Superradiance and multiple scattering of photons in atomic gases

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We study the influence of cooperative effects such as superradiance and subradiance on the scattering properties of dilute atomic gases. We show that cooperative effects lead to an effective potential between two atoms that decays as $1/r$. In the case of superradiance, this potential is attractive for close enough atoms and can be interpreted as a coherent mesoscopic effect. We consider a model of multiple scattering of a photon among superradiant pairs and calculate the elastic mean free path and the group velocity. We study first the case of a scalar wave which allows us to obtain and to understand basic features of cooperative effects and multiple scattering. We then turn to the general problem of a vector wave. In both cases, we obtain qualitatively similar results and derive, for the case of a scalar wave, analytic expressions for the elastic mean free path and for the group velocity for an arbitrary detuning (near resonance).

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I. INTRODUCTION

Coherent multiple scattering of photons in cold atomic gases is an important problem since it presents a path toward the onset of the Anderson localization transition, a long-standing and still open issue. The large resonant scattering cross section of photons reduces the elastic mean free path to values comparable to the photon wavelength, for which the weak-disorder approximation breaks down, thus signaling the onset of the Anderson localization transition [1,2]. Another advantage of cold atomic gases is that sources of decoherence and inelastic scattering such as Doppler broadening are often negligible. Moreover, propagation of photons in atomic gases differs from the case of electrons in disordered metals or of electromagnetic waves in suspensions of classical scatterers, for which mesoscopic effects and Anderson localization have been thoroughly investigated [1]. This problem is thus of great interest since it may raise new issues in the Anderson problem, such as a change of universality class and therefore new critical behavior. New features displayed by the photon-atom problem are the existence of internal degrees of freedom (Zeeman sublevels) and cooperative effects such as subradiance or superradiance [3], which lead to effective interactions between atoms [4]. These two differences are expected to lead to qualitative changes of both mesoscopic quantities and Anderson localization. Some of the effects of Zeeman degeneracy have been investigated in the weak-disorder limit [5] using a set of finite phase coherence times [6], which reduce mesoscopic effects, such as coherent backscattering [1,7].

The influence of cooperative effects and more specifically of superradiance on the multiple scattering of photons has been recently investigated [8]. It has been shown that in atomic gases superradiance and subradiance lead to a potential between two atoms, analogous to the one considered in [9,10], which decays as the inverse of the distance between them. In the case of superradiance, this potential is attractive for close enough atoms, and can be interpreted as a coherent mesoscopic effect. The contribution of superradiant pairs to multiple-scattering properties of a dilute gas has been calculated by using an effective propagator that describes a scalar wave being scattered by a pair of two-level atoms. Simple expressions for the photon elastic mean free path and group velocity have been derived at resonance and found to be significantly different from those of independent atoms. To be more specific, near resonance, as well as at resonance, the superradiant effect leads to a finite and positive group velocity, unlike the one obtained for light interaction with independent atoms.

In this paper we provide, for the case of a scalar wave, closed expressions for the suprerradiant contribution to the elastic mean free path and the group velocity for an arbitrary (near resonance) detuning, and calculate the dependence of the transport time on it. In addition, we estimate the maximal interatomic separation in a superradiant pair, which accounts for possible mechanisms that may break the pair. We also compare the effective approach presented in [8] to a more realistic one that takes into account the vectorial nature of the wave.

The paper is organized as follows: In Sec. II we describe the model, which consists of pairs of two-level atoms placed in an external radiation field where the Doppler shift and recoil effects are negligible. In order to investigate the influence of the cooperative effects of such pairs on the multiple scattering of photons we briefly review, in Sec. III, Dicke states and some of their properties. Then we calculate the average interaction potential of a pair of atoms in a Dicke state by averaging over the random orientations of pairs of atoms with respect to the wave vector of a photon incident on the atomic cloud. Next, we study the scattering of a photon by such pairs and, in Sec. IV, compare the results to the case where a classical wave is being scattered by a pair of atoms. This comparison allows us to find an unexpected connection between superradiance and mesoscopic effects. In Secs. V and VI, we consider the multiple scattering of photons by pairs of atoms and calculate the elastic mean free path and the group velocity of photons in the random medium. Finally, our analysis is compared to other approaches in Sec. VII and its results are summarized in Sec. VIII.

II. MODEL

Atoms are taken to be degenerate, two-level systems denoted by $|g\rangle=|j_g=0,m_g=0\rangle$ for the ground state and $|e\rangle=|j_e$

$=1, m_e=0,\pm 1\rangle$ for the excited state, where j is the total angular momentum and m is its projection on a quantization axis, taken as the $\hat{z}$ axis. The energy separation between the two levels, including radiative shift, is $\hbar\omega_0$, and the natural width of the excited level is $\hbar\Gamma$. This simple picture of a two-level atom neglects the rather complicated energy structure of a real atom, which reflects various internal interactions, e.g., Coulomb interactions, spin-orbit interactions, hyperfine interactions, etc. But, due to selection rules which limit the allowed transitions between states, in some cases a certain state may couple to only one other. Thus, the two-level atom approximation is close to reality and not merely a mathematical convenience.

We consider a pair of such atoms in an external radiation field and the corresponding Hamiltonian is $H=H_0+V$, with

$$H_0 = \frac{\hbar\omega_0}{2}\sum_{l=1}^{2}(|e\rangle\langle e| - |g\rangle\langle g|)_l + \sum_{\mathbf{k}\varepsilon}\hbar\omega_k a^\dagger_{\mathbf{k}\varepsilon}a_{\mathbf{k}\varepsilon}. \tag{1}$$

$a_{\mathbf{k}\varepsilon}$ ($a^\dagger_{\mathbf{k}\varepsilon}$) is the annihilation (creation) operator of a mode of the field of wave vector $\mathbf{k}$, polarization $\hat{\varepsilon}_{\mathbf{k}}$, and angular frequency $\omega_k=c|\mathbf{k}|$. The interaction V between the radiation field and the dipole moments of the atoms is given by

$$V = -\sum_{l=1}^{2}\mathbf{d}_l\cdot\mathbf{E}(\mathbf{r}_l), \tag{2}$$

where $\mathbf{E}(\mathbf{r})$ is the electric field operator

$$\mathbf{E}(\mathbf{r}) = i\sum_{\mathbf{k}\varepsilon}\sqrt{\frac{\hbar\omega_k}{2\epsilon_0\Omega}}(a_{\mathbf{k}\varepsilon}\hat{\varepsilon}_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}} - a^\dagger_{\mathbf{k}\varepsilon}\hat{\varepsilon}^*_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{r}}). \tag{3}$$

Ω is a quantization volume and $\mathbf{d}_l$ is the electric dipole moment operator of the lth atom. As an odd operator, which changes sign upon inversion, $\mathbf{d}_l$ may be written as

$$\mathbf{d}_l = \langle g|\mathbf{d}|e\rangle\Delta^-_l + \langle e|\mathbf{d}|g\rangle\Delta^+_l \tag{4}$$

where the atomic raising and lowering operators are

$$\Delta^+_l = (|e\rangle\langle g|)_l \quad \Delta^-_l = (|g\rangle\langle e|)_l. \tag{5}$$

We assume that the typical speed of the atoms, $v\simeq\sqrt{k_BT_0/\mu}$, is small compared to $v_{max}=\Gamma/k$ but large compared to $v_{min}=\hbar k/\mu$, where μ is the mass of the atom and T_0 is the temperature, so that it is possible to neglect the Doppler shift and recoil effects. Indeed, for a temperature of $T_0=10^{-3}$ K, the typical speed of the atom is $v\simeq 0.3$ m/s. Since, for a wave number of $k=10^7$ m^{-1} and a natural width of $\Gamma=10^7$ s^{-1}, $v_{max}\simeq 1$ m/s and $v_{min}\simeq 0.01$ m/s, both assumptions are satisfied.

