Collective Lamb Shift in Single Photon Dicke Superradiance

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The collective Lamb shift and associated radiative decay of a large cloud of radius R containing N atoms uniformly excited by one photon of wavelength λ is analyzed. It is shown that the time evolution of the symmetric state prepared by single photon absorption in the limit $R \gg \lambda$ is similar to that encountered in the Dicke limit of small sample ($R \ll \lambda$) superradiance. The theory includes virtual (counterrotating) terms naturally and thus provides a simple calculation of the collective Lamb shift of a single Dicke state.

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The Lamb shift [1] and Dicke superradiance [2] are two of the most intriguing effects in atomic physics and quantum optics. Lamb measured the electromagnetic level shift in hydrogen and provided the stimulus for renormalized quantum field theory. Dicke gave us a simple formalism for calculating the collective spontaneous emission from a small cloud of N atoms yielding fascinating results; e.g., one symmetric excitation of such a cloud will decay N times faster than a single isolated atom. We call this "single photon superradiance."

In the Dicke limit $R \ll \lambda$ appropriate to NMR (sample radius R, resonant radiation wavelength λ) this single photon superradiance is obtained when the symmetric N atom Dicke state, given by

$$|+\rangle = \frac{1}{\sqrt{N}} \sum_j |\downarrow_1 \downarrow_2 \ldots \uparrow_j \ldots \downarrow_N\rangle \tag{1a}$$

decays to the ground state $|\downarrow_1 \downarrow_2 \ldots \downarrow_N\rangle$, with a rate proportional to N, where the atomic excited (ground) state of the jth atom is denoted by $\uparrow_j$ ($\downarrow_j$). For most laser experiments $R \gg \lambda$, and conditional preparation [3,4] by a single photon of wave vector $\vec{k}_0$ yields the state

$$|+\rangle_{k_0} = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}_0 \cdot \vec{r}_j} |\downarrow_1 \downarrow_2 \ldots \uparrow_j \ldots \downarrow_N\rangle, \tag{1b}$$

where $\vec{r}_j$ denotes the vector position of the jth atom. The physics contained in the phase factors is best understood by noting that $\vec{k}_0 \cdot \vec{r}_j = \omega \hat{n}_0 \cdot \vec{r}_j / c \equiv \omega t_i$ ($\hat{n}_0 = \vec{k}_0/|k_0|$ and ω is the atomic resonant frequency) showing that atoms at various locations $\vec{r}_j$ are excited at different times t_i. This message was conveyed in the subtitle of Ref. [3] with the phrase: "timing is everything." Hence we call the state (1b) a timed Dicke state, and the corresponding complete set is called the timed Dicke basis, see Table I and Fig. 1; there the notation $|+\rangle_{k_0} \equiv |B_0\rangle$ is introduced.

The focus of the present Letter is a toy model yielding the dynamical evolution of the atomic system described by $|B_0\rangle$, associated with real (decay) and virtual (level shift) photon emission. Specifically we find to a good approximation that the probability amplitude β_0 for the large sample state $|B_0\rangle$ obeys the simple small sample superradiance type equation

$$\dot{\beta}_0 = -(\Gamma + \Gamma_N + i\mathcal{L}_N)\beta_0, \tag{2a}$$

where Γ is the single atom decay rate and the collective decay rate of the atomic cloud containing N atoms in a volume V of radius R is given by $\Gamma_N = \frac{\Gamma}{2\pi} \frac{N-1}{V} \lambda^2 R$, while the N atom Lamb shift [3,4] of $|B_0\rangle$ is given by

$$\mathcal{L}_N = -\frac{\Gamma}{\pi}\left(\ln\frac{K^2 - k_0^2}{k_0^2} - N \ln\frac{K + k_0}{k_0}\right) + \frac{\Gamma_N}{\pi} \frac{\lambda}{4R} S, \tag{2b}$$

where K is the Bethe cutoff, $k_0 = \omega/c$, and S is an uninteresting shape factor of order one; see Eq. (15). The first term in Eq. (2b) is the usual single (two level) atom level shift; the second term is a collective shift common to the ground state; see Eqs. (16) and (17). The last term is the interesting one.

Several aspects of (2a) and (2b) are noteworthy. First of all the simple Eq. (2a) for β_0 has been, and continues to be,

TABLE I. The ground state and first excited states of extended medium single photon superradiance. In the convenient spin notation $|\downarrow\rangle(|\uparrow\rangle)$ represents an atom in the ground (excited) state. Position in the bra denotes atomic number e.g. $|\downarrow_1 \downarrow_2 \ldots \downarrow_N\rangle$ is written $|\downarrow\downarrow \ldots \downarrow_N\rangle$. The wave vector $\vec{k}_0$ is determined by the preparation photon and $ck_0 = \omega$ is the atomic resonant frequency.

