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Light propagation in a solid with resonant atoms at random positions

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Propagation of light in media with a small density of resonant atoms at quenched random positions is studied. It is found that the transport speed is reduced substantially due to dwell effects when the atoms are close to resonance. Besides propagation of real photons, the transport channel of nonradiative transfer due to the resonance dipole–dipole interaction between excited and ground state atoms is considered. The transport equation including both transport channels is derived. The diffusion coefficient and the dielectric constant are found. The weight of the nonradiative channel increases with the density of resonant atoms, n , and becomes essential when $n\lambda^3$ approaches unity, where λ is the wavelength of the light.

Multiple elastic scattering of light in disordered media is a theme of intensive current research. Its extreme limit is Anderson localization of visible light, which is still a challenge for the field (for a recent review, see ref. [1]). Many interesting precursor effects have been found, such as speckle correlations [2], speed of propagation [3], the role of the skin layer [4]. Good conditions for Anderson localization are to be expected for a system of atoms at quenched random positions, with the light frequency being close to the resonance frequency of the atoms [5]. Indeed, under these conditions a large cross section will be achieved. However, in contrast to classical scatterers, in this system one should take into account nonradiative excitation transfer due to the resonance dipole–dipole interaction. This is just the interaction responsible for the formation of Frenkel excitons in an insulating crystal, see e.g. ref. [6]. The resonance dipole–dipole interaction also plays an important role in the radiation transfer occurring in binary collisions of atoms in gases [7]. It is the resonance dipole–dipole interaction and nonradiative excitation transfer, in the background of strong scattering of photons, that make an essential difference between light propagation in media with resonant atoms and usual scalar wave prop-

agation. For the latter the possibility of localization near resonance frequencies is shown in ref. [8].

In the present paper we shall study transport of electromagnetic waves, such as light, in a medium with a small fraction of resonant atoms at quenched random positions. There are two transport mechanisms. First, an excited atom can decay to its ground state by emitting a photon. Transport of the photon, subject to resonance scattering, leads to the possibility of absorption by another atom, thus transferring the excitation. Second, the excitation may be transferred nonradiatively by the resonance dipole–dipole interaction, which is equivalent to including the longitudinal component of the electromagnetic field. The importance of this mechanism increases with increasing density of atoms, n . This channel influences the diffusion of light and becomes essential when the average number of atoms per cubic wavelength, $n\lambda^3$, becomes of order unity ($\lambda = 2\pi/k$ is the wavelength of the light). In this case the resonance dipole–dipole interaction $d^2/\bar{R}^3$ at mean interparticle separation $\bar{R}$ becomes comparable with the inverse radiative lifetime of the excited atom, d^2/λ^3 , where d is the dipole moment of the atomic transition and units are such

that $\hbar = c = 1$.

Below we focus our attention on the case $n\lambda^3 \ll 1$, where the dipole-dipole interaction can be considered in the pair approximation, and derive the transport equation for the density of the excited atoms. We consider the isotropic case where all directions for the transition dipole moment are equivalent.

Let us start by considering transport of light, omitting for the moment the longitudinal component of the electromagnetic field, i.e. the resonance dipole-dipole interaction. The interaction of the transverse field with the atoms is described by the Hamiltonian $\mathcal{H}_{\text{int}} = -\sum_i \mathbf{d}_i \cdot \mathbf{E}(\mathbf{R}_i)$, where $\mathbf{d}_i$ is the dipole momentum operator of the atom located at position $\mathbf{R}_i$, and where $\mathbf{E}(\mathbf{R}_i)$ is the electric field. In the simplest case of an s-p transition the transition between the groundstate and the excited state of the atom has an angular momentum index $\lambda = -1, 0, 1$. Its matrix element has the form $\langle 0 | \mathbf{d}_i | \lambda \rangle = d \mathbf{e}_\lambda$, where d is the dipole moment of the transition, and where the $\mathbf{e}_\lambda$ are three orthogonal unit vectors.

