Influence of center-of-mass correlations on spontaneous emission and Lamb shift in dense atomic gases

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Abstract. Local field effects on the rate of spontaneous emission and Lamb shift in a dense gas of atoms are discussed taking into account correlations of atomic center-of-mass coordinates. For this the exact retarded propagator in the medium is calculated in independent scattering approximation and employing a virtual-cavity model. The resulting changes of the atomic polarizability lead to modifications of the medium response which can be of the same order of magnitude but of opposite sign than those due to local field corrections of the dielectric function derived by Morice, Castin, and Dalibard [Phys. Rev. A 51, 3896 (1995)].

1 Introduction

The experimental progress in cooling and trapping of atoms and the observation of Bose-Einstein condensation in atomic vapors has lead to a growing theoretical interest in the interaction of light with dense atomic gases. In the present paper it is analyzed how the spatial distribution of nearest neighbors in a dense gas, characterized by two-particle correlations, affects the interaction of an excited atom with the electromagnetic field vacuum. In particular modifications of the rate of spontaneous emission and the Lamb shift are calculated and the resulting modifications of the dielectric function discussed.

While the interaction of light with a dilute gas is well described in terms of macroscopic quantities with the well-known Maxwell-Bloch equations, this is no longer true when the gas becomes dense. Here two new types of effects arise: When the density of atoms becomes large enough such that the resonant absorption length is less than the characteristic medium dimension, re-absorption and multiple scattering of spontaneous photons need to be taken into account. Secondly with increasing density dipole-dipole interactions between nearest neighbors become important. Here the macroscopic picture of a homogeneous polarization breaks down and it is necessary to introduce local field corrections.

The most famous local-field correction leads to the Lorentz-Lorenz (LL) relation between atomic polarizability \( \alpha(\omega) \) and dielectric function \( \varepsilon(\omega) \). The LL-correction removes the unphysical contact interaction of two atoms at

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1 This paper is dedicated to the memory of Dan Walls
the same position which arises in a continuum picture of a homogeneous polarization and is independent of any specifics of the atoms. Recently Maurice, Castin, and Dalibard have derived a generalization of the LL relation that takes into account center-of-mass correlations for finite distances at which point specific properties of the atomic gas enter. They showed in particular that the tendency of bosonic atoms to bunch, when the critical temperature of condensation is approached, leads to a measurable change of the complex refractive index. Even more pronounced effects such as a dramatic line-narrowing were recently predicted by Ruostekoski and Javanainen for a Fermi gas in the case ideal case or in the presence of a BEC transition.

On the other hand, it is known since the early work of Purcell, that the microscopic environment of an excited atom can also change its interaction with the field vacuum. Thus local field effects should lead to a modification of the atomic polarizability itself, in particular the spontaneous emission rate and the Lamb shift. Different macroscopic models for Lorentz-Lorentz-type corrections of the spontaneous emission rate of an atom embedded in a lossless dielectric have been developed and experimentally tested. In the presence of losses, as is the case in dense gases of the same kind of atoms, macroscopic and microscopic approaches have indicated however that local-field effects due to nearest neighbors may be equally important as the LL correction of the contact interaction. In the present paper I analyze these effects using a Greens function approach.

According to Fermi’s golden rule the rate of spontaneous emission $\Gamma$ and the Lamb shift $\Delta$ are given by the (regularized) exact retarded propagator $D$ of the electric field at the position $r_0$ of the probe atom:

$$\Gamma = \frac{2}{\hbar} d \cdot \text{Re} \left[ D(r_0, r_0; \omega_0) \right] \cdot d,$$

$$\Delta = \frac{1}{\hbar} d \cdot \text{Im} \left[ D(r_0, r_0; \omega_0) \right] \cdot d.$$

$d$ is the dipole vector and $\omega_0$ the true transition frequency of the atom. From the known propagator in free space and after regularisation one finds for an isolated atom:

$$\Gamma = \Gamma_0 = \frac{d^2 \omega_0^3}{3\pi \hbar \epsilon_0 c^2}, \quad \Delta = 0.$$

