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Interplay between radiation pressure force and scattered light intensity in the cooperative scattering by cold atoms


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The interplay between the superradiant emission of a cloud of cold two-level atoms and the radiation pressure force is discussed. Using a microscopic model of coupled atomic dipoles driven by an external laser, the radiation field and the average radiation pressure force are derived. A relation between the far-field scattered intensity and the force is derived, using the optical theorem. Finally, the scaling of the sample scattering cross-section with the parameters of the system is studied.

Keywords: cold atoms; Dicke superradiance; cooperative scattering

1. Introduction

Cooperative effects occur when the behavior of a many body system is determined by their collective interactions with each other and thus manifest themselves in a large variety of physical systems. In this paper, we focus on the specific case of a collection of atoms illuminated by a laser. In this situation, the electro-magnetic field mediates resonant dipole–dipole interactions between the atoms, leading to a cooperative response of the system, which quantitatively differs from the single atom response. Such effects are imprinted on physical observables that can be experimentally measured such as e.g. the emission diagram or the radiation pressure force acting on the cloud.

When a single atom is illuminated by a laser, the scattering process results in a force proportional to the number of scattered photons. Indeed, as an atom absorbs a photon from the laser of wave vector \( \mathbf{k}_0 \), it acquires a momentum \( \hbar \mathbf{k}_0 \), but the average momentum change during the emission process is zero.

For a collection of atoms, the picture changes drastically as it was first noticed in a pioneering work by Dicke [1] where he showed enhanced spontaneous emission decay rates in small and large samples due to constructive interferences of collective emission. In the situation of an incident laser scattering on a cloud of atoms, the atoms cooperate to scatter the light leading to a directional emission. This phenomenon is due to the synchronization of the atomic dipoles with the laser. The collective effects become even stronger as the atomic medium becomes optically dense and the radiation of the atoms starts to alter significantly the wave propagation. Among the other collective effects that arise, one can mention the collective Lamb shift [2,3], Mie resonances [4], subradiance [5], the refractive index of a dilute Bose gas [6] as well as a reduction of the radiation pressure force [7,8].

Since the radiated light results from the interference of the waves emitted by each dipole, the simple relation between emitted photon and atomic recoil is lost. For example, a striking feature of cooperativity is the modification of the atomic recoil due to the presence of the neighboring atoms [9,10], an effect that cannot be deduced from single-atom physics.

We here discuss the particular relation between the directional superradiant emission, and the reduction of the radiation pressure force. The atomic cloud is described as a microscopic ensemble of coupled atomic dipoles, and both the radiated field and the force are expressed as a function of these dipoles. The optical theorem is derived in this framework, and is shown to lead to a direct relation between intensity scattered and radiation pressure force for the cloud center-of-mass.

2. Cooperative scattering model

The atomic cloud is described as a system of two-level \((g \text{ and } e)\) atoms, with resonant frequency \( \omega_a \) and position...
the single-photon Rabi frequency, where \( \Omega \) is the electric-dipole transition matrix element and \( V \) is the photon mode volume. The special case where a single photon (mode \( k \)) can be assumed to be present in the system, was extensively investigated in [2,11,12], and later extended to include a low-intensity laser in [7,13,14]. The system atoms+photons is then described by a state of the form [15]:

\[
|\Psi\rangle = \alpha(t)|g_1 \ldots g_N\rangle|0\rangle_k + \exp(-i\Delta_0 t) \sum_{j=1}^{N} \beta_j(t)|g_1 \ldots e_j \ldots g_N\rangle|0\rangle_k + \sum_{k} \gamma_k(t)|g_1 \ldots g_N\rangle|1\rangle_k + \sum_{k,m,n=1}^{N} \epsilon_{m<n, k(t)}|g_1 \ldots e_m \ldots e_n \ldots g_N\rangle|1\rangle_k.
\]

(2)