For a small density of resonant atoms the dielectric function becomes

$$\epsilon = 1 + \frac{4\pi n d^2}{\omega_0 - \omega - i\gamma_R \sqrt{\epsilon}}, \quad (1)$$

where

$$\gamma_R = \frac{2}{3} d^2 \omega^3 \quad (2)$$

is the radiative halfwidth of the excited state of an isolated atom. The factor $\sqrt{\epsilon}$ describes renormalization of this result due to renormalization of the speed of light in the medium.

The dressed propagator of the transverse electric field reads

$$\begin{aligned} G_{\alpha\beta}(\mathbf{p}, \omega) &= \omega^2 \frac{\delta_{\alpha\beta} - p_\alpha p_\beta / p^2}{\omega^2 - p^2 - \Sigma} \\ &= \omega^2 \frac{\delta_{\alpha\beta} - p_\alpha p_\beta / p^2}{\epsilon \omega^2 - p^2}. \end{aligned} \quad (3)$$

Taking into account eq. (1) we have for the "self-energy"

$$\Sigma = \frac{4\pi n d^2 \omega^2}{\omega + i\gamma_R \sqrt{\epsilon} - \omega_0}. \quad (4)$$

As it was mentioned above, we did not yet take into account the longitudinal part of the field. It will be included below as a small perturbation, which is correct

in the limit $n\lambda^3 \ll 1$. Note that in this limit, where the pair approximation works well for the dipole interaction, the condition $\Sigma \ll \omega^2$ is satisfied, and we have $\gamma_R \sqrt{\epsilon} \approx \gamma_R$.

The intensity of emitted photons is attenuated by the interaction with atoms. The scattering mean free path l_R is determined by the imaginary part of the pole of eq. (3) at $p = \sqrt{\epsilon} \omega \equiv K + i/2l_R$. The result reads

$$l_R = \frac{\omega^2 [(\omega - \omega_0)^2 + \gamma_R^2]}{6\pi n \gamma_R^2}. \quad (5)$$

As expected, this result agrees with the expression $l_R = 1/n\sigma_R$, where σ_R is the well-known cross section of light scattered from an isolated atom.

Let us now turn to the calculation of the diffusion of photons. It is described by the evolution of the density matrix for photons. In our situation, where the mean free path is much larger than the wavelength, this density matrix is local in space. For later use we shall consider, however, the density matrix of excited atoms. In a situation where many scatterings take place, this object satisfies the same transport equation as the density matrix of photons. The only difference will be the expression for the source term, a point that is not relevant to us. We consider the situation where one incident photon has excited an atom. This excitation is transferred over the system. Let $\psi(\mathbf{R}, \lambda, \omega)$ denote a frequency component of the amplitude that at position $\mathbf{R}$ at time t an atom is in the excited state with polarization λ . In a non-stationary situation we study the density matrix of excited atoms at two slightly different frequencies $L_{\lambda\mu}(\mathbf{R}, \Omega) = \langle \psi^*(\mathbf{R}, \mu, \omega - \frac{1}{2}\Omega) \psi(\mathbf{R}, \lambda, \omega + \frac{1}{2}\Omega) \rangle$. In the ladder approximation it satisfies the Bethe-Salpeter equation

$$\begin{aligned} L_{\lambda\mu}(\mathbf{R}, \Omega) &= S_{\lambda\mu}(\mathbf{R}) \\ &+ \int d\mathbf{R}' \sum_{\lambda'\mu'} M_{\lambda\mu, \lambda'\mu'}(\mathbf{R} - \mathbf{R}', \Omega) L_{\lambda'\mu'}(\mathbf{R}', \Omega). \end{aligned} \quad (6)$$

Here S is a source resulting from the excitation of atoms by the incoming beam. For the case of a plane wave with polarization ϵ_{in} , incident under an angle θ_{in} on a system where the atoms are confined to the half-space $z \geq 0$, the source reads

$$S_{\lambda\mu}(z) = n\omega_0^2 d^2 \epsilon_{\text{in}} \cdot \mathbf{e}_\lambda \epsilon_{\text{in}}^* \cdot \mathbf{e}_\mu^* \exp\left(-\frac{z}{l_R \cos \theta_{\text{in}}}\right). \quad (7)$$

