Cooperative Spontaneous Emission as a Many Body Eigenvalue Problem

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We study emission of a single photon from a spherically symmetric cloud of N atoms (one atom is excited, N-1 are in ground state) and present an exact analytical expression for eigenvalues and eigenstates of this many body problem. We found that some states decay much faster then the single-atom decay rate, while other states are trapped and undergo very slow decay. When size of the atomic cloud is small compared with the radiation wave length we found that the radiation frequency undergoes a large shift.

Recent quantum optical experiments and calculations [1, 2] focus on the problem in which a single photon is stored in a gas cloud and then retrieved at a later time. The directionality and spectral content of the cooperatively reemitted photon is then of interest.

Furthermore synchrotron radiation experiments involving N nuclei excited by weak γ ray pulse have features in common with the present problem [3]. For example, in such experiments a thin disk of nuclei can easily be prepared in a superposition in which the atoms are all in the ground state together with a small probability of a uniform excitation of the state, similar to Eq. (29), added in. The simplest example of two-atom cooperative decay has been studied in many publications [4]. The N-atom problem has been also investigated by several authors [5]. Time evolution and directionality of the radiation emitted from a system of two-level atoms which are excited by a plane-wave pulse have been discussed in [6].

Having motivated our interest in the problem we now turn to the analysis of the correlated spontaneous emission from N atoms in free-space. We consider a system of two level (a and b) atoms, initially one of them is in the excited state a and $E_a - E_b = \hbar\omega$. Initially there are no photons. Atoms are located at positions $\mathbf{r}_j$ ($j = 1, ..., N$). In the dipole approximation the interaction of atoms with photons is described by the Hamiltonian

$$\hat{H}_{\rm int} = \sum_{\mathbf{k}} \sum_{j=1}^{N} g_k \left[\hat{\sigma}_j \hat{a}_k^{\dagger} \exp(i(\nu_k - \omega)t - i\mathbf{k}\cdot\mathbf{r}_j) + {\rm adj} \right], \tag{1}$$

where $\hat{\sigma}_j$ is the lowering operator for atom j, $\hat{a}_k$ is the photon operator and g_k is the atom-photon coupling constant for the k mode. We look for a solution of the Schrödinger equation for the atoms and the field as a superposition of Fock states

$$\Psi = \sum_{j=1}^{N} \beta_j(t) |b_1 b_2 ... a_j ... b_N > |\, 0 > +$$

$$\sum_{\mathbf{k}} \gamma_{\mathbf{k}}(t) |b_1 b_2 ... b_N > |1_{\mathbf{k}} > . \tag{2}$$

States in the first sum correspond to zero number of photons, while in the second sum the photon occupation number is equal to one and all atoms are in the ground state b. For simplicity we neglect the effects of photon polarization. Substitute of Eq. (2) into the Schrödinger equation yields the following equations for $\beta_j(t)$ and $\gamma_k(t)$ (we put $\hbar = 1$)

$$\dot{\beta}_j(t) = -i \sum_{\mathbf{k}} g_k \gamma_k(t) \exp[-i(\nu_k - \omega)t + i\mathbf{k}\cdot\mathbf{r}_j], \tag{3}$$

$$\dot{\gamma}_{\mathbf{k}}(t) = -i \sum_{j=1}^{N} g_k \beta_j(t) \exp[i(\nu_k - \omega)t - i\mathbf{k}\cdot\mathbf{r}_j]. \tag{4}$$

Integrating Eq. (4) over time gives $[\gamma_k(0) = 0]$

$$\gamma_{\mathbf{k}}(t) = -i \int_0^t dt' \sum_{j=1}^{N} g_k \beta_j(t') \exp[i(\nu_k - \omega)t' - i\mathbf{k}\cdot\mathbf{r}_j]. \tag{5}$$

Substituting this into (3) we obtain equation for $\beta_j(t)$

$$\dot{\beta}_j(t) = -\sum_{\mathbf{k}} \sum_{j'=1}^{N} \int_0^t dt' g_k^2 \beta_{j'}(t') e^{i(\nu_k - \omega)(t'-t) + i\mathbf{k}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})}. \tag{6}$$