The first term in Equation (2) corresponds to the initial ground state without photons, the second term is the sum over the states where a single atom has been excited by the classical field. The third term corresponds to the atoms that returned to the ground state having emitted a photon in the mode \( k \), whereas the last one corresponds to the presence of two excited atoms and one virtual photon with ‘negative’ energy. It is due to the counter-rotating terms in Hamiltonian (1) and this disappears when the rotating wave approximation is made. In the linear regime \( \alpha \approx 1 \) and in the Markov approximation, valid if the decay time is larger than the photon time-of-flight through the atomic cloud, the scattering problem reduces to the following differential equation [13,14,16]

\[
\dot{\beta}_j = \left( i\Delta_0 - \frac{\Gamma}{2} \right) \beta_j - i\frac{\Omega_0}{2} \exp(i k_0 \cdot r_j) - \frac{\Gamma}{2} \sum_{m \neq j} \exp(i k_0 |r_j - r_m|) \beta_m
\]

with initial condition \( \beta_j(0) = 0 \), for \( j = 1, \ldots, N \). Here, \( \Gamma = g_{0}^{2} \kappa_{0}^{2}/\pi c = d^{2}k_{0}^{2}/2\pi c\epsilon_0 h \) is the single-atom spontaneous decay rate. The kernel in the last term of Equation (3) has a real component, \(-\Gamma/2\sum_{m \neq j} \langle \sin(x_{jm})/x_{jm} \rangle \) (where \( x_{jm} = k_0 |r_j - r_m| \), describing the collective atomic decay, and an imaginary component, \( i\Gamma/2\sum_{m \neq j} \cos(x_{jm})/x_{jm} \), describing the collective Lamb shift [16–18]. Notice that while Equation (3) is here deduced from a quantum mechanical model, it can also be obtained classically, treating the two-level atoms as weakly excited classical harmonic oscillators [15,19].

3. Radiated field

The radiation field operator \( \hat{a}_k \) evolves according to the following Heisenberg equation

\[
\frac{d\hat{a}_k}{dt} = \frac{1}{i\hbar} \left[ \hat{a}_k, \hat{H} \right]
\]

\[
= -ig_k \exp[i(\omega_k - \omega_s)t] \sum_{m=1}^{N} \hat{\sigma}_m \exp(-i\hat{k} \cdot \hat{r}_m),
\]

(4)

where the fast oscillating term proportional to \( \exp[i(\omega_k + \omega_s)t] \) has been neglected. The scattered field is obtained by performing the sum over all the modes, considering only the positive-frequency part of the electric field operator

\[
\hat{E}_s(r,t) = \sum_k \hat{E}_s(r)\hat{a}_k(t) \exp(i\hat{k} \cdot \hat{r} - i\omega_k t),
\]

(5)

where \( \hat{E}_s = (\hbar \omega_0/2\epsilon_0 V)^{1/2} \). Integrating Equation (4) with respect to time, with \( \omega_s(0) = 0 \), inserting it in Equation (5), and assuming the usual Markov approximation, one obtains [14]

\[
\hat{E}_s(r,t) \approx -\frac{dk_{0}^{3}}{4\pi \epsilon_0} \exp(-i\omega_0 t) \sum_{m=1}^{N} \exp(i k_0 |r - r_m|) \hat{\sigma}_m(t).
\]

(6)

When applied on the state (2), neglecting virtual transitions, it yields \( \hat{E}_s|\Psi\rangle = E_s \exp(-i\omega_0 t)|g_1 \ldots g_N\rangle \), with

\[
E_s(r,t) = -\frac{\hbar \Gamma}{2d} \sum_{m=1}^{N} \beta_m(t) \exp(i k_0 |r - r_m|) - k_0 |r - r_m| \hat{\sigma}_m(t).
\]

(7)

Hence, the radiated field appears as a sum of spherical waves radiated by the atomic dipoles. In the far-field limit, one has \( k_0 |r - r_m| \approx k_0 r - k \cdot r_m \), with \( k = k_0 r/r \), so the field (7) radiated in a direction \( k \) reads

\[
E_{s}(\text{far})(k,t) \approx -\frac{\hbar \Gamma}{2d} \exp(i k_0 r) \sum_{m=1}^{N} \beta_m(t) \exp(-i k \cdot r_m).
\]