In the present situation the kernel $M = M^R$ of the transport equation reads in Fourier space

$$M_{\lambda\mu;\lambda'\mu'}^R(\mathbf{q}, \Omega) = \frac{1}{n} \Sigma(\omega_+) \Sigma^*(\omega_-) \times \int \frac{d\mathbf{p}}{(2\pi)^3} G_{\lambda\lambda'}(\mathbf{p}_+, \omega_+) G_{\mu'\mu'}^*(\mathbf{p}_-, \omega_-) \quad (8)$$

where $\omega_\pm = \omega \pm \frac{1}{2}\Omega$ and $\mathbf{p}_\pm = \mathbf{p} \pm \frac{1}{2}\mathbf{q}$.

Next we study the diffusion approximation. For $\mathbf{q} = \Omega = 0$ it holds that $\delta_{\lambda\mu}$ is an eigenvector of M^R with unit eigenvalue, i.e. $\sum_{\lambda'\mu'} M_{\lambda\mu;\lambda'\mu'}^R(\mathbf{0}, 0) \delta_{\lambda'\mu'} = \delta_{\lambda\mu}$. This optical theorem ensures that for a homogeneous system the density of excited atoms, $\text{tr} L \equiv \sum_\lambda L_{\lambda\lambda}$, is a conserved quantity. If both Ω and $\mathbf{q}$ are small, the leading eigenvalue can be found from a first order perturbation expansion

$$A^R(\mathbf{q}, \Omega) \approx \frac{1}{3} \sum_{\lambda\mu} M_{\lambda\mu;\lambda\mu}^R(\mathbf{q}, \Omega) = 1 + i\Omega(l_R + 1/2\gamma_R) - \frac{1}{3}q^2 l_R^2. \quad (9)$$

Combining these results we obtain

$$\text{tr} L(\mathbf{q}, \Omega) = \frac{2\gamma_R}{1 + 2\gamma_R l_R} \frac{\text{tr} S(\mathbf{0}, 0)}{Dq^2 - i\Omega}, \quad (10)$$

with diffusion coefficient

$$D = \frac{1}{3} \frac{\gamma_R l_R^2}{\gamma_R l_R + \frac{1}{2}}. \quad (11)$$

The general definition $D = l^2/3\tau$ implies a mean time per collision

$$\tau = \tau_{\text{tr}} + \tau_{\text{dw}}, \quad (12)$$

where $\tau_{\text{tr}} = l_R/c$ is the characteristic time for light to travel from one atom to another atom, and where $\tau_{\text{dw}} = 1/2\gamma_R$ is the dwell time of the light due to the excitation of the resonant atom. (We have momentarily restored the light velocity c .)

Recently it was shown that for scattering of light waves on classical dielectric particles that are close to a geometric resonance a reduction in the speed of propagation may occur due to dwell effects of the waves inside the resonant dielectric particles [3]. Let

us also introduce the transport speed $v = l_R/\tau$. Equation (12) yields

$$v = c \frac{\tau_{\text{tr}}}{\tau_{\text{tr}} + \tau_{\text{dw}}} = c \frac{1}{1 + c/2\gamma_R l_R}. \quad (13)$$

The same equation for the transport velocity was obtained in ref. [9] for the resonance scattering of scalar waves on microscopic objects. Very far from resonance the transport velocity equals the vacuum speed of light, as expected. Near resonance, however, v may become many orders of magnitude smaller. Indeed, at resonance one has $\gamma_R l_R/c \sim \omega^2 \gamma / nc^3 \sim (\gamma/\omega_0)/n\lambda^3$. Even in the very dilute case the ratio between line width and resonance frequency is usually much smaller than the number of atoms per cubic wavelength, implying strong scattering and a substantial reduction in the propagation speed.