We proceed by making the Markov approximation a-la Weisskopf and Wigner to obtain

$$\dot{\beta}_i(t) = -\gamma \sum_{j=1}^{N} \Gamma_{ij} \beta_j(t), \tag{7}$$

where for $i \neq j$

$$\Gamma_{ij} = \frac{\sin(k_0|\mathbf{r}_i - \mathbf{r}_j|)}{k_0|\mathbf{r}_i - \mathbf{r}_j|} - i\frac{\cos(k_0|\mathbf{r}_i - \mathbf{r}_j|)}{k_0|\mathbf{r}_i - \mathbf{r}_j|}, \tag{8}$$

$\Gamma_{ii} = 1$, $k_0 = \omega/c$ and γ is the single atom spontaneous decay rate

$$\gamma = \frac{V_{\rm ph} k_0^2 g_{k_0}^2}{\pi c},$$

$V_{\rm ph}$ is the photon volume.

We point out that a rigorous treatment of the problem beyond the rotating wave approximation Hamiltonian (1) also yields Eqs. (7) and (8) [7, 8]. Imaginary part of Γ_{ij}

in Eq. (8) appears due to a short range interaction between atoms which is induced by electromagnetic field and causes a frequency shift [9]. The frequency shift becomes substantial when size of the atomic cloud is smaller then the wave length, this will be clear from Eq. (26) below.

One can rewrite Eq. (7) in a matrix form

$$\dot{B} = -\gamma\Gamma B, \tag{9}$$

where the vector B and the decay matrix Γ are given by

$$B = \begin{pmatrix} \beta_1(t) \\ \beta_2(t) \\ \vdots \\ \beta_N(t) \end{pmatrix}, \quad \Gamma = ||\Gamma_{ij}|| = \begin{pmatrix} 1 & \Gamma_{12} & \cdots & \Gamma_{1N} \\ \Gamma_{21} & 1 & \cdots & \Gamma_{2N} \\ \vdots & \vdots & \ddots & \vdots \\ \Gamma_{N1} & \Gamma_{N2} & \cdots & 1 \end{pmatrix}. \tag{10}$$

The matrix Γ is symmetric $\Gamma_{ij} = \Gamma_{ji}$. Let $|\lambda_i >$ be eigenvectors of Γ and λ_i $(i = 1, ..., N)$ are the corresponding eigenvalues. If initially the system is prepared in an eigenstate $B(0) = |\lambda_i >$, then according to Eq. (9) the state evolution is given by

$$B(t) = e^{-\gamma\lambda_i t}B(0), \tag{11}$$

that is state evolves independently of other states and decays with the rate $\gamma\mathrm{Re}\lambda_i$, where γ is the spontaneous decay rate of a single atom. The general solution of the Schrödinger equation is

$$\Psi = C_1 e^{-\gamma\lambda_1 t}|\lambda_1 > + C_2 e^{-\gamma\lambda_2 t}|\lambda_2 > + \ldots +$$

$$C_N e^{-\gamma\lambda_N t}|\lambda_N > + \sum_{\mathbf{k}} \gamma_{\mathbf{k}}(t)|b_1 b_2 ... b_N > |1_{\mathbf{k}} >, \tag{12}$$

where C_1, C_2, ..., C_N are constants determined by the initial conditions. Real part of λ_i are positive numbers and, hence, the general solution corresponds to an exponential decay of the initial state.

One should note that since $\mathrm{tr}(\Gamma) = N$ we obtain

$$\sum_{i=1}^{N} \lambda_i = \mathrm{tr}(\Gamma) = N, \tag{13}$$

which is useful and insightful result.