(8)
The scattered intensity in a direction \( \mathbf{k} \) is then derived as
\[
I_s(\mathbf{k}) = \frac{\epsilon_0 c \hbar^2 \Gamma^2}{2(d k_0 r)^2} \left[ \sum_{m=1}^{N} \beta_m(t) \exp(-i \mathbf{k} \cdot \mathbf{r}_m) \right]^2
\]
\[
= \frac{\epsilon_0 c \hbar^2 \Gamma^2}{2(d k_0 r)^2} \times \left( \sum_{m=1}^{N} |\beta_m|^2 + \sum_{j \neq m} \beta_j \beta_m^* \exp(-i \mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_m)) \right).
\]

Integrating this intensity over all directions leads to the total scattered power
\[
P_s = \frac{d^2 k_0^4 c}{2 \pi \epsilon_0} \left( \sum_{m=1}^{N} |\beta_m|^2 + \sum_{m \neq j} \beta_j \beta_m^* \frac{\sin(k_0 |\mathbf{r}_j - \mathbf{r}_m|)}{k_0 |\mathbf{r}_j - \mathbf{r}_m|} \right),
\]
where we have used the equality
\[
\int d\mathbf{k} e^{i \mathbf{k} \cdot \mathbf{d}} = 4\pi \frac{\sin(k_0 |d|)}{k_0 |d|}.
\]

In Equation (11), the first term corresponds to the *incoherent* sum of the single atom radiated power. The second term is an interference term; in the limit of a cloud small compared to the wavelength, the dipole moments have the same phase and this latter term is responsible for a superradiant build-up of the radiated power \( \propto N^2 \) (see, e.g. [1]).

### 4. Radiation pressure force

As for the radiation force operator acting on the \( j \)th atom, it is derived from Equation (1) as
\[
\hat{F}_j = -\nabla_j \hat{H} = \hat{F}_{aj} + \hat{F}_{ej}.
\]
A first contribution associated with the absorption of photons of the pump appears [7,13]:
\[
\hat{F}_{aj} = i\hbar k_0 \frac{\Omega_0}{2} \left[ \hat{\sigma}_j \exp[i(\Delta \omega t - k_0 \cdot \mathbf{r}_j)] - \text{h.c.} \right],
\]
whereas the second contribution comes from the emission of the photons in any direction \( \mathbf{k} \):
\[
\hat{F}_{ej} = i\hbar \sum_{\mathbf{k}} \mathbf{k} g_k \left\{ \hat{a}_k^\dagger \hat{\sigma}_j \exp[i(\omega_k - \omega_\alpha) t - i \mathbf{k} \cdot \mathbf{r}_j] - \hat{\sigma}_j^\dagger \hat{a}_k \exp[-i(\omega_k - \omega_\alpha) t + i \mathbf{k} \cdot \mathbf{r}_j] \right\}.
\]

In Equation (15), the counter-rotating terms proportional to \( \exp[\pm i(\omega_k + \omega_\alpha) t] \) were neglected.

As we are interested in comparing the radiation pressure force to the single-atom case, we define the average radiation force \( \bar{\hat{F}} = (1/N) \sum_j \hat{F}_j = (\hat{F}_{tot}/N)\hat{e}_z \) that measures acceleration of the cloud center-of-mass given by \( \mathbf{a}_{CM} = \bar{\hat{F}} / m \), with \( m \) the single-atom mass. Note that this average force is \( N \) times smaller than the total force \( \hat{F}_{tot} \) acting on the whole cloud of atoms. Since we consider clouds with rotational symmetry around the laser axis, this force is in the same direction as the incident field wave vector \( \mathbf{k}_0 = k_0 \hat{e}_z \). This average force is measured by time-of-flight techniques in cold atomic clouds released, for instance, from magneto-optical traps (MOTs) and has recently revealed cooperative effects in the scattering by extended atomic samples [8,20]. Like the scattered radiation, this force is an observable that contains signatures of the cooperative scattering by the atoms [7,8]. The average absorption force along the \( z \)-axis, resulting from the recoil received upon absorption of a photon from the incident laser, reads
\[
\hat{F}_a = \frac{i}{2N} \hbar k_0 \Omega_0 \sum_j \left[ \hat{\sigma}_j \exp[i(\Delta \omega t - k_0 \cdot \mathbf{r}_j)] - \text{h.c.} \right].
\]