Let us now include the longitudinal component mentioned above. This is equivalent to considering excitation transfer due to the resonance dipole-dipole interaction

$$\mathcal{U}_{ij}^{\lambda\lambda'} = V_{ij}(\delta_{\lambda\lambda'} - 3n_\lambda n_{\lambda'}) \equiv \frac{d^2}{R_{ij}^3}(\delta_{\lambda\lambda'} - 3n_\lambda n_{\lambda'}). \quad (14)$$

Here $\mathbf{n} = \mathbf{R}_{ij}/R_{ij}$ is the unit vector pointing from atom i to atom j . Equation (14) represents the matrix element of the dipole-dipole operator for transfer of the excitation from atom i to atom j .

The Green function of the excited atom is given by $K_{0\lambda\lambda'} = K_0 \delta_{\lambda\lambda'}$ with

$$K_0 = \frac{1}{\omega - \omega_0 + i\gamma_R}. \quad (15)$$

For deriving the transport properties we are interested in calculating the matrix

$$\Gamma = \frac{1}{1 - K_0 \mathcal{U}} = \sum_{m=0}^{\infty} K_0^m \mathcal{U}^m. \quad (16)$$

This quantity cannot be evaluated in general. Moreover, there is no small parameter in which a perturbation expansion can be performed. In case of a small concentration of atoms the resonance dipole-dipole interaction gives a small contribution as compared to transport by photons. It can therefore be treated in the pair approximation. First, let us derive the Green

function for scattering by atom i . Making a configurational average ($\sum_i \rightarrow n \int d^3 R_1$) one obtains

$$\begin{aligned} K'_{\lambda\lambda'} &= K_0 \delta_{\lambda\lambda'} + K_0^3 \sum_i (\mathcal{U}_{i1} \mathcal{U}_{i1})_{\lambda\lambda'} \\ &+ K_0^5 \sum_i (\mathcal{U}_{i1} \mathcal{U}_{i1} \mathcal{U}_{i1} \mathcal{U}_{i1})_{\lambda\lambda'} \\ &+ K_0^5 \sum_{i,2} (\mathcal{U}_{i1} \mathcal{U}_{i1} \mathcal{U}_{i2} \mathcal{U}_{i2})_{\lambda\lambda'} + \dots \\ &= \left(\frac{1}{K_0} - n \int d\mathbf{R}_1 \frac{K_0 \mathcal{U}_{i1}^2}{1 - K_0^2 \mathcal{U}_{i1}^2} \right)^{-1}_{\lambda\lambda'}. \end{aligned} \quad (17)$$

In a self-consistent approach this leads to the renormalized Green function

$$K = \frac{1}{\omega - \omega_0 + i\gamma_R - \Sigma_D}. \quad (18)$$

The self-energy

$$\Sigma_D \delta_{\lambda\lambda'} = n \int d\mathbf{R} C_{\lambda\lambda'}(\mathbf{R}) \quad (19)$$

involves

$$\begin{aligned} C_{\lambda\lambda'}(\mathbf{R}_{ij}) &= \left(\frac{K \mathcal{U}_{ij}^2}{1 - K^2 \mathcal{U}_{ij}^2} \right)_{\lambda\lambda'} \\ &= \frac{K V_{ij}^2}{1 - K^2 V_{ij}^2} (\delta_{\lambda\lambda'} - n_\lambda n_{\lambda'}) + \frac{4K V_{ij}^2}{1 - 4K^2 V_{ij}^2} n_\lambda n_{\lambda'}. \end{aligned} \quad (20)$$

We find

$$\Sigma_D = -\frac{8}{9} i\pi^2 n d^2 \equiv -i\gamma_D. \quad (21)$$

An analogous result for the case of binary collisions in a gas of resonant atoms was obtained by Vdovin and Galitskii [7].

For calculating the diffusion coefficient we should find the analogues of eqs. (6) and (8), introducing retarded and advanced Green functions with frequencies shifted over $\pm \frac{1}{2}\Omega$,

$$\begin{aligned} K &= \frac{1}{\omega - \omega_0 + \frac{1}{2}\Omega + i\gamma}, \\ \bar{K} &= \frac{1}{\omega - \omega_0 - \frac{1}{2}\Omega - i\gamma}. \end{aligned} \quad (22)$$

They have a total width

$$\gamma = \gamma_R + \gamma_D. \quad (23)$$

The same change occurs in eqs. (1) and (4).