Next we solve the eigenvalue Eq. (7) analytically for a dense spherically symmetric cloud with atomic density $\rho(r)$ ($\int \rho(r)d\mathbf{r} = N$). Γ_{ij} changes in a scale of $1/k_0$. Assuming there are many atoms in the volume $1/k_0^3$ we can replace summation by integration, then Eq. (7) reads ($\beta = e^{-\gamma\lambda t}\beta(\mathbf{r})$)

$$\int d\mathbf{r}'\rho(r')\left[K_0(\mathbf{r}, \mathbf{r}') + iK_1(\mathbf{r}, \mathbf{r}')\right]\beta(\mathbf{r}') = \lambda\beta(\mathbf{r}), \tag{14}$$

where

$$K_0(\mathbf{r}, \mathbf{r}') = \frac{\sin(k_0|\mathbf{r} - \mathbf{r}'|)}{k_0|\mathbf{r} - \mathbf{r}'|}, \quad K_1(\mathbf{r}, \mathbf{r}') = -\frac{\cos(k_0|\mathbf{r} - \mathbf{r}'|)]}{k_0|\mathbf{r} - \mathbf{r}'|}.$$

It turns out that for $k_0 R \gg 1$, where R is the characteristic size of atomic cloud, the eigenfunctions of the integral Eq. (14) are determined by the real part of the kernel, $K_0(\mathbf{r}, \mathbf{r}')$, that is by equation

$$\int d\mathbf{r}'\rho(r')K_0(\mathbf{r}, \mathbf{r}')\beta(\mathbf{r}') = \lambda\beta(\mathbf{r}). \tag{15}$$

Eq. (15) can be solved using the identity [10]

$$\frac{\sin(k_0|\mathbf{r} - \mathbf{r}'|)}{k_0|\mathbf{r} - \mathbf{r}'|} = 4\pi \sum_{n=0}^{\infty} \sum_{m=-n}^{n} j_n(k_0 r) Y_{nm}(\hat{r}) Y_{nm}^*(\hat{r}') j_n(k_0 r')$$

and the orthogonality condition

$$\int d\Omega_r Y_{nm}^*(\hat{r}) Y_{ks}(\hat{r}) = \delta_{nk}\delta_{ms},$$

where Y_{nm} are spherical harmonics, $\hat{r}$ is a unit vector in the direction of $\mathbf{r}$ and $j_n(x)$ are the spherical Bessel functions. Solutions of Eq. (15) are

$$\beta_{nm}(\mathbf{r}) = j_n(k_0 r) Y_{nm}(\theta, \varphi), \tag{16}$$

$$\lambda_{nm} = \int d\mathbf{r}\rho(r) j_n^2(k_0 r), \tag{17}$$

where θ and φ are angles describing direction of $\mathbf{r}$ in the spherical coordinate system. Eigenvalues (17) are independent of m. That is the eigenvalues λ_{nm} are $(2n+1)$-fold degenerate. If atoms are uniformly distributed inside a sphere of radius R (that is $\rho(r) = N/V$, for $r < R$) we obtain [11]

$$\lambda_{nm} = \frac{3N}{2}\left[j_n^2(k_0 R) - j_{n-1}(k_0 R) j_{n+1}(k_0 R)\right]. \tag{18}$$

In the limit $k_0 R \gg n$ we find

$$\beta_{nm}(\mathbf{r}) \approx \frac{1}{r}\sin\left(k_0 r - \frac{\pi}{2}n\right) Y_{nm}(\theta, \varphi), \tag{19}$$

$$\lambda_{nm} \approx \frac{3N}{2(k_0 R)^2}. \tag{20}$$

Fig. 1 shows λ_{nm}/N as a function of n obtained from Eq. (18) at different $\lambda/R \ll 1$ ($\lambda = 2\pi/k_0$). The states with $n < k_0 R$ are degenerate and decay with the rate $3\gamma N/2(k_0 R)^2$, while decay of the states with $n > k_0 R$ is suppressed.

