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Spontaneous Emission Suppression and Stimulated Raman Transitions in a Four Level System

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We investigate the spontaneous emission process during stimulated Raman transitions in a system containing two ground states and two excited states. The coupling of two excited states to a common ground state via the vacuum electric field modifies the time evolution of the excited state amplitudes from that of a single excited state. The excited state amplitudes are adiabatically eliminated to obtain an evolution equation for the reduced system of two ground states with zero photons in the vacuum. We find that spontaneous emission cannot be suppressed without also suppressing the two-photon Rabi frequency. For particular values of the dipole moments and the laser intensity, however, the two-photon Rabi frequency to spontaneous emission rate is improved by a factor of two as compared to two excited states not coupled via the vacuum electric field.

It is well known that spontaneous emission plays a fundamental role in the decoherence of quantum gates in ion trap quantum computers [1]. Many of these quantum processors use two hyperfine ground states as the qubit and use stimulated Raman transitions to perform arbitrary single qubit rotations [2]. Even though the detuning from the excited state reduces spontaneous emission during a stimulated Raman transition, its effect on gate fidelity becomes important for fault tolerant quantum computing [3].

In this work, we explore spontaneous emission from a four level system consisting of two excited states and two ground states in the presence of two classical laser fields and the vacuum electric field. We begin in part 1 by deriving the decay of two excited states coupled to a common ground state by the vacuum. It is shown that the time evolution of the amplitudes of the excited states are no longer simple exponentials, and the decay process can either be enhanced or suppressed by particular choices of initial conditions. In part 2 we derive the evolution equations for the reduced two level system of ground states for a stimulated Raman process in the absence of the vacuum. We follow the standard approach of adiabatically eliminating the excited states in the presence of two off-resonant laser fields. In part 3 we combine the results of parts 1 and 2 to find the evolution equations for the ground states in the presence of spontaneous emission.

Part 1. Decay of Two Excited States Coupled to a Common Ground State by the Vacuum

The energy level diagram for two excited states coupled through the vacuum to a common ground state is shown in Figure 1. Our analysis here follows that of Zhu, Chan, and Lee [4].

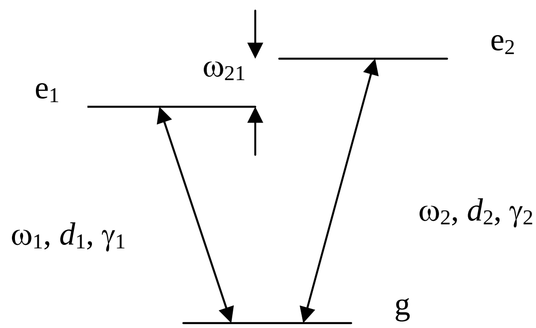


Figure 1

The excited states $|e_1\rangle$ and $|e_2\rangle$ are separated by ω_{21} and are each separated by ω_1 and ω_2 from the ground state $|g\rangle$. We assume $\omega_{21} << \omega_1, \omega_2$; $\omega_2 > \omega_1 > \omega_{21} > 0$; . The Hamiltonian for this system is $H = H_0 + H_I$ where $H_0 = \hbar\omega_{e1}|e_1\rangle\langle e_1| + \hbar\omega_{e2}|e_2\rangle\langle e_2| + \hbar\omega_g|g\rangle\langle g| + \sum_k \hbar\omega_k a_k^+ a_k$ and $H_I = -\left(\vec{d}_1 + \vec{d}_2\right)\cdot \vec{E}_{QED}$. $\vec{E}_{QED} = \sum_k \hat{\varepsilon}_k E_{\omega_k}\left(ia_k - ia_k^+\right)$ is the electric field operator, $E_{\omega_k} = \left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}$ in gaussian units, a_k^+ (a_k) are the creation (annilation) operators for the k^{th} vacuum mode, and $\vec{d}_j = \hat{d}_j\left(d_j|e_j\rangle\langle g| + d_j^*|g\rangle\langle e_j|\right)$ is the dipole operator for the transition from $g \rightarrow e_j$.

The interaction Hamiltonian in the interaction picture is given by:

(1) $$H_I' = \hbar\sum_k \begin{Bmatrix} i\Omega_k^{(1)} e^{i(\omega_1-\omega_k)t} a_k |e_1\rangle\langle g| - i\Omega_k^{(1)*} e^{-i(\omega_1-\omega_k)t} a_k^+ |g\rangle\langle e_1| \\ + i\Omega_k^{(2)} e^{i(\omega_2-\omega_k)t} a_k |e_2\rangle\langle g| - i\Omega_k^{(2)*} e^{-i(\omega_2-\omega_k)t} a_k^+ |g\rangle\langle e_2| \end{Bmatrix}$$

where $\Omega_k^{(j)} \equiv -d_j \frac{\hat{d}_j \cdot \hat{\varepsilon}_k}{\hbar} E_{\omega_k}$ is the vacuum Rabi frequency for the k^{th} vacuum mode.

