Population dynamics in cold gases resulting from the long-range dipole–dipole interaction

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Abstract
We consider the effect of long-range dipole–dipole interaction on the excitation exchange dynamics of cold two-level atomic gases in the conditions where the size of the atomic cloud is large as compared to the wavelength of the dipole transition. We show that this interaction results in population redistribution across the atomic cloud and in specific spectra of the spontaneous photons emitted at different angles with respect to the direction of atomic polarization.

1. Introduction

Cold atomic gases give the possibility of addressing collective atomic quantum states, where an elementary atomic excitation is coherently distributed among a large number of individual atoms. Such systems have been considered in the context of quantum information [1], coherence protection [2], individual photon manipulations [3, 4] etc. For cold Rydberg gases that have large dipole moments, one has to allow for the effects associated with strong dipole–dipole interaction among atoms [5–7]. Considering the regime typical of current experimental setting, one usually takes into account the regular static dipole–dipole interaction \( V \sim 1/R^3 \) among pairs of the atoms, which from the viewpoint of quantum electrodynamics is a result of exchange by a virtual strongly off-resonant vacuum photon with a typical wavelength of the order of interatomic distance \( R \) [8]. This sort of interatomic coupling can be attributed neither to the short-range interaction, nor to the long-range ones, since the integral \( \int_0^\infty V(R)R^2\,dR \) corresponding to the average binary interaction logarithmically diverges at both upper and lower limits. This circumstance results in a number of interesting dynamic phenomena [9], such as the incomplete decay of a single atom population at the limit of long times, which resembles the Anderson localization effect. At the same time, for the description one cannot make use of the continuous media model and the mean field approximation, which are applicable only for the long-range interactions.

Among experiments with Rydberg atoms, there are some [6] that have been done in the regime where the size of a cloud of cold Rydberg atoms is of the order of or larger than the wavelength of the resonant dipole active transition. In such conditions, the role of the radiation trapping, super-radiance [10, 11] and the dynamic long-range dipole–dipole interactions \( \sim 1/R \) is dominant, and the continuous media model becomes applicable. In other words quantum states of the atomic ensemble and that of the resonant radiation field get strongly entangled, whereas the eigenstates of the compound atoms+field system correspond to atomic states dressed in the resonant radiation field. Driven by curiosity we will now consider the atomic population dynamics in the simplest version of such a process. We make use of the mean-field approximation, and concentrate on an exactly soluble case of an infinite uniform continuous two-level media, ignoring the contribution of the static dipole–dipole interaction not conforming to the requirements of the mean-field model. We note here that the simple model studied in this work can be eventually extended in order to contribute, for instance, to the recent efforts to entangle photons via long-range interactions between the Rydberg states of multilevel atoms [4].
We consider a static continuous media corresponding to the gas of $N$ two-level particles with the transition frequency $\omega$ and polarization $\mathbf{d}$ along the $z$-direction that are uniformly scattered in space at fixed positions. Since the static dipole–dipole interaction between the atoms can be viewed as a process intermediated by virtual photon exchange, the effective Hamiltonian that governs the evolution of the combined system is
\[
\hat{H} = \sum_k \hbar c k \hat{\sigma}_k^+ \hat{\sigma}_k^- + \frac{\hbar \omega}{2} \hat{\sigma}_k^z + \hbar v_k (\hat{\sigma}_k^+ \hat{\sigma}_k^- + \hat{\sigma}_k^+ \hat{\sigma}_k^-),
\]
where
\[
\hat{\sigma}_k^i = \sum_j \delta_k^i e^{i\mathbf{d} \cdot \mathbf{r}_j}, \quad i = +, -, z,
\]
are the collective operators of the exciton $\mathbf{k}$, $v_k = U \sqrt{2} \sin \alpha$ is the coupling and $\alpha$ is the angle between the direction of the polarization $\mathbf{d}$ and the normal to the photon polarization plane. The coupling $U \sqrt{2} = d \sqrt{4\pi \hbar c k}$ is a product of the photon vacuum field strength $\mathbb{E} = \sqrt{4\pi \hbar c k}/V$ in a volume $V = N/n$ and the collective atomic dipole moment $d \sqrt{N}$, where $d$ is the atomic transition dipole moment, $c$ is the speed of light, and $k = |\mathbf{k}|$.