III. DICKE STATES

A. Interaction potential and lifetime

The absorption of a photon by a pair of atoms in their ground state leads to a configuration where the two atoms, one excited and the second in its ground state, have multiple exchange of a photon, giving rise to an effective interaction potential and to a modified lifetime as compared to independent atoms. These two quantities are obtained from the matrix elements of the evolution operator $U(t)$ between states such as $|g_1e_2;0\rangle$. There are six unperturbed and degenerate states with no photon, given by $\{|g_1e_{2i};0\rangle, |e_{1j}g_2;0\rangle\}$ in a standard basis where $i,j=-1,0,1$. The symmetries of the Hamiltonian, namely, its invariance by rotation around the axis between the two atoms, and by reflection with respect to a plane containing this axis, allows us to use combinations of these states that are given by

$$|\phi^\epsilon_i\rangle = \frac{1}{\sqrt{2}}[|e_{1i}g_2;0\rangle + \epsilon|g_1e_{2i};0\rangle] \tag{6}$$

with $\epsilon=\pm 1$, so that

$$\langle\phi^{\epsilon'}_j|U(t)|\phi^\epsilon_i\rangle = \delta_{ij}\delta_{\epsilon\epsilon'}S^\epsilon_i(t) \tag{7}$$

and

$$S^\epsilon_i(t) = \langle e_{1i}g_2;0|U(t)|e_{1i}g_2;0\rangle + \epsilon\langle g_1e_{2i};0|U(t)|e_{1i}g_2;0\rangle. \tag{8}$$

The states $|\phi^\epsilon_i\rangle$ may be rewritten in terms of the well-known Dicke states $|LM\rangle$, where L is the cooperation number and M is half of the total atomic inversion [3]. For two atoms, the singlet Dicke state is

$$|00\rangle = \frac{1}{\sqrt{2}}[|e_1g_2\rangle - |g_1e_2\rangle] \tag{9}$$

and the triplet Dicke states are

$$|11\rangle = |e_1e_2\rangle,$$

$$|10\rangle = \frac{1}{\sqrt{2}}[|e_1g_2\rangle + |g_1e_2\rangle],$$

$$|1-1\rangle = |g_1g_2\rangle. \tag{10}$$

The states $|11\rangle$ and $|1-1\rangle$ correspond, respectively, to both atoms in their excited states and both atoms in their ground states. The singlet state $|00\rangle$ and the triplet state $|10\rangle$ both correspond to one atom in the excited state and the other in the ground state, but $|00\rangle$ is antisymmetric where $|10\rangle$ is symmetric under an exchange of the atoms. Therefore, we may rewrite (6) as $|\phi^+_i\rangle=|10;0\rangle$ and $|\phi^-_i\rangle=|00;0\rangle$.

For times such that $t\gg r/c$, where r is the distance between the two atoms, up to second order in the coupling to the radiation, (8) reads

$$S^\epsilon_i(t) \simeq 1 - \frac{it}{\hbar}\left(\Delta E^\epsilon_i - i\frac{\hbar\Gamma^\epsilon_i}{2}\right). \tag{11}$$

The two real quantities ΔE^ϵ_i and Γ^ϵ_i are, respectively, the interaction potential and the probability per unit time of emission of a photon by the two atoms in the state $|\phi^\epsilon_i\rangle$. The calculation of these two quantities requires second-order perturbation theory with respect to the interaction (2). For this purpose we define an initial state where one atom is excited and the other is in its ground state without any photon, and a final state where the two atoms are exchanged. We also define intermediate states of two types: both atoms in their ground states with one virtual photon present and both atoms

in their excited states with one virtual photon present. Summing the corresponding diagrams [11] gives

$$\Delta E_i^{\epsilon} = \epsilon \frac{3\hbar\Gamma}{4}\left[- p_i \frac{\cos k_0 r}{k_0 r} + q_i \left(\frac{\cos k_0 r}{(k_0 r)^3} + \frac{\sin k_0 r}{(k_0 r)^2} \right) \right] \tag{12}$$

and

$$\frac{\Gamma_i^{\epsilon}}{\Gamma} = 1 - \frac{3}{2}\epsilon \left[- p_i \frac{\sin k_0 r}{k_0 r} + q_i \left(\frac{\sin k_0 r}{(k_0 r)^3} - \frac{\cos k_0 r}{(k_0 r)^2} \right) \right], \tag{13}$$

where we have defined $k_0 = \omega_0 / c$,

$$p_i = 1 - \hat{\mathbf{r}}_i^2, \quad q_i = 1 - 3\hat{\mathbf{r}}_i^2, \tag{14}$$

and $\hat{\mathbf{r}} = (1, \theta, \varphi)$ is a unit vector along the direction joining the two atoms. For a $\Delta m = m_e - m_g = 0$ transition,

$$p_0 = \sin^2\theta, \quad q_0 = 1 - 3\cos^2\theta, \tag{15}$$

while for a $\Delta m = \pm 1$ transition,

$$p_{\pm} = \frac{1}{2}(1 + \cos^2\theta), \quad q_{\pm} = \frac{1}{2}(3\cos^2\theta - 1). \tag{16}$$

At short distance $k_0 r \ll 1$, we obtain that $\Gamma_i^+ \simeq 2\Gamma$ for the superradiant state $|\phi_i^+\rangle = |10;0\rangle$ and $\Gamma_i^- \simeq 0$ for the subradiant state $|\phi_i^-\rangle = |00;0\rangle$.

B. Average interaction potential

For a photon of wave vector $\mathbf{k}$ incident on an atomic cloud, the potential between two atoms that we shall denote by V_e is obtained from (12) by averaging over the random orientations of the pairs of atoms with respect to $\mathbf{k}$. Since, according to (15) and (16), $\langle q_i \rangle = 0$ and $\langle p_i \rangle = 2/3$, we obtain for the average potential V_e

$$\epsilon V_e(r) = \langle \Delta E_i^{\epsilon} \rangle = - \epsilon \frac{\hbar\Gamma}{2} \frac{\cos k_0 r}{k_0 r} \tag{17}$$

and the average inverse lifetimes of Dicke states are

$$\langle \Gamma_i^{\epsilon} \rangle = \Gamma \left(1 + \epsilon \frac{\sin k_0 r}{k_0 r} \right), \tag{18}$$

which retain the same features as (13) for $k_0 r \ll 1$.

Let us now characterize the interaction potential V_e. Whereas for a single pair of atoms the potential (12) is anisotropic and decays at short distance as $1/r^3$, a behavior that originates from the transverse part of the photon propagator, we obtain that, on average over angular configurations, the potential (17) between two atoms in a Dicke state $|L0\rangle$ in vacuum becomes isotropic and decays as $1/r$. This behavior coincides with the one obtained by considering the interaction of two-level atoms with a scalar wave. This could have been anticipated since in that case the transverse contribution q_i to the photon propagator averages to 0. A related behavior for the orientation average interaction potential has been also obtained for the case of an intense radiation field [9], and it has recently been investigated in order to study effects of a long-range and attractive potential between atoms in a Bose-Einstein condensate for far-detuned light [10]. This latter potential, which is fourth order in the coupling to the radiation, corresponds to the interaction energy between two atoms in their ground states in the presence of at least one photon. The average potential V_e we have obtained is different from that case: it is second order in the coupling to the radiation and it corresponds to the interaction energy of Dicke states $|L0\rangle$ in vacuum.