We assume that the initial state vector is in a superposition of excited states, namely:

(2) $$|\Psi(0)\rangle = c_{e1}(0)|e_1\rangle|0\rangle + c_{e2}(0)|e_2\rangle|0\rangle$$

At all other times $t > 0$, the state vector can be written in the interaction picture as:

(3) $$\left|\Psi(t)\right\rangle = c_{e1}(t)\left|e_1\right\rangle\left|0\right\rangle + c_{e2}(t)\left|e_2\right\rangle\left|0\right\rangle + \sum_k c_{gk}(t)\left|g\right\rangle\left|1_k\right\rangle$$

The evolution of the state vector is determined by the Schrödinger equation:

(4) $$i\hbar\frac{d}{dt}\left|\Psi(t)\right\rangle = H_I'\left|\Psi(t)\right\rangle$$

which leads to evolution equations for the coefficients c_{e1}, c_{e2}, and c_{gk}.

(5) $$\begin{aligned} i\dot{c}_{e1} &= \sum_k i\Omega_k^{(1)} e^{i(\omega_1-\omega_k)t} c_{gk} \\ i\dot{c}_{e2} &= \sum_k i\Omega_k^{(2)} e^{i(\omega_2-\omega_k)t} c_{gk} \\ i\dot{c}_{gk} &= -i\Omega_k^{(1)*} e^{-i(\omega_1-\omega_k)t} c_{e1} - i\Omega_k^{(2)*} e^{-i(\omega_2-\omega_k)t} c_{e2} \end{aligned}$$

Formally integrating $\dot{c}_{gk}$ and substituting the result into the first two equations in (5) yields:

(6) $$\begin{aligned} \dot{c}_{e1} &= -\frac{\gamma_1}{2}c_{e1} - \frac{\cos(\theta_d)e^{i\phi_d}}{2}\sqrt{\gamma_1\gamma_2}\,c_{e2}e^{-i\omega_{21}t} \\ \dot{c}_{e2} &= -\frac{\gamma_2}{2}c_{e2} - \frac{\cos(\theta_d)e^{-i\phi_d}}{2}\sqrt{\gamma_1\gamma_2}\,c_{e1}e^{i\omega_{21}t} \end{aligned}$$

where θ_d is the angle between the dipole moments $\vec{d}_1$ and $\vec{d}_2$, $\phi_d = \arg(d_1) - \arg(d_2)$ is the phase difference between $\vec{d}_1$ and $\vec{d}_2$, and $\gamma_j \equiv \frac{4}{3}\frac{\left|d_j\right|^2}{\hbar c^3}\omega_j^3$ is the spontaneous emission rate from state e_j to the ground state (see Appendix).

From (6) we see that the coupling provided by the vacuum for the excited states to a common ground states produces an effective coupling between the excited states themselves. This is similar to the adiabatic elimination step for stimulated Raman transitions. There, two ground states each having a coupling to a common excited state obtains an effective coupling between the ground states with the excited state only virtually populated (see part 2).

In the above derivation, nowhere did we assume the smallness of ω_{21} in reference to the decay rates $\gamma_{1,2}$. However, when ω_{21} becomes large compared to the decay rates, the coupling between excited states becomes highly oscillatory, and we obtain regular exponential spontaneous decay. Also note that if the dipole moments are orthogonal, the excited state coupling vanishes.

Equation (6) can be solved analytically by the use of Laplace transforms after first transforming to a rotating frame to remove the explicit time dependence. Let

(7) $$c_{e1} \equiv c_{e1}' e^{-i\omega_{21} t/2}$$
$$c_{e2} \equiv c_{e2}' e^{i\omega_{21} t/2}$$

(6) then becomes

(8) $$\dot{c}_{e1}' = \left(-\frac{\gamma_1}{2} + \frac{i}{2}\omega_{21}\right) c_{e1}' - \frac{\cos(\theta_d) e^{i\phi_d}}{2}\sqrt{\gamma_1\gamma_2}\, c_{e2}'$$
$$\dot{c}_{e2}' = \left(-\frac{\gamma_2}{2} - \frac{i}{2}\omega_{21}\right) c_{e2}' - \frac{\cos(\theta_d) e^{-i\phi_d}}{2}\sqrt{\gamma_1\gamma_2}\, c_{e1}'$$

Taking the Laplace transform of (8) and solving the algebraic equation yields:

(9) $$C_1(s) = \frac{2}{f(s)}\left\{(2s + \gamma_2 + i\omega_{21}) c_{e1}(0) - \sqrt{\gamma_1\gamma_2}\cos(\theta_d) e^{i\phi_d} c_{e2}(0)\right\}$$
$$C_2(s) = \frac{2}{f(s)}\left\{(2s + \gamma_1 - i\omega_{21}) c_{e2}(0) - \sqrt{\gamma_1\gamma_2}\cos(\theta_d) e^{-i\phi_d} c_{e1}(0)\right\}$$
$$\text{where } f(s) = (2s + \gamma_1 - i\omega_{21})(2s + \gamma_2 + i\omega_{21}) - \gamma_1\gamma_2\cos^2(\theta_d)$$

The inverse Laplace transform of (9) gives the time domain solution for the $c_{ej}(t)$ amplitudes as a function of initial conditions. Figure 2 shows the time evolution of the population in both excited states for the initial conditions $c_{e1}(0) = 1$, $c_{e2}(0) = 0$. The population in e_2 starts at 0 and grows prior to its final decay at a slower rate.