Let us notice that in eq. (17) we can ignore the contribution of the transverse field (mixed terms) only in the case $|\omega - \omega_0| \gg \gamma$, where the distances $r \approx (d^2/|\omega - \omega_0|)^{1/3} \ll \lambda$ are important. For these distances the longitudinal part exceeds the transverse one. In the opposite case, $|\omega - \omega_0| \simeq \gamma$, the mixed terms and also so-called dependent scattering terms, omitted in eq. (1), give contributions comparable with those of the resonance dipole-dipole interaction, and our results are valid only qualitatively. (A more complete approach starts from all two-scatterer contributions to the self-energy, as considered for scattering from dielectric spheres in ref. [10]. The expression for the kernel then follows via the Ward identity. We present here a simplified version of this approach.)

Let us now turn to the calculation of the density matrix of excited atoms. The kernel in eq. (6) will describe both transport channels,

$$M = M^R + M^D, \quad (24)$$

with the "radiative" kernel defined in eq. (8) with γ replacing γ_R . The "dipolar" kernel has two contributions

$$M^D(\mathbf{R}, \Omega) = M_T^D(\mathbf{R}, \Omega) + \delta(\mathbf{R}) M_S^D(\Omega). \quad (25)$$

M_T^D describes real transport,

$$\{M_T^D(\mathbf{R}, \Omega)\}_{\lambda\mu, \lambda'\mu'} = n B_{\lambda\lambda'}(\mathbf{R}, \Omega) \bar{B}_{\mu\mu'}(\mathbf{R}, \Omega), \quad (26)$$

with amplitudes taking into account the exchange forth and back,

$$\begin{aligned} B_{\lambda\lambda'}(\mathbf{R}_{ij}, \Omega) &= \left(\frac{K \mathcal{U}_{ij}}{1 - K^2 \mathcal{U}_{ij}^2} \right)_{\lambda\lambda'} \\ &= \frac{K V_{ij}}{1 - K^2 V_{ij}^2} (\delta_{\lambda\lambda'} - n_\lambda n_{\lambda'}) + \frac{-2K V_{ij}}{1 - 4K^2 V_{ij}^2} n_\lambda n_{\lambda'}, \end{aligned} \quad (27)$$

and $\bar{B}$ defined similarly in terms of $\bar{K}$. The second term in eq. (25) describes the influence of turning back,

$$\{M_S^D(\Omega)\}_{\lambda\mu,\lambda'\mu'} = nK\bar{K} \int d\mathbf{R}'' C_{\lambda\lambda'}(\mathbf{R}'', \Omega) \bar{C}_{\mu\mu'}(\mathbf{R}'', \Omega). \quad (28)$$

In momentum representation at $\mathbf{q} = \Omega = 0$ one finds again that $\delta_{\lambda\mu}$ is an eigenvalue of the matrix M with unit eigenvalue. Indeed, we can express the eigenvalue as $\gamma_R/\gamma + K\bar{K}(K - \bar{K})^{-1}(\Sigma_D - \bar{\Sigma}_D) = 1$. This result remains valid for general potentials $V(R)$ in eq. (14), at least for those that they decay as $V \sim 1/R^3$ or faster, for large R .

Let us now study the diffusion approximation. If both Ω and $\mathbf{q}$ are small, the leading eigenvalue $A = A^R + A^D$ can again be found from a first order perturbation expansion,

$$A^R(\mathbf{q}, \Omega) = \frac{\gamma_R}{\gamma} [1 + i\Omega(l_R + 1/2\gamma) - \frac{1}{3}\mathbf{q}^2 l_R^2]. \quad (29)$$