C. Scattering properties

In order to study the scattering properties of Dicke states we introduce the collision operator $T(z) = V + VG(z)V$, where V is given by (2) and $G(z) = (z - H)^{-1}$ is the resolvent where the Hamiltonian H is the sum of (1) and (2). The matrix element that describes the transition amplitude from the initial state $|i\rangle = |1-1;\mathbf{k}\hat{\varepsilon}\rangle$, where the two atoms are in their ground states in the presence of a photon of frequency $\omega = c|\mathbf{k}|$ and polarization $\hat{\varepsilon}$, to the final state $|f\rangle = |1-1;\mathbf{k}'\hat{\varepsilon}'\rangle$ is

$$T = \langle f | T(z = \hbar(\omega - \omega_0)) | i \rangle \tag{19}$$

where $|\mathbf{k}| = |\mathbf{k}'|$. By using the closure relation we may write T as the sum of a superradiant and a subradiant contribution, $T = T^+ + T^-$ [12], with

$$T^{\pm} = \langle f | V | \phi^{\pm} \rangle \langle \phi^{\pm} | G(z = \hbar(\omega - \omega_0)) | \phi^{\pm} \rangle \langle \phi^{\pm} | V | i \rangle, \tag{20}$$

where $|\phi^{\pm}\rangle$ are the Dicke states $|L0\rangle$ in vacuum. The two matrix elements in (20) represent the absorption and the emission of a real photon by the pair of atoms. They are easily obtained from (2)–(5) and lead to the following expressions for the scattering amplitudes:

$$T^+ = A e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \cos\left(\frac{\mathbf{k}\cdot\mathbf{r}}{2} \right) \cos\left(\frac{\mathbf{k}'\cdot\mathbf{r}}{2} \right) G^+ \tag{21}$$

and

$$T^- = A e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \sin\left(\frac{\mathbf{k}\cdot\mathbf{r}}{2} \right) \sin\left(\frac{\mathbf{k}'\cdot\mathbf{r}}{2} \right) G^-. \tag{22}$$

We have defined $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$, and

$$A = \frac{\hbar\omega}{\epsilon_0 \Omega} d^2 (\hat{d}\cdot\hat{\varepsilon})(\hat{d}^*\cdot\hat{\varepsilon}'^*), \tag{23}$$

where the reduced matrix element and the corresponding unit vector are

$$d = \frac{\langle j_e \| \mathbf{d} \| j_g \rangle}{\sqrt{2j_e + 1}}, \quad \hat{d} = \frac{1}{d} \langle j_e m_e | \mathbf{d} | j_g m_g \rangle. \tag{24}$$

The propagators $G^{\pm}$ are the expectation values of the resolvent in the Dicke states $|\phi^{\pm}\rangle$, namely, $G^{\pm} = \langle \phi^{\pm} | G(\hbar\delta) | \phi^{\pm} \rangle$, where close to resonance $\delta = \omega - \omega_0 \ll \omega_0$. The propagators result from the sum of an infinite series of virtual photon exchanges between the two atoms in the pair and are given in terms of (12) and (13) by

$$G^{\pm} = \left(\hbar\delta - \Delta E^{\pm} + i\hbar \frac{\Gamma^{\pm}}{2} \right)^{-1}. \tag{25}$$

The average propagator is then obtained by averaging $G^{\pm}$ over the random orientations of the pairs of atoms with respect to the wave vector $\mathbf{k}$ of the incident photon. However, we shall consider in a first stage the effective propagator obtained for the case of a scalar wave. This amounts to writing for the effective propagator the expression

$$G_e^{\pm} = \left[\hbar \left(\delta + i\frac{\Gamma}{2} \pm \frac{\Gamma}{2} \frac{e^{ik_0 r}}{k_0 r} \right) \right]^{-1}, \tag{26}$$

where we have used (17) and (18) for the average potential and for the average inverse lifetimes. This expression constitutes *a priori* a rough approximation of the exact average. We shall calculate later, in Sec. VI, the exact expression of the average propagator and show that it is rather complicated, whereas the approximate expression using a scalar wave gives similar qualitative results. Therefore, it allows for a better understanding of relevant physical quantities such as the elastic mean free path and group velocity. From now on, we thus use the scalar wave approximation in order to provide, in a rather simple way, the main features of multiple scattering by superradiant pairs.

With the help of (26), the scattering amplitudes are

$$T_e^{+} = A e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \cos\left(\frac{\mathbf{k}\cdot\mathbf{r}}{2} \right) \cos\left(\frac{\mathbf{k}'\cdot\mathbf{r}}{2} \right) G_e^{+} \tag{27}$$

and

$$T_e^{-} = A e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \sin\left(\frac{\mathbf{k}\cdot\mathbf{r}}{2} \right) \sin\left(\frac{\mathbf{k}'\cdot\mathbf{r}}{2} \right) G_e^{-}. \tag{28}$$

At short distances $k_0 r \ll 1$, the subradiant amplitude T_e^{-} becomes negligible as compared to the superradiant term T_e^{+}. Therefore, the potential (17) is attractive and decays as $1/r$. More precisely, at short distances the effective propagator G_e^{-} diverges for $\delta/\Gamma = 1/(2k_0 r)$ and G_e^{+} is purely imaginary for $\delta/\Gamma = -1/(2k_0 r)$. Thus, for $\delta/\Gamma < 1/(2k_0 r)$ the imaginary part of the subradiative term (28) is negligible as compared to the imaginary part of the superradiative term (27) and for $|\delta|/\Gamma < 1/(2k_0 r)$ both the real part and the imaginary parts of (28) are negligible as compared to (27).

We can interpret these results by saying that, at short distances ($k_0 r \ll 1$), the time evolution of the initial state

$$|\psi(0)\rangle = |e_1 g_2; 0\rangle = \frac{1}{\sqrt{2}} [|\phi^{+}\rangle + |\phi^{-}\rangle], \tag{29}$$

corresponds, for times shorter than $1/\Gamma$, to a periodic exchange of a virtual photon between the two atoms at the Rabi frequency

$$\Omega_R = \frac{\langle \Delta E^{-} \rangle - \langle \Delta E^{+} \rangle}{\hbar}, \tag{30}$$

which is much larger than Γ since, with the help of (17),

$$\Omega_R \simeq \frac{\Gamma}{k_0 r}. \tag{31}$$

For larger times, the two atoms return to their ground states and a real photon ($\mathbf{k}'\hat{\varepsilon}'$) is emitted. At large distances ($k_0 r \gg 1$), the Rabi frequency becomes smaller than Γ, so that the excitation energy makes only a few oscillations between the two atoms, thus leading to a negligible interaction potential.

We finally notice that the angular distribution of the light scattered by two atoms in a superradiant state is nearly identical to that of a single atom. This follows from the fact that at short distance $k_0 r \ll 1$, we can neglect higher-order multipolar corrections so that the corresponding additional phase shift $k_0 r \cos\vartheta$ between waves emitted by the two atoms becomes negligible (ϑ is the angle between the direction of the emitted photon and the axis between the two atoms).

