

Transport properties of waves in absorbing random media with microstructure

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Diffusion of waves in absorbing random media with microstructure is studied. A resonant correction to the transport mean free path l_T is calculated. It is shown that in the certain range of wavelengths this correction leads to a growth of the l_T compared to its nonresonant value. Significant changes in the transport velocity, the transport mean free path, and the diffusion constant caused by absorption are also found. In addition, we show that the use of the on-shell transfer matrix can lead to erroneous results. [S0163-1829(96)03922-7]

I. INTRODUCTION

In a medium with randomly positioned anisotropic scatterers a wave propagates a distance of the order of the transport mean free path l_T before it forgets completely about its initial direction of propagation. In the weakly scattering regime, when the wavelength is much smaller than l_T , wave intensity satisfies the diffusion equation with the rate of flow given by the diffusion coefficient D . At the same time the ‘‘energy’’ flux spreads through the medium with a transport velocity v_E . These three parameters fully characterize wave transport through a random medium and are interconnected by the relation $D = l_T v_E / 3$.¹⁻³ If scatterers are treated as pointlike, one finds that v_E is equal to the phase velocity c_p and $l_T = l_{sc} / (1 - \langle \cos \theta \rangle)$, where $l_{sc} = (n\sigma)^{-1}$ is the scattering mean free path, n is a density of scatterers, σ is a scattering cross section and θ is a scattering angle. Properties of the transport parameters may be notably changed by an account for the microstructure of the medium. It has been shown by the Amsterdam group⁴⁻⁷ (AG) that when scatterers of a finite size are considered v_E is no longer equal to the phase velocity and depends upon the wave vector of the incident wave k_0 . They find that in the low-density limit $v_E \approx c_p / [1 + na(k_0)]$. The correction $a(k_0)$ exhibits strong resonant structure being studied as a function of k_0 , and leads to the renormalization of the diffusion constant. It has been deduced by the AG that these resonances in the correction to v_E result from Mie resonances⁸ in the transfer matrix of waves scattered by finite-size scatterers. Analogous corrections have been obtained by Cwilich and Fu⁹ and by Kogan and Kaveh.¹⁰

In the present paper we show that there is another source of the renormalization of D . Microstructure resonances lead not only to the correction to the transport velocity but to the correction to the transport mean free path as well. It is intuitively clear that such a correction to l_T should occur, since Mie resonances provide an additional anisotropy of the scattering matrix and, therefore, should modify $\langle \cos \theta \rangle$. We would like to outline here why this correction has never been accounted for. The transport mean free path can be found

once the diffusion constant and the energy velocity are known. The diffusion constant can be evaluated from the diffusive pole of the average intensity of waves $I(\mathbf{q}, \omega)$ where \mathbf{q} and ω are the transferred momentum and frequency, respectively. $I(\mathbf{q}, \omega)$ itself is calculated from the Bethe-Salpeter (BS) equation. To find a diffusion constant one has to perform an expansion in the BS equation up to the lowest order of \mathbf{q} and ω , since in the diffusion approximation $\mathbf{q}, \omega \rightarrow 0$. Then, terms collected near the lowest order of ω provide the correction $a(k_0)$. It has, however, been concluded by AG that all terms in \mathbf{q} expansion cancel after an application of the energy conservation law in the form of the Ward identity (WI). In the present study we explicitly show that WI cannot be used to cancel terms near the lowest order of \mathbf{q} . We identify these terms as a correction to the transport mean free path.

The effect of absorption on resonant corrections to the diffusion constant is another important issue of the present study. Since in most experiments absorption is weak, it has been usually neglected.^{4-7,9-14} However, the underestimation of the role of absorption may be misleading. We show, that even weak absorption leads to significant changes in the functional behavior of both corrections.

We also argue that the application of the on-shell approximation for a transfer matrix of an individual scatterer can lead to physically incorrect results. The correction $a(k_0)$ contains partial derivatives of the transfer matrix $t_{\mathbf{k}\mathbf{k}'}(E)$ with respect to frequency E , while the correction to l_T has analogous derivatives but with respect to \mathbf{k} . Both derivatives are evaluated on the energy ‘‘shell’’ $|\mathbf{k}| = |k_0| = E/c_p$. Since the density of scatterers is low, the substitution $\partial/\partial E \approx \partial/(c_p \partial k) + O(n)$ is usually made^{4-7,9-13} and, thus, the on-shell transfer matrix is used in calculations. However, we have recently argued that this assumption is incorrect.¹⁴ For example, we show below that within the on-shell approximation absorption can lead to the growth of the diffusion constant that can become larger than its nonresonant value. This effect increases with increasing of absorption in the medium. Such unphysical prediction does not occur when the off-shell approximation is applied.

