Inhibited spontaneous emission and energy oscillation between two-level atoms

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The theoretical model presented here is motivated by experimental works demonstrating the suppression of spontaneous emission by atoms placed inside a cavity with a fundamental frequency above that for the atomic transition. In my model, two distinguishable two-level atoms, one of them initially excited, the other in the ground state, are coupled to a nonresonant radiation mode, with no direct interaction between the atoms. The exchange of energy between the atoms is investigated, and it is found that it oscillates from one atom to the other with angular frequency Ω related to the radiative lifetime τ by $\Omega\tau=(3/2\pi^2)(\omega_0/\omega_0-\omega)(\omega_0/\omega)^3$, with ω_0 and ω, the frequencies for the radiation mode and the atomic transition, respectively.

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A two-level "atom" with energy levels $(0,\hbar\omega)$ interacting with the free electromagnetic field in empty space, may decay spontaneously from its upper energy level to its ground state by giving its energy to the electromagnetic (e.m.) field. On the other hand, if the atom interacts with an e.m. field whose first excited state [1] is with energy $\hbar\omega_0$ such that $\hbar\omega_0 > h\omega$, then the atom cannot exchange its energy with the field and stays in its upper state, as has been demonstrated experimentally [2]. In this paper I investigate the oscillation of energy between two distinguishable two-level atoms, one of them initially excited, the other in its ground state, with no direct interaction between them, coupled to such an e.m. field that spontaneous emission does not take place.

In my model, the two atoms, each having energy levels $(0,\hbar\omega)$, interact with one radiation mode, of frequency $\omega_0 > \omega$. We will use Schrödinger picture operators to describe the field and the two-level atoms. The e.m. field mode with frequency ω_0 is described by the creation annihilation operators $a^\dagger, a$, obeying the commutation relation $[a^\dagger, a]=1$, with the Hamiltonian

$$H_f=\hbar\omega_0(a^\dagger a) . \tag{1}$$

The electric-field operator is given by

$$\mathbf{E}(\mathbf{r})=(2\pi\hbar\omega)^{1/2}\mathbf{u}(\mathbf{r})(a^\dagger+a) , \tag{2}$$

where $\mathbf{u}(\mathbf{r})$ describes the spatial dependence of the field, obeying $\nabla^2\mathbf{u}+(\omega_0^2/c^2)\mathbf{u}=0$ inside a given volume with suitable boundary conditions, and normalization $\int \mathbf{u}^2(\mathbf{r})d^3\mathbf{r}=1$ over that volume. The two-level atoms are described by angular momentum operators l_1,l_2,l_3, and j_1,j_2,j_3 for atoms 1 and 2, respectively, with commutation relations $[l_1,l_2]=il_3$, $[j_1,j_2]=ij_3$, and cyclic permutations thereof, and the atomic Hamiltonian

$$H_a=H_{a1}+H_{a2}=\hbar\omega(l_3+\tfrac{1}{2})+\hbar\omega(j_3+\tfrac{1}{2}) . \tag{3}$$

With the help of step-up, step-down operators $l_\pm=(l_1\pm il_2)$, $j_\pm=(j_1\pm ij_2)$, one can write for the atomic dipole moment operators

$$\boldsymbol{\mu}_1=\mathbf{d}_1 l_+ +\mathbf{d}_1^* l_- , \quad \boldsymbol{\mu}_2=\mathbf{d}_2 j_+ +\mathbf{d}_2^* j_- , \tag{4}$$

where it is assumed the "atoms" have no permanent dipole, and the components of the vector $\mathbf{d}_1$ ($\mathbf{d}_2$) are the dipole matrix elements between the levels of atom 1 (2) for the three spatial Cartesian coordinates. The atoms interact with the e.m. field by electric dipole coupling with Hamiltonian:

$$H'=-\boldsymbol{\mu}_1\cdot\mathbf{E}(\mathbf{r}_1)-\boldsymbol{\mu}_2\cdot\mathbf{E}(\mathbf{r}_2)=H_1'+H_2' , \tag{5}$$

where $\mathbf{r}_1,\mathbf{r}_2$ are the position vectors of the atoms. Substituting Eqs. (3) and (4) into Eq. (5), one obtains

$$H_1'=\hbar\gamma_1(a^\dagger l_- +al_+)+\hbar\gamma_1(a^\dagger l_+ +al_-) , \tag{6a}$$

$$H_2'=\hbar\gamma_2(a^\dagger j_- +aj_+)+h\gamma_2(a^\dagger j_+ +aj_-) , \tag{6b}$$

with the coupling coefficients γ_i $(i=1,2)$, assumed to be real, given by

$$\gamma_i=(2\pi\omega_0/\hbar)^{1/2}\mathbf{d}_i\cdot\mathbf{u}(r_i) . \tag{7}$$