The exact retarded propagator in a medium can formally be obtained from a scattering series. This series is here calculated neglecting multiple scattering of photons by the same atoms and taking into account only two-particle correlations of center-of-mass coordinates. It is shown that the alterations of the atomic polarizability $\alpha$ due to local-field corrections of spontaneous emission and Lamb shift lead to modifications of $\varepsilon$ which are of the same order of magnitude as those found by Maurice, Castin and Dalibard and Ruostekoski and Javanainen.
2 Scattering series for the retarded propagator

A dense medium affects the interaction of an excited probe atom with the surrounding electromagnetic vacuum by multiple scattering of virtual photons emitted and re-absorbed by that atom. These scattering processes can formally be described by the exact retarded Greens-function (GF)

\[ D(r_1, r_2; \tau) = \delta(\tau) \langle 0 \vert [\mathbf{E}(r_1, t_1), \mathbf{E}(r_2, t_2)] \vert 0 \rangle, \]

where \( \tau = t_1 - t_2 \) and \( \mathbf{E} \) is the operator of the electric field interacting with all atoms. The free-space or vacuum GF in the frequency domain is given by

\[ \mathbf{G}^0(\mathbf{x}, \omega) = -\frac{k^2 e^{i(k + s) \mathbf{x}}}{4\pi x} \left[ P(i k x) 1 + Q(i k x) \frac{x \cdot \mathbf{x}}{x^2} \right] + \frac{1}{3} \delta(\mathbf{x}) 1, \]

where \( D^0 = i \hbar \mathbf{G}^0/\epsilon_0 \). Here \( k = \omega/c, \ x = |\mathbf{x}| \) and

\[ P(z) = 1 - \frac{1}{z} + \frac{1}{z^2}, \quad Q(z) = -1 + \frac{3}{z} - \frac{3}{z^2}. \]

\( \mathbf{G}^0(\mathbf{x}, \omega) \) diverges as \( x \rightarrow 0 \) which is related to the large-\( q \) behavior in reciprocal space. This will lead to corresponding divergences in the exact propagator \( \mathbf{G}(\mathbf{x}, \omega) \) which can however be removed by introducing a regularisation \( \mathbf{G}(\mathbf{q}, \omega) \rightarrow \mathbf{G}(\mathbf{q}, \omega) f(\Lambda, q) \) with a wave-number cut-off \( \Lambda \). For the purpose of the present paper I will assume that the large-\( q \) behavior of the Greens function is properly regularized and ignore all contributions containing the regularisation parameter \( \Lambda \). The subject of regularisation will be discussed in more detail at a different place.

The net effect of all possible multiple scattering events can be described by a scattering series for the exact propagator. We here assume not too large densities, such that dependent scattering can be neglected. i.e. there can be as many scattering events as possible but never twice from the same atom. In this so-called independent scattering approximation (ISA), the scattering series can be expressed in the form

\[ \mathbf{G}(0, \omega) = \mathbf{G}^0(0, \omega) - \sum_{i \neq \emptyset} \mathbf{G}^0(\mathbf{x}_{\emptyset i}, \omega) \cdot \mathbf{a}_i(\omega) \cdot \mathbf{G}^0(\mathbf{x}_{\emptyset i}, \omega) + \]

\[ + \sum_{i \neq j \neq \emptyset} \mathbf{G}^0(\mathbf{x}_{\emptyset i \emptyset j}, \omega) \cdot \mathbf{a}_i(\omega) \cdot \mathbf{G}^0(\mathbf{x}_{\emptyset i \emptyset j}, \omega) \cdot \mathbf{a}_j(\omega) \cdot \mathbf{G}^0(\mathbf{x}_{\emptyset i \emptyset j}, \omega) + \cdots, \]

where \( \mathbf{a}_j(\omega) \) is the polarizability tensor of the \( j \)th atom of the host material, the summation is over all atomic positions, and \( \mathbf{x}_{ij} = \mathbf{r}_i - \mathbf{r}_j \). It should be noted that the polarizability of the excited probe atom does not enter the scattering series. However, the probe atom can affect the spatial distribution of the surrounding scatterers and it is necessary to keep track of its presence.