IV. COOPERATIVE EFFECTS AND COHERENT BACKSCATTERING

It is interesting to derive the previous results in another way that emphasizes the analogy with coherent backscattering [1,2]. To that purpose, we write the scattering amplitude T defined previously in (19) as a superposition of two "classical," scalar amplitudes T_1 and T_2 [13], each of them being a sum of single-scattering and double-scattering contributions, that is,

$$T_1 = \frac{t}{1 - t^2 G_0^2} (e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_1} + t G_0 e^{i(\mathbf{k}\cdot\mathbf{r}_1 - \mathbf{k}'\cdot\mathbf{r}_2)}) \tag{32}$$

and

$$T_2 = \frac{t}{1 - t^2 G_0^2} (e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_2} + t G_0 e^{i(\mathbf{k}\cdot\mathbf{r}_2 - \mathbf{k}'\cdot\mathbf{r}_1)}). \tag{33}$$

Here

$$t = \frac{4\pi}{k_0} \frac{\Gamma/2}{\delta + i\Gamma/2} \tag{34}$$

is the amplitude of a scalar wave scattered by a single atom at the origin, and the prefactor $t/(1-t^2 G_0^2)$, where

$$G_0 = -\frac{e^{ik_0 r}}{4\pi r} \tag{35}$$

accounts for the summation of the series of virtual photon exchange between the two scatterers. We can single out in the total amplitude $T = T_1 + T_2$ the single-scattering contribution T_s and write the intensity associated with the higher-order scattering term shown in Fig. 1 as

$$|T - T_s|^2 = 2 \left| \frac{t^2 G_0}{1 - t^2 G_0^2} \right|^2 [1 + \cos(\mathbf{k} + \mathbf{k}') \cdot (\mathbf{r}_1 - \mathbf{r}_2)]. \tag{36}$$

The structure of relation (36) is very reminiscent of that of the so-called coherent backscattering intensity, which occurs in the multiple elastic scattering of light. But although they are analogous, (36) differs from coherent backscattering. In the latter case, averaging over the spatial positions $\mathbf{r}_1$ and $\mathbf{r}_2$ makes the interference term $\cos(\mathbf{k}+\mathbf{k}')\cdot(\mathbf{r}_1-\mathbf{r}_2)$ vanish in general, with two exceptions:

(1) $\mathbf{k}+\mathbf{k}' \simeq \mathbf{0}$. In the direction exactly opposite to the direction of incidence, the intensity is twice the classical value.

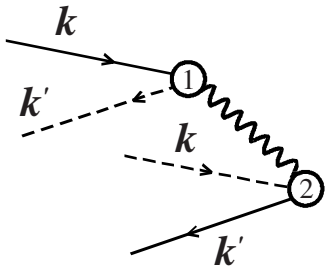

FIG. 1. Schematic representation of the two amplitudes that describe double scattering of a scalar wave. The wavy line accounts for the exchange of a virtual photon between the two atoms. This diagram is analogous to the coherent backscattering in quantum mesoscopic physics.

This phenomenon is known as coherent backscattering.

(2) $\mathbf{r}_1=\mathbf{r}_2$. These are closed multiple-scattering trajectories which are at the origin of the phenomenon of weak localization.

In (36) the interference term, i.e., the second term in the square brackets, reaches its maximum value 1 for $\mathbf{r}_1=\mathbf{r}_2$ so that we obtain from (32), (33), and (27) that $T_1=T_2 \propto (1/2)T_e^+$, up to a proportionality factor [13]. Thus, the total amplitude is given by the superradiant term with no subradiant contribution.

V. MULTIPLE SCATTERING AND COOPERATIVE EFFECTS

A. Effective self-energy

We consider now multiple scattering of a photon by superradiant pairs built out of atoms separated by a distance r and coupled by the attractive interaction potential V_e. This situation corresponds to a dilute gas that is assumed to satisfy

$$r \ll \lambda_0 \ll n_i^{-1/3}, \tag{37}$$

where n_i is the density of pairs and $\lambda_0=2\pi/k_0$ is the atomic transition wavelength. The limiting case (37) corresponds to a situation where the two atoms that form a superradiant pair, through exchange of a virtual photon, constitute an effective scatterer, and cooperative interactions between otherwise well-separated pairs are negligible. Let us stress that we study here a simplified model where only pairs of atoms have been taken into account. A more realistic model should include higher-order terms that account for cooperative effects between more than two atoms, but we do not consider such higher-order terms, i.e., including superradiant clusters of three or more atoms. The purpose of the current model is to examine the contribution of superradiant pairs to the transport properties of the gas. We use the Edwards model [1,14] to describe the medium as a discrete collection of N_i superradiant pairs in a volume Ω. Each pair, located at $\mathbf{R}_l$, is characterized by its scattering potential $u(\mathbf{R}-\mathbf{R}_l)$. Therefore, the disorder potential is given by

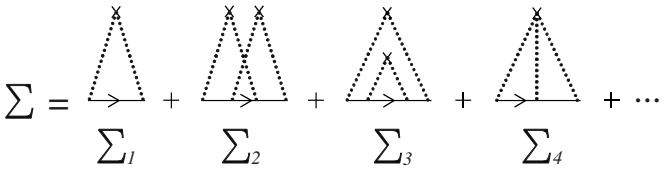

FIG. 2. Pertubative expansion of the self-energy in a power series in the parameter $n_i u_0^2$. Solid lines account for the free photon Green's function g_0. Pairs of dotted lines, connected by $\times$, stand for the two-point correlation function B. The first term Σ_1, proportional to $n_i u_0^2$, accounts for independent scattering events, while the second term Σ_2, proportional to $n_i^2 u_0^4$, describes interference effects between pairs of scatterers.

$$U(\mathbf{R}) = \sum_{l=1}^{N_i} u(\mathbf{R}-\mathbf{R}_l). \tag{38}$$

We assume that the scattering potential is short range compared to the wavelength, and we approximate it by a (conveniently regularized) δ function potential $u(\mathbf{R})=u_0\delta(\mathbf{R})$. In the limit of a high density of weakly scattering pairs, but with a constant value of $n_i u_0^2$, it can be shown [1] that the correlation function defined by

$$B(\mathbf{R}-\mathbf{R}') = n_i \int d\mathbf{R}'' u(\mathbf{R}''-\mathbf{R}) u(\mathbf{R}''-\mathbf{R}') \tag{39}$$

becomes

$$B(\mathbf{R}-\mathbf{R}') = n_i u_0^2 \delta(\mathbf{R}-\mathbf{R}'). \tag{40}$$

In other words, in this limit, the Edwards model reduces to a Gaussian white noise model characterized by the condition (40).

The Green's function g of a scattered photon is related to the free photon Green's function g_0, i.e., in the absence of disorder potential, by the equation [1]

$$g = g_0 + g_0 U g. \tag{41}$$

Averaging (41) over disorder and using the properties of the Gaussian model discussed above yields the Dyson equation

$$\langle g \rangle_d = g_0 + g_0 \Sigma \langle g \rangle_d \tag{42}$$

where $\langle \cdots \rangle_d$ denotes averaging over the random potential. The function Σ, known as the self-energy, represents the sum of all irreducible scattering diagrams. The pertubative expansion of the self-energy in a power series controlled by the parameter $n_i u_0^2$ is represented in Fig. 2.

For small values of $n_i u_0^2$, the main contribution is obtained by keeping only the first term Σ_1 which describes independent scattering events. Therefore, the first contribution to the self-energy is proportional to the density of scatterers and to the average scattering amplitude, and it is given, for $k_0 r \ll 1$, by

$$\Sigma_1 = \frac{6\pi n_i}{k_0} A_{j_g j_e} \hbar \Gamma \overline{G}_e^+ \tag{43}$$

where

$$A_{j_g j_e} = \frac{1}{3}\frac{2j_e+1}{2j_g+1}. \tag{44}$$

The latter quantity is obtained by averaging A in (27) over Zeeman sublevels m that appear in its definition given by (23) and (24).

