## Effect of Superradiance on Transport of Diffusing Photons in Cold Atomic Gases

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We show that in atomic gases cooperative effects like superradiance and subradiance lead to a potential between two atoms that decays like 1/r. 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, such as photon elastic mean free path and group velocity, is significantly different from that of independent atoms. We discuss the conditions under which these effects may be observed and compare our results to recent experiments on photon transport in cold atomic gases.

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The issue of coherent multiple scattering of photons in cold atomic gases is important since it presents a path towards the onset of 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 Anderson localization transition [1,2]. Another advantage of cold atomic gases is that sources of decoherence and inelastic scattering such as Doppler broadening can be neglected. 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 then of great interest since it may raise new issues in the Anderson problem such as 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 that lead to effective interactions between atoms [3]. These two differences may lead to qualitative changes of both mesoscopic quantities and Anderson localization. Some of the effects of a Zeeman degeneracy have been investigated in the weak disorder limit [4] using a set of finite phase coherence times [5] which reduce mesoscopic effects, such as coherent backscattering [1,6]. The aim of this Letter is to investigate the influence of cooperative effects and more specifically of superradiance on the multiple scattering of photons. We show that two atoms in a Dicke superradiant state [7] interact by means of a potential which, once averaged over disorder configurations, is attractive at short distances and decays like 1/r. This potential, analogous to the one considered in [8,9], has important consequences on transport properties since the contribution of superradiant pairs of atoms in a dilute gas provides smaller values of both group velocity and diffusion coefficient so that the photons become closer to the edge of Anderson localization.

Atoms are taken as degenerate two-level systems denoted by  $|g\rangle = |j_g = 0$ ,  $m_g = 0$ ) for the ground state and  $|e\rangle = |j_e = 1$ ,  $m_e\rangle$  for the excited state, where j is the total angular momentum and m is its projection on the 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  $\hbar\Gamma$ . We consider a pair of such atoms in an external radiation field and the corresponding Hamiltonian is  $H = H_0 + V$ , with

$$H_0 = \frac{\hbar\omega_0}{2} \sum_{l=1}^{2} [|e\rangle\langle e| - |g\rangle\langle g|]_l + \sum_{\mathbf{k}\epsilon} \hbar\omega_k a_{\mathbf{k}\epsilon}^{\dagger} a_{\mathbf{k}\epsilon}, \quad (1)$$

 $a_{\mathbf{k}\varepsilon}$  ( $a_{\mathbf{k}\varepsilon}^{\dagger}$ ) 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 may be written as

$$V = -\mathbf{d_1} \cdot \mathbf{E}(\mathbf{r_1}) - \mathbf{d_2} \cdot \mathbf{E}(\mathbf{r_2}), \tag{2}$$

where  $\mathbf{d}_l$  is the electric dipole moment operator of the *l*th atom and  $\mathbf{E}(\mathbf{r})$  is the electric field operator.

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  $\{|g_1e_{2i};0\rangle, |e_{1i}g_2;0\rangle\}$  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 one to find combinations of these states that are given by  $|\phi_i^{\epsilon}\rangle = \frac{1}{\sqrt{2}}[|e_{1i}g_2;0\rangle + \epsilon|g_1e_{2i};0\rangle]$  with  $\epsilon=\pm 1$ , so that  $\langle \phi_j^{\epsilon'}|U(t)|\dot{\phi}_i^{\epsilon}\rangle=\delta_{ij}\delta_{\epsilon\epsilon'}S_i^{\epsilon}(t)$  and

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$$S_i^{\epsilon}(t) = \langle e_{1i}g_2; 0|U(t)|e_{1i}g_2; 0\rangle$$
  
+  $\epsilon \langle g_1e_{2i}; 0|U(t)|e_{1i}g_2; 0\rangle.$  (3)

The states  $|\phi_i^{\epsilon}\rangle$  are the well-known Dicke states, otherwise defined as  $|LM\rangle$ , where L is the cooperation number and M is half of the total atomic inversion [7] so that  $|\phi_i^+\rangle = |10\rangle$  and  $|\phi_i^-\rangle = |00\rangle$ . For large times,  $t \gg r/c$ , where r is the distance between the two atoms, up to second order in the coupling to the radiation, we obtain that

$$S_i^{\epsilon}(t) \simeq 1 - \frac{it}{\hbar} \left[ \Delta E_i^{\epsilon} - i \frac{\hbar \Gamma_i^{\epsilon}}{2} \right].$$
 (4)