II. DERIVATION OF THE GENERAL EXPRESSION FOR D

In the present section we outline a general method for obtaining the diffusion constant of classical waves in random media since it is important to understand from where different terms in the expression for D come from. We consider the Fourier transform of the scalar field $\psi(\mathbf{r}, t)$ satisfying the wave equation,

$$\left\{ \nabla^2 + \left(\frac{E}{c} \right)^2 \epsilon(\mathbf{r}) \right\} \psi(\mathbf{r}, E) = 0, \quad (1)$$

where c is the wave speed in a space free of scatterers. The wave propagates in an infinite absorbing random medium.

The dielectric constant $\epsilon(\mathbf{r})$ comprises the properties of individual scatterers randomly distributed inside the medium. The statistical properties of the dielectric constant as a function of the position vector \mathbf{r} will be specified later in this section. Field generated at any point \mathbf{r} in space by the point source located at \mathbf{r}_0 can be expressed in terms of the Green function of Eq. (1), $\psi(\mathbf{r}, t) = G(\mathbf{r}, \mathbf{r}_0, t)$. As a result of macroscopic homogeneity, the function $|G(\mathbf{r}, \mathbf{r}', t)|^2$ averaged over disorder has translational invariance, i.e., it depends upon $|\mathbf{r} - \mathbf{r}'|$ only, and $\langle |G(\mathbf{r}, \mathbf{r}', t)|^2 \rangle$ is the wave intensity $I(\mathbf{r} - \mathbf{r}', t)$ due to a point source at \mathbf{r}' . In the weakly scattering regime the space-time Fourier transform of the disorder averaged intensity $I(\mathbf{q}, \omega)$, defined as

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

must have a diffusive pole as $\omega, \mathbf{q} \rightarrow 0$ (e.g., $I(\mathbf{q}, \omega) \propto [-i\omega + D\mathbf{q}^2]^{-1}$). In Eq. (2) the notations $\mathbf{k}_\pm = \mathbf{k} \pm \mathbf{q}/2$ and $E^\pm = E \pm \omega/2 \pm i0$ were introduced. From this pole one can evaluate the diffusion constant. The quantity of our primary interest, however, is not $I(\mathbf{q}, \omega)$ but rather the function $\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E)$, which on one hand, inherits all the analytical properties of $I(\mathbf{q}, \omega)$ including the diffusive pole, and, on the other, an exact form of the equation it satisfies is known.

The function $\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E)$ can be expressed in terms of the Fourier transform of the averaged one-field Green function via the BS equation^{1,2}

$$\begin{aligned} \left[-\frac{2E\omega}{c^2} + 2\mathbf{q} \cdot \mathbf{k} \right] \Phi_{\mathbf{k}}(\mathbf{q}, \omega; E) \\ = \Delta G_{\mathbf{k}}(\mathbf{q}, \omega) + \int_{\mathbf{k}'} U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega) \Phi_{\mathbf{k}'}(\mathbf{q}, \omega), \end{aligned} \quad (3)$$

where disorder averaged *retarded* $G_{\mathbf{k}}^-$, and *advanced* $G_{\mathbf{k}}^+$, Green functions have the form

$$\begin{aligned} G_{\mathbf{k}}^\pm(E^\pm) &\equiv \langle G_{\mathbf{k}, \mathbf{k}}^\pm(E^\pm) \rangle \\ &= \delta(\mathbf{k} - \mathbf{k}') \left[\left(\frac{E^\pm}{c} \right)^2 - \mathbf{k}^2 - \Sigma_{\mathbf{k}}^\pm(E^\pm) \right]^{-1}. \end{aligned} \quad (4)$$

Here $\Sigma_{\mathbf{k}}^\pm$ is a self-energy and we denote $\int_{\mathbf{k}} = (2\pi)^{-3} \int d\mathbf{k}$, $\Delta G_{\mathbf{k}}(\mathbf{q}, \omega; E) \equiv G_{\mathbf{k}_+}^+(E^+) - G_{\mathbf{k}_-}^-(E^-)$. All possible scattering processes are included in the kernel $U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega)$, which has the following form:

$$U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega) = \Delta G_{\mathbf{k}}(\mathbf{q}, \omega) K_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega) - \Delta \Sigma_{\mathbf{k}}(\mathbf{q}, \omega) \delta_{\mathbf{k}, \mathbf{k}'}, \quad (5)$$

with $\Delta \Sigma_{\mathbf{k}} = \Delta_{\mathbf{k}_+}(E^+) - \Delta_{\mathbf{k}_-}(E^-)$. In the case of classical waves the irreducible vertex function $K_{\mathbf{k}\mathbf{k}'}$ is related to the self-energy $\Sigma_{\mathbf{k}}$ by the *generalized Ward identity*¹⁵