We assume that at time $t = 0$, all but one particle is in the ground state. The consideration is equally applicable to the case where a group of particles located in a volume with a typical size $a$, is small as compared to the resonant transition wavelength $2\pi c/\omega$, is in a coherent superposition of the individual excited states, such that the total number of excitations is one. In order to take advantage of the uniform distribution, we take states
\[
|\mathbf{k}\rangle = \frac{1}{\sqrt{N}} \hat{\sigma}_k^+ |0\rangle
\]
(3)
corresponding to a given wavevector $\mathbf{k}$ as the basis set in the collective Hilbert space. Here $|0\rangle$ denotes the vacuum where all particles are in the ground state, and the condition that the average distance $n^{-1/3}$ among the neighbouring particles corresponding to the density $n$ is much shorter than the transition wavelength $2\pi c/\omega$ is implicit.

Each state, (3), interacts only with the photon of the same wavevector, and the amplitudes $\psi_k$ and $\varphi_k$ of the particle+field compound state $|\mathbf{k}\rangle_c = \psi_k |\mathbf{k}, 0\rangle + \varphi_k |0, \mathbf{k}\rangle$ (where the second quantum number corresponds to photons) satisfy the Schrödinger equation
\[
\hat{H}_k \psi_k = U \sqrt{2} \sin \alpha \varphi_k + \frac{1}{(2\pi)^3} \delta(t)
\]
\[
\hat{H}_k \varphi_k = \hbar c k - \omega \varphi_k + U \sqrt{2} \sin \alpha \psi_k,
\]
where the Dirac delta function $\delta(t)$ stands for the initial condition corresponding to the excitation location in the origin, that is at the point $\mathbf{r} = 0$.

The exact solution of (4) and (5)
\[
\psi_k = -e^{i\omega_k c t} \frac{\cos [\Omega t] + (\omega_k - \omega)c \sin [\Omega t]}{2\pi} \frac{\Omega}{\Omega + \omega}
\]
(6)
\[
\varphi_k = -\frac{U \sqrt{2} \sin \alpha c t}{(2\pi)^3} \frac{\sin [\Omega t]}{\Omega}
\]

Figure 1. Transformation of the integration contour for the inverse Fourier transformation integrals (7) and (8). The initial contour $C_1$ can be moved to the lower part of the complex plane of $k$. The part of the contour $C_2$ circumventing the branching points (9) accounts for the contribution of the long-range dipole–dipole interaction, while the remaining part of the contour accounts for the regular static dipole–dipole interaction $\sim 1/R^3$.

with the Rabi frequency $\Omega = \sqrt{\left(\frac{\omega - \omega_k}{2}\right)^2 + k (\frac{U}{\hbar})^2 \sin^2 \alpha}$ suggests
\[
\psi(\mathbf{r}) = \int k^2 e^{-ik \cdot \mathbf{r}} \varphi(\mathbf{r}) \frac{\cos[\Omega t] + (\omega - \omega_k) \sin[\Omega t]}{2\Omega} \frac{dt}{(2\pi)^3}
\]
(7)
\[
\varphi(\mathbf{r}) = \int k^2 e^{-ik \cdot \mathbf{r}} \psi(\mathbf{r}) \frac{\sin[\Omega t]}{\Omega} \frac{dt}{8\pi^3} \frac{U \sqrt{2} \sin(\alpha)}{\Omega}
\]
(8)
for the coordinate-dependent amplitudes, with $dt = dk d\Gamma$, where $d\Gamma$ denotes the integration over the solid angle and the integration over $k$ starts at point $0$ and goes to $+\infty$.