The two real quantities  $\Delta E_i^{\epsilon}$  and  $\Gamma_i^{\epsilon}$  are, respectively, the interacting potential and the probability per unit time of emission of a photon by the two atoms in a Dicke state  $|\phi_i^{\epsilon}\rangle$ . A standard calculation [10] gives

$$\Delta E_i^{\epsilon} = \epsilon \frac{3\hbar\Gamma}{4} \left[ -p_i \frac{\cos k_0 r}{k_0 r} + q_i \left( \frac{\cos k_0 r}{(k_0 r)^3} + \frac{\sin k_0 r}{(k_0 r)^2} \right) \right]$$
(5)

and

$$\frac{\Gamma_i^{\epsilon}}{\Gamma} = 1 - \frac{3}{2} \epsilon \left[ -p_i \frac{\sin k_0 r}{k_0 r} + q_i \left( \frac{\sin k_0 r}{(k_0 r)^3} - \frac{\cos k_0 r}{(k_0 r)^2} \right) \right], \quad (6)$$

where  $k_0 = \omega_0/c$ . We have defined  $p_i = 1 - \hat{\mathbf{r}}_i^2$  and  $q_i = 1 - 3\hat{\mathbf{r}}_i^2$ ,  $\hat{\mathbf{r}}$  being a unit vector along the two atoms. At short distance  $k_0 r \ll 1$ , we obtain that  $\Gamma_i^+ = 2\Gamma$  for the superradiant state  $|\phi_i^+\rangle = |10\rangle$  and  $\Gamma_i^- = 0$  for the subradiant state  $|\phi_i^-\rangle = |00\rangle$ .

For a photon of wave vector  $\mathbf{k}$  incident on an atomic cloud, the potential we shall denote by  $V_e$  is obtained by averaging upon the random orientations of the pairs of atoms. Since  $\langle q_i \rangle = 0$  and  $\langle p_i \rangle = 2/3$  regardless of i, we obtain for the average potential  $V_e$ 

$$\epsilon V_e(r) = \langle \Delta E_i^{\epsilon} \rangle = -\epsilon \frac{\hbar \Gamma}{2} \frac{\cos k_0 r}{k_0 r}$$
 (7)

and the average inverse lifetimes of Dicke states are

$$\langle \Gamma_i^{\epsilon} \rangle = \Gamma \left[ 1 + \epsilon \frac{\sin k_0 r}{k_0 r} \right], \tag{8}$$

which retains the same features as (6) for  $k_0 r \ll 1$ .

Let us characterize the interaction potential  $V_e$ . Whereas for a single pair of atoms, the potential (5) is anisotropic and decays at short distance like  $1/r^3$ , a behavior that originates from the transverse part of the photon propagator, we obtain that on average over angular configurations, the potential (7) between two atoms in a Dicke state M=0 becomes isotropic and decays like 1/r. This behavior is also obtained by considering the interaction of two-level atoms with a scalar wave. This could have been anticipated since the transverse contribution  $q_i$  to the photon propagator averages to 0. A similar expression for the interacting potential has been obtained for the case of an intense radiation field [8,9]. But this latter potential is fourth order in the coupling to the radiation and it corresponds to the

interaction energy between two atoms in their ground state in the presence of at least one photon. The average potential  $V_e$  we have obtained is different. It is second order in the coupling to the radiation and it corresponds to the interaction energy of Dicke states M=0 in vacuum.

We turn now to scattering properties of Dicke states. The collision operator is given by T(z) = V + VG(z)V, where V is given by (2) and G(z) is the resolvent whose expectation value in a Dicke state M=0 is obtained by a summation of the series of exchange of a virtual photon between the two atoms. The matrix element that describes the transition from the initial state  $|i\rangle = |1-1; \mathbf{k}\hat{\varepsilon}\rangle$  where the two atoms are in their ground state in the presence of a photon  $(\mathbf{k}\hat{\varepsilon})$  to the final state,  $|f\rangle = |1-1; \mathbf{k}'\hat{\varepsilon}'\rangle$ , is the sum of the superradiant and subradiant contributions,  $T=T^++T^-$ , with  $T^\pm=\langle f|V|\phi^\pm\rangle\langle\phi^\pm|G(\omega-\omega_0)|\phi^\pm\rangle\times\langle\phi^\pm|V|i\rangle$  [11]. A standard derivation leads to the following expressions for the average amplitudes  $T_e^\pm$ 