The additional average, denoted by $\overline{\cdots}$, is taken over distances r up to a maximal value r_m which accounts for all possible mechanisms that may break the pairs.

The value of r_m can be estimated by comparing the kinetic energy K of a superradiant pair to its average potential energy V_e^+. We have $K \simeq \hbar^2/\mu r^2$ and from (17) we obtain that $V_e^+ \simeq -\hbar\Gamma/2k_0 r$. Minimizing the average energy

$$E(r) \simeq \frac{\hbar^2}{\mu r^2} - \frac{\hbar\Gamma}{2k_0 r} \tag{45}$$

with respect to r yields

$$k_0 r_m = 4\frac{\hbar k_0^2}{\mu\Gamma} \tag{46}$$

or

$$k_0 r_m = 4\frac{v_{min}}{v_{max}}, \tag{47}$$

where the speeds v_{min} and v_{max} have been defined in Sec. II. For typical values $\Gamma = 10^7\ \mathrm{s}^{-1}$ and $k_0 = 10^7\ \mathrm{m}^{-1}$ we obtain that $k_0 r_m \simeq 0.05$. Thus, we can use the results obtained in Sec. III C and consider the superradiant term only.

For $j_g = 0$ and $j_e = 1$, $A_{01} = 1$, and using (26) we rewrite (43) as

$$\Sigma_1 = \frac{6\pi n_i}{k_0}\frac{1}{r_m}\int_0^{r_m} \frac{dr}{\delta/\Gamma + 1/(2k_0 r) + i}. \tag{48}$$

We stress again that, in our approach, a pair of atoms in a superradiant state is considered as a single scatterer, and the effective medium parameters are derived from Σ_1 as will be shown in the next sections. In contrast to our treatment, others [16,19] consider multiple scattering of a real photon by *independent* atoms and use the second term Σ_2, which describes interference effects between the scatterers, to calculate corrections to the elastic mean free path and to the refractive index of the medium. A further comparison between these two points of view is given in Sec. VII.

B. Elastic mean free path

The elastic mean free path l_e is obtained from the imaginary part of the self-energy, namely,

$$\frac{k_0}{l_e} = -\,\mathrm{Im}\,\Sigma_1. \tag{49}$$

Let us stress that (49) is equivalent, in the case of a dilute gas, to the known formula

$$l_e = \frac{1}{n_i \sigma_e}, \tag{50}$$

where the total cross section σ_e is obtained for $k_0 r \ll 1$ from (27) by means of the optical theorem

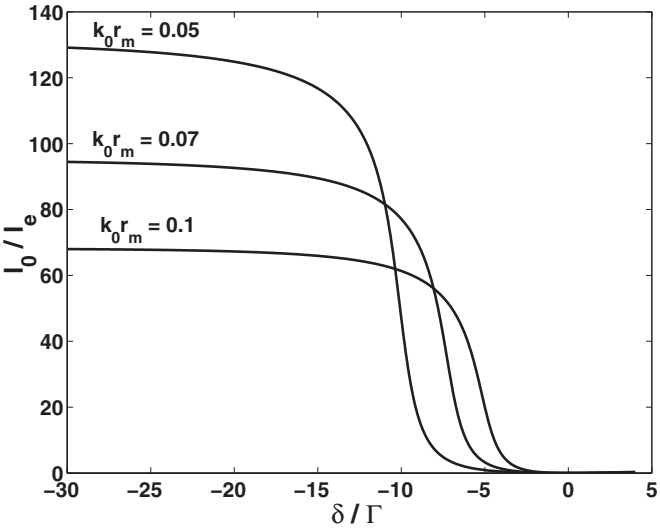

FIG. 3. Ratio between the elastic mean free paths l_0 and l_e as a function of the reduced detuning δ/Γ for $k_0 r_m = 0.05$, 0.07, and 0.1. Away from resonance, for blue detuning, the elastic mean free path l_e becomes smaller than l_0 in a ratio roughly given by $1/(k_0 r_m)^2$. At resonance, the ratio between the elastic mean free paths is given by (57).

$$\sigma_e = -\frac{2\Omega}{\hbar c}\,\mathrm{Im}\langle \overline{T}_e^+(\mathbf{k}=\mathbf{k}', \hat{\varepsilon}=\hat{\varepsilon}')\rangle_m \tag{51}$$

and $\langle\cdots\rangle_m$ represents an averaging over Zeeman sublevels. The equivalence in this case is proven easily if one uses (44) and the usual expression for the inverse lifetime

$$\Gamma = \frac{d^2 k_0^3}{3\pi\epsilon_0\hbar}, \tag{52}$$

where the reduced matrix element is defined in (24). Therefore, from (48) and (49) we obtain that

$$\frac{1}{l_e(\delta)} = \frac{6\pi n_i}{k_0^2} f_1\left(k_0 r_m, \frac{\delta}{\Gamma}\right) \tag{53}$$

where we have defined the function

$$f_1(u,v) = \frac{1}{2u}\int_0^{2u} \frac{dx}{1+(v+1/x)^2}. \tag{54}$$

The integral is easily carried out analytically and the explicit expression is given in Appendix A. It is interesting to compare l_e to the elastic mean free path l_0 that corresponds to near-resonant elastic scattering of a photon by a single atom. The latter quantity is obtained by replacing Γ by $\Gamma/2$ in (53) (since the inverse lifetime of a single atom is half the one related to a superradiant pair) and $1/x$ by 0 in (54) (since the interatomic distance is taken to be infinite for a single atom) and it is given by

$$l_0(\delta) = \frac{k_0^2}{6\pi n_i}\left[1+\left(\frac{2\delta}{\Gamma}\right)^2\right]. \tag{55}$$

In Fig. 3 the ratio between these two quantities is plotted as

a function of the reduced detuning δ/Γ from resonance for several values of $k_0 r_m$.

At resonance, we obtain from (53) that

$$l_e(0) = \frac{k_0^2}{8\pi n_i} \frac{1}{(k_0 r_m)^2} \tag{56}$$

and hence

$$\frac{l_0(0)}{l_e(0)} = \frac{4}{3}(k_0 r_m)^2 \ll 1. \tag{57}$$

Away from resonance, for blue detuning, the elastic mean free path l_e becomes smaller than l_0 in a ratio roughly given by $1/(k_0 r_m)^2$. This is a direct consequence of the existence of the attractive potential V_e.