We now assume a homogeneous medium of density \( \rho \) with randomly oriented two-level atoms such that \( a_i(\omega) = a(\omega) 1 \). In this case we may replace
the sums over atomic positions by integrals. For this we introduce normalized joint probabilities $p_2(r_8, r_1), p_3(r_8, r_1, r_2)$ etc. to find one atom at position $r_1$ and the probe atom at $r_8$; to find two atoms at positions $r_1$ and $r_2$ and the probe atom at $r_8$ etc., with $p_1(r) = 1$. This leads to
\[
G(0) = G^0(0) - \varrho \alpha \int d^3r_i \ p_2(r_8, r_i) \ G^0(x_{i8}) \cdot G^0(x_i) + \\
+ \varrho^2 \alpha^2 \int d^3r_i d^3r_j \ p_3(r_8, r_i, r_j) \ G^0(x_{i8}) \cdot G^0(x_ij) \cdot G^0(x_{j8}) + \ldots
\]
where we have suppressed the frequency argument for notational simplicity.

For a dilute gas the positions of the atoms can be treated as independent and one can factorize all particle correlations, which amounts to $p_m \equiv 1$. The scattering series can then easily be solved. The poles of $G(q, \omega) = \int d^3x e^{iq \cdot x} G(x, \omega)$ determine the dielectric function for which one finds the well-known dilute-medium result
\[
\varepsilon(k) = \frac{\varrho_0}{k^2} = 1 + \varrho \alpha(\omega), \quad (\omega = kc)
\]
Properly regularizing $G(q, \omega)$ and transforming the result back to coordinate space eventually yields the spontaneous emission rate and the Lamb shift relative to the vacuum
\[
\Gamma = I_0 \text{Re} [\sqrt{\varepsilon}], \quad \Delta = \frac{I_0}{2} \text{Im} [\sqrt{\varepsilon}].
\]
Here $\varepsilon$ is the dielectric function of the gas at the true transition frequency. For an atom embedded in a dielectric host with real dielectric function the result for $\Gamma$ is identical to that obtained by Nienhuis and Alkemade based on a density-of-states argument $[\text{25}, \text{26}]$.

3 Local field effects and center-of-mass correlations

In order to obtain a non-perturbative result for the retarded propagator in a dense gas, one has to sum all contributions of the scattering series (\text{8}) without factorizing the center-of-mass correlations. Apart from some special cases such as hard-sphere scatterers $[\text{24}]$ it is not possible to bring the scattering series in an exact closed form and approximations are needed. The first approximation I use here is to take into account only two-particle correlations by using a Kirkwood-type factorization $[\text{28}]$ of higher-order contributions
\[
p_3(1, 2, 3) = p_2(1, 2) \ p_2(2, 3) \ p_2(1, 3), \quad \text{etc.}
\]
Furthermore it is assumed that only correlations between successive scatterers matter, which is however correct up to second order in the density. With this the scattering series (\text{8}) can be represented in the form
\[
G(0) = G^0(0) - \varrho \alpha \ H^0(x_{i8}) \cdot G^0(x_i) + \\
+ \varrho^2 \alpha^2 \ H^0(x_{i8}) \cdot H^0(x_ij) \cdot H^0(x_{j8}) - \\
- \varrho^3 \alpha^3 \ H^0(x_{i8}) \cdot H^0(x_{ij}) \cdot H^0(x_{jk}) \cdot H^0(x_{k8}) + \ldots
\]
\[
G(0) = G^0(0) - \varrho \alpha \ H^0(x_{i8}) \cdot G^0(x_i) + \\
+ \varrho^2 \alpha^2 \ H^0(x_{i8}) \cdot H^0(x_ij) \cdot H^0(x_{j8}) - \\
- \varrho^3 \alpha^3 \ H^0(x_{i8}) \cdot H^0(x_{ij}) \cdot H^0(x_{jk}) \cdot H^0(x_{k8}) + \ldots
\]
where the spatial integration has been suppressed and

\[ \mathbf{H}^\mu(r_1, r_2, \omega) = p_2(r_1, r_2) \mathbf{G}^\mu(r_1, r_2, \omega) \]

(13)
is the retarded propagator modified by the two-particle correlation \( p_2 \). Note that the first order term contains only a single function \( \mathbf{H}^\mu \).