$$T_e^+ = Ae^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}} \cos\left(\frac{\mathbf{k} \cdot \mathbf{r}}{2}\right) \cos\left(\frac{\mathbf{k}' \cdot \mathbf{r}}{2}\right) G_e^+ \tag{9}$$

and

$$T_e^- = Ae^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}} \sin\left(\frac{\mathbf{k} \cdot \mathbf{r}}{2}\right) \sin\left(\frac{\mathbf{k}' \cdot \mathbf{r}}{2}\right) G_e^-.$$
 (10)

We have defined  $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ ,  $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ , and  $A = \frac{\hbar \omega}{\epsilon_0 \Omega} d^2 (\hat{e}_j \cdot \hat{\epsilon}) (\hat{e}_j^* \cdot \hat{\epsilon}'^*)$  (d is a reduced matrix element and  $\Omega$  the quantization volume). The average propagators  $G_e^{\pm}$  associated, respectively, to the superradiant and subradiant states are,

$$G_e^{\pm} = \langle \phi^{\pm} | G(\delta) | \phi^{\pm} \rangle = \frac{1}{\hbar (\delta + i \frac{\Gamma}{2} \pm \frac{\Gamma}{2} \frac{e^{ik_0 r}}{k \cdot r})}, \quad (11)$$

where close to resonance,  $\delta = \omega - \omega_0 \ll \omega_0$  and where we have used (7) and (8) for the average potential and for the average inverse lifetimes. At short distances  $k_0 r \ll 1$ , the subradiant amplitude  $T_e^-$  becomes negligible as compared to the superradiant term (9). Therefore, the potential (7) is attractive and decays like 1/r. We can interpret these results by saying that, at short distances  $(k_0 r \ll 1)$ , the time evolution of the initial state  $|\psi(0)\rangle = |e_1, g_2; 0\rangle =$  $\frac{1}{\sqrt{2}}[|\phi^+\rangle + |\phi^-\rangle]$  corresponds for times shorter than  $1/\Gamma$ to a periodic exchange of a virtual photon between the two atoms at the Rabi frequency  $(\langle \Delta E^- \rangle - \langle \Delta E^+ \rangle)/\hbar \simeq$  $\Gamma/(k_0 r)$  which is much larger than  $\Gamma$ . For larger times, the two atoms return to their ground state and a real photon  $(\mathbf{k}'\hat{\epsilon}')$  is emitted. At large distances  $(k_0r\gg 1)$ , the Rabi frequency becomes smaller than  $\Gamma$ , so that the excitation energy makes only a few oscillations between the two atoms, thus leading to a negligible interaction potential

It is interesting to derive the previous results in another way that emphasizes the analogy with weak localization corrections [1,2].

To that purpose, we write the scattering amplitude T defined previously as a superposition of two scalar amplitudes  $T_1$  and  $T_2$  [13], each of them being a sum of single scattering and double scattering contributions, that is

$$T_1 = \frac{t}{1 - t^2 G_0^2} \left[ e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{r}_1} + t G_0 e^{i(\mathbf{k} \cdot \mathbf{r}_1 - \mathbf{k}' \cdot \mathbf{r}_2)} \right]$$
(12)

and

$$T_2 = \frac{t}{1 - t^2 G_0^2} \left[ e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{r}_2} + t G_0 e^{i(\mathbf{k} \cdot \mathbf{r}_2 - \mathbf{k}' \cdot \mathbf{r}_1)} \right]. \tag{13}$$

Here  $t = (2\pi\Gamma/k_0)/(\delta + i\Gamma/2)$  is the amplitude of a scalar wave scattered by a single atom at the origin and the prefactor  $t/(1-t^2G_0^2)$  where  $G_0 = -e^{ik_0r}/4\pi r$  accounts for the summation of the series of virtual photon exchange between the two scatterers. We single out in the total amplitude  $T = T_1 + T_2$ , the single scattering contribution  $T_s$ , and write the intensity associated to the double scattering term shown in Fig. 1 as

$$|T - T_s|^2 = 2 \left| \frac{t^2 G_0}{1 - t^2 G_0^2} \right|^2 [1 + \cos(\mathbf{k} + \mathbf{k}') \cdot (\mathbf{r}_1 - \mathbf{r}_2)].$$
(14)

We recognize in the bracket the well-known Cooperon interference term which is at the basis of coherent effects in quantum mesoscopic systems such as weak localization and coherent backscattering [1,2,6]. The interference term reaches its maximum value 1 for  $\mathbf{r}_1 = \mathbf{r}_2$  so that we obtain from (12) and (13) that  $T_1 = T_2 \propto (1/2)T_e^+$ , up to a proportionality factor [13]. Thus, the total amplitude is exactly given by the superradiant term with no subradiant contribution.