$|C_0\rangle = |\downarrow\downarrow\downarrow \ldots \downarrow_N\rangle$
$|B_0\rangle = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}_0 \cdot \vec{r}_j} |\downarrow\downarrow\downarrow \ldots \uparrow_j \ldots \downarrow_N\rangle$
$|B_1\rangle = \frac{1}{\sqrt{2}}[e^{i\vec{k}_0 \cdot \vec{r}_1} |\uparrow\downarrow\downarrow \ldots \downarrow_N\rangle - e^{i\vec{k}_0 \cdot \vec{r}_2} |\downarrow\uparrow\downarrow \ldots \downarrow_N\rangle]$
$|B_2\rangle = \frac{1}{\sqrt{6}}[e^{i\vec{k}_0 \cdot \vec{r}_1} |\uparrow\downarrow\downarrow \ldots \downarrow_N\rangle + e^{i\vec{k}_0 \cdot \vec{r}_2} |\downarrow\uparrow\downarrow \ldots \downarrow_N\rangle - 2e^{i\vec{k}_0 \cdot \vec{r}_3} |\downarrow\downarrow\uparrow \ldots \downarrow_N\rangle]$
$\vdots$
$|B_{N-1}\rangle = \frac{1}{\sqrt{N(N-1)}}[e^{i\vec{k}_0 \cdot \vec{r}_1} |\uparrow\downarrow\downarrow \ldots \downarrow_N\rangle + e^{i\vec{k}_0 \cdot \vec{r}_2} |\downarrow\uparrow\downarrow \ldots \downarrow_N\rangle + \ldots$
$+ e^{i\vec{k}_0 \cdot \vec{r}_{N-1}} |\downarrow\downarrow\downarrow \ldots \uparrow_{N-1}, \downarrow_N\rangle - (N-1) e^{i\vec{k}_0 \cdot \vec{r}_N} |\downarrow\downarrow\downarrow \ldots \uparrow_N\rangle]$

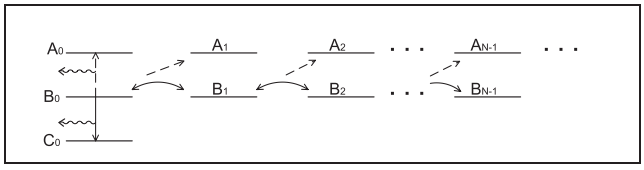

FIG. 1. The timed Dicke states corresponding to the first three energy levels of a uniformly excited cloud of N atoms. The important B_l states are given in Table I. The solid lines indicate the Fano-Agarwal coupling from the initially prepared B_0 state. The dashed lines indicate virtual transitions in which an atom jumps from the ground state to an excited state while emitting a photon. The B_l states have one excited atom and the A_l states have two excited atoms. The A_ℓ state can be computed from the B_ℓ states by applying $\sum_j e^{i\vec{k}_0\cdot\vec{r}_j}\sigma_j^+$.

the subject of debate and confusion. The good news is that Eq. (2a) is very simple. The bad news is that it only applies for large number density, N/V, and for a specifically prepared atomic configuration, i.e., $|B_0\rangle$. This has been a frequent topic of discussion and source of confusion. For example, if we consider the case of $N = 3$, in the large sample limit, then β_0 evolves according to Fano-Agarwal coupling such that

$$\dot{\beta}_0 = -\Gamma_{0,0}\beta_0 - \Gamma_{0,1}\beta_1 - \Gamma_{0,2}\beta_2, \tag{2c}$$

where β_1 (β_2) is the amplitude of the $|B_1\rangle$ ($|B_2\rangle$) state of Table I. The decay rates $\Gamma_{0,\ell}$ are discussed in the following. Only in the large sample limit is $\Gamma_{0,\ell}\beta_\ell$ small for $\ell \neq 0$, see Eqs. (9)–(12) and associated discussion.

Second, Friedberg and Manassah argued in an interesting paper [5(d)] that the virtual photon-Lamb shift processes [5] can complicate the decay and negate the validity of Eq. (2a). In their words: "[The neglect of virtual photon processes] supports the illusory Dicke picture of small sample superradiance according to which the symmetric state is superradiant ... ." Counterarguments and debate followed [6]. Hence, establishing the validity of (2a) in a clear and convincing way is our first goal.