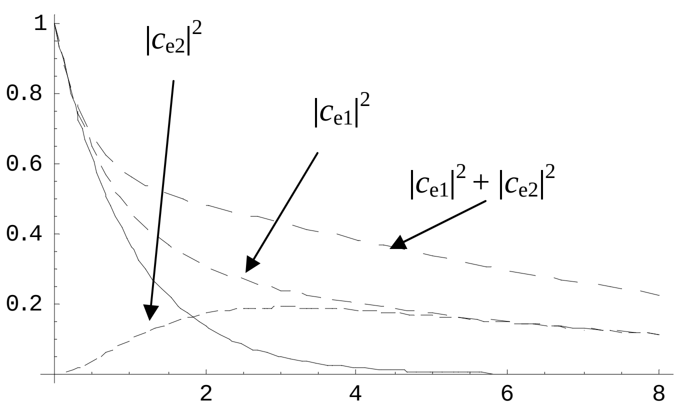


Figure 2. $c_{e1}(0) = 1$, $c_{e2}(0) = 0$. Solid line is regular spontaneous decay for reference. $\gamma_1 = \gamma_2 = 1$, $\omega_{21} = 0.5$, $\phi_d = 0$, $\cos(\theta_d) = -1$.

Figure 3 shows the total population in the excited states for different sets of initial conditions. It should be noted that the initial conditions play a very strong role in whether the spontaneous emission is suppressed or enhanced.

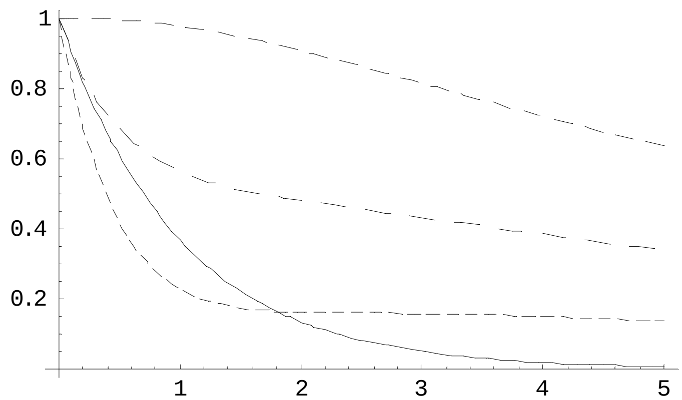


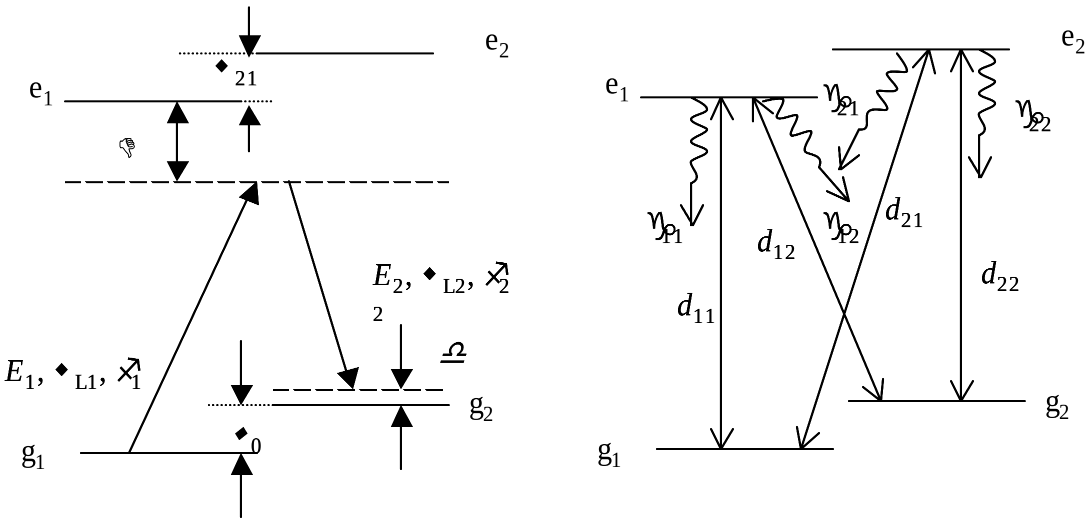
Figure 3. Time evolution of total population in the excited states for different initial conditions. Solid line is regular spontaneous decay for reference. Dashed lines from top to bottom: $c_{e1}(0) = c_{e2}(0) = 1/\sqrt{2}$; $c_{e1}(0) = 1$, $c_{e2}(0) = 0$; $c_{e1}(0) = -0.6$ $c_{e2}(0) = 0.74 - 0.3i$. $\gamma_1 = \gamma_2 = 1$, $\omega_{21} = 0.5$, $\phi_d = 0$, $\cos(\theta_d) = -1$.