C. Group velocity

Another important physical quantity that characterizes multiple scattering of a photon is its group velocity v_g given in terms of the refractive index η by the usual relation

$$\frac{c}{v_g} = \eta + \omega \frac{d\eta}{d\omega}. \tag{58}$$

The refractive index for a dilute medium is

$$\eta = (1 + n_i \,\mathrm{Re}\, \alpha)^{1/2}, \tag{59}$$

where the dynamic atomic polarizability α is proportional to the self-energy

$$\alpha = -\frac{1}{n_i}\left(\frac{c}{\omega}\right)^2 \Sigma_1. \tag{60}$$

Thus, we obtain that

$$\eta = \left[1 - \left(\frac{c}{\omega}\right)^2 \mathrm{Re}\, \Sigma_1\right]^{1/2}. \tag{61}$$

Substituting (61) into (58) yields

$$\frac{c}{v_g} = \frac{1}{\eta}\left(1 - \frac{c^2}{2\omega}\frac{d}{d\omega}\,\mathrm{Re}\, \Sigma_1\right). \tag{62}$$

From the self-energy (48), we notice that $\eta \simeq 1$ for all values of the detuning δ/Γ and in a large range of densities n_i, so that

$$\frac{c}{v_g(\delta)} \simeq 1 - \frac{n_i}{n_c} f_2\left(k_0 r_m, \frac{\delta}{\Gamma}\right), \tag{63}$$

where we have defined the characteristic density

$$n_c = \frac{k_0^3}{6\pi}\frac{\Gamma}{\omega_0} \tag{64}$$

and the function

$$f_2(u,v) = \frac{1}{2u}\int_0^{2u} dx \frac{1 - (v + 1/x)^2}{[1 + (v + 1/x)^2]^2}. \tag{65}$$

The integration is easily performed and the explicit expression is given in Appendix A. By replacing Γ by $\Gamma/2$ in (63) and $1/x$ by 0 in (65), we obtain the group velocity v_0 of light interacting with independent two-level atoms,

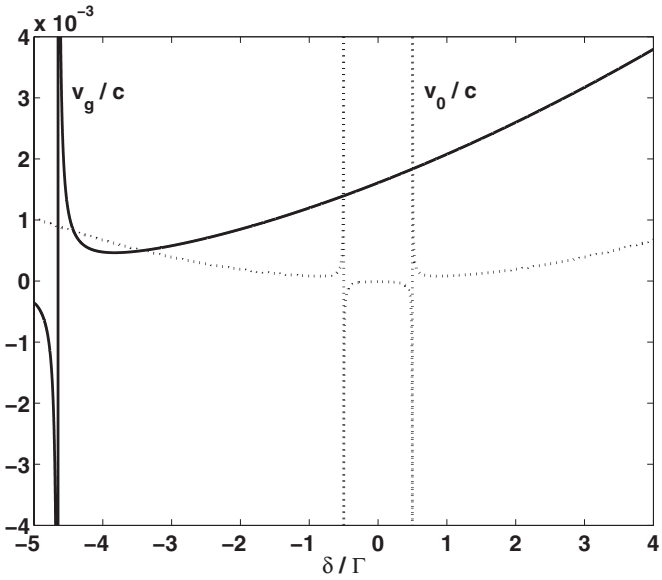

FIG. 4. Group velocities v_g (solid line) and v_0 (dotted line) as a function of the reduced detuning δ/Γ for $n_i/n_c = 10^5$ and $k_0 r_m = 0.1$. The group velocity v_0 diverges at two symmetric values of order unity of the reduced detuning and it takes negative values in between. The group velocity v_g, near resonance, remains finite and positive.

$$\frac{c}{v_0(\delta)} = 1 - \frac{n_i}{n_c}\frac{1 - (2\delta/\Gamma)^2}{[1 + (2\delta/\Gamma)^2]^2}. \tag{66}$$

For the typical values $\Gamma = 10^7\ \mathrm{s}^{-1}$, $k_0 = 10^7\ \mathrm{m}^{-1}$, and $n_i = 10^{10}\ \mathrm{cm}^{-3}$, we obtain that $n_i/n_c \simeq 10^5$.

Figure 4 displays the group velocities v_g and v_0 plotted as a function of the reduced detuning δ/Γ for $n_i/n_c = 10^5$ and $k_0 r_m = 0.1$.

v_g appears to diverge at quite a large and negative value of the detuning $\delta/\Gamma \simeq -1/(2k_0 r_m)$. But near resonance it is well behaved, meaning that it remains finite and positive. At resonance, according to (63), the group velocity is

$$\frac{c}{v_g(0)} = 1 + 4\pi \frac{n_i}{k_0^3}\frac{\omega_0}{\Gamma}(k_0 r_m)^2. \tag{67}$$

This expression of v_g differs substantially from the one obtained for v_0. For densities $n_i > n_c$, the group velocity v_0 diverges at two symmetric values of order unity of the detuning and it takes negative values in between (i.e., also at resonance), as can be seen in Fig. 4. This problem was recognized a long time ago [15] and an energy velocity has been defined which describes energy transport through a diffusive medium [16,17]. However, the diffusion coefficient, which will be discussed in the next section, is derived from the group velocity and not from the energy velocity [1]. Moreover, a closed expression for the energy velocity v_E has been obtained only for the case of resonant Mie scattering [18]. The expression is similar to (67) and is given by

$$\frac{c}{v_E} = 1 + 9\pi \frac{n_i}{k_0^3} \frac{\omega_0}{\Gamma}. \tag{68}$$

It is then interesting to notice that the inclusion of cooperative effects even at the lowest order, i.e., taking into account superradiant pairs, allows one to obtain a group velocity that is well behaved at resonance, unlike the case of resonant scattering by independent atoms.

D. Diffusion coefficient and transport time

Diffusive transport of photons through a gas is characterized by the photon diffusion coefficient

$$D(\delta) = \frac{1}{3} v_g(\delta) l_e(\delta) \tag{69}$$

which combines the elastic mean free path and the group velocity, both derived from the complex-valued self-energy (48). The diffusion coefficient D is of great importance since it enters into expressions of various measured physical quantities, such as the transmission and the reflection coefficients of a disordered medium [1]. In addition to these average quantities, an incident pulse that probes a nearly static configuration of scatterers may provide an instantaneous picture of the medium that displays a random distribution of bright and dark spots. This snapshot, known as a speckle pattern, can be characterized by the angular-correlation function and the time-correlation function of the light intensity (diffusing wave spectroscopy). In the first case, the correlation function of the transmission coefficient between two distinct directions of the transmitted wave is measured. In the second case, the intensity of the transmitted wave is measured at different times, so that the motion of the scatterers must be taken into account. As pointed out before, in both cases the diffusion coefficient plays an important role, as it enters in the relevant expressions. Moreover, the critical behavior of transport close to the Anderson localization transition at strong disorder is also obtained from the scaling form of D. Its expression, deduced from (53) and (63), depends on the range r_m and on the detuning δ/Γ. Since the group velocity and the elastic mean free path are significantly modified for superradiant states, we thus expect the diffusion coefficient to be different from its value obtained for independent atoms.

We define the transport time by

$$\tau_{tr}(\delta) = \frac{l_e(\delta)}{v_g(\delta)}. \tag{70}$$

At resonance and for $n_i \gg n_c$, it can be rewritten with the help of (56) and (67) as

$$\tau_{tr}(0) = \frac{1}{2\Gamma}, \tag{71}$$

in accordance with our assumption of superradiant states. Near resonance, the transport time depends weakly on the detuning. But, away from it, τ_{tr} depends on the detuning and thus on frequency, as can seen from Fig. 5 where the inverse of the transport time τ_{tr}^{-1}/Γ is plotted as a function of the reduced detuning δ/Γ for $n_i = 10^{10}$ cm^{-3}, $\Gamma = 10^7$ s^{-1}, and $k_0 = 10^7$ m^{-1} for several values of $k_0 r_m$.