More importantly, the present approach provides a useful tool for calculating the Lamb shift of the state $|B_0\rangle = |r = \frac{1}{2}N, m = -\frac{1}{2}N + 1\rangle$. To put this in perspective we quote from the classic paper of Friedberg, Hartman and Manassah (FHM): they say [7] that the calculation "of the energy shift of such a state is not easily computed." Instead, they calculate a Lamb shift associated with an average over states B_ℓ of Table I. In fact the present approach allows us to readily calculate the Lamb shift (2b) for state $|B_0\rangle$ with the same level of effort required to calculate the cooperative decay (1b).

Finally, we note that the calculations yielding the most interesting many body contribution to the Lamb shift going as $\Gamma_N\lambda/R$ scales as R^{-3} and is free from the usual divergences and cutoffs encountered in calculating the single atom Lamb shift [see discussion after (14)]. That is, in order to arrive at the single atom level shift the (infinite) self-energy had to be subtracted off as was shown by Bethe. In the present problem, the need for renormalization is mitigated by the summation over atomic phase factors of the form $\exp i(\vec{k} - \vec{k}_0)\cdot\vec{r}_j$, as is discussed later in connection with Eqs. (9)–(12).

In concluding our introduction, we note that while the small sample Dicke limit ($R \ll \lambda$) has an appealing simplicity, it neglects the near dipole component of the field and various effects of short wavelength virtual photons. Such effects can have important consequences, e.g., can destroy superradiance. One way to overcome the undesired effects of nearby atoms is to replace the small sample by an extended cloud. Unfortunately this tends to destroy superradiance since it brings in subradiant states (i.e. $|B_1\rangle \ldots |B_N\rangle$ of Fig. 1 and we lose the charm and the simplicity of the Dicke limit. Nevertheless, to a good approximation we here report simple small sample superradiance for large samples.

Hence we focus on the collective Lamb shift in an extended sample whose radius R is large compared to the resonant wavelength λ. This is a challenging problem and we shall make several assumptions which allow us to bring out the physics most simply. To that end, we use a scalar photon theory ignoring the polarization and vector character of the field, make the Markov approximation, and ignore related effects such as the Lorenz-Lorentz effect. None of these idealizations are very serious, the essential physics of the collective Lamb shift remains intact, especially in the important limit of very large R. Even with these approximations the problem is complicated, but the present toy model (and approximate analysis) lays out the problem clearly and will (hopefully) clarify issues and stimulate interest. Having set the stage we proceed to derive Eqs. (2a)–(2c) and develop the physics of the collective Lamb shift as elucidated by the calculation.

The interaction Hamiltonian, in the interaction picture, is given by

$$V(t) = \sum_{j,k}\hbar g_k \sigma_j a_k^\dagger e^{-i\vec{k}\cdot\vec{r}_j} e^{i(\nu_k-\omega)t} + \text{adj} + \sum_{j,k}\hbar g_k \sigma_j^\dagger a_k^\dagger e^{i\vec{k}\cdot\vec{r}_j} e^{i(\nu_k+\omega)t} + \text{adj}, \tag{3}$$

where the coupling constant $g_k = \omega\wp/\sqrt{\hbar\epsilon_0 v\nu_k}$ with $\wp$ being the dipole matrix element, ω the atomic frequency, v the photon quantization volume, ϵ_0 the permittivity of free space and $\nu_k = ck$ is the frequency of the kth radiation mode. The lowering (raising) operators for the jth atom and the kth field mode are $\sigma_j(\sigma_j^\dagger)$ and $a_k(a_k^\dagger)$. The relevant state vector, for calculation of $\beta_0(t)$ in the notation of Fig. 1, is

$$|\psi\rangle = \sum_l \beta_l |B_l, 0\rangle + \sum_k \gamma_k |C_0, 1_k\rangle + \sum_{k,l}\alpha_{l,k}|A_l, 1_k\rangle. \tag{4}$$

The probability amplitudes obey the equations

$$\dot{\beta}_l = -i\sum_{k,j}\langle B_l, 0|g_k\sigma_j^\dagger a_k|C_0, 1_k\rangle e^{i\vec{k}\cdot\vec{r}_j - i(\nu_k-\omega)t}\gamma_k - i\sum_{k,l',j}\langle B_l, 0|g_k\sigma_j a_k|A_{l'}, 1_k\rangle e^{i\vec{k}\cdot\vec{r}_j - i(\nu_k+\omega)t}\alpha_{l',k} \quad (5)$$

$$\dot{\gamma}_k = -i\sum_{l,j}\langle C_0, 1_k|g_k\sigma_j a_k^\dagger|B_l, 0\rangle e^{-i\vec{k}\cdot\vec{r}_j + i(\nu_k-\omega)t}\beta_l, \quad (6)$$

$$\dot{\alpha}_{l,k} = -i\sum_{l',j}\langle A_l, 1_k|g_k\sigma_j^\dagger a_k^\dagger|B_{l'}, 0\rangle e^{-i\vec{k}\cdot\vec{r}_j + i(\nu_k+\omega)t}\beta_{l'}. \quad (7)$$