$$-\frac{2\omega E}{c^2} A_{\mathbf{k}}(\mathbf{q}, \omega) = \int_{\mathbf{k}'} U_{\mathbf{k}'\mathbf{k}}(\mathbf{q}, \omega) \quad (6)$$

with

$$\begin{aligned} A_{\mathbf{k}}(\mathbf{q}, \omega) &= \frac{c^2}{2E^2} \left\{ \Sigma_{\mathbf{k}_+}(E^+) + \Sigma_{\mathbf{k}_-}(E^-) + \int_{\mathbf{k}'} K_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega) \right. \\ &\quad \left. \times [G_{\mathbf{k}'_+}^+(E^+) + G_{\mathbf{k}'_-}^-(E^-)] \right\}. \end{aligned} \quad (7)$$

It is easy to see that for $\mathbf{q}, \omega = 0$, Eq. (6) reduces to the optical theorem for monochromatic light.^{1,2}

The structure of the BS equation suggests that the function $\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E)$ should have isotropic and anisotropic parts. Based on this, it is convenient to start the solution of the BS equation with the definition of a function

$$P_E(\mathbf{q}, \omega) = \sum_{\mathbf{k}} \Phi_{\mathbf{k}}(\mathbf{q}, \omega; E), \quad (8)$$

which is isotropic and may be regarded as the Fourier transform of the “ E component” of the averaged intensity excited at \mathbf{r}' at $t=0$. The anisotropic properties of the average intensity can be described by the “correlation current” $J_E(\mathbf{q}, \omega)$

$$J_E(\mathbf{q}, \omega) = \sum_{\mathbf{k}} (\mathbf{k} \cdot \mathbf{q}) \Phi_{\mathbf{k}}(\mathbf{q}, \omega; E). \quad (9)$$

The next step is to derive a system of equations that would relate these two functions and then solve for P_E . After integration of the Eq. (3) and application of the WI in the form of Eq. (4) we obtain the continuity equation

$$-\frac{E\omega}{c^2} P_E + J_E + \frac{E\omega}{c^2} \int_{\mathbf{k}} A_{\mathbf{k}}(\mathbf{q}, \omega) \Phi_{\mathbf{k}}(\mathbf{q}, \omega) = -\frac{ik_0}{4\pi}, \quad (10)$$

where we have used the fact that $\int_{\mathbf{k}} \Delta G_{\mathbf{k}} = -ik_0/2\pi$. In order to deal with the integral in the left-hand side of Eq. (10) we can make use of the diffusion approximation ($\mathbf{q} \rightarrow 0$) and expand $\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E)$ to the first order in \mathbf{q}

$$\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E) = \Delta G_{\mathbf{k}}(0, 0) [A_{\mathbf{k}} + (\mathbf{k} \cdot \mathbf{q}) B_{\mathbf{k}}]. \quad (11)$$

Integration of Eq. (11) with respect to \mathbf{k} yields the ‘‘isotropic’’ coefficients $A_{\mathbf{k}}$. Multiplying both sides of Eq. (11) by $(\mathbf{k} \cdot \mathbf{q})$ and then integrating with respect to \mathbf{k} gives the ‘‘anisotropic’’ coefficient $B_{\mathbf{k}}$

$$A_{\mathbf{k}} = \frac{2\pi i}{k_0} \Delta G_{\mathbf{k}}(0, 0) P_E; \quad (12)$$

$$B_{\mathbf{k}} = \frac{2\pi i}{k_0} \left(\frac{3\mathbf{k} \cdot \mathbf{q}}{k^2 q^2} \right) \Delta G_{\mathbf{k}}(0, 0) J_E.$$

Combining Eqs. (12) with Eq. (11) gives

$$\Phi_{\mathbf{k}}(\mathbf{q}, \omega; E) = \frac{2\pi i}{k_0} \Delta G_{\mathbf{k}}(0, 0) \left\{ P_E + \frac{3\mathbf{k} \cdot \mathbf{q}}{k^2 q^2} J_E \right\}. \quad (13)$$

Substituting Eq. (13) back into Eq. (10) and expanding all terms in powers of ω and \mathbf{q} up to the lowest order, we find the continuity equation

$$J_E - \frac{E\omega}{c^2} [1 + a(E)] P_E = -\frac{ik_0}{4\pi}, \quad (14)$$

where the function $a(E)$ represents a correction to the wave velocity found by AG.⁴⁻⁷ It is defined as

$$a(E) = -\frac{2\pi i c_p}{E} \int_{\mathbf{k}} A_{\mathbf{k}}(0, 0) \Delta G_{\mathbf{k}}(0, 0). \quad (15)$$

