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# Resonantly induced dipole-dipole interactions in the diffusion of scalar waves

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The induced dipole-dipole interaction can be understood in terms of recurrent elastic scattering of waves. We implement this notion into transport theory of scalar waves in inhomogeneous dielectric media and calculate rigorously the pivotal cooperative effect of pairs of polarizable dipoles.

Multiple scattering of classical waves is a field of wide interest. It manifests itself in various different areas of physics. Many experiments have been performed on diffusive transport in strongly scattering media. To describe these studies conventional transport theory ignores any form of recurrent scattering, being valid for weak scattering only. The phenomenon of recurrent scattering is equivalent to what is known as induced dipole-dipole coupling in atomic physics.[1]

In this paper we derive a transport theory using the exact solution for two particles as a building block for multiple scattering, thereby including all induced dipole-dipole interactions (IDDI's). Apart from the dipole-dipole energy density we shall also calculate other transport properties related to diffusion. Our calculations were possible because we solved the highly nontrivial problem of keeping track of energy conservation in transport theory. Explicit energy conservation is essential to maintain long-range diffusion.

In transport theory[2,3] the building block for the amplitude is the self-energy $\Sigma_{\mathbf{p}}(E)$; for the intensity it is the irreducible vertex $U_{\mathbf{pp}'}(E)$. ($\mathbf{p}$,$\mathbf{p}'$ indicate momentum channels; $E$ is the frequency). Energy conservation is expressed by a relation between amplitude and intensity according to (a "Ward" identity)

$$-\frac{1}{E}\mathrm{Im}\,\Sigma_{\mathbf{p}}(E) = -\frac{1}{E}\int\frac{d^3\mathbf{p}'}{(2\pi)^3}\,\mathrm{Im}\,G_{\mathbf{p}'}(E)\,U_{\mathbf{pp}'}(E)\,. \quad (1)$$

The Green function $G_{\mathbf{p}}(E) \equiv \left[E^2 - p^2 - \Sigma_{\mathbf{p}}(E)\right]^{-1}$ describes the propagation of the (ensemble averaged) amplitude in momentum space. We set $c_0 = 1$.

All diagrams involving two particles in the self-energy and vertex are shown in Fig. 1. For uncorrelated point dipoles with number density $n$ the self-energy follows from $nt + \Sigma^{(2)}_{\mathbf{p}}$, where $\Sigma^{(2)}_{\mathbf{p}}$ is given by[4]

$$\Sigma^{(2)}_{\mathbf{p}}(E) = n^2\int d^3\mathbf{r}\,\frac{t^3g^2}{1-t^2g^2} + n^2\int d^3\mathbf{r}\,\frac{t^4g^3}{1-t^2g^2}\,e^{i\mathbf{p}\cdot\mathbf{r}}. \quad (2)$$

The vertex is given by the Boltzmann approximation $n|t|^2$ plus the sum of all possible pairs of two-particle corrections,

$$U^{(2)}_{\mathbf{pp}'}(\text{irred. ladders}) = n^2|t|^2\int d^3\mathbf{r}\,|tg|^2\left(\frac{1}{|1-t^2g^2|^2}-1\right), \quad (3)$$

$$U^{(2)}_{\mathbf{pp}'}(\text{loops}) = n^2|t|^2\int d^3\mathbf{r}\left(\frac{1}{|1-t^2g^2|^2}-1\right), \quad (4)$$

$$U^{(2)}_{\mathbf{pp}'}(\text{loop-ladders}) = n^2|t|^2\int d^3\mathbf{r}\,2\mathrm{Re}\,(tg)\left(\frac{1}{|1-t^2g^2|^2}-1\right)\left(e^{i\mathbf{p}\cdot\mathbf{r}}+e^{i\mathbf{p}'\cdot\mathbf{r}}\right), \quad (5)$$

$$U^{(2)}_{\mathbf{pp}'}(\text{crossed}) = n^2|t|^2\int d^3\mathbf{r}\,\frac{|tg|^2}{|1-t^2g^2|^2}e^{i(\mathbf{p}+\mathbf{p}')\cdot\mathbf{r}}, \quad (6)$$

$$U^{(2)}_{\mathbf{pp}'}(\text{forward-crossed}) = n^2|t|^2\int d^3\mathbf{r}\left(\frac{1}{|1-t^2g^2|^2}-1\right)e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}}\,. \quad (7)$$