Long-range dipole–dipole interaction corresponds to the exchange by photons close to the resonance domain $k \approx \omega/c$. In this domain, the integrands (7) and (8) have two branching points
\[
cos^2 - 2U^2 \sin^2 \alpha \pm i 2U \sin \alpha \sqrt{\hbar^2 \omega - U^2 \sin^2 \alpha}
\]
(9)
in the complex plane of $k$ where $\Omega$ assumes zero value. Therefore the contribution of the long-range interaction is given by an integral along a contour presented in figure 1 circumventing these points, which can be expressed in terms of the Bessel functions $J_n(x)$. In order to shorten the notations we employ the units where $\omega/c = 1$ henceforth. In the limit $U \ll 1$ of a coupling, which is weak as compared to the energy of atomic transition quantum, the integration over $dk$ yields
\[
\psi(\mathbf{r}) = \frac{\int d\Gamma U \sqrt{2} \sin(\alpha)}{2\pi^2} \frac{U \Theta_H(r_0) \Theta_H(t - r_0)}{r_0^{1/2} (t - r_0)^{-1/2} J_1(2U \sqrt{2} \sin(\alpha)) e^{-i(\theta - U \theta)}}
\]
(10)
\[
\varphi(\mathbf{r}) = \int d\Gamma i U \sqrt{2} \sin(\alpha) \Theta_H(t - r_0)
\]
(11)
up to terms of higher orders in $U$. Here $\Theta_H(x)$ is the Heaviside-$\Theta$ functions, while $r_0$ and $U_0$ stand for $r \cos \Theta$ and $U \sin \alpha$, respectively.

The main steps for performing the integration over $k$ and arriving at equations (10) and (11) are the following. We
assume that the main contribution comes from the domain $k \sim \frac{c}{2}$ and we make the corresponding simplifications in (7) and (8). Then we choose as a new variable $x, k \rightarrow \frac{1}{2}k - \frac{x}{\sqrt{1+x^2}}$, and we perform integration by parts, employing the identity
\[ \int_0^\infty \cos(ax) \sin(b \sqrt{1+x^2}) \frac{dx}{\sqrt{1+x^2}} = \frac{\pi}{2} J_0(b^2 - a^2), \]
that is valid iff $b^2 > a^2$ and zero elsewhere.

Integration over the solid angle in (10) and (11) has to be done numerically in the polar coordinates associated with the radius vector $r$, among these coordinates there exist relations (12) and (13). The photon polarization $E$ is on the plane perpendicular to $k$.

Figure 2. Two sets of spherical coordinates in the space of the wavevectors $k$: the coordinates $(k, \alpha, \phi)$ associated with the direction of polarization $d$, and the coordinates $(k, \Theta, \Phi)$ associated with the radius vector $r$. For a given angle $\theta$ between the direction of the polarization and the radius vector, among these coordinates there are two size scales in the problem: the distance $1/k$ and the wavelength $\lambda$. We note that there are two size scales in the problem: the distance $1/k$ and the wavelength $\lambda$. For a given angle $\theta$ between the direction of the polarization and the radius vector, among these coordinates there are two size scales in the problem: the distance $1/k$ and the wavelength $\lambda$.

Figure 3. Probability of the excitation $w(r) = |\psi(r)|^2$ for the infinite media and for times $T = tU^2 \ll 1$ given by equation (15).

2. Population dynamics at the wavelength scale

We note that there are two size scales in the problem: the wavelength $2\pi$ (that is $2\pi/k$ in the dimensional units) and a distance $1/U$ (that is $c/U$ in the dimensional units) at which light propagates during the Rabi period at resonance. Population distribution at these scales has to be analysed separately.