Here the phase velocity in the medium is equal to $c_p = c(1 - \text{Re} \Sigma_{\mathbf{k}}^+ / k_0)^{-1/2}$. We now have to derive an equation for the current J_E . We multiply Eq. (3) by $\mathbf{k} \cdot \mathbf{q}$ and integrate it with respect to \mathbf{k} using the relation (11) to obtain

$$J_E = \frac{6\pi i}{k_0} \int_{\mathbf{k}} \int_{\mathbf{k}'} \frac{(\mathbf{k} \cdot \mathbf{q})(\mathbf{k}' \cdot \mathbf{q})}{k'^2 q^2} U_{\mathbf{k}\mathbf{k}'}(0, 0) \Delta G_{\mathbf{k}'}(0, 0) \\ = P_E \left\{ \frac{2}{3} k_0^2 q^2 - \frac{2\pi i}{k_0} \int_{\mathbf{k}} \int_{\mathbf{k}'} (\mathbf{k} \cdot \mathbf{q}) U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, 0) \Delta G_{\mathbf{k}'}(0, 0) \right\}. \quad (16)$$

The second term on the right-hand side of Eq. (16) is important in the calculation of the diffusion constant. It has been concluded by the Amsterdam group⁴⁻⁷ and Barabanenkova and Ozrin^{11,15} that after application of the generalized WI in Eq. (16) this term is *exactly* equal to zero. If it would have the form $\int_{\mathbf{k}} \int_{\mathbf{k}'} (\mathbf{k} \cdot \mathbf{q}) U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, 0) \Delta G_{\mathbf{k}'}(\mathbf{q}, 0)$ the statement made in Refs. 4-7, 11, and 15 would be correct. It is, however, *impossible* to use WI in the second term of the right-hand side of the Eq. (16) due to appearance of $\Delta G_{\mathbf{k}}(0, 0)$

instead of $\Delta G_{\mathbf{k}}(\mathbf{q}, 0)$ in it. Instead, one can expand $U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, 0)$ up to the lowest power of \mathbf{q} and then solve Eqs. (14) and (16) for P_E yielding

$$P_E = \frac{c^2}{4\pi c_p [1 + a(E)]} \frac{1}{-i\omega + Dq^2}, \quad (17)$$

where the diffusion coefficient has the form

$$D = D_0 \left(\frac{c}{c_p} \right)^2 \frac{1 + \Delta(E)}{1 + a(E)}, \quad (18)$$

and the bare diffusion constant D_0 is given by

$$D_0 = \frac{1}{3} c_p \left\{ \frac{3\pi}{k_0^2} \int_{\mathbf{k}} \int_{\mathbf{k}'} K_{\mathbf{k}\mathbf{k}'}(0, 0) \Delta G_{\mathbf{k}}(0, 0) \Delta G_{\mathbf{k}'}(0, 0) \right. \\ \left. \times \frac{(\mathbf{k} \cdot \mathbf{q})(\mathbf{k}' \cdot \mathbf{q})}{k^2} [1 - \delta_{\mathbf{k}, \mathbf{k}'}] \right\}^{-1}. \quad (19)$$

The expression in curly brackets in Eq. (19) represents a conventional transport mean free path l_T .¹⁶ $\Delta(E)$ is an additional correction to the diffusion constant. It can be written as

$$\Delta(E) = -\frac{3\pi i}{k_0} \int_{\mathbf{k}} \int_{\mathbf{k}'} \frac{(\mathbf{k} \cdot \mathbf{q})}{k_0^2 q^2} \left(\frac{\partial U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}', 0)}{\partial \mathbf{q}'} \Big|_{\mathbf{q}'=0} \cdot \mathbf{q} \right) \\ \times \Delta G_{\mathbf{k}'}(0, 0). \quad (20)$$

An analogous renormalization factor for electrons has been found by Burin.¹⁷ The interpretation of the Eq. (18) is transparent enough. The diffusion coefficient for waves in any kind of random medium can be expressed as $D = \nu_E l/3$, where l is a general form of a transport mean free path, different from the conventional l_T given by Eq. (19). Lagendijk and van Tiggelen⁴⁻⁷ have shown that the speed ν_E in the case of classical waves is equal to

$$\nu_E(E) = \frac{c^2}{c_p} \frac{1}{1 + a(E)}. \quad (21)$$

The correction $\Delta(E)$ then provides a renormalization of the transport mean free path,