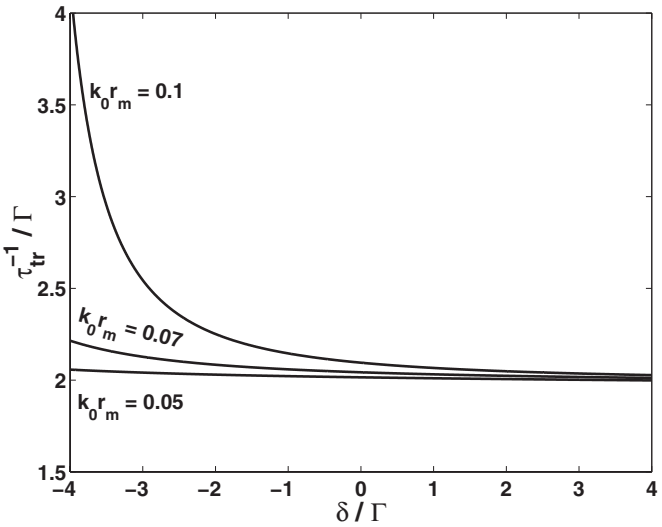

FIG. 5. Inverse of the transport time τ_{tr}^{-1}/Γ as a function of the reduced detuning δ/Γ for $n_i = 10^{10}$ cm^{-3}, $\Gamma = 10^7$ s^{-1}, and $k_0 = 10^7$ m^{-1} for $k_0 r_m = 0.05$, 0.07, and 0.1. Near resonance, the transport time depends weakly on the detuning. But, away from it, τ_{tr} depends on the detuning and thus on frequency.

VI. AVERAGE SELF-ENERGY

So far, we have used the effective approach introduced in Sec. III C, where we have considered the case of a scalar wave being scattered by a pair of two-level atoms. In this simple approach, the propagator of a scalar wave (26) has been calculated and the self-energy (43) has been obtained by averaging (26) over the distance between the two atoms in a pair. This effective approach leads to simple expressions for the elastic mean free path (53) and the group velocity (63) of the wave. In this section we calculate these quantities for a given Δm transition and $k_0 r \ll 1$, while taking into account the vectorial nature of the wave. With this purpose, we average the propagator (25) over the random orientations of the pairs of atoms (with respect to the wave vector of the incident photon) as well as over the distance between the two atoms in a pair. Therefore, the average self-energy is now given by

$$\Sigma_1' = \frac{6\pi n_i}{k_0} \frac{1}{4\pi r_m} \int \hbar \Gamma G^+(\mathbf{r}) d\mathbf{r}, \tag{72}$$

where the averaging is over the interatomic axis $\mathbf{r}$ (over both magnitude and orientations). The evaluation of (72) for a $\Delta m = 0$ transition is rather cumbersome and it is presented in Appendix B. By following the procedure described in the previous section, we obtain the corresponding elastic mean free path l_e' and the group velocity v_g'. In Fig. 6 the ratio between l_0 given by (55) and l_e' is plotted as a function of the reduced detuning δ/Γ for several values of $k_0 r_m$.

As in the effective approach, at resonance l_e' is found to be larger than l_0, but away from resonance it becomes smaller.

In Fig. 7 the group velocity v_g' is plotted as a function of the reduced detuning δ/Γ for $n_i/n_c = 10^5$ and $k_0 r_m = 0.1$.

Around resonance, the group velocity v_g' is finite and positive, as in the scalar case, but much larger as compared to

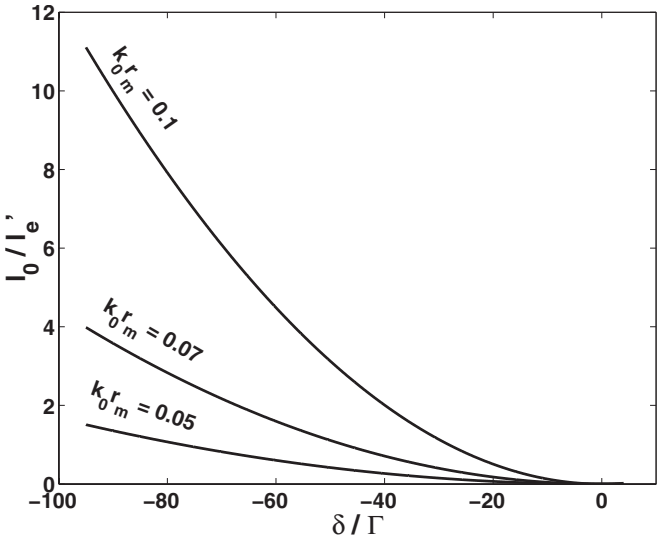

FIG. 6. Ratio between the elastic mean free paths l_0 and l_e' as a function of the reduced detuning δ/Γ for $k_0 r_m$=0.05, 0.07, and 0.1. At resonance, l_e' is larger than l_0, but away from resonance it becomes smaller.

(63) and it is close to c. Thus, we may conclude that in both approaches the superradiant effect leads to a finite and positive group velocity, unlike the one obtained for light interaction with independent atoms. However, the group velocity of a scalar wave is much smaller compared to the one of a photon.

VII. DISCUSSION

In this section we compare our analysis to other approaches [16,19] where resonant multiple scattering of light has been considered. There, using a multiple-scattering expansion for the calculation of the self-energy up to second order in $n_i u_0^2$, a correction to the elastic mean free path and to the refractive index has been obtained. In the latter approach, no distinction has been made between the external photon that performs multiple scattering on all atoms and virtual photons exchanged between two atoms in a superradiant state, leading to the average interaction potential V_e. This distinction needs to be made for dilute enough atomic gases since in that case the average distance $n_i^{-1/3}$ between atoms is large. Moreover, in this case, the dipole-dipole interaction induced by the external photon depends on the detuning, a situation that corresponds to the case of intense radiation presented in [9] but not to the current experiments made on cold atomic clouds [20].

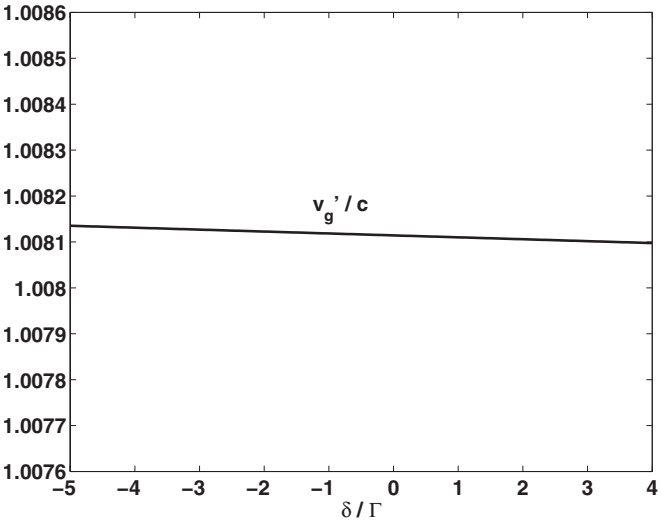

FIG. 7. Group velocity v_g' as a function of the reduced detuning δ/Γ for $n_i/n_c=10^5$ and $k_0 r_m$=0.1. Around resonance, the group velocity v_g' is finite and positive and it is close to c.

VIII. CONCLUSIONS

We have considered multiple scattering of a photon by pairs of atoms that are in a superradiant state. On average over disorder configurations, an attractive interaction potential builds up between close enough atoms, which decays as $1/r$. The contribution of superradiant pairs, resulting from this potential, to scattering properties is significantly different from that of independent atoms. This shows up in the behaviors of the group velocity, the elastic mean free path and the diffusion coefficient which are different from their values obtained for independent atoms. We have considered the case of a scalar wave and have shown that it allows to define an effective long-range and attractive potential for pairs of atoms in a superradiant state. Then, we have studied the case of a vector wave and have shown that the results obtained in the scalar case remain qualitatively valid. We have considered a simplified model where only pairs of atoms have been taken into account. A more realistic model should include higher-order terms that account for cooperative effects between more than two atoms [21]. The purpose of the current model is to show that already for a dilute gas in the weak-disorder limit, cooperative effects modify significantly the transport properties of light.