| Figure 3. Probability of the excitation $w(r) = |\psi(r)|^2$ for the infinite media and for times $T = tU^2 \ll 1$ given by equation (15). |
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Let us first consider the wavelength scale. For short times $t \ll 1/U^2$, the Bessel function can be cast in Taylor series and the first order yields
\[ \psi(r) = \frac{tU^2}{2\pi^2} \int \sin^2(\alpha) \Theta_H(\cos \Theta) e^{ir \cos \Theta} d\Gamma \]
for the amplitude in (11). With the help of equation (12) one can calculate the integral exactly and find
\[ \psi(r) = \frac{iU^2}{4\pi} (3r^2 - 2e^{-ir}(r^2 + ir + 1) + 2) \]
\[ + \frac{iU^2}{4\pi} (r^2 + 2e^{-ir}(r^2 - 3ir - 3) + 6) \cos(2\theta) \]
which results in the population $|\psi(r)|^2$ quadratically increasing in time, shown in figure 3. Note that at long distances the probability corresponding to (15) decreases as $r^{-2}$, which means that the integral of the population over the volume logarithmically diverges at the upper limit being truncated just at the radius $r = t$. In other words, some part of the excitation is getting transferred by the resonant photons at a large distance $r \sim t$.

The distribution of figure 3 persists as long as $tU^2$ remains a small number. Its structure can be easily understood when one notes that according to the second equation (6), the amplitudes of emitted photons are located in the domain of resonance where the the detunings $(1 - k)$ are small. These photons, after being emitted, result in discarding of the corresponding spacial harmonics from the initial $\delta$-like excitation profile $\psi(r)$ containing all harmonics with equal amplitudes. But such a discarding implies an establishment of the population distribution given by the discarded harmonics taken with the opposite signs. As a result, the sum of the $\delta$-like distribution and the latter distributions of $\psi(r)$ no longer contains harmonics strongly coupled to the cooperative resonant radiation. In the course of time the harmonics with higher detuning became involved in the process, and the net population of figure 3 increases. For an initial distribution of a small but finite size $a$, the relative amount of this population is given by the ratio of the length $U^2$ of the $k$-interval of emitted photons and the length $\sim 1/a$ of the $k$-interval occupied by the initial distribution.

In the course of time $t \gg U^{-2}$, the distribution $|\psi(r)|^2$ in the domain $r \sim 1$ gets modified, as shown in figure 4.

One can find an analytic expression for the asymptotic form of $\psi(r)$ at $t \rightarrow \infty$. The main contribution to the
integral (11) comes from the domain of small $\cos \Theta$, that results from the exchange by the resonant photons with $k$ almost orthogonal to $r$. Integration over $d\Gamma$ can be carried out in the reference system associated with $r$ with the allowance for the approximate relation $\sin 2\alpha \simeq 1 - (\sin \Theta \cos \Phi)^2$ and yields

$$\psi(r) = \frac{i}{2\pi \sqrt{z^2 + \rho^2}} e^{-i \frac{\rho^2}{2z^2 + \rho^2}} J_0 \left( \frac{tU^2 \rho^2}{2(z^2 + \rho^2)} \right) - \frac{i}{2\pi \sqrt{z^2 + \rho^2}} e^{i \frac{\rho^2}{2z^2 + \rho^2}} J_0 \left( \frac{tU^2 \rho^2}{2(z^2 + \rho^2)} \right).$$

(16)

One sees that the population distribution decreases with the radius as $1/r^2$ and has an angular dependence $|\psi(\theta)|^2 = \left| e^{i \frac{\rho^2}{2z^2 + \rho^2}} J_0 \left( \frac{tU^2 \rho^2}{2(z^2 + \rho^2)} \right) - e^{-i \frac{\rho^2}{2z^2 + \rho^2}} J_0 \left( \frac{tU^2 \rho^2}{2(z^2 + \rho^2)} \right) \right|^2$, shown in figure 5. Angular shrinking of the distribution in the course of time can be understood if we note that the photons emitted in the direction close to that of the particle polarization are weekly coupled due to the smallness of the projection of the electric strength vector to the particle dipole moment. Therefore the typical evolution time for their interaction with the atomic cloud is longer.

3. Population dynamics and emitted radiation at large distance

What happens at the large scale with typical distances $r \sim 1/U$? The results of numerical integration of (11) are shown in figure 6 for two different times. One sees rather irregular structures to some extent resembling the time dependence of fractional revivals$^3$, which result from the interference of different spacial harmonics with the wavevectors close to unity. One should note that this regime implies that the size of the system is larger or comparable with the distance $r \sim 1/U$.