In these formulas $g(r) = -\exp(iEr)/4\pi r$ is the bare Green function in real space. The $t$ matrix of one individual oscillating dielectric dipole with eigenfrequency $E_0$ and linewidth $E_0^2\Gamma$ is given by [5] $t(E) \equiv -4\pi E_0^{-1}[\Delta - i]^{-1}$, with $\Delta \equiv 2(E_0 - E)/\Gamma E_0^2$; $\Delta > 0$ ($< 0$) signifies frequencies below (above) the resonance. The same expression describes the semiclassical interaction (polarizability) of an atom with light in the dipole approximation.[6,7]

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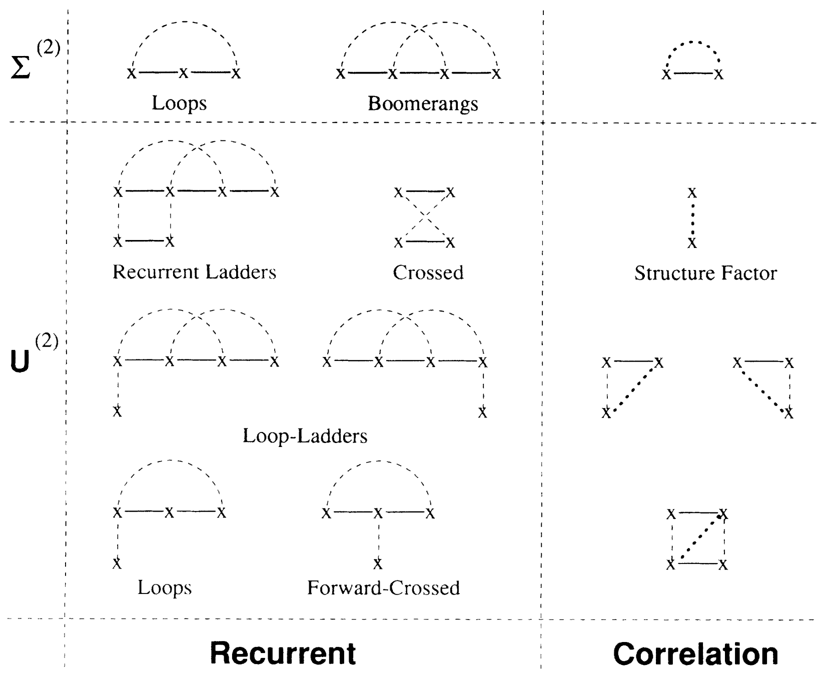


FIG. 1. All connected diagrams involving two particles in the self-energy $\Sigma$ and the irreducible vertex $U$. Only the simplest diagram of every class has been displayed. Neither high orders in recurrent scattering nor complex conjugates are shown. Crosses denote $t(E)$, bold lines $g(E,\mathbf{r})$, dashed lines connect identical particles, dotted lines denote $h(\mathbf{r})$.

From the optical theorem for the $T$ matrix of two point dipoles,[4] conservation of energy in transport, that is, Eq. (1), can be shown to be satisfied explicitly in first and second order of the dipole density. All integrals in Eqs. (2) – (7) are finite, indicating that the IDDI is short range on the scale of the mean free path.

We will now present our main results. The diffusion constant is given by $D(E) = \frac{1}{3} v_E \ell$ containing a dynamic transport velocity $v_E$ and a stationary transport mean free path $\ell$. We first focus on stationary properties. The transport mean free path is given by $\ell = 1/n\sigma_p$ with $\sigma_p$ the radiative pressure cross section. It differs from the scattering mean free path $\ell_s = 1/n\sigma_s$ (in terms of the scattering cross section $\sigma_s$) by a well-known cosine weighting of the differential cross-section.[3] The differential cross section is defined as $d\sigma/d\Omega \equiv n^{-1} U_{\mathbf{kk}'}/(4\pi)^2$ ($|\mathbf{k}| = |\mathbf{k}'| = E$) and is modified for the IDDI according to

$$\frac{d\sigma}{d\Omega}(\theta) \equiv \frac{1}{E_0^2}\left[\frac{1}{\Delta^2+1} + \eta\,\mathcal{H}(\theta,\Delta)\right]. \tag{8}$$

The second term is the IDDI correction and constitutes the sum of Eqs. (3) – (7). The modification introduces an angular anisotropy and is seen to be proportional to the "number of particles per optical volume," $\eta \equiv 4\pi n/E_0^3$. The first term is the conventional cross section for single scattering. If $\langle\cdots\rangle$ denotes the angular average, the scattering and pressure cross section become[8]

$$\sigma_s = \frac{4\pi}{E_0^2}\left[\frac{1}{\Delta^2+1} + \eta\,\langle\mathcal{H}(\theta,\Delta)\rangle\right], \tag{9}$$

$$\sigma_p = \frac{4\pi}{E_0^2}\left[\frac{1}{\Delta^2+1} + \eta\,\langle(1-\cos\theta)\mathcal{H}(\theta,\Delta)\rangle\right]. \tag{10}$$