Since two-particle correlations between atoms of the host material can be different from correlations between the (excited) probe atom and a host atom, we will distinguish these two in the following. This also includes the case of an atomic impurity in an environment of a different species. The scattering series can then be written in the form

\[ \mathbf{G}(0, \omega) = \mathbf{G}^\mu(0, \omega) - \hbar \alpha \int d^3r_1 \mathbf{H}_e^\mu(r_0, r_1, \omega) \cdot \mathbf{G}^\mu(r_1, r_2, \omega) + \]

\[ + \int d^3r_1 d^3r_2 \mathbf{H}_e^\mu(r_0, r_1, \omega) \cdot \mathbf{T}^{(2)}(r_1, r_2, \omega) \cdot \mathbf{H}_e^\mu(r_2, r_3, \omega), \]

(14)

where \( \mathbf{T}^{(2)}(r_1, r_2, \omega) \) is the part of the scattering matrix that contains at least two scattering processes in the host material. \( \mathbf{H}_e^\mu \) is the free propagator modified by the correlation of the excited probe atom with an atom of the background gas. Although in ISA the \( \mathbf{T} \)-matrix does not contain scattering events from the probe atom, it would in general still depend on its presence through the position correlations. With the earlier assumption that only correlations between successive scatterers matter, this dependence is lost. I.e. we treat the scattering of photons in the gas as if the place of the probe atom would be filled with the host material, which is equivalent to the virtual cavity model of Knoester and Mukanel [3]. From the discussion of impurities in cubic dielectric host materials by DeVries and Lagendijk [4] it is however expected that this approximation does not affect the results in leading order of the density. Finally we will restrict ourselves to the case of a homogeneous and isotropic gas, such that the two-particle correlation depends only on the distance between the atoms \( p_2(r_1, r_2) = p_2(|r_1 - r_2|) \) where \( x_{12} = |r_1 - r_2| \). In this case the scattering matrix obeys a simple Dyson equation in reciprocal space

\[ \mathbf{T}(q, \omega) = \hbar \alpha(\omega) \mathbf{H}_p^\mu(q, \omega) \hbar \alpha(\omega) - \hbar \alpha(\omega) \mathbf{H}_e^\mu(q, \omega) \cdot \mathbf{T}(q, \omega) \]

(15)

\( \mathbf{H}_p^\mu \) is the free propagator modified by the two-particle correlations of the host material.

It is convenient at this point to introduce the irreducible correlation \( \mathbf{H}_p^\mu \) according to \( p_2^\mu = 1 + \mathbf{H}_p^\mu \). One then has

\[ \mathbf{H}_p^\mu(q, \omega) = - \left( \frac{k^2}{3q^2} + \frac{2k^2}{3} \right) \frac{1 - q \cdot q}{q^2 - k^2 - i0} + f_1(q, \omega) \mathbf{1} + f_2(q, \omega) \frac{q \cdot q}{q^2}, \]

(16)

with

\[ f_1(q, \omega) = -k^3 \int_0^\infty dx \frac{\exp(-xq)}{x} h_0^\mu(x) \left[ j_0(qx) P(x) + j_1(qx) \frac{Q(x)}{x} \right], \]

(17)

\[ f_2(q, \omega) = k^3 \int_0^\infty dx \frac{\exp(-xq)}{x} h_0^\mu(x) j_2(qx) Q(x). \]