$$l(E) = l_T [1 + \Delta(E)]. \quad (22)$$

III. RESULTS AND DISCUSSION

In order to perform detailed investigations of the diffusion coefficient in the form of Eq. (18) an exact analytical form of the kernel $U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, 0)$ must be known. The limit of low densities of scatterers is a good approximation to use to find $U_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, 0)$, since it is realized in most experimental setups.^{4,18} In this approximation the self-energy and the irreducible vertex are expressed in terms of the scattering matrix for individual scatterer $t_{\mathbf{k}\mathbf{k}'}$ and density of scatterers n as $\Sigma_{\mathbf{k}}^{\pm}(\mathbf{k}_{\pm}, \omega; E^{\pm}) = n t_{\mathbf{k}_{\pm}, \mathbf{k}_{\pm}}(E^{\pm})$ and $K_{\mathbf{k}\mathbf{k}'}(\mathbf{q}, \omega) = n t_{\mathbf{k}_{+}, \mathbf{k}_{+}}(E^{+}) t_{\mathbf{k}'_{-}, \mathbf{k}'_{-}}(E^{-})$. The density of scatterers n must be small enough to allow the weak scattering approximation to be valid. Then, the expressions for corrections $a(E)$ and $\Delta(E)$ in the low-density limit are

$$\Delta(E) = n \frac{\partial \operatorname{Re} t_{\mathbf{k},\mathbf{k}}}{\partial k^2} + n \frac{iE}{4\pi c_p} \langle (1 + \mu) K(\mathbf{k}, \mathbf{k}') \rangle_\mu,$$

$$a(E) = n \frac{\partial \operatorname{Re} t_{\mathbf{k},\mathbf{k}}}{\partial E^2} - in \sum_{\mathbf{k}'} \Delta G_{\mathbf{k}'} |t_{\mathbf{k},\mathbf{k}'}|^2 \frac{\partial \phi_{\mathbf{k},\mathbf{k}'}}{\partial E^2} \quad (23)$$

with

$$K(\mathbf{k}, \mathbf{k}') = i \operatorname{Im} \left\{ \left[\frac{\partial t_{\mathbf{k},\mathbf{k}'}}{\partial k^2} + \frac{1 - \mu}{k^2} \frac{\partial t_{\mathbf{k},\mathbf{k}'}}{\partial \mu} \right] t_{\mathbf{k},\mathbf{k}'}^* \right\}, \quad (24)$$

where $\mu \equiv \mathbf{k} \cdot \mathbf{k}' / k^2$ is a cosine of the scattering angle. In deriving these equations it has been taken into account that in

the case of elastic collision, the scattering matrix $t_{\mathbf{k},\mathbf{k}'}$ depends on the modulus of the momenta $|\mathbf{k}|^2 = |\mathbf{k}'|^2$ and on the cosine of the scattering angle $\mu = \mathbf{k} \cdot \mathbf{k}' / k^2$ only. We have also denoted the phase shift of the scattering matrix as $\phi_{\mathbf{k},\mathbf{k}'}(E)$ according to $t_{\mathbf{k},\mathbf{k}'}(E^+) = |t_{\mathbf{k},\mathbf{k}'}| \exp(i\phi_{\mathbf{k},\mathbf{k}'})$. In the low-density approximation both $\Delta(E)$ and $a(E)$ become true corrections of the order of n to D_0 . Since $\Delta(E)$ and $a(E)$ contain partial derivatives with respect to k and E , respectively, the *off-shell* transfer matrix has to be used in calculations. An exact form of the off-shell transfer matrix for a dielectric sphere of the index of refraction M and radius R is given in Ref. 14,

$$t_{k,k'}(E^\pm) = \frac{4\pi R^2 j_1(|k-k'|)(1-M^2)\xi_\pm^2(\xi_\pm^2-k^2)}{|k-k'|(M^2\xi_\pm^2-k'^2)} + \frac{4\pi R^2(M^2-1)\xi_\pm^2(\xi_\pm^2-k^2)}{(M^2\xi_\pm^2-k'^2)(M^2\xi_\pm^2-k^2)}$$

$$\times \sum_l (2l+1) \frac{j_l'(kR)h_l^{(\mp)}(\xi_\pm R) - j_l(kR)h_l^{(\mp)'}(\xi_\pm R)}{Mj_l'(M\xi_\pm R)h_l^{(\mp)}(\xi_\pm R) - j_l(M\xi_\pm R)h_l^{(\mp)'}(\xi_\pm R)} P_l(\cos\theta) [M\xi_\pm j_{l+1}(M\xi_\pm R)j_l(kR)$$