ACKNOWLEDGMENTS

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APPENDIX A

In this appendix, we establish expressions (53) and (63) for the elastic mean free path and the group velocity. At resonance, simple expressions for the elastic mean free path (56) and the group velocity (67) are obtained by a pertubative expansion with respect to the small parameter $k_0 r_m$.

1. Elastic mean free path

The elastic mean free path is given by (53) in terms of the function f_1 defined in (54). The integral in (54) is easily carried out analytically, and it leads to

$$\frac{1}{l_e(\delta)} = \frac{6\pi n_i}{k_0^2}\frac{1}{aC_1}\left(\frac{b}{2a}A_1 + \frac{a-2}{2a}B_1 + C_1\right), \tag{A1}$$

where

$$a = 1 + (\delta/\Gamma)^2, \quad b = \delta/\Gamma, \tag{A2}$$

$$A_1 = \ln\left(\frac{\frac{1}{4}x_m^2}{a + bx_m + \frac{1}{4}x_m^2}\right), \tag{A3}$$

$$B_1 = \frac{\pi}{2} - \tan^{-1}\left(b + \frac{1}{2}x_m\right), \tag{A4}$$

and

$$C_1 = \frac{1}{x_m} = k_0 r_m \ll 1. \tag{A5}$$

At resonance ($\delta=0$) we have $a=1$, $b=0$ and by expanding (A4) with respect to $k_0 r_m$ we obtain

$$B_1 \simeq \frac{2}{x_m}\left(1 - \frac{4}{3x_m^2}\right). \tag{A6}$$

Thus,

$$l_e(0) = \frac{k_0^2}{8\pi n_i}\frac{1}{(k_0 r_m)^2} \tag{A7}$$

as given in (56).

2. Group velocity

The group velocity is given by (63) in terms of the function f_2 defined in (65). The integral in (65) is easily carried out analytically and it yields

$$\frac{c}{v_g(\delta)} = 1 - \frac{n_i}{n_c}\frac{F_1}{a^2 C_1}, \tag{A8}$$

where

$$F_1 = b\left(\frac{1}{a} - \frac{1}{4}\right)A_1 + \frac{a-2}{4}A_1' + \left(\frac{3}{2} - \frac{2}{a}\right)B_1 - bB_1' + \left(1 - \frac{a}{2}\right)C_1, \tag{A9}$$

$$A_1' = -\frac{b + \frac{1}{2}x_m}{a + bx_m + \frac{1}{4}x_m^2}, \tag{A10}$$

and

$$B_1' = -\frac{\frac{1}{2}}{1 + \left(b + \frac{1}{2}x_m\right)^2}. \tag{A11}$$

At resonance ($\delta=0$) we have $a=1$, $b=0$ and by expanding (A10) and (A4) with respect to $k_0 r_m$ we obtain

$$A_1' \simeq \frac{2}{x_m}\left(\frac{4}{x_m^2} - 1\right) \tag{A12}$$

and

$$B_1 \simeq \frac{2}{x_m}\left(1 - \frac{4}{3x_m^2}\right). \tag{A13}$$

Thus,

$$\frac{c}{v_g(0)} = 1 + \frac{2}{3}\frac{n_i}{n_c}(k_0 r_m)^2, \tag{A14}$$

as given in (67).

APPENDIX B

The aim of this appendix is to calculate the average self-energy (72) for a $\Delta m=0$ transition in the case where $k_0 r \ll 1$. First, we average the superradiative propagator (25) over the orientation of the inter-atomic axis and obtain analytical expressions for its real and imaginary parts. Then, by averaging over the interatomic distance up to r_m, we obtain the average self-energy (72).

For a $\Delta m=0$ transition and $k_0 r \ll 1$, the superradiative propagator (25) may be written with the help of (12) and (13) as

$$\hbar\Gamma G^+ = \left[\frac{\delta}{\Gamma} + \frac{3}{4}\left(\frac{3\cos^2\theta - 1}{(k_0 r)^3} + \frac{\frac{1}{2}(1+\cos^2\theta)}{k_0 r}\right) + i\right]^{-1}, \tag{B1}$$

where the interatomic axis is $\mathbf{r}=(r,\theta,\varphi)$. Averaging over the orientations

$$\hbar\Gamma\langle G^+\rangle = \frac{1}{4\pi}\int \hbar\Gamma G^+ d\cos\theta\, d\varphi \tag{B2}$$

yields for the imaginary part

$$\hbar\Gamma\ \mathrm{Im}\langle G^+\rangle = -\frac{P+Q}{\beta^2} \tag{B3}$$

and for the real part

$$\hbar\Gamma\ \mathrm{Re}\langle G^+\rangle = W_- P + W_+ Q, \tag{B4}$$

where we have defined

$$P = \frac{1}{8A_2\beta\cos(\gamma/2)}\ln\left(\frac{1 + 2\beta\cos(\gamma/2) + \beta^2}{1 - 2\beta\cos(\gamma/2) + \beta^2}\right), \tag{B5}$$

$$Q = \frac{1}{4A_2\beta \sin(\gamma/2)}\left(\frac{\pi}{2} + \tan^{-1}\frac{1-\beta^2}{2\beta \sin(\gamma/2)}\right), \quad \text{(B6)}$$

and

$$W\pm = -\sqrt{A_2}(\cos\gamma \mp 1). \quad \text{(B7)}$$

The auxiliary parameters are given by

$$\beta = \left(\frac{C_2}{A_2}\right)^{1/4}, \quad \gamma = \cos^{-1}\left(-\frac{B_2}{2\sqrt{A_2C_2}}\right), \quad \text{(B8)}$$

where

$$A_2 = \frac{9}{16(k_0r)^2}\left(\frac{3}{(k_0r)^2} + \frac{1}{2}\right)^2, \quad \text{(B9)}$$

$$B_2 = \frac{3}{4k_0r}\left(\frac{3}{(k_0r)^2} + \frac{1}{2}\right)\left[\frac{2\delta}{\Gamma} + \frac{3}{2k_0r}\left(\frac{1}{2} - \frac{1}{(k_0r)^2}\right)\right], \quad \text{(B10)}$$

and

$$C_2 = 1 + \frac{1}{4}\left[\frac{2\delta}{\Gamma} + \frac{3}{2k_0r}\left(\frac{1}{2} - \frac{1}{(k_0r)^2}\right)\right]^2. \quad \text{(B11)}$$

Finally, we average (B3) and (B4) over the interatomic distance up to r_m,

$$\hbar\Gamma\ \mathrm{Im}\langle\overline{G^+}\rangle = -\frac{1}{r_m}\int_0^{r_m} dr\frac{P+Q}{\beta^2} \quad \text{(B12)}$$

and

$$\hbar\Gamma\ \mathrm{Re}\langle\overline{G^+}\rangle = \frac{1}{r_m}\int_0^{r_m} dr(W_-P + W_+Q). \quad \text{(B13)}$$

The integrals can be evaluated numerically and give the average self-energy (72) since

$$\frac{1}{4\pi r_m}\int \hbar\Gamma G^+(\mathbf{r})d\mathbf{r} = \hbar\Gamma\langle\overline{G^+}\rangle. \quad \text{(B14)}$$

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