In Fig. 2 we display the line profiles of both cross sections for a typical experimental value[9] of $\eta$. Exactly on the resonance ($\Delta = 0$) are $\langle\mathcal{H}\rangle = 0.3756$ and $\langle(1-\cos\theta)\mathcal{H}\rangle = 0.8605$ so that both cross sections are increased by IDDI's. The increase continues beyond the resonance ($\Delta < 0$); below the resonance scattering is diminished. These results indicate that the maximum in the scattering cross section shifts towards the *blue*. For $\eta = 0.7$ the maximum of the scattering cross section is 1.28 times the unitary limit $4\pi/E_0^2$ of a single dipole, and occurs at $\Delta = -0.1$.

The area underneath the line profile of the scattering cross section turns out not to be changed by IDDI's. This is a manifestation of a Kramers-Kronig sum rule for the

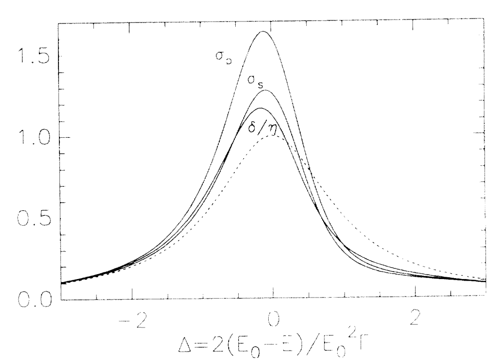


FIG. 2. Influence of scalar recurrent scattering from two polarizable dipoles on typical transport quantities near the eigenfrequency (resonance) $E_0$, for a density $\eta \equiv 4\pi n/E_0^3 = 0.7$. $\sigma_s$, scattering cross section (in units of the unitary limit $4\pi/E_0^2$), inversely proportional to scattering mean free path; $\sigma_p$, pressure cross section (in same units), inversely proportional to transport mean free path; $\delta/\eta$, relative "stored" energy per dipole (in units of the inverse dimensionless linewidth $1/E_0\Gamma$), inversely proportional to transport velocity. The dashed line is the Lorentzian profile to which all three quantities would coincide without recurrent scattering.

dielectric function (related to the self-energy).[3,6] Thus, the IDDI narrows the line profile of the scattering cross section with conservation of the area underneath.

The dynamics in wave diffusion is described by the transport velocity $v_E$. It can be written as [10,11] $v_E v_p = 1/[1+\delta(E)]$ where $v_p$ is the phase velocity ($c_0 = 1$). The parameter $\delta(E)$ is associated with "stored energy,"[11,12] and will be changed by the dipole-dipole binding energy. It can be calculated in two independent ways. Both calculations were found to coincide within applied numerical accuracy, showing that all diagrams have been included in our approach. One method uses the fact that stored energy is obtained from derivatives of the conservation law (1) with respect to small imaginary part of the potential and was successfully applied to find the Boltzmann result for $\delta(E)$.[11] The second method[13,14] is more convenient for the present purposes. In that case,

$$\delta(E)\mathrm{Im}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\,G_{\mathbf{p}}(E)\,E^2$$

$$= -\mathrm{Im}\int\frac{d^3\mathbf{p}}{(2\pi)^3}G_{\mathbf{p}}(E)\,\Sigma_{\mathbf{p}}(E)\,. \quad (11)$$

In evaluating the right-hand side of Eq. (11) one UV singularity must be regularized precisely as has been done in Ref. 5 in order to be consistent with the $t$ matrix used here. In lowest order of the dipole density we obtain $\delta_1(\Delta) = -(n/E_0^3\Gamma)\,\mathrm{Im}\,t$, corresponding to the internal energy of independent dipoles.[12] In second order we find

$$\delta_2(\Delta) = -\frac{n^2}{E_0^3\Gamma}\,\mathrm{Im}\int d^3\mathbf{r}\,\frac{t^3g^2}{1-t^2g^2} - \frac{n^2}{E_0\Gamma}\,\mathrm{Im}\frac{t^2}{4}\,. \quad (12)$$

Except for the second term, this surprising and extremely simple form for $\delta_2$ is indeed recognized as an induced dipole-dipole-type contribution to the total potential energy in the medium.[1] (The second term arises because $\delta$ describes the potential energy relative to the radiative energy outside which is also modified by the presence of dipoles.) This important notion demonstrates that the IDDI is still correctly described by scalar waves, and is not intrinsically a vector effect. The quantity $\delta = \delta_1 + \delta_2$ is shown in Fig. 2, for a density $\eta = 0.7$. It is seen that the amount of scattering $\sigma_s$ remains roughly proportional to the relative amount of stored energy $\delta$, as in the Boltzmann approximation. Like the sum rule for the scattering cross section, the integral of $\delta_2(\Delta)$ can also be shown to vanish by analycity. The non-Lorentzian line shape of $\delta$ in the frequency domain corresponds to a nonexponential decay of the excited oscillator in the time domain. This may be of interest to atomic physicists.