$K_1(\mathbf{r}, \mathbf{r}')$ term in Eq. (14) yields an imaginary contribution in λ_{nm} (frequency shift) and also modifies the real part. To solve Eq. (14) we use the identity [10]

$$\frac{\exp(ik_0|\mathbf{r} - \mathbf{r}'|)}{k_0|\mathbf{r} - \mathbf{r}'|} = 4\pi i \sum_{k=0}^{\infty} \sum_{s=-k}^{k} Y_{ks}(\hat{r}) Y_{ks}^*(\hat{r}')$$

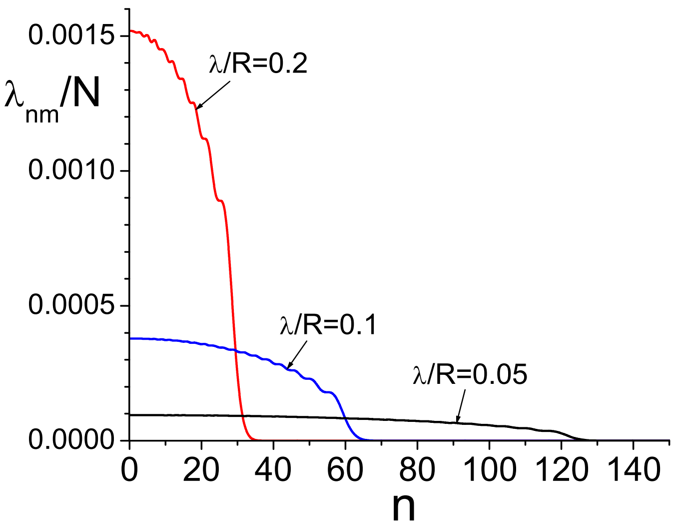

FIG. 1: λ_{nm}/N as a function of n obtained from Eq. (18) at different λ/R.

$$\times \begin{cases} j_k(k_0r')h_k^{(1)}(k_0r), & r > r' \\ j_k(k_0r)h_k^{(1)}(k_0r'), & r \le r' \end{cases}, \tag{21}$$

where $h_k^{(1)}(z)$ are the spherical Bessel functions, $h_k^{(1)}(z) = \sqrt{\pi/2z}H_{k+1/2}^{(1)}(z)$, $H_{k+1/2}^{(1)}(z)$ are the Hankel functions of the first kind.

If atoms are uniformly distributed inside a sphere of radius R the answer for eigenfunctions is given by

$$\beta(\mathbf{r}) = j_n\left(ak_0r\right)Y_{nm}(\hat{r}), \tag{22}$$

where

$$a = \sqrt{1 - \frac{3Ni}{k_0^3R^3\lambda_n}}, \tag{23}$$

and the eigenvalues λ_n are determined from the following equation for a

$$a = \frac{j_n\left(ak_0R\right)}{j_{n-1}\left(ak_0R\right)}\frac{h_{n-1}^{(1)}(k_0R)}{h_n^{(1)}(k_0R)}. \tag{24}$$

In the Dicke limit $k_0R \ll 1$ Eqs. (22) and (24) yield

$$\beta_{nlm}(\mathbf{r}) = j_n\left(A_{nl}\frac{r}{R}\right)Y_{nm}(\hat{r}), \tag{25}$$

$$\lambda_{nl} \approx -\frac{3iN}{A_{nl}^2k_0R} + \frac{6N(k_0R)^{2n}}{A_{nl}^4[(2n-1)!!]^2}, \tag{26}$$

where A_{nl} are nonnegative zeroes of the Bessel function $j_{n-1}(x)$. In particular, $A_{0l} = (2l-1)\pi/2$ and $A_{1l} = \pi l$, $l = 1, 2, 3 \ldots$. One can see that $\mathrm{Im}(\lambda_{nl})$ (frequency shift) becomes large for $k_0R \to 0$. In the Dicke limit only eigenvalues with $n = 0$ have large real part and decay fast with the rate $\Gamma_l = 96N\gamma/\pi^4(2l-1)^4$ (Dicke superradiance [12]), while eigenvalues with $n > 0$ are suppressed by a factor $(k_0R)^{2n}$. Those states are trapped. Please note that $\sum_{l=1}^{\infty}\Gamma_l = N\gamma$, as expected from general arguments. To check validity of our analytical results we solved numerically the eigenvalue problem for the matrix (10) with atoms randomly distributed inside a sphere. In the Dicke limit our numerical simulations show excellent agreement with Eq. (26) already for a few hundred atoms.