Similarly, the average emission force writes \( \hat{F}_e = (1/N) \sum_j \hat{F}_{ej} \). Inserting the expression for \( \hat{a}_k \) from Equation (4) into Equation (15), and approximating the discrete sum over the modes \( \mathbf{k} \) by an integral, it is possible to obtain, as was done for the radiation field operator \( \hat{E}_S \) of Equation (7), the following expression for the average emission force along the \( z \)-axis [7]:
\[
\hat{F}_e = \frac{-\hbar k_0 \Gamma}{2\pi N} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \cos \theta \times \sum_{j,m=1}^{N} \left[ \exp[-i \mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_m)] \hat{\sigma}_j^\dagger \hat{\sigma}_j + \text{h.c.} \right].
\]

Neglecting virtual photon contributions, the expectation values of the absorption and emission forces for state (2) are
\[
\langle \hat{F}_a \rangle = -\frac{\hbar k_0 \Omega_0}{N} \sum_{j=1}^{N} \text{Im} \left[ \beta_j \exp(-i \mathbf{k}_0 \cdot \mathbf{r}_j) \right],
\]
\[
\langle \hat{F}_e \rangle = -\frac{\hbar k_0 \Gamma}{4\pi N} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \cos \theta \times \sum_{j,m=1}^{N} \left( \beta_j \beta_m^* \exp[-i \mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_m)] \right)
\]
\[
= -\frac{\hbar k_0 \Gamma}{N} \sum_{j,m=1}^{N} \frac{(z_j - z_m)}{|\mathbf{r}_j - \mathbf{r}_m|} f_j(k_0|\mathbf{r}_j - \mathbf{r}_m|) \text{Im} (\beta_j \beta_m^*). \]

where we used the identity
\[
\int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \cos \theta \exp[-i \mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')] = 4\pi i \frac{z - z'}{|\mathbf{r} - \mathbf{r}'|} f_j(k_0|\mathbf{r} - \mathbf{r}'|).
\]

\( f_j(z) \) here refers to the first-order spherical Bessel function.

Note that the decomposition into absorption (18) and emission (19) forces is fully compatible with classical expressions of the optical force [21], where the force arises as the product between the atomic dipole and the *total* field [22] (i.e. including the radiation from the other atoms).
5. Optical theorem

Let us now discuss the formulation of the optical theorem in the framework of collective scattering. To that purpose, we consider an infinite slab illuminated by a plane wave. In the far-field limit, the field in a direction \( \hat{k} \) is

\[
E(\mathbf{r}) = \frac{E_0}{2} \left[ \exp(ik_0z) + E_s^{(\text{far})}(r, \hat{k}) \right] \exp(-i\omega_0t) \\
= \frac{E_0}{2} \left[ \exp(ik_0z) - \frac{\exp(ik_0r)}{k_0r} f(\hat{k}) \right] \exp(-i\omega_0t),
\]

where the scattering amplitude for the scattered field \( f \) is given by

\[
f(\hat{k}) = \frac{\Gamma}{\Omega_0} \sum_j \beta_j \exp(-i(k_0 \hat{k} \cdot \mathbf{r}_j)).
\]

As a consequence, the scattered intensity at a large distance \( r \) from the cloud is

\[
I_s = I_0 \frac{|f(\hat{k})|^2}{k_0^2 r^2},
\]

while the total scattering cross-section is obtained by integrating over all the solid angle

\[
\sigma_{\text{sca}} = \frac{1}{k_0^2} \int d\hat{k} |f(\hat{k})|^2.
\]