Integrating (6) and (7) to obtain $\gamma_k(t)$ and $\alpha_{l,k}(t)$ and inserting into (5) we obtain

$$\dot{\beta}_0 = -\sum_{k,l} g_k^2\int_0^t dt'\Big\{\sum_{i,j}\langle B_0|\sigma_j^\dagger|C_0\rangle\langle C_0|\sigma_i|B_l\rangle \times e^{-i(\nu_k-\omega)(t-t')}\beta_l(t') + \sum_{i,j,l'}\langle B_0|\sigma_j|A_{l'}\rangle\langle A_{l'}|\sigma_i^\dagger|B_l\rangle e^{-i(\nu_k+\omega)(t-t')}\beta_l(t')\Big\}. \quad (8)$$

Evaluating the matrix elements Eq. (8) becomes

$$\dot{\beta}_0(t) = -\sum_k g_k^2\int_0^t dt'\Big\{(\tilde{\mathcal{R}}_{0,0}(t') + \tilde{\tilde{\mathcal{R}}}_{0,0}(t'))\beta_0(t') + \sum_{l=1}^{N-1}\mathcal{R}_{0,l}(t')\beta_l(t')\Big\}, \quad (9)$$

where the rate coefficients $\mathcal{R}_{0,l}$ are given by

$$\tilde{\mathcal{R}}_{0,0} = e^{-i(\nu_k-\omega)(t-t')} + (N-1)e^{-i(\nu_k+\omega)(t-t')}, \quad (10)$$

$$\tilde{\tilde{\mathcal{R}}}_{0,0} = \frac{1}{N}\sum_{i\neq j}e^{i(\vec{k}_0-\vec{k})\cdot(\vec{r}_i-\vec{r}_j)}e^{-i(\nu_n-\omega)(t-t')} + \frac{1}{N}\sum_{i\neq j}e^{i(\vec{k}_0+\vec{k})\cdot(\vec{r}_i-\vec{r}_j)}e^{-i(\nu_n+\omega)(t-t')}, \quad (11)$$

$$\mathcal{R}_{0,\ell} = \frac{1}{\sqrt{Nl(l+1)}}\sum_i e^{-i(\vec{k}_0-\vec{k})\cdot\vec{r}_i}S(\ell,\vec{k},\omega) + \frac{1}{\sqrt{Nl(l+1)}}\sum_i e^{-i(\vec{k}_0+\vec{k})\cdot\vec{r}_i}S(\ell,-\vec{k},-\omega), \quad (12)$$

where

$$S(\ell,\vec{k},\omega) = \sum_{j=1}^{l}[e^{i(\vec{k}_0-\vec{k})\cdot\vec{r}_j} - e^{i(\vec{k}_0-\vec{k})\cdot\vec{r}_\ell}]e^{-i(\nu_k-\omega)(t-t')}$$

in which the first (second) term in (11) and (12) corresponds to real (virtual) processes.

Considering the off-diagonal factors $\mathcal{R}_{0,l}$, we first note that for a large sample,

$$\sum_i e^{i(\vec{k}_0\pm\vec{k})\cdot(\vec{r}_j)} \to \delta(\vec{k}\pm\vec{k}_0),$$

and, in this limit, the Sterms in (12) tend to zero. Thus the $\mathcal{R}_{0,0}$ term dominates dominates; i.e., the contribution from the off-diagonal terms proportional to $\beta_1, \beta_2, \ldots, \beta_{N-1}$ is small and, for the present purposes, may be neglected. This is in keeping with the spirit of the present toy model neglecting photon polarization and Lorenz-Lorentz effects, etc. Using

$$\delta(\vec{k}\pm\vec{k}_0) = \frac{(2\pi)^2}{k^2}\int_{-R}^{R}dr e^{i(k-k_0)r}\delta(\hat{\Omega}_{\vec{k}} - \hat{\Omega}_{\pm\vec{k}}), \quad (13)$$

where $\hat{\Omega}_{\vec{k}}$ is the angular unit vector, making the Markov approximation, setting $\Gamma = \wp^2\omega^3/2\pi\hbar\epsilon_0 c^3$, and replacing the sum over $\vec{k}$ by an integral, Eq. (9) becomes