The suppression of spontaneous emission can be thought of as the system evolving into a superposition of excited states corresponding to a dark state that have a net dipole moment much smaller than the dipole moments between eigenstates of the base Hamiltonian H_0. This particular superposition's dipole moment is a coherent sum of the individual dipole moments, and destructive interference occurs to suppress this matrix element to a smaller value [5].

From here others have optimized the suppression of spontaneous decay by finding initial conditions that maximize the excited state population for some particular time *t* [5]. Because the evolution is reversible, in principle it should be possible then continue to suppress the decay by applying microwave π pulses at intervals *t*. Zhu, Chan, and Lee in their treatment of this problem calculate the spontaneous emission spectrum [4]. They find regions in the spectrum where dark lines occur and explore situations where the spectrum can be narrowed. During a Raman transition, however, we do not have the liberty to pick the excited state initial amplitudes. Rather, the ground state amplitudes themselves determine the excited state amplitudes. In part 3 we solve the problem of two excited states decaying in the process of a Raman transition.

Part 2. Stimulated Raman Transitions in the Absence of the Vacuum

Consider the energy level diagram for the four level system shown in Figure 4.



Laser fields, frequency splittings

Figure 4

Dipole moments, decay rates

The interaction Hamiltonian for this system is:

$$H_I = -\left(\vec{d}_{11} + \vec{d}_{12} + \vec{d}_{21} + \vec{d}_{22}\right)\cdot\left(\vec{E}_1 + \vec{E}_2 + \vec{E}_{QED}\right)$$

where

(10)
$$\vec{d}_{ij} \equiv \hat{d}_{ij} d_{ij} \left| e_i \right\rangle\left\langle g_j \right| + \text{h.c.}$$

$$\vec{E}_j \equiv E_j \hat{\varepsilon}_j 2\cos\left(-\omega_{Lj} t + \phi_j\right)$$

$\vec{E}_{QED}$ is defined in part 1. $\gamma_{ij} \equiv \frac{4}{3}\frac{\left|d_{ij}\right|^2}{\hbar c^3}\left(\omega_{ei} - \omega_{gj}\right)^3$. We assume ω_{21}, $\Delta << \omega_{ei} - \omega_{gj}$; $\delta << \omega_0 << \Delta$; $\gamma << \Delta$. Consequently, $\gamma_{ij} \cong \beta \left|d_{ij}\right|^2$ where β is independent of i and j. Again, we do not make any assumptions of the size of ω_{21} compared to γ_{ij}. We will ignore $\vec{E}_{QED}$ in this part and come back to it in part 3. Define the Rabi frequencies as follows.

(11) $$\Omega_{ijk} \equiv -\frac{d_{ij} E_k \hat{d}_{ij} \cdot \hat{\varepsilon}_k}{\hbar}$$

We assume $\Omega_{ijk} << \Delta$. At all times $t > 0$, the state vector in the interaction picture can be written as:

(12) $$\left|\Psi(t)\right\rangle = c_{e1}(t)\left|e_1\right\rangle + c_{e2}(t)\left|e_2\right\rangle + c_{g1}(t)\left|g_1\right\rangle + c_{g2}(t)\left|g_2\right\rangle$$

The Schrödinger equation gives the evolution of the coefficients $c_{(e,g)j}$. Using the rotating wave approximation to neglect highly oscillatory terms at twice the optical frequencies, we obtain the following.

(13)
$$\begin{aligned} i\dot{c}_{e1} &= \left(\Omega_{111}e^{i\phi_1} + \Omega_{112}e^{i\phi_2}e^{i(\delta+\omega_0)t}\right)e^{i\Delta t}c_{g1} \\ &\quad + \left(\Omega_{122}e^{i\phi_2}e^{i\delta t} + \Omega_{121}e^{i\phi_1}e^{-i\omega_0 t}\right)e^{i\Delta t}c_{g2} \\ i\dot{c}_{e2} &= \left(\Omega_{211}e^{i\phi_1} + \Omega_{212}e^{i\phi_2}e^{i(\delta+\omega_0)t}\right)e^{i(\Delta+\omega_{21})t}c_{g1} \\ &\quad + \left(\Omega_{222}e^{i\phi_2}e^{i\delta t} + \Omega_{221}e^{i\phi_1}e^{-i\omega_0 t}\right)e^{i(\Delta+\omega_{21})t}c_{g2} \end{aligned}$$

To simplify the analysis, we let $C_e \equiv \begin{bmatrix} c_{e1} \\ c_{e2} \end{bmatrix}$ and $C_g \equiv \begin{bmatrix} c_{g1} \\ c_{g2} \end{bmatrix}$. Then $i\dot{C}_e = UM^+C_g$ where