The total Hamiltonian for the system of the atoms and the field is now defined by

$$H=H_0+H'=H_{a1}+H_{a2}+H_f+H' . \tag{8}$$

At time $t=0$, atom 1 is in its upper state, atom 2 is in its ground state, the field is in its nth energy state, and the system is described by a normalized state $|ugn\rangle$. The probability at time t that a transition has taken place to the state $|gun\rangle$, the atoms having interchanged their energy values, is determined by the overlap

$$S(t)=\langle gun|e^{-iHt/\hbar}ugn\rangle$$

$$=\sum_i\langle gun|\Psi_i\rangle\langle\Psi_i|ugn\rangle e^{-iE_it/h} , \tag{9}$$

where Ψ_i are the eigenstates of H with eigenvalues E_i.

To find the eigenstates of H we diagonalize its matrix in the basis of H_0 eigenstates, dealing at first with the case that the system is initially in the state $|ug0\rangle$, with the field unexcited. Using the well-known properties of angular momentum and creation and anihilation opera-

tors, we find from Eqs. (6) the following matrix elements of H':

$$\langle ug0|H'|gg1\rangle=\hbar\gamma_1,\quad \langle ug0|H'|uu1\rangle=\hbar\gamma_2\ ,$$

which together with their Hermitian conjugates are the only nonzero elements involving $|ug0\rangle$.

While the states $|ug0\rangle$ and $|gu0\rangle$ belong to the same H_0 eigenvalue $E^{(0)}=\hbar\omega$, and $|gg1\rangle$ to $E^{(0)}=\hbar\omega_0$, the state $|uu1\rangle$ belongs to the much higher value $E^{(0)}=\hbar\omega+h\omega_0$, and therefore makes small contributions to the H eigenstates which in the limit $H'\to 0$ become combinations of $|ug0\rangle$ and $|gu0\rangle$. We will neglect this contribution altogether, the H matrix becoming block diagonal, and the submatrix of interest to us given by

$$H=\begin{bmatrix}\hbar\omega & 0 & \hbar\gamma_1\\ 0 & \hbar\omega & \hbar\gamma_2\\ \hbar\gamma_1 & \hbar\gamma_2 & \hbar\omega_0\end{bmatrix} \tag{10}$$

with columns and rows labeled according to $|ug0\rangle$, $|gu0\rangle$, and $|gg1\rangle$.

We make now an assumption of weak coupling, such that $\gamma_{1,2}/(\omega_0-\omega)\ll 1$, and defining $\nu=\omega_0-\omega$, and $\varepsilon_i=E_i/h-\omega$, we obtain

$$\begin{aligned}&\varepsilon_1=0,\quad \varepsilon_2=-(\gamma_1^2+\gamma_2^2)/\nu,\\ &\varepsilon_3=\upsilon+(\gamma_1^2+\gamma_2^2)/\nu\ .\end{aligned} \tag{11}$$

To simplify the analysis, we will take below $\gamma_1=\gamma_2=\gamma$ and write for the normalized eigenstates

$$\begin{aligned}&2^{1/2}\Psi_1=|ug0\rangle-|gu0\rangle\ ,\\ &N_2^{1/2}\Psi_2=|ug0\rangle+|gu0\rangle-(2\gamma/\nu)|gg1\rangle\ ,\\ &N_3^{1/2}\Psi_3=|gg1\rangle+(\gamma/\nu)(|ug0\rangle+|gu0\rangle)\ ,\end{aligned} \tag{12}$$

where N_2 and N_3 are normalization constants. If the factor $(1/\nu)$ in Eq. (11) is replaced by $[(1/\nu)+1/2\omega+\nu]$, one recovers the second-order time-independent perturbation theory values for the energies, the small difference being due to our neglect of coupling to the state $|uu1\rangle$ in obtaining Eq. (10) for the H matrix (this equation remains, however, valid for $\nu\to 0$, when perturbation approximation is inapplicable).

We now substitute Eqs. (11) and (12) into Eq. (9) to obtain the probability for energy exchange between the atoms during time t:

$$W_{12}(t)=|S(t)|^2=1-\cos(2\gamma^2/\nu)t\ , \tag{13}$$

where we have omitted the contribution of fast oscillating terms with small amplitudes, proportional to $(\gamma/\nu)^2[\cos(\nu+\gamma^2/2\nu)t+\cos\nu t]$, since it was assumed that $\gamma/\nu\ll 1$ in obtaining Eq. (11), and $\nu\neq 0$, $\gamma^2/\nu\ll\nu$, as we are treating the case of interaction with an off-resonance mode.