In the limit $k_0R \gg n$ we find

$$\lambda_n \approx \frac{3N}{2(k_0R)^2}\left[1 - \frac{(-1)^n}{2k_0R}\sin(2k_0R) - i\frac{(-1)^n}{2k_0R}(\cos(2k_0R) - 1)\right]. \tag{27}$$

In such a limit the contribution from the K_1 term in the kernel is smaller by a factor $1/k_0R$ then those from the K_0 piece.

Figs. 2 and 3 show real and imaginary part of λ_{0l} as a function of k_0R obtained by solving Eq. (24) numerically.

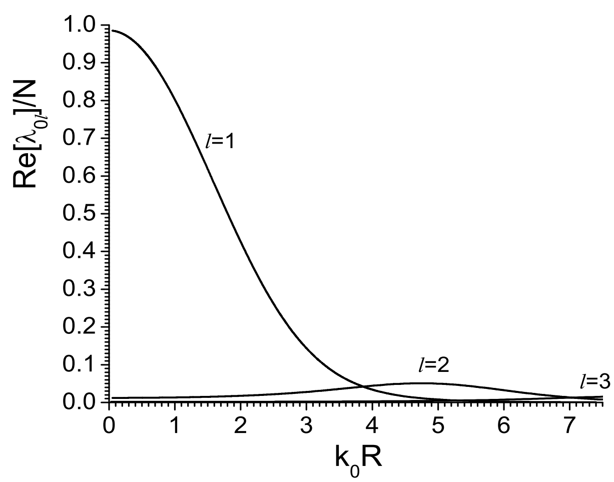

FIG. 2: Real part of λ_{0l} as a function of k_0R for $l = 1$, 2 and 3.

We next discuss the angular distribution of the emitted radiation produced by an eigenstate β_{nlm}. Taking into account $\beta_j(t) = e^{-\gamma\lambda_{nl}t}\beta_{nlm}(\mathbf{r}_j)$ we obtain from Eq. (5) for a uniform cloud of radius R

$$\gamma_{\mathbf{k}}(t) = g_{\mathbf{k}}\frac{N}{V}\frac{\left[1 - e^{-\gamma\lambda_{nl}t + i(\nu_k - \omega)t}\right]}{\omega - \nu_k + i\gamma\lambda_{nl}} \times \int_0^R drr^2 \int d\Omega_r \beta_{nlm}(\mathbf{r})\exp[-i\mathbf{k}\cdot\mathbf{r}]. \tag{28}$$

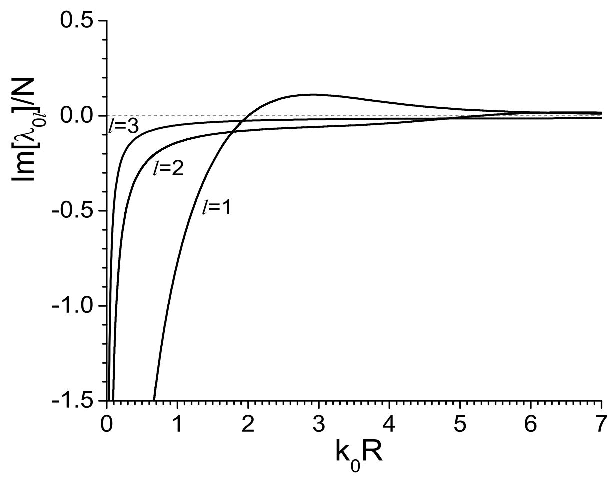

FIG. 3: Imaginary part of λ_{0l} as a function of $k_0 R$ for $l = 1$, 2 and 3.

Substitute Eq. (22) yields $\gamma_{\mathbf{k}}(t) \propto Y_{nm}(\hat{k})$. That is the angular distribution of the emitted radiation is given by the same spherical function $Y_{nm}(\theta, \varphi)$ which describes anisotropy of the initial eigenstate.