$$- k j_l(M\xi_\pm R)j_{l+1}(kR)] + \frac{4\pi R^2(M^2-1)\xi_\pm^2}{(M^2\xi_\pm^2-k'^2)} \sum_l \frac{Mj_l(kR)j_l'(M\xi_\pm R) - j_l'(kR)j_l(M\xi_\pm R)}{Mj_l'(M\xi_\pm R)h_l^{(\mp)}(\xi_\pm R) - j_l(M\xi_\pm R)h_l^{(\mp)'}(\xi_\pm R)}$$

$$\times (2l+1)P_l(\cos\theta) [\xi_\pm h_{l+1}^{(\mp)}(\xi_\pm R)j_l(kR) - kh_l^{(\mp)}(\xi_\pm R)j_{l+1}(kR)], \quad (25)$$

where $\xi_\pm = E^\pm / c_p$, $j_l(x)$ are spherical Bessel functions of the l th order, $h_l^{(+)}(x)$ and $h_l^{(-)}(x)$ are spherical Neumann functions of the first and second kind, respectively, $P_l(\mu)$ are Legendre polynomials of the l th order, and $j_l'(x) = dj_l(x)/dx$. A conventional *on-shell* t matrix can be obtained from Eq. (25) by making the substitution $k = k' = \xi_\pm$,

$$t_{k,k'}(E^+) = -\frac{2\pi i}{E} \sum_l (2l+1)P_l(\mu)b_l^*(E), \quad (26)$$

where $b_l(x)$ is a Van de Hulst coefficient for the TE mode of the vector Mie sphere.¹⁹

Equations (23)–(26) give the analytical form of the corrections to the diffusion coefficient. Now we can study the functional behavior of D as a function of a dimensionless size parameter $x = k_0 R$ (below the value of $R = 1$ cm is used). Absorption in the medium can be introduced as an imaginary part of the complex index of refraction $M = M_r - iM_i$. The range of values of M_i which can be used in calculations is, however, limited. Eq. (18) for the diffusion constant is obtained within the diffusion approximation which implies that $l_T/l_a \ll 1$, where $l_a = (2M_r M_i k_0)^{-1}$, otherwise conventional diffusion would break down. Thus, we obtain a condition $M_i \ll R/(2M_r l_T x)$. A minor inconvenience hidden here is that the transport mean free path itself is a function of a size parameter. Thus, for different values of x we obtain different values of M_i satisfying the validity condition. Using the fact that the scattering mean free path is not changed significantly by absorption we have estimated the value of $R/(2M_r l_T x)$ as a function of x for $M = M_r$ and then have used the largest value of M_i that does not violate the above condition. In order to be able to make a comparison with previously ob-

tained results^{4–7,9} we use $M_r = 2.73$ in the evaluations. The appropriate value of M_i that covers the range of the size parameter values $0 \leq x \leq 5$ is $M_i = 0.005$. Our goal, however, is not limited to the study of the effect of absorption on resonances. A demonstration of necessity of the use of the off-shell approximation for the t matrix is an equally important issue. For this reason we show our results for both approximations.

We plot the on-shell and off-shell versions of the total correction to the diffusion constant, given in the case of low density by $(D - D_0)/D_0 \approx -a(x) + \Delta(x)$, in Figs. 1 and 2 respectively, for the relative index of refraction $M = 2.73 - i0.005$ (thick lines). The figures also show the corresponding correction in the absence of absorption $M = 2.73$ (thin lines). We would like to stress here that we consider very weak absorption and even in this case we find significant changes in the corrections to the diffusion constant. The profound difference between two approximations for t matrix can be seen from these graphs. The most striking changes occur in the on-shell correction: it becomes positive at values of the size parameter $x \approx 2, 2.5, 3, 3.5, 4, 4.5$, and 5, which correspond to the principal Mie resonances. On the other hand, even such weak absorption substantially washes off resonances in the off-shell version of the total correction, which, however, remains negative for the whole range of values of the size parameter. In order to understand which correction leads to these changes we will look at properties of $a(x)$ and $\Delta(x)$ separately.

We plot the on-shell version of the correction $a(x)$ in Fig. 3 and the off-shell version of $a(x)$ in Fig. 4 for $M = 2.73 - i0.005$ (thick lines) and $M = 2.73$ (thin lines). It can be