(14) $$U = \begin{bmatrix} e^{i\Delta t} & 0 \\ 0 & e^{i(\Delta+\omega_{21})t} \end{bmatrix}$$

is a unitary diagonal matrix, and

(15) $$M^+ = \begin{bmatrix} \Omega_{111}e^{i\phi_1} + \Omega_{112}e^{i\phi_2}e^{i(\delta+\omega_0)t} & \Omega_{122}e^{i\phi_2}e^{i\delta t} + \Omega_{121}e^{i\phi_1}e^{-i\omega_0 t} \\ \Omega_{211}e^{i\phi_1} + \Omega_{212}e^{i\phi_2}e^{i(\delta+\omega_0)t} & \Omega_{222}e^{i\phi_2}e^{i\delta t} + \Omega_{221}e^{i\phi_1}e^{-i\omega_0 t} \end{bmatrix}$$

It follows from the hermiticity of the Hamiltonian that $i\dot{C}_g = MU^+C_e$. We can adiabatically eliminate the excited states by moving to a rotating frame. Let $C_e = UC_e'$ define C_e'. Then $i\dot{C}_e = iU_2UC_e' + iU\dot{C}_e' = UM^+C_g$ where $U_2 \equiv \begin{bmatrix} i\Delta & 0 \\ 0 & i(\Delta+\omega_{21}) \end{bmatrix}$ is a purely imaginary diagonal matrix. We can neglect the $iU\dot{C}_e'$ term from the assumption that $\Delta >> \gamma$. Multiplying on the left by $-iU^{-1}U_2^{-1}$ and using the fact that diagonal matrices commute, we obtain $C_e' = QM^+C_g$ where $Q \equiv -iU_2^{-1} = \begin{bmatrix} -\frac{1}{\Delta} & 0 \\ 0 & -\frac{1}{\Delta+\omega_{21}} \end{bmatrix}$ is a real diagonal matrix. Substituting this result into the evolution equation for C_g and using the fact that U is unitary yields:

(16) $$i\dot{C}_g = MU^+UC_e' = MQM^+C_g$$

From (16) we see that MQM^+ acts as an effective Hamiltonian for the reduced 2-level system of C_g. MQM^+ will have low frequency terms (oscillating at frequencies δ) as well as terms oscillating at frequencies ω_0, $2\omega_0$. We can ignore the higher frequency terms in favor of the low frequency terms by applying the rotating wave approximation and using $\delta << \omega_0$. The result is:

(17a) $$R \equiv RWA(MQM^+) = \begin{bmatrix} \delta_{s1} & \Omega^* e^{i\delta t} \\ \Omega e^{-i\delta t} & \delta_{s2} \end{bmatrix}$$

where

(17b) $$\delta_{s1} \equiv -\frac{|\Omega_{111}|^2}{\Delta} - \frac{|\Omega_{112}|^2}{\Delta} - \frac{|\Omega_{211}|^2}{\Delta + \omega_{21}} - \frac{|\Omega_{212}|^2}{\Delta + \omega_{21}}$$

(17c) $$\delta_{s2} \equiv -\frac{|\Omega_{121}|^2}{\Delta} - \frac{|\Omega_{122}|^2}{\Delta} - \frac{|\Omega_{221}|^2}{\Delta + \omega_{21}} - \frac{|\Omega_{222}|^2}{\Delta + \omega_{21}}$$

(17d) $$\Omega \equiv -e^{i(\phi_1 - \phi_2)} \left(\frac{\Omega_{111}\Omega_{122}{}^*}{\Delta} + \frac{\Omega_{211}\Omega_{222}{}^*}{\Delta + \omega_{21}} \right)$$

δ_{sj} are the stark shifts of the ground state levels. Their relative difference shifts the transition frequency ω_0, and these can be neglected by a redefinition of ω_0 and using the assumption that $\Omega_{ijk} << \Delta$. Ω is the Rabi frequency for the 2-level system. Its magnitude and phase are set by the laser intensities and the laser relative phases $\phi_1 - \phi_2$.

The two level problem can be solved by moving to a rotating frame at frequency δ to remove any explicit time dependence. The problem then becomes one of solving two coupled linear ordinary differential equations with constant coefficients. These problems are easily solved with Laplace transform techniques. There are many works on this subject, and the reader is referred to [6] for an in-depth analysis. We will not pursue this subject further here.

Part 3. Stimulated Raman Transitions in the Presence of the Vacuum

Spontaneous emission can be added to the Raman process by adding the effects of the vacuum electric field coupling to the system. We have already worked out this affect in part 1 to obtain the evolution equations (6). In that treatment, the vacuum couples states $|e_j\rangle|0\rangle$ to states $|g_j\rangle|1_k\rangle$. There is no coupling produced by the vacuum between states $|g_j\rangle|0\rangle$ and any other state. Because the Schrödinger equation is linear and both evolution equations were calculated in the interaction picture, we can simply add *i* times equation (6) to equation (13) to obtain the following result.