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 fulfills  $r \ll \lambda_0 \ll n_i^{-1/3}$ , where  $n_i$  is the density of pairs and  $\lambda_0 = 2\pi/k_0$  is the atomic transition wavelength. Based on this inequality, we may consider the two atoms that form a superradiant pair through exchange of a virtual photon as an effective scatterer and neglect cooperative interactions

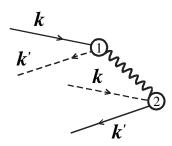


FIG. 1. Diagrammatic representation of the two amplitudes that describe double scattering of a scalar wave. The wavy line accounts for the photon exchange between the two atoms. This diagram is known in quantum mesoscopic physics as a Cooperon.

between otherwise well-separated pairs. The photon behavior is described by the configuration average of its Green's function, whose expression is obtained from a standard derivation [1]. In the limit of large enough densities of weakly scattering pairs, it reduces to the calculation of a self-energy given in terms of the average propagator (11) by

$$\overline{\Sigma}_{e}^{(1)} = \frac{6\pi\hbar\Gamma n_{i}}{k_{0}}\overline{G}_{e}^{+} = \frac{6\pi n_{i}}{k_{0}r_{m}} \int_{0}^{r_{m}} \frac{dr}{\frac{\delta}{\Gamma} + \frac{1}{2k_{0}r} + i}.$$
 (15)

The average, denoted by ..., is taken over distances r up to a maximal value  $r_m \ll k_0^{-1}$  which accounts for all possible mechanisms that may break those pairs. In the considered limit, the density of the gas can be assimilated to that of the pairs. The imaginary part of  $\overline{\Sigma}_e^{(1)}$  defines the elastic mean free path  $l_e$  by  $k_0/l_e = -\mathrm{Im}\overline{\Sigma}_e^{(1)}$ , namely,

$$\frac{1}{l_e} = \frac{3\pi n_i}{k_0^2} \frac{1}{k_0 r_m} \int_0^{2k_0 r_m} \frac{dx}{1 + (\frac{\delta}{\Gamma} + \frac{1}{r})^2}.$$
 (16)

It is interesting to compare  $l_e$  to the mean free path  $l_0 = \frac{k_0^2}{6\pi n_i}[1+(2\delta/\Gamma)^2]$  that corresponds to near resonant elastic scattering of a photon by independent atoms. At resonance  $(\delta=0)$ , we have  $l_0/l_e=\frac{4}{3}(k_0r_m)^2\ll 1$ . Away from resonance, the elastic mean free path  $l_e$  becomes smaller than  $l_0$  and for blue detuning it is reduced in a ratio roughly given by  $1/(k_0r_m)^2$ .

Another important physical quantity is the group velocity  $v_g$  given in terms of the refraction index  $\eta$  by  $c/v_g = \eta + \omega \frac{d\eta}{d\omega}$ . Since  $\overline{\Sigma}_e^{(1)}$  is proportional to the polarizability, the refraction index depends on its real part, namely  $\eta = [1 - (c/\omega)^2 \operatorname{Re} \overline{\Sigma}_e^{(1)}]^{1/2}$ . From (15), we notice that  $\eta \simeq 1$  for all values of the detuning  $\delta/\Gamma$  and in a large range of densities  $n_i$  so that

$$\frac{c}{v_g} = 1 - \frac{n_i}{n_c} \frac{1}{2k_0 r_m} f\left(k_0 r_m, \frac{\delta}{\Gamma}\right), \tag{17}$$

where we have defined  $n_c = (k_0^3/6\pi) \frac{\Gamma}{\omega_0}$  and the function

$$f(k_0 r_m, \Delta) = \int_0^{2k_0 r_m} dx \frac{1 - (\Delta + \frac{1}{x})^2}{[1 + (\Delta + \frac{1}{x})^2]^2}.$$
 (18)