Our calculations can and have been generalized for pair correlation of the scatterers. This is relevant in dielectric scattering, but is negligible in light-atom scattering. A pair-correlation function $h(\mathbf{r})$ is introduced as $f^{(2)}(1,2) = f^{(1)}(1)\,f^{(1)}(2)\,[1+h(\mathbf{r}_{12})]$, where $f^{(N)}$ denotes the $N$-particle distribution function. Seven extra two-particle diagrams show up that are connected by correlation only (Fig. 1), one in the self-energy and six in the irreducible vertex.[2] The diagrams that were already connected by recurrent scattering must be given a weighting factor $1 + h(\mathbf{r})$. Explicit energy balance can again be shown to be established. Not unexpectedly, the diagrammatic analysis reveals that only one simple weighting factor $1 + h(\mathbf{r})$ enters into the integral of Eq. (12). In Fig. 3 we display some calculations for "hard spheres" $h(\mathbf{r}) = -\Theta(a - |\mathbf{r}|)$. We infer that the shape of the scattering cross section is hardly affected. On the other hand, the binding energy is more sensitive to the exact value of $E_0 a$. The pressure cross section increases considerably with increasing excluded volume. This is mainly due to the structure factor of the two-particle system, which becomes less than 1 at forward scattering due to excluded volume, and therefore lowers the transport mean free path,[15] hence enhances the pressure cross section.

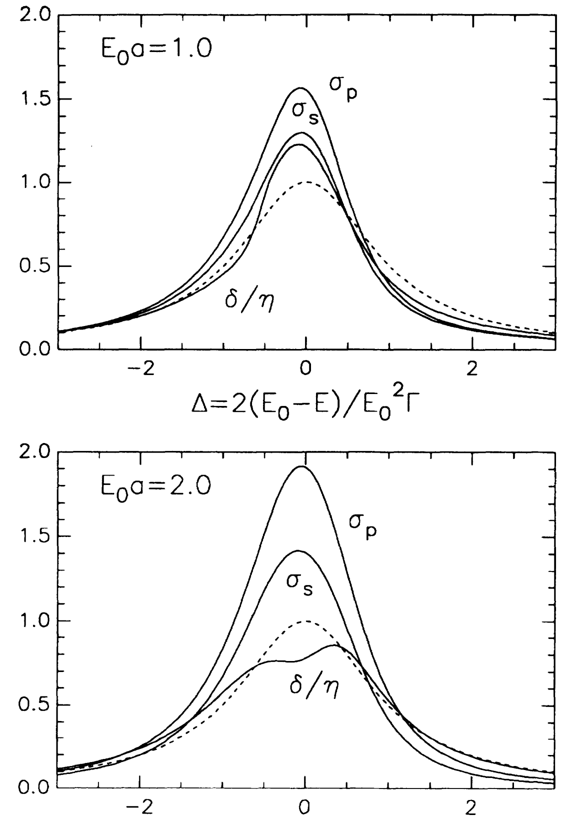


FIG. 3. As in Fig. 2 but now for different excluded volumes with radius $a$, $E_0a = 1.0, 2.0$, and a value $\eta = 0.5$.

Our method has been presented using the language of classical scalar waves and should be directly applicable to acoustic waves. With a minor modification our findings are immediately applicable to low-energy electron-impurity scattering in the solid state. The presence of longitudinal waves makes the treatment of vector waves considerably more complicated. Given the importance of light propagation, this generalization should be pursued. Hopefully this work can serve as a guide.

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[8] Since only two simple diagrams dominate the angular distribution, $\langle \cos\theta \mathcal{H} \rangle \approx -(\pi/6)\left(\Delta^2+1\right)^{-2} + (4/3)(1-\ln 2)\Delta\left(\Delta^2+1\right)^{-3}$.

[9] In induced dipole-dipole coupling between atoms the quantity $\eta$ can be orders of magnitude larger, which is beyond the range of our theory.

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