(18) $$i\dot{C}_e = UM^+ C_g + iTC_e$$

where *T* is a hermitian matrix and is given by:

(19a) $$T \equiv \begin{bmatrix} -a_1 & -be^{-i\omega_{21}t} \\ -b^* e^{i\omega_{21}t} & -a_2 \end{bmatrix}$$

(19b) $$a_1 \equiv \frac{1}{2}(\gamma_{11} + \gamma_{12})$$

(19c) $a_2 \equiv \frac{1}{2}(\gamma_{21} + \gamma_{22})$

(19d) $b \equiv \frac{1}{2}\sqrt{\gamma_{11}\gamma_{21}} \cos\theta_{d1} e^{i\phi_{d1}} + \frac{1}{2}\sqrt{\gamma_{12}\gamma_{22}} \cos\theta_{d2} e^{i\phi_{d2}}$

where $\cos\theta_{dj} \equiv \hat{d}_{1j} \cdot \hat{d}_{2j}$ and $\phi_{dj} \equiv \phi_{d1j} - \phi_{d2j}$.

Following a similar treatment to that in part 2, we move to a rotating frame to remove the time dependence at frequencies near Δ. Using (14) and the definition of U_2 in part 2, we let $C_e = UC_e$' and obtain:

(20) $i\dot{C}_e = iU_2 UC_e' + iU\dot{C}_e' = UM^+ C_g + iTUC_e'$

Neglecting the time derivative of C_e' relative to U_2C_e', using the definition of Q in part 2, and solving for C_e', we find:

(21) $C_e' = QM^+ C_g + PQM^+ C_g$

where

(22)
$$P \equiv (I - iQT')^{-1} - I$$
$$T' \equiv U^{-1}TU = \begin{bmatrix} -a_1 & -b \\ -b^* & -a_2 \end{bmatrix}$$

Notice that the transformation $C_e = UC_e$' in (20), when solved, removes the explicit time dependence $e^{\pm i\omega_{21}t}$ in T without any approximation on the smallness of ω_{21}. This is very important because in part 1 we showed that it was a requirement that ω_{21} be on the same order or smaller than the natural line width in order to observe modifications to spontaneous decay. Here, this restriction is no longer necessary. It is an artifact of the unitary transformation U that these oscillations are removed. We may expect then that the spontaneous decay from the excited states during a Raman transition may also be modified even for excited state splittings comparable to the detuning Δ. Remember that the only requirement on ω_{21} is that it be small compared to the gross structure splittings $\omega_e - \omega_g$. Thus, it may be possible to use two different fine structure levels as the two excited state levels in a possible experiment.

We now can proceed to adiabatically eliminate the excited state to obtain an evolution equation for the reduced two level system of ground states and zero photons in the vacuum. The evolution equations for the ground states with zero photons in the vacuum are unchanged by the presence of the vacuum, namely $i\dot{C}_g = MU^+ C_e$. Substituting (21), we obtain:

(23) $i\dot{C}_g = (MQM^+ + MPQM^+)C_g$

By defining $S \equiv MPQM^+$, then $V \equiv R+S$ acts like an effective Hamiltonian for the reduced two level system of ground states with zero photons in the vacuum. It should be noted here that S is not hermitian. The real parts of the diagonal elements contribute to the stark shifts of the ground state levels and can be neglected by a redefinition of ω_0. The imaginary parts of the diagonal elements lead to decay into non-zero photon states in the vacuum. The off-diagonal elements perturb the Rabi frequency. In the limit that $\gamma_{ij} \to 0$, then $P \to 0$ and $S \to 0$, and we recover the regular two level system evolving according to a hermitian effective Hamiltonian R as in part 2. Taking the rotating wave approximation similar to part 2 and keeping only lowest order terms in γ/Δ, we obtain for the imaginary parts of the diagonal elements of S:

(24a)
$$\begin{aligned}\mathrm{Im}(S_{11}) = &-\frac{a_1}{\Delta^2}\left(\left|\Omega_{111}\right|^2 + \left|\Omega_{112}\right|^2\right) - \frac{a_2}{(\Delta+\omega_{21})^2}\left(\left|\Omega_{211}\right|^2 + \left|\Omega_{212}\right|^2\right)\\ &-\frac{2}{\Delta(\Delta+\omega_{21})}\mathrm{Re}\left(b\Omega_{211}\Omega_{111}{}^* + b\Omega_{212}\Omega_{112}{}^*\right)\end{aligned}$$

(24b)
$$\begin{aligned}\mathrm{Im}(S_{22}) = &-\frac{a_1}{\Delta^2}\left(\left|\Omega_{121}\right|^2 + \left|\Omega_{122}\right|^2\right) - \frac{a_2}{(\Delta+\omega_{21})^2}\left(\left|\Omega_{221}\right|^2 + \left|\Omega_{222}\right|^2\right)\\ &-\frac{2}{\Delta(\Delta+\omega_{21})}\mathrm{Re}\left(b\Omega_{221}\Omega_{121}{}^* + b\Omega_{222}\Omega_{122}{}^*\right)\end{aligned}$$

where a_1, a_2, and b are defined in equations (19). The average decay rate for the population to leave the zero-photon states is minus the sum of these imaginary diagonal elements (twice the average of (24a) and (24b)).