We refer now to the case that the field is excited, the system being initially in the state $|ugn\rangle$ with $n>0$. It is found from Eqs. (6) that the interaction Hamiltonian H' has the following nonzero matrix elements involving $|ugn\rangle$:

$$\begin{aligned}&\langle ugn|H'|gg(n+1)\rangle=(n+1)^{1/2}\hbar\gamma_1\ ,\\ &\langle ugn|H'|uu(n+1)\rangle=(n+1)^{1/2}\hbar\gamma_2\ ,\\ &\langle ugn|H'|uu(n-1)\rangle=n^{1/2}\hbar\gamma_2\ ,\\ &\langle ugn|H'|gg(n-1)\rangle=n^{1/2}\hbar\gamma_1\ ,\end{aligned}$$

and their Hermitian conjugates, with analogous elements involving $|gun\rangle$. States $|uu(n+1)\rangle$ and $|gg(n-1)\rangle$ belong to H_0 eigenvalues $E^{(0)}=2\omega+(n+1)\omega_0$, and $E^{(0)}=(n-1)\omega_0$, respectively, which are relatively far from the value $E^{(0)}=\omega+n\omega_0$ for $|ugn\rangle$ and $|gun\rangle$, and we will neglect the mixture with these states, as we did with regard to $|uu1\rangle$ in the calculation for $n=0$, bringing the H matrix into a block diagonal form. It is convenient to define here $\varepsilon_i=E_i/\hbar-n\omega_0-\omega$, and obtain for the H submatrix involving $|ugn\rangle$, $|gun\rangle$, $|uu(n+1)\rangle$, and $|gg(n+1)\rangle$, the values

$$\begin{aligned}&\varepsilon_1=0,\quad \varepsilon_2=-2\gamma^2/\nu\ ,\\ &\varepsilon_3=\upsilon+2(n+1)\gamma^2/\nu\ ,\quad \varepsilon_4=-\nu-2n\gamma^2/\nu\ ,\end{aligned} \tag{14}$$

where it is noted that the difference between E_1 and E_2 is independent of n. On substituting the eigenvalues, Eq. (14), and the corresponding eigenstates into Eq. (9), one recovers Eq. (13) for the probability of the transition from $|ugn\rangle$ to $|gun\rangle$. Energy oscillates between the atoms with the same frequency (γ^2/ν), regardless of whether the radiation mode is excited or in its ground state.

The angular frequency of energy oscillation, $\Omega=\gamma^2/(\omega_0-\omega)$, can be expressed in terms of the lifetime [3] of the upper atomic level for spontaneous emission to the lower level, $1/\tau=4d^2\omega^3/3\hbar c^3$ where d is dipole matrix element between the two atomic states. In Eq. (7) we take the dipole parallel to the electric field, put $|\mathbf{d}\cdot\mathbf{u}(\mathbf{r})|^2=d^2/V$ where V is the effective volume of the mode, $V=(\omega_0/\pi c)^3$, and with $\gamma^2=(2\pi\omega_0/\hbar)d^2/V$, obtain finally

$$\begin{aligned}\Omega&=\gamma^2/(\omega_0-\omega)\\ &=(1/\tau)(3/2\pi^2)(\omega_0/\omega_0-\omega)(\omega_0/\omega)^3\ .\end{aligned} \tag{15}$$

For the case that the cavity fundamental frequency is twice the atomic frequency, $\omega_0=2\omega$, the energy oscillates with a frequency comparable to the reciprocal of τ. The strong dependence of Ω on the ratio (ω_0/ω) indicates that one need only consider coupling to the fundamental cavity mode, with negligible interference from coupling to higher modes.

Generalization to the case of a coherently excited collection of two-level atoms coupled by the field to a second distinguishable collection in their ground state can be carried out by combining each set of atoms into an angular momentum oscillator [4].

This paper focuses on a process which is only relevant under conditions of absence of spontaneous emission: the direct transfer of energy between atoms with a rate proportional to the second power of the coupling constant, while in "normal" conditions the first-order process of emission of a photon by the excited atom is much more

significant, with some probability for a subsequent absorption by the other atom.

Equations (13) and (15) which describe the oscillation of energy between the atoms could be of interest for experimentalists. Preparing two different species of atoms in the upper and lower energy states, respectively, of the resonant pair of levels, and passing them through a suitable cavity, one could measure the fraction of energy transferred between the species as a function of time spent inside the cavity. Earlier experiments [2] have shown the suppression of spontaneous emission in certain conditions, and such a demonstration of the consequences of this supression is of some importance.

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[1] Such as the field inside a waveguide with a frequency cutoff at, or a cavity with a fundamental frequency, ω_0. This frequency lower limit may be only relevant to radiation with a particular polarization, and it is assumed the atom is suitably polarized.

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[3] E. G. Harris, *A Pedestrian Approach to Quantum Field Theory* (Wiley-Interscience, New York, 1972).

[4] I. R. Senitzky, Phys. Rev. A **3**, 421 (1971).