(18)
$j_n(z)$ are spherical Bessel functions and $P$ and $Q$ have been defined in eq. (3). In the first term of eq. (18), we have made use of $p_{3}(x)\delta(x) = 0$ which corresponds to the LL correction of the contact interaction. The solution of eq. (15) is now easy to obtain. The poles $q_0$ of $\mathbf{T}$ which determine the dielectric function follow from the equation

$$q_0^2 - k^2 = g_0(\omega) \left( \frac{1}{3} q_0^2 + \frac{2}{3} k^2 - f_1^2(q_0, \omega)(q_0^2 - k^2) \right) - i0 = 0.$$  \hspace{1cm} (19)

Since in lowest order of $g_0$ one has $q_0^2 \approx k^2 + i0$, we may replace $f_1^2(q_0, \omega)$ by $f_1^2(k, \omega)$ which yields

$$\varepsilon(k) = \frac{q_0^2}{k^2} = 1 + \frac{g_0(\omega)}{1 - g_0(\omega)/3 + g_0(\omega)f_1^2(k, \omega)}.$$ \hspace{1cm} (20)

This result is identical to that of Maurice, Castin and Dalibard \cite{lag} in ISA. It is interesting to note that although the free GF $\mathbf{H}^0$ contained also longitudinal components (proportional to $f_0^2 q \cdot q / q^2$), they exactly cancel in the expression for the dielectric function.

To obtain the spontaneous emission rate and Lamb shift, we consider only the leading order corrections in the density where we can replace $f_{1,2}^2(q_0, \omega)$ by $f_{1,2}^2(k, \omega)$ and introduce a regularisation of the large-$q$ behavior. This yields after some algebra for the orientation averaged retarded GF:

$$G(x = 0, \omega) = -\frac{i k^3}{6\pi} \left[ 1 + g_0 \left( \frac{7}{6} - f_1^2 \right) + \varepsilon_0 a^2 \left( \frac{17}{24} - \frac{7}{3} f_1^2 + f_1'^2 - \frac{7}{6} f_1^2 + 2 f_1^2 f_1^0 \right) + \cdots \right].$$ \hspace{1cm} (21)

One recognizes that there is again no contribution from the longitudinal terms $f_2$ up to second order in $q$. Furthermore the two-particle correlation $f_3^2$ between ground-state atoms enters only in second order of the density, while there is a first-order contribution from $f_1^2$. This is physically intuitive since correlations between ground-state atoms enter only after two scattering events, while correlations involving the probe atom are already important in first order. From the above results one finds in leading order of the density

$$\Gamma = \Gamma_0 \left[ 1 + \frac{7}{6} g_0 a' - g(a' f_1'' - a'' f_1') + \cdots \right],$$ \hspace{1cm} (22)

$$\Delta = \frac{\Gamma_0}{2} \left[ \frac{7}{6} g_0 a'' - g(a' f_1'' + a' f_1') + \cdots \right]$$ \hspace{1cm} (23)

where we have introduced the real and imaginary parts of $f_1 = f_1'' + i f_1'$. Eqs. (22) and (23) are the main result of the present paper. The first-order corrections to spontaneous emission rate and Lamb shift, which are independent on the $f$'s are the Lorentz-Lorenz local field corrections derived in \cite{lag}. 

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In order to illustrate the implications of eqs. (22) and (23) to the medium response, I will discuss in the following section the dielectric function of a dense gas of two-level atoms using some simple model functions for the center-of-mass correlations.

4 Modifications of medium response

We have shown that local field corrections change not only the relation between atomic polarizability and dielectric function of the medium but also the atomic polarizability itself (with respect to the thin-medium case). To illustrate the net effect of both corrections to the dielectric function, I now consider a classical, homogeneous gas of radiatively broadened (cold) two-level atoms with randomly oriented dipole vectors. The dimensionless atomic polarizability of such atoms is isotropic and has in free space the strength

$$\alpha^3 = \frac{6\pi}{k_0^3}$$  

where $k_0$ is the resonance wavenumber, $\delta = (\omega_{ab} - \omega)/\gamma_{ab}$ is the normalized detuning from the true resonance and $\gamma_{ab} = I_0/2$ is the free-space dipole decay rate. With this we find in lowest order of the dimensionless density $\bar{n} = n/k_0^3$

$$\Gamma = I_0 \left[ 1 + 6\pi \bar{n} f_1'' + \cdots \right], \quad \Delta = \frac{I_0}{2} \left[ 7\pi \bar{n} - 6\pi \bar{n} f_1'' + \cdots \right]$$  