$$\begin{aligned} A^D(\mathbf{q}, \Omega) &\approx \frac{n}{3} \int d\mathbf{R} \left(2 \frac{K\bar{K}V^2(e^{i\mathbf{q}\cdot\mathbf{R}} + K\bar{K}V^2)}{(1 - K^2V^2)(1 - \bar{K}^2V^2)} \right. \\ &\quad \left. + \frac{4K\bar{K}V^2(e^{i\mathbf{q}\cdot\mathbf{R}} + 4K\bar{K}V^2)}{(1 - 4K^2V^2)(1 - 4\bar{K}^2V^2)} \right) \\ &= \frac{\gamma_D}{\gamma} (1 + i\Omega/2\gamma - \frac{1}{3}\mathbf{q}^2 l_D^2), \end{aligned} \quad (30)$$

with the radiative mean free path in the presence of the excitation transfer channel

$$l_R = \frac{\omega^2[(\omega - \omega_0)^2 + \gamma^2]}{6\pi n\gamma\gamma_R}, \quad (31)$$

and with l_D determined by the expression

$$l_D^2 = (d^2/\gamma)^{2/3} f((\omega - \omega_0)/\gamma), \quad (32)$$

where

$$f(x) = \left(\frac{1}{2} + 1/2^{1/3}\right) (1 + x^2)^{-5/6} \frac{\sin(\frac{5}{3} \arctan x)}{\sin(2 \arctan x)}. \quad (33)$$

For small frequency detuning, $|\omega - \omega_0| \ll \gamma$, we have a mean free path of the order of the wavelength, $l_D \sim \lambda$. For large frequency detuning, $|\omega - \omega_0| \gg \gamma$, eq. (33) leads to $l_D \sim (d^2/|\omega - \omega_0|)^{1/3}$, which is much smaller than for small frequency detuning. It shows that only those pairs of atoms are involved, for which the separation is such that the detuning matches the interaction strength $\mathcal{U}$.

Combining eqs. (29) and (30) we obtain a diffusion coefficient in the form

$$D = \frac{1}{3} \frac{\gamma_R l_R^2 + \gamma_D l_D^2}{\gamma_R l_R + \frac{1}{2}}. \quad (34)$$

In the limit of a atom small density, $n\lambda^3 \ll 1$, $\gamma_D \ll \gamma_R$ holds and the excitation transfer mechanism only gives a small correction to the transport by real photons. However, we can already see the tendency in the behavior. For small frequency detuning, $|\omega - \omega_0| \ll \gamma$, we have $l_R = 1/n\sigma_R$ with the photon scattering cross section σ_R of the order λ^2 , while $l_D \sim \lambda$. With increasing density n , the contribution of the excitation transfer channel enhances the role of the dipole-dipole transport (the term $\gamma_D l_D^2$ in eq. (34)).

At large detuning the excitation transfer contribution $\gamma_D l_D^2$ to the diffusion coefficient is not significant. However, the presence of γ_D , originating from the resonance dipole-dipole interaction, in the expression for γ (see eq. (23)) leads to a relative decrease of l_R and D with rising n .

When $n\lambda^3$ approaches unity all length scales at resonance become comparable, $l_R \sim l_D \sim \lambda$. This emphasizes the importance of the excitation transfer for the problem of light localization ^{#1}.

In conclusion, we have considered the propagation of light in a medium with a small density of resonant atoms. It is found that close to resonance the transport speed is considerably diminished, since the light is captured for a large time by the atoms. The classical analog of this effect was observed for light scattering from resonant dielectric particles [3]. Whereas in the latter case the reduction of speed can be as large as a factor 10–100, in the quantum situation this reduction is typically of the order 10^5 – 10^7 .

Next we have included the nonradiative transport channel due to the resonance dipole-dipole interaction. In our situation it leads to a small, but principal effect. We have presented a new set of diagrams, which generalizes the ladder diagrams for transport of real photons, to situations with an additional transport channel of the form (14).

^{#1} Weak localization effects lead to a correction to the diffusion coefficient $\delta D \sim -D(\lambda/l_R)^2$. This contribution is larger than the contribution of the excitation transfer channel when $n\lambda^3 \ll 1$. However, it is density independent and hence it does not change the trend under variation of the density.

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