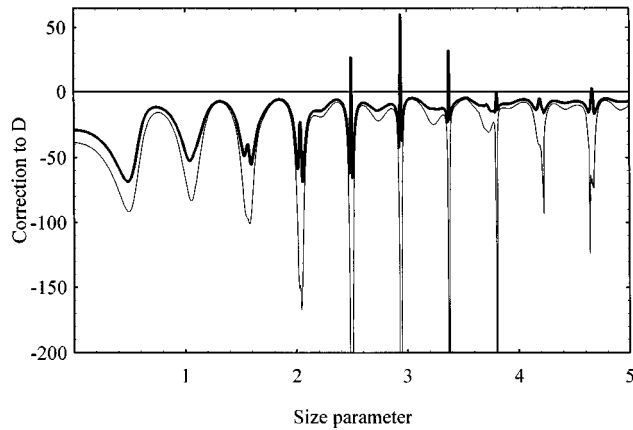


FIG. 1. The total correction to the diffusion constant, $\Delta(x) - a(x)$, calculated within the *on-shell* approximation, with and without absorption as a function of the size parameter. The thin line corresponds to the value of $M=2.73$, the thick line corresponds to the value of $M=2.73-i0.005$. In order to preserve the details of resonances in the presence of absorption we shorten the vertical scale of this and other figures. The magnitude of peaks left out on the figure can be as large as 600. The total correction becomes positive in the presence of absorption for the values of $x \approx 1.5, 2$, and 2.5 .

seen, that the effect of absorption on resonances is similar in both cases. Even weak absorption decreases the magnitude of resonances at least by 10–25 times. On the other hand, the value of $a(x)$ remains positive definite in agreement with the statement of Ref. 7 that the quantity $k_0^2 a(E)$ represents positive definite “potential energy” inside the scatterers.

The effect of absorption on the on-shell (Fig. 5) and off-shell (Fig. 6) versions of the correction $\Delta(x)$ is different. Special attention has to be paid to the off-shell version of $\Delta(x)$ in the absence of absorption. $\Delta(x)$ is positive for the wide range of values of x , thus leading to the growth of the transport mean free path. These changes occur in the vicinity of the first five principal Mie resonances located at values of $x \approx 1, 1.5, 2, 2.5$, and 3 . These peculiarities can be understood if we look at properties of Mie resonances. The transport mean free path given by Eq. (19) can be rewritten in its

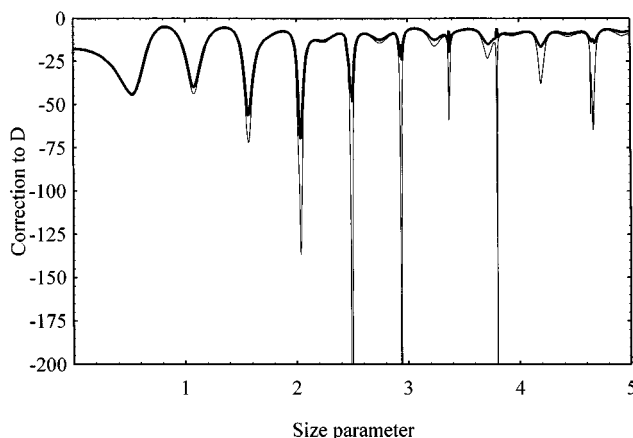


FIG. 2. The same as in Fig. 1 but the total correction is calculated within the *off-shell* approximation. The total correction stays negative in both cases.

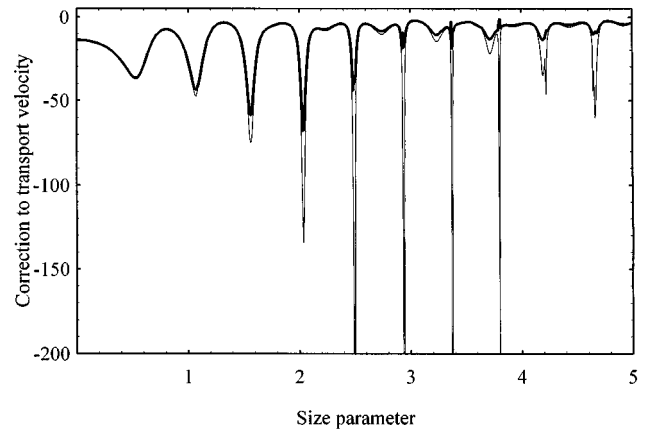


FIG. 3. The correction to the transport velocity $a(x)$ calculated within the *on-shell* approximation for the same values of M as in all previous figures, $M=2.73-i0.005$ (thick line) and $M=2.73$ (thin line).

traditional form $l_T = l_{sc}/(1 - \langle \mu \rangle)$, where l_{sc} is scattering mean free path. It is known that in the vicinity of principal Mie resonances scattering is primarily in the forward direction.¹⁹ Thus, the average cosine of the scattering angle increases leading to the growth of the transport mean free path. Analogous features cannot be found in the on-shell version of $\Delta(x)$. It is always negative and it basically replicates features of $a(x)$ shown by a thin line in Fig. 3. When absorption is introduced in the medium, resonances are washed out in the case of the off-shell version of $\Delta(x)$, which becomes now negative for values of $x=2.5$ and 3 . The functional behavior of the on-shell version of $\Delta(x)$ is altered much more seriously. It becomes positive due to inversion of the principal Mie resonances that is shown in Fig. 6, while the rest of it is hardly affected by absorption.