This expression of  $v_g$  diverges at a large and negative value of the detuning  $\frac{\delta}{\Gamma} \simeq -1/(2k_0r_m)$  and beyond it takes both positive and negative values. Otherwise it is well behaved, meaning that it remains finite and positive for all values of the density  $n_i$ . At resonance, the group velocity is

$$\frac{c}{v_g} = 1 + \frac{4\pi n_i}{k_0^3} \frac{\omega_0}{\Gamma} (k_0 r_m)^2.$$
 (19)

The present expression of  $v_g$  differs substantially from the one obtained for light interaction with independent two-level atoms. There, for densities  $n_i > n_c$  where  $n_c$  defined above is usually overwhelmingly small, the group

velocity is known to diverge at two symmetric values of the detuning of order unity and takes negative values in between (i.e., also at resonance). For instance, in a gas of <sup>85</sup>Rb atoms, where  $n_i = 6 \times 10^{10}$  cm<sup>-3</sup>,  $\lambda_0 = 780$  nm, and  $\frac{\Gamma}{2\pi} = 5.9$  MHz, we have  $n_i/n_c \simeq 10^5$ . The validity of the concept of group velocity in such systems has thus been often questioned [14] and an energy velocity has been defined which describes energy transport through a diffusive medium [15].

Transport of photons through a diffusing gas is characterized by the diffusion coefficient  $D = \frac{1}{3}v_g l_e$  that combines the elastic mean free path and the group velocity [1,16], both derived from the complex valued self-energy (15). The diffusion coefficient D is of great importance since it enters in expressions of all measured physical quantities such as reflection and transmission coefficients, angular correlations of speckle patterns, time correlation functions of the intensity (diffusing wave spectroscopy), etc. [1]. Moreover, the critical behavior of transport close to Anderson localization transition at strong disorder is also obtained from the scaling form of D. Its expression, deduced from (16) and (17), depends on the range  $r_m$  and on the detuning  $\delta/\Gamma$ . Since the group velocity and the elastic mean free path are significantly modified for the case of superradiant pairs, we thus expect the diffusion coefficient to be different from its value obtained for independent atoms. We also define the transport time by  $\tau_{\rm tr}(\delta) = l_e/v_g = 3D/v_g^2$ . At resonance, it can be rewritten with the help of (19) as  $\tau_{tr}(0) = \frac{1}{2\Gamma}$  which is consistent with our considering of superradiant pairs. We would like nevertheless to call attention to the fact that, away from resonance,  $\tau_{\rm tr}(\delta)$  depends on frequency.

We now compare our results to recent measurements of the diffusion coefficient D and of the group velocity  $v_{\varrho}$ obtained for multiple scattering of light at resonance, in a cold atomic gas of  $^{85}$ Rb [17]. Since the range  $r_m$  cannot be directly determined, we first use Eqs. (16) and (17) to obtain an expression independent of  $k_0 r_m$  given by the ratio  $\frac{(v_g/c)^2}{3D} = 8\pi n_c/k_0^2 c = 2\Gamma/c^2$ . For <sup>85</sup>Rb atoms, this ratio equals  $8.2 \times 10^{-10}$  s/m<sup>2</sup>, which is in good agreement with the value  $4.8 \times 10^{-10}$  s/m<sup>2</sup> obtained from measurements of D and  $v_g$ . Finally, from the previous numerical expression we deduce for the maximal range of interaction  $r_m$  the value  $k_0 r_m \simeq 0.51$  also consistent with our assumption of superradiant states. Therefore, multiple scattering of photons by superradiant pairs provides a relevant mechanism that needs to be considered, in addition to others, e.g., scattering by independent atoms, for description of multiple scattering properties of dilute cold atomic gases.

We have considered multiple scattering of a photon on pairs of atoms that are in a superradiant state. On average over disorder configurations, an attractive interaction potential builds up between close enough atoms that decays like 1/r. The contribution of superradiant pairs, resulting from this potential, to scattering properties is significantly different from that of independent atoms. It leads to a well defined but much smaller group velocity as compared to c and correlatively to a smaller diffusion coefficient. For densities considered in recent experiments on cold  $^{85}$ Rb atoms, the quantity  $k_0 l_e$  that describes eventually the closeness to a localization transition, is reduced at moderate detunings, by 1 order of magnitude. This effect is expected to be even stronger for larger densities which could then be close to the localization edge.

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