However, this distance can be relatively large, exceeding the typical size of the atomic cloud. In such a situation, instead of considering the population distribution at distances $r \geq 1/U$,

$^3$ Note that fractional revivals discussed by I Sh Averbukh and N F Perelman in [Phys. Lett. A 139 449] are typical of the time evolution of quantum systems with discrete spectra that have a small quadratic inequidistance. Here a similar phenomenon occurs in the continuous spectrum, which requires a special and more detailed consideration.

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Figure 4. Contour plot of the population distribution for long times $T = tU^2$.

Figure 5. Asymptotic dependence (17) of the angular part of the population distribution on time $T = tU^2$. In the inset we show the contour plot of the population distribution calculated with the asymptotic formula for $T = 15$.

Figure 6. Population distribution along the radial direction at distances $\rho \sim c/U$ for two values of the parameter $TU = 6$ (a) and $TU = 9$ (b) resulting from the numerical integration of equation (11).
where the frequency deviation from the resonance $\delta \omega = \varepsilon / U \sin \alpha$.

one may calculate the frequency spectrum of photons detected at a distance much larger as compared to the typical size of the atomic cloud.

The calculations relevant to the case of large distances rely on the Fourier transformed (4) and (5),

$$\epsilon \psi_k = U \sqrt{k} \sin(\alpha) \varphi_k + \frac{1}{(2\pi)^3}$$

while the resulting amplitude

$$\varphi_k = -\frac{\sqrt{k} U \sin \alpha}{8\pi^3(-\varepsilon^2 + k \varepsilon - \varepsilon + k U^2 \sin^2 \alpha)}$$

to have a photon with the wavevector $k$ at an angle $\alpha$ to the direction of the polarization has to be averaged over an interval around $k = 1$ of a width $\sim 2\pi / L$ corresponding to the uncertainty resulting from the finite size $L$ of the system. The averaging has to be performed with a weight function $n(k)$ given by the Fourier transformed particle density profile

$$\varphi_\alpha = \int \frac{n(k) U \sin \alpha \, dk}{8\pi^3(-\varepsilon^2 + k \varepsilon + U^2 \sin^2 \alpha)}.$$  

For an analytic profile $n(\kappa)$ this yields the spectral intensity

$$|\varphi_\alpha|^2 = \frac{1}{16\pi^4(\delta \omega)^2} n^2 \left( \frac{\delta \omega}{\delta \omega} - \frac{1}{\delta \omega} \right),$$

where the frequency deviation from the resonance $\delta \omega = \varepsilon / U \sin \alpha$ is scaled by the interaction.

In figure 7, we present the probability of the photon detection as a function of the frequency calculated for the particle density distribution $n(r) = 1 / \cosh^2(|r| / L)$ with $L = 4$. This distribution stands as a physical approximation to the delta function spatial distribution of excited atoms we have assumed throughout the derivation. One sees, that though the cooperative coupling between the particles and the electromagnetic field does not govern the population distribution $|\psi(r)|^2$ over the ensemble of two-level particles of a finite size $L < c / U$, it manifests itself in the spectrum of emitted photons that can be registered at distances large as compared to $L$.

4. Conclusions

We conclude by presenting the overall picture of the process shown in figure 8. An atomic cloud, initially in the ground state, has been excited in a small domain close to the centre. In the course of time the excitations get redistributed over all the volume of the cloud due to the long-range dipole–dipole interaction. The typical rate of the process corresponds to the $U^2 \rightarrow 4\pi d^2 n / \hbar$. For short times, the population distribution is given by (15) and for $t U^2 \gg 1$ it takes the asymptotic form (16). The process is associated with the cooperative spontaneous emission of photons. The typical scale corresponding to the spectrum of emitted photons is given by the interaction strength $U \rightarrow d \sqrt{4\pi \alpha / \hbar}$, and at a large distance from the atomic cloud it has the two-peak structure of equation (22). As a general conclusion, one may state that the presence of two essentially different energy scales ($U$ and $U^2$) can be considered as a typical manifestation of collective atomic phenomena.

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