$$\dot{\beta}_0 = -\Gamma\beta_0 - \Big(\frac{\Gamma}{2\pi}\frac{N-1}{V}\lambda^2 R\Big)\beta_0(t) + i\frac{\Gamma}{\pi k_0}\times\int_0^K kdk\Big[\frac{1}{k-k_0}+\frac{N-1}{k+k_0}\Big]\beta_0(t) + i\frac{\Gamma}{k_0}\frac{N-1}{V}\times\int_0^\infty\frac{dk}{k}\int_{-R}^{R}dr e^{i(k-k_0)r}\Big[\frac{1}{k-k_0}+\frac{1}{k+k_0}\Big]\beta_0(t), \quad (14)$$

where K is the cutoff. We proceed to carry out the first integral by subtracting off the electron self-energy terms; i.e., replace $(k\pm k_0)^{-1}$ by $(k\pm k_0)^{-1} - k_0^{-1}$ as it appears in the square brackets. The second integral is finite [due to the $\sum\exp i(\vec{k}-\vec{k}_0)\cdot(\vec{r}_i-\vec{r}_j)$ factor in (11)] and yields a simple result in the $k_0R \gg 1$ limit. Equation (1) then yields

$$\dot{\beta}_0 = -(\Gamma+\Gamma_N)\beta_0 + i[\frac{\Gamma}{\pi}(\ln\frac{K^2-k_0^2}{k_0^2} - N\ln\frac{K+k_0}{k_0}) - \frac{\Gamma_N}{\pi}\frac{\lambda}{4R}S]\beta_0, \quad (15)$$

where Γ_N is given by Eq. (1b) and $S \cong 1 - \cos(2k_0R)/\pi k_0R$.

A similar calculation for the ground state yields

$$\dot{\gamma}_0 = -i\Big(\frac{\Gamma}{\pi}N\ln\frac{K+k_0}{k_0}\Big)\gamma_0 \quad (16)$$

so the relative level shift between $|B_0\rangle$ and $|C_0\rangle$ is,

$$\delta\omega_\beta - \delta\omega_\gamma = -\frac{\Gamma}{\pi}\ln\frac{K^2-k_0^2}{k_0^2} + \frac{\Gamma_N}{\pi}\frac{\lambda}{4R}S. \quad (17)$$

The amplitude of the $|B_0\rangle$ state (1a) has been shown to decay predominantly to $|C_0\rangle$ even though the collective Lamb shift can be large. This supports the approach and results of Refs. [3]. Furthermore, the techniques developed to calculate the collective decay rate are extended to compute, for the first time, the Lamb shift of the $|B_0\rangle$ state. The many particle (atom-atom) contribution to the Lamb shift,

$\mathcal{L}_n \sim \Gamma_N \lambda/4R \sim N\Gamma(\lambda/R)^3$ can be much larger than the single particle shift (which is of order Γ); e.g., for a gas at 1 torr $N/R^3 \sim 10^{16}$ atom/cm^3 and taking $\lambda = 1\mu$ yields $\mathcal{L}_N \sim 10^4\Gamma$.

It is important to note that cooperative spontaneous emission is essentially a many body eigenvalue problem [8]. For instance in the timed Dicke basis Eqs. (9)–(12) (in the Markov approximation) may be easily extended to obtain

$$\dot{\beta}_m(t) = -\sum_l \Gamma_{m,l}\beta_l(t). \tag{18}$$

We have shown that in the large N limit $\dot{\beta}_0 \simeq -\Gamma_{0,0}\beta_0$.

Of course, there are many other choices of basis states, for example, the single atom $|j\rangle$ basis that has been studied extensively [8]. The decay eigenvalue problem in this basis has been shown to be

$$\dot{\beta}_j(t) = -\Gamma\beta_j(t) + i\Gamma \sum_{i=1, i\neq j}^{N} \frac{\exp(ik_0|\vec{r}_j - \vec{r}_i|)}{k_0|\vec{r}_j - \vec{r}_i|}\beta_i(t), \tag{19}$$

which is a useful expression for many problems is but not the natural basis for the case in which the atoms start out in $|B_0\rangle$. Connection with past [9] and future experiments as well as other related theoretical work [10] will be published elsewhere.

In conclusion, it is to be emphasized that it is the timed Dicke basis that yields the simple results contained in Eq's. (2), and clearly demonstrates the utility of the basis. Specifically, when the sample is large enough $|B_0\rangle$ decays to $|C_0\rangle$ with only a small percentage coupled into states $|B_\ell\rangle$. In such a case the mathematics is simplified, and we regain the essential charm of the Dicke formalism in the $R \ll \lambda$ limit [11]; this provides a useful tool for investigating properties such as the collective decay rate and Lamb shift.

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