To simulate numerically the slab illuminated by a plane wave, we consider a cylinder of transverse size large compared to its thickness and to the wavelength, with a random homogeneous distribution of atoms. Figure 1 shows the emission diagram of the scattered field for resonant excitation and a cylindrical cloud of atoms. The energy conservation imposes that

\[
\sigma_{\text{ext}} = \sigma_{\text{sca}} + \sigma_{\text{abs}},
\]

where \( \sigma_{\text{ext}} \) and \( \sigma_{\text{abs}} \) are the cross-sections for extinction and absorption, respectively. The extinction cross-section is then obtained from the optical theorem. In the forward direction the total field is

\[
E_{\text{fwd}}(\theta = 0) = \frac{E_0}{2} \left[ \exp(ik_0z) - \frac{\exp(ik_0r)}{k_0r} f(0) \right] \exp(-i\omega_0t).
\]

In the slab configuration, the cloud radiates mainly in a narrow forward cone – the angle of the cone of emission is given by the inverse of the cloud transverse size. Hence, observing the field in a plane far from the atoms and within the forward cone of emission, the radius expands as \( r \approx z + (x^2 + y^2)/2z \), and one obtains

\[
E_{\text{fwd}}(r) \approx \frac{E_0}{2} \left[ 1 - \frac{f(0)}{k_0z} \exp[ik_0(x^2 + y^2)/(2z)] \right] \times \exp[i(k_0z - \omega_0t)].
\]

So the intensity reads

\[
|E_{\text{fwd}}(r)|^2 \approx \frac{|E_0|^2}{4} \left[ 1 - \frac{2}{k_0z} \text{Re} \left[ f(0) \exp[ik(x^2 + y^2)/(2z)] \right] \right],
\]

since we have neglected the quadratic term \( |E_s|^2 \). The measured intensity is the incident intensity minus the extinction intensity. In Equation (28), the integration over \( x, y \) yields a factor \( 2\pi z/k_0 \), and one gets

\[
\sigma_{\text{ext}} = -\frac{4\pi}{k_0^2} \text{Im}[f(0)].
\]

Hence, from Equation (24) one obtains the relation

\[
-\text{Im}[f(0)] = \frac{1}{4\pi} \int d\hat{k} |f(\hat{k})|^2 + \frac{k_0^2}{2\pi} \sigma_{\text{abs}}.
\]

In our microscopic description of the light–atom interaction there is no absorption, so that \( \sigma_{\text{abs}} = 0 \). An illustration of the validity of the optical theorem is given in Figure 2 for resonant light scattering by a slab containing two-level scatterers with a uniform density distribution. From Equations (22) and (29), and introducing the wavevector \( \mathbf{k} = k_0 \hat{k}(\theta, \phi) \), we obtain the relation

\[
- \frac{\Omega_0}{T} \sum_j \text{Im} \left[ \beta_j \exp(-i k_0 \cdot \mathbf{r}_j) \right] \\
= \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \times \sum_{j,m} \left[ \beta_j \beta_m^* \exp[-i k_0 \hat{k} \cdot (\mathbf{r}_j - \mathbf{r}_m)] \right].
\]
We observe from Equation (32) that the average radiation force along the z-axis reads:

\[
F_z = \frac{\hbar k_0 \Gamma}{4\pi N} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta (1 - \cos \theta) \left\{ \sum_{j,m=1}^N \left( \beta_j \beta_m^* \exp[-i k_0 |r_j - r_m|] \right) \right\},
\]

Equation (32)

Consequently, using Equations (18) and (19), the average force along the \(z\)-axis reads:

\[
F_z = \frac{\hbar k_0 \Gamma}{4\pi N} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta (1 - \cos \theta) I_s(\theta, \phi),
\]

Equation (33)

where the scattered far-field intensity is \(I_s(\theta, \phi) = 2\epsilon_0 |E_s(\theta, \phi)|^2\). This highlights the fact that the radiation pressure force, that pushes the atoms along the direction of the incident beam, is proportional to the net radiation flux of the scattered intensity.