Substituting these expressions into the dielectric function, eq. (23), one eventually finds for the shift relative to the dilute-medium resonance and the effective linewidth up to first order in $\bar{n}$:

$$\gamma_{eff} = \frac{I_0}{2} \left[ 1 - 6\pi \bar{n} f_1'' + 6\pi \bar{n} f_1'' + \cdots \right],$$  

$$\Delta_{eff} = \frac{I_0}{2} \left[ -2\pi \bar{n} + 6\pi \bar{n} f_1'' + 7\pi \bar{n} - 6\pi \bar{n} f_1'' + \cdots \right]$$

The second term in the expression for the linewidth is due to local field corrections of the dielectric function and the third one due to the changed spontaneous emission rate. It is interesting to note that both contributions are of the same order of magnitude but differ in sign. Thus local field effects to the vacuum interaction may compensate the line-narrowing / broadening effects resulting from local field corrections of the dielectric function in lowest order of the density. In the expression for the line-shift, eq. (24), one recognizes the familiar Lorentz-Lorenz shift $-2\pi \bar{n}$. The second term emerges again from local field corrections of the dielectric function and the two last terms are due to modifications of the Lamb shift.

To illustrate the effect of the center-of-mass correlations let us consider a gas with repulsive interaction such that the two-particle correlation $h_2$ is
close to $-1$ over an effective correlation distance $z$ and then approaches zero. As simple model functions we use a Gaussian and a hyper-Gaussian

$$h_2^{(a)}(x) = -\exp \left[ -\frac{x^2}{z^2} \right] \quad \text{and} \quad h_2^{(b)}(x) = -\exp \left[ -\frac{x^8}{z^8} \right] \quad (28)$$

where the distance $\tilde{x} = x/\lambda$ and the correlation length $\tilde{z} = z/\lambda$ are normalized to the resonance wavelength $\lambda$. I have plotted in the following figures the real and imaginary parts of $f_1$ for both correlations. It is worth noting that for values of the correlation length larger than the resonance wavelength, $f' \rightarrow 7/12$, while $f''$ becomes a linear function of $\tilde{z}$.

If the correlation length $\tilde{z}$ scales with the density according to $\tilde{z} \sim \tilde{\rho}^{-1/3}$ the above behavior can be associated to the density dependence of the correction terms. In the following plots the real and imaginary parts of $\tilde{z}^{-3} f_1 \sim \tilde{\rho} f_1$ are shown as function of $\tilde{z}^{-3} \sim \tilde{\rho}$. One recognizes that $\tilde{\rho} f''_1$ scales for small $\tilde{\rho}$ as $\tilde{\rho}^{1/3}$, while $\tilde{\rho} f'_1$ is approximately linear in the density. Thus in the low-density limit linewidth changes will dominate line shifts.
5 summary

In the present paper local field effects on spontaneous emission and Lamb shift in a dense atomic gas have been discussed taking into account two-particle center-of-mass correlations. It has been shown that the corresponding changes of the atomic polarizability can lead to modifications of the medium response, which are of the same order as those resulting from direct corrections of the dielectric function found by Maurice, Castin and Dalibard and Ruostekoski and Javanainen. They are however of opposite sign and thus may compensate the leading order corrections in the density. The present approach is based on an independent scattering approximation and a virtual cavity assumption. It is thus only applicable for densities which are sufficiently smaller than the cubic wavenumber, \( \bar{q} \ll 1 \). Also the existence of a distinguished probe atom has been assumed, which is valid only for classical gases. Nevertheless the results indicate that local field corrections to the atomic polarizability may change or even reverse the predicted line-shifts and linewidth modifications found for Bose gases near the condensation temperature and for low-temperature Fermi gases.

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