Let us next apply the proceeding to the problem of a uniformly excited sample in which one atom is excited and the state is given by

$$|\Psi(0)\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{i\mathbf{k}_0 \cdot \mathbf{r}_j} |b_1, b_2, ..., a_j, ..., b_N\rangle |0\rangle. \quad (29)$$

This is an interesting many body problem comprised of N atoms collectively emitting a single photon [1]. For example, this state can be prepared by the experiments of the type described in Ref. [2]. We are interested in the state of the system at time t in the limit $k_0 R \gg 1$. We obtain the state evolution by expanding $|\Psi(0)\rangle$ in terms of the eigenvectors $|\lambda_{nm} >$

$$|\lambda_{nm} >= \sum_{j=1}^{N} j_n(k_0 r_j) Y_{nm}(\hat{r}_j) |b_1, b_2, ..., a_j, ..., b_N\rangle |0\rangle. \quad (30)$$

We do this using the relation

$$\exp(i\mathbf{k}_0 \cdot \mathbf{r}_j) = 4\pi \sum_{n=0}^{\infty} \sum_{m=-n}^{n} i^n j_n(k_0 r_j) Y^*_{nm}(\hat{k}_0) Y_{nm}(\hat{r}_j), \quad (31)$$

where $\hat{k}_0$ and $\hat{r}_j$ are unit vectors in the directions $\mathbf{k}_0$ and $\mathbf{r}_j$ respectively. The state (29) evolves as

$$|\Psi(t)\rangle_{\text{atomic}} = \frac{4\pi}{\sqrt{N}} \sum_{n=0}^{\infty} \sum_{m=-n}^{n} i^n Y^*_{nm}(\hat{k}_0) e^{-\lambda_n \gamma t} |\lambda_{nm} > \quad (32)$$

where $\lambda_n \gamma \approx 3\gamma N / 2(k_0 R)^2 \equiv \Gamma_N$, see Eq. (27). Hence we have for the atomic state

$$|\Psi(t)\rangle_{\text{atomic}} = e^{-\Gamma_N t} |\Psi(0)\rangle \quad (33)$$

and therefore

$$\beta_j(t) = \frac{1}{\sqrt{N}} e^{-\Gamma_N t} e^{i\mathbf{k}_0 \cdot \mathbf{r}_j}. \quad (34)$$

Substituting this into (5) we obtain

$$\gamma_{\mathbf{k}}(t) = \frac{g_k}{\sqrt{N}} \frac{[\exp(-\Gamma_N t + i(\nu_k - \omega)t) - 1]}{\omega - \nu_k - i\Gamma_N} \sum_{j=1}^{N} e^{-i(\mathbf{k}_0 - \mathbf{k}) \cdot \mathbf{r}_j}, \quad (35)$$

which yields the following probability of a photon being emitted with the wave vector $\mathbf{k}$ [2, 6]

$$|\gamma_{\mathbf{k}}(\infty)|^2 = \frac{g_k^2}{N} \frac{1}{(\omega - \nu_k)^2 + \Gamma_N^2} \left| \sum_{j=1}^{N} e^{-i(\mathbf{k}_0 - \mathbf{k}) \cdot \mathbf{r}_j} \right|^2. \quad (36)$$

The decay rate Γ_N can be understood as the following. Dicke-like arguments [12] for a coherent decay would yield the decay rate of $N\gamma$. However, the spontaneous decay rate is due to emission in all 4π sr directions, while the state (29) emits photon in a small diffraction angle λ/R. As a result, the Dicke superradiance rate $N\gamma$ is reduced by the ratio of the solid diffraction angle λ^2/R^2 to 4π sr, this yields Γ_N.

In summary, we studied correlated spontaneous emission from N atoms in a spherically symmetric cloud which is a $N-$body problem. For a dense atomic gas we obtained analytical expression for the eigenstates and eigenvalues of the system. We found that some states decay much faster then the single-atom decay rate, while other states are trapped and undergo very slow decay. In the limit $\lambda \gg R$ only states with $n = 0$ decay fast ($\Gamma_N \sim N\gamma$), all other states are trapped. In the opposite limit $\lambda \ll R$ many states decay with the same rate $\Gamma_N \approx N\gamma\lambda^2/R^2$. We also found that for $\lambda \gg R$ the eigenvalues have a large imaginary part which corresponds to a frequency shift of emitted radiation.

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