In the case of an isotropic emission (e.g. single-atom case, or cloud much smaller than the wavelength), the scattered intensity \(I_s\) is independent of the angle and we get \(F_z = (4\pi \hbar^2/\epsilon_0) I_s\); the direct proportionality between scattered power and radiation pressure force is recovered.

The cooperative effect of light scattering in such small samples is then encoded in the total scattered intensity \(I_s\). In the case of superradiant scattering for larger samples, a pronounced emission into the forward direction decreases the radiation force, as observed for example in [8].

6. Scaling of the scattering cross-section

In this section we are interested in understanding how the scattering cross-section scales with the parameters of the system. We consider the case of a slab with uniform density distribution. The slab contains \(N\) atoms and its size along the \(x, y, z\) axes is denoted by \(L_x, L_y, L_z\) respectively. The numerical simulations presented in Figure 3 show how the scattering cross-section depends on the optical thickness of the cloud \(b_0 = 4\pi N/(\kappa_0^2 L_x L_y)\). For dilute clouds of atoms we find:

\[
\sigma_{\text{scat}} = 2.15 \times L_x L_y \left[ 1 - \exp\left( -\frac{b_0}{2.15} \right) \right].
\]

Equation (34)

When the slab is optically thick, i.e. \(b_0 \gg 1\), we observe that the cross-section appears to approach \(2 \times L_x L_y\). This factor of two corresponds to the well-known ‘extinction paradox’ [25,26] for which the extinction cross-section is twice as large as the one predicted by geometrical optics due to the diffraction contribution. The residual deviations from the factor of 2 between the scattering and geometrical cross-sections might be associated with a still moderate size of our sample [27], or to dipole blockade effects [28,29]. For spherical dielectric spheres, \(\sigma_{\text{ext}}\) shows an oscillatory behavior.
around $2\sigma_{\text{geo}}$ ($\sigma_{\text{geo}} = L_x L_y$ for our square geometry), which is damped for increasing sizes of the sphere [30,31]. When $b_0 \ll 1$ the scattering cross-section can be written as $\sigma_{\text{sca}} = (L_x L_y) b_0 = N \sigma_0$, where $\sigma_0 = \lambda^2/\pi$ is the resonant scattering cross-section for a single atom in the scalar wave description (it differs from the well-known cross-section for vectorial light $\sigma_0 = 3\lambda^2/(2\pi)$). In this limit, the interpretation is clear: at low optical thickness the cooperative effects are negligible and the scattering of light is given by the response of $N$ independent atoms. We refer the reader to [32] for a study of the areal scaling of the light scattering by varying the size of a dense, cold atomic cloud.

Before concluding, we would like to underline the importance of the role of diffraction. Since we are using a microscopic description of the system, diffraction effects for the scattered field are already included in our model. However, free propagation of the incident field needs to be added for a fully consistent description. In this respect, the incident plane wave considered so far in the paper is a peculiar case. We will focus on these aspects in forthcoming studies to precisely understand the role of diffraction. This will naturally lead us to compare our coherent microscopic model of coupled dipoles to stochastic incoherent models commonly used to describe photon propagation in random media. Understanding coherent light propagation in disordered resonant scatterers is of prime importance for both the atomic physics and the waves in complex media communities.

7. Conclusion

We here discussed the superradiant emission of a cloud of cold atoms, when the interference of the waves radiated by the atomic dipoles builds up a coherent emission. Despite the fact that the simple relation between absorbed photons and radiation pressure force existing in the single-atom case was lost, the optical theorem allowed one to recover a simple relation between the total scattered intensity and the displacement of the cloud center-of-mass. The measure of the force of the center of mass of the atomic cloud contains (partial) information on the scattered intensity, even for large values of optical thickness of the cloud. We have computed the total scattering cross-section which approaches a value close to twice the geometrical cross-section of the sample, in line with the well-known extinction paradox. Finally, understanding the role of diffraction paves the way for further studies to compare our coherent microscopic model to well-established stochastic incoherent models describing photon propagation in random media.

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