We can rewrite Ω_{ijk} in terms of γ_{ij} as follows.

(25a) $$\Omega_{ijk} = \alpha\, e^{i\phi_{dij}} \sqrt{\gamma_{ij}}\, E_k \hat{d}_{ij} \cdot \hat{\varepsilon}_k$$

where

(25b) $$\alpha \equiv \left(\frac{3c^3}{4\hbar\omega^3}\right)^{1/2}_{cgs}, \quad \left(\frac{3\pi\varepsilon_0 c^3}{\hbar\omega^3}\right)^{1/2}_{SI}$$, $\omega = \omega_e - \omega_g$ is the gross structure transition frequency.

Substituting (25a) into $\mathrm{Re}\left(b\Omega_{2j1}\Omega_{1j1}{}^* + b\Omega_{2j2}\Omega_{1j2}{}^*\right)$ and using the definition of b, (19) yields:

(26a)
$$\begin{aligned}\mathrm{Re}\left(b\Omega_{2j1}\Omega_{1j1}{}^* + b\Omega_{2j2}\Omega_{1j2}{}^*\right) = &\frac{\alpha^2}{2}\left(\left|E_1\right|^2\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_1\right)\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_1\right) + \left|E_2\right|^2\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_2\right)\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_2\right)\right)\\ &\times\left(\cos\theta_{dj}\gamma_{1j}\gamma_{2j} + \cos\theta_{d\bar{j}}\cos(\phi_{d1}-\phi_{d2})\sqrt{\gamma_{11}\gamma_{12}\gamma_{21}\gamma_{22}}\right)\end{aligned}$$

(26b) where $$\bar{j} \equiv \begin{cases} 2 & j=1 \\ 1 & j=2 \end{cases}$$

We can rewrite (24) using (19) and (25a) to obtain:

$$\text{(27)}\quad \begin{aligned}\mathrm{Im}(S_{jj}) = -\frac{\alpha^2}{2}\Bigg\{ & \frac{\gamma_{1j}(\gamma_{11}+\gamma_{12})}{\Delta^2}\left(E_1^{\,2}\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_1\right)^2 + E_2^{\,2}\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_2\right)^2 \right) \\ & + \frac{\gamma_{2j}(\gamma_{21}+\gamma_{22})}{(\Delta+\omega_{21})^2}\left(E_1^{\,2}\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_1\right)^2 + E_2^{\,2}\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_2\right)^2 \right) \\ & + \frac{2}{\Delta(\Delta+\omega_{21})}\left(E_1^{\,2}\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_1\right)\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_1\right) + E_2^{\,2}\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_2\right)\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_2\right)\right) \\ & \times\left(\cos\theta_{dj}\gamma_{1j}\gamma_{2j} + \cos\theta_{d\bar{j}}\cos\phi_d\right)\sqrt{\gamma_{11}\gamma_{12}\gamma_{21}\gamma_{22}} \quad \Bigg\}\end{aligned}$$

where $\phi_d \equiv \phi_{d1} - \phi_{d2}$. Consider a system where $\hat{d}_{ij}\cdot\hat{\varepsilon}_k = 1\ \forall i, j, k$; $\hat{d}_{1j}\cdot\hat{d}_{2j} = 1 \Rightarrow \cos\theta_{dj} = 1$; $\cos(\phi_{d1}-\phi_{d2}) = 1$; and $\gamma_{ij} = \gamma$. Then (27) simplifies to the following.

$$\text{(28)}\quad \mathrm{Im}(S_{jj}) = -\alpha^2\gamma^2\left(E_1^{\,2}+E_2^{\,2}\right)\left(\frac{1}{\Delta^2} + \frac{1}{(\Delta+\omega_{21})^2} + \frac{2}{\Delta(\Delta+\omega_{21})}\right)$$

We can maximize (28) by choosing $\Delta = -\omega_{21}/2$. Then, $\mathrm{Im}(S_{jj}) = 0$, and spontaneous emission is suppressed completely. Under these circumstances, we must take into account higher order terms in γ/Δ. There is only one set of higher order terms. These are shown below.

$$\text{(29)}\quad \begin{aligned}\mathrm{Im}^{(2)}(S_{jj}) = -\frac{a_1a_2 - |b|^2}{\Delta^2(\Delta+\omega_{21})^2}\Big\{ & a_2\left|\Omega_{1j1}\right|^2 + a_2\left|\Omega_{1j2}\right|^2 + a_1\left|\Omega_{2j1}\right|^2 + a_1\left|\Omega_{2j2}\right|^2 \\ & - 2\,\mathrm{Re}\left(b\Omega_{2j1}\Omega_{1j1}^{\,*} + b\Omega_{2j2}\Omega_{1j2}^{\,*}\right)\Big\}\end{aligned}$$

Under the conditions for (27) to be zero, then $a_1a_2 - |b|^2$ would also be zero, and the spontaneous emission is truly zero.

