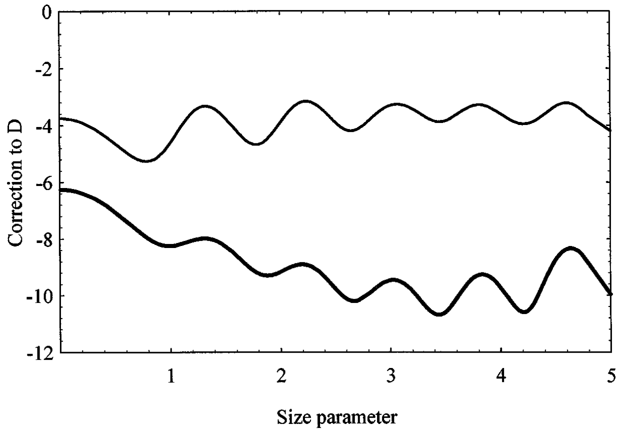

Fig. 4. Our correction to the diffusion constant in the off-shell (thick curve) and on-shell (thin curve) approximations for $M = 1.5$.

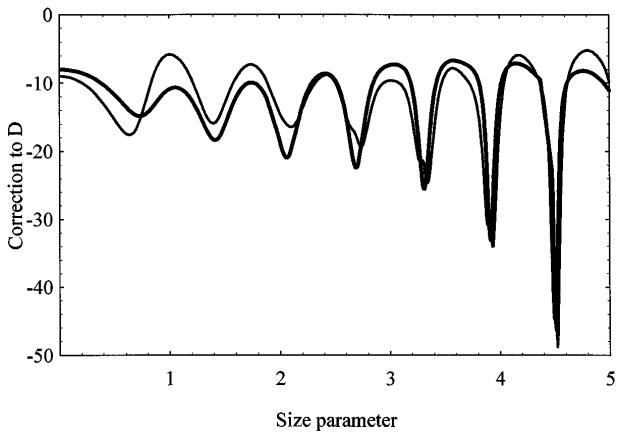

Fig. 5. Same corrections as in Fig. 4 but for $M = 2$.

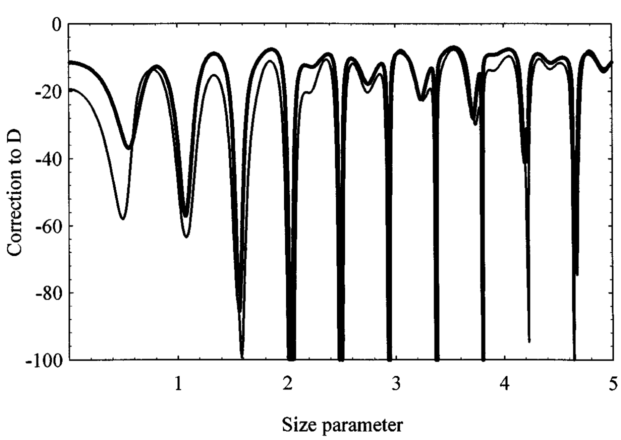

Fig. 6. Same corrections as in Fig. 4 but for $M = 2.73$.

in the hydrodynamic media considered in Ref. 4 for the case $M = z$, where z is an impedance.

Three corrections to the diffusion constant—$\alpha(x)$, obtained by the Amsterdam group[1–3]; $\Delta(x)$, obtained by Cwilich and Fu[4]; and our correction, given by Eqs. (16)—are shown in Figs. 1–3. It is important to point out that the on-shell t matrix has been used in the evaluation of both $\alpha(x)$ (Refs. 1–3) and $\Delta(x)$ (Ref. 4), whereas Eqs. (26), implementing the off-shell t matrix, have been employed in the evaluation of our correction. To make a thorough comparison among different corrections, the results in Figs. 1–3 have been plotted not only for the same value of $M = 2.73$ (Fig. 3) as in Refs. 1–3 but also for $M = 1.5$ (Fig. 1) and for $M = 2$ (Fig. 2). The last choice is rather arbitrary and is based only on the fact that for $M > 2$ all three corrections exhibit strongly resonant behavior that makes a detailed comparison among different corrections difficult. One the other hand, one would expect Mie resonances to be washed out for values of $M < 2$, as can be seen in Fig. 1. It is evident from the figures that, despite the similarities in the vicinity of the principal Mie resonances at $x \approx 1, 1.5, 2, 2.5, 3, 3.5, 4$, and 4.5, our correction exhibits different functional behavior from that previously known. Moreover, the magnitude of our correction for the principal Mie resonances is much larger than the magnitude of both $\alpha(x)$ and $\Delta(x)$. For example, for the fourth Mie resonance located at $x = 2$ ($M = 2.73$) the magnitude of our correction is of the order of 700, whereas the magnitude of $\alpha(x)$ is of the order of 500.

The importance of the off-shell approximation is demonstrated in Figs. 4–6, where we have plotted our correction calculated with the help of on-shell (thin line) and off-shell (thick line) t matrices for the same values of M as those in Figs. 1–3. The changes caused by the off-shell approximation for the transfer matrix are significant for all values of x and for all three indices of refraction. This effect can be attributed to the specific structure of the derivative of $b_l(p, \xi)$ with respect to p involved in $\Delta(x)$, in contrast to that of $\partial b_l(p, \xi)/\partial \xi$ at constant p involved in $\alpha(x)$. The energy derivative contains differentiation of both the numerator and the denominator of the van de Hulst coefficient and therefore is proportional to $\partial b_l(x)/\partial x$, which leads to sharp resonances in $\alpha(x)$. The derivative with respect to p involves only the numerator of $b_l(p, \xi)$, and it can be shown to be proportional to $b_l(x)$. The magnitudes of the resonances in $b_l(x)$ are much smaller than those in $\partial b_l(x)/\partial x$ for $M > 2$ (both terms can be of the same order for $M < 2$), and they are much less sensitive to the value of the index of refraction. In the case of the on-shell approximation, however, $\Delta(x)$ depends on $\partial b_l(x)/\partial x$ and therefore exhibits resonances as strong as those of $\alpha(x)$. As a result, the functional behavior of the total correction to D, $\alpha(x) + \Delta(x)$, is significantly altered.

5. CONCLUSION

In conclusion, we have calculated the general expression for the renormalization of the diffusion coefficient for classical waves propagating in a random medium with microstructural resonances. The diffusion constant is estimated in the low-density limit for scalar waves. Numerical calculations require the off-shell scattering matrix, which is obtained. The renormalization terms show a functional behavior significantly different from that obtained by other authors, where the on-shell matrix was implemented. The importance of the off-shell approximation for the evaluation of the diffusion constant is demonstrated. The ultimate goal is to make a comparison of the obtained results with experimental data (see, for example, Ref. 21). However, the comparison of our results with experiment is limited by the low-density approximation used in the calculations. When propagation occurs in a medium with a high relative index of refraction, both renormalization terms can become large (see Figs. 3 and 6), thus requiring low enough densities to provide the condition $|D - D_0|/D \ll 1$ that ensures the applicability of the low-density approximation. However, the filling fractions used in published experiments range from 15% to 35%, which are too big to satisfy the low-density approximation in the vicinity of resonances. Thus Eqs. (16) cannot be used for direct comparison between theory and experiment, which is possible only if higher powers in the density of scatterers are taken into account.

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