Let us return to the total correction to the diffusion constant. In the on-shell approximation absorption “stimulates” diffusion since the renormalized value of D becomes greater than D_0 . This result, however, can be hardly justified physically. Contrary, the growth of the transport mean free path in

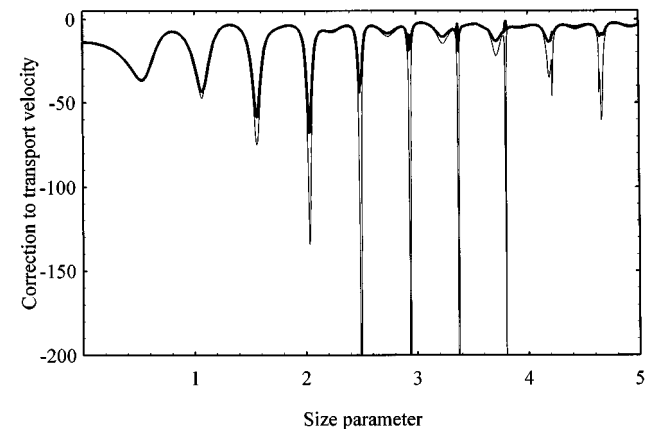


FIG. 4. The correction to the transport velocity $a(x)$ calculated within the *off-shell* approximation for the same values of M as in all previous figures, $M=2.73-i0.005$ (thick line) and $M=2.73$ (thin line). As in the on-shell case of the magnitude of resonances is significantly decreased.

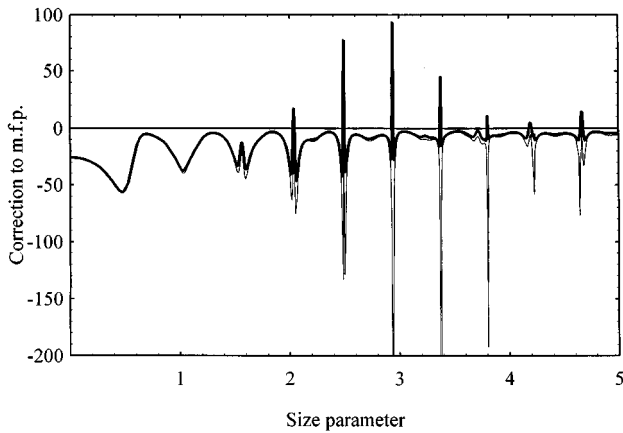


FIG. 5. The correction to the transport mean free path $\Delta(x)$ calculated within the *on-shell* approximation for the same values of M as in all previous figures, $M=2.73-i0.005$ (thick line) and $M=2.73$ (thin line). The principal Mie resonances are inverted by absorption.

the case of the off-shell t matrix is not strong enough to stimulate diffusion in the medium, since $\Delta(x)$ is always less than $a(x)$. Even in the presence of absorption, when $a(x)$ is strongly decreased, $\Delta(x)$ is decreased as well, thus keeping the total correction to D negative. It, therefore, supports our suggestion that it is incorrect to use the on-shell t matrix in the evaluation of the diffusion constant.

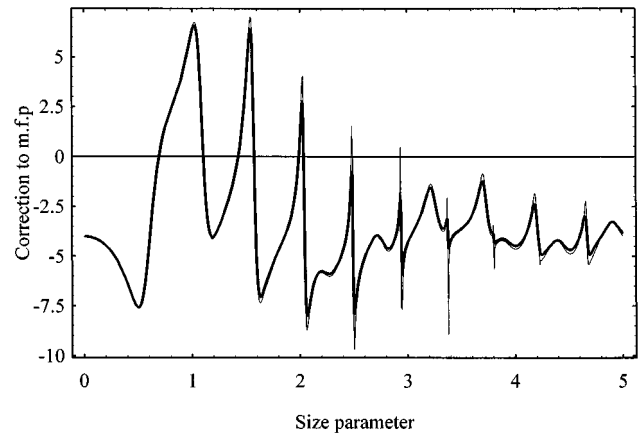


FIG. 6. The correction to the transport mean free path $\Delta(x)$ calculated within the *off-shell* approximation for the same values of M as in all previous figures, $M=2.73-i0.005$ (thick line) and $M=2.73$ (thin line). This correction is positive even in the absence of absorption. In this case absorption decreases the magnitude of resonances.

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