Spontaneous emission suppression in the above system is not useful, however, because the Rabi frequency Ω is also zero. We desire to maximize the magnitude of the Rabi frequency while minimizing the decay. From (17d) and (25a), we obtain for a measure of the magnitude of the Rabi frequency:

$$\text{(30)}\quad \begin{aligned}\frac{|\Omega|^2}{\alpha^2E_1^{\,2}E_2^{\,2}} = & \frac{\gamma_{11}\gamma_{12}}{\Delta^2}\left(\hat{d}_{11}\cdot\varepsilon_1\right)^2\left(\hat{d}_{12}\cdot\varepsilon_2\right)^2 + \frac{\gamma_{21}\gamma_{22}}{(\Delta+\omega_{21})^2}\left(\hat{d}_{21}\cdot\varepsilon_1\right)^2\left(\hat{d}_{22}\cdot\varepsilon_2\right)^2 \\ & + \frac{2\cos\phi_d}{\Delta(\Delta+\omega_{21})}\sqrt{\gamma_{11}\gamma_{12}\gamma_{21}\gamma_{22}}\left(\hat{d}_{11}\cdot\varepsilon_1\right)\left(\hat{d}_{21}\cdot\varepsilon_1\right)\left(\hat{d}_{12}\cdot\varepsilon_2\right)\left(\hat{d}_{22}\cdot\varepsilon_2\right)\end{aligned}$$

To simplify the analysis further, assume $\gamma_{ij} = \gamma$. The average decay rate for the population, Γ can be expressed by:

$$\frac{\Gamma}{\alpha^2\gamma^2} = \sum_{k=1}^{2} E_k^{\,2}\left\{\frac{\left(\hat{d}_{11}\cdot\hat{\varepsilon}_k\right)^2}{\Delta^2} + \frac{\left(\hat{d}_{12}\cdot\hat{\varepsilon}_k\right)^2}{\Delta^2} + \frac{\left(\hat{d}_{21}\cdot\hat{\varepsilon}_k\right)^2}{\left(\Delta+\omega_{21}\right)^2} + \frac{\left(\hat{d}_{22}\cdot\hat{\varepsilon}_k\right)^2}{\left(\Delta+\omega_{21}\right)^2}\right.$$

$$(31) \qquad + \frac{1}{\Delta\left(\Delta+\omega_{21}\right)}\left[\left(\hat{d}_{21}\cdot\hat{\varepsilon}_k\right)\left(\hat{d}_{11}\cdot\hat{\varepsilon}_k\right)\left(\cos\theta_{d1}+\cos\theta_{d2}\cos\phi_d\right)\right.$$

$$\left.\left. + \left(\hat{d}_{22}\cdot\hat{\varepsilon}_k\right)\left(\hat{d}_{12}\cdot\hat{\varepsilon}_k\right)\left(\cos\theta_{d2}+\cos\theta_{d1}\cos\phi_d\right)\right]\right\}$$

and (30) can be simplified as well.

$$(32) \qquad \frac{|\Omega|^2}{\alpha^2 E_1^{\,2} E_2^{\,2}\gamma^2} = \frac{\left(\hat{d}_{11}\cdot\varepsilon_1\right)^2\left(\hat{d}_{12}\cdot\varepsilon_2\right)^2}{\Delta^2} + \frac{\left(\hat{d}_{21}\cdot\varepsilon_1\right)^2\left(\hat{d}_{22}\cdot\varepsilon_2\right)^2}{\left(\Delta+\omega_{21}\right)^2} + \frac{2\cos(\phi_d)}{\Delta\left(\Delta+\omega_{21}\right)}\left(\hat{d}_{11}\cdot\varepsilon_1\right)\left(\hat{d}_{21}\cdot\varepsilon_1\right)\left(\hat{d}_{12}\cdot\varepsilon_2\right)\left(\hat{d}_{22}\cdot\varepsilon_2\right)$$

We wish to minimize (31) while maximizing (32).

It can be shown that $\cos\theta_{dj}\left(\hat{d}_{2j}\cdot\hat{\varepsilon}_k\right)\left(\hat{d}_{1j}\cdot\hat{\varepsilon}_k\right)\geq 0$. Therefore, the part in [] in (31) is ≥ 0. It turns out that the best Rabi rate to decay rate ratio occurs when the [] part in (31) vanishes and the cos ϕ_d term in (32) is maximized. This occurs when $\Delta < 0$, $\hat{d}_{ij}\cdot\hat{\varepsilon}_k = 1\ \forall i, j, k$; $\hat{d}_{1j}\cdot\hat{d}_{2j} = 1 \Rightarrow \cos\theta_{dj} = 1$, and $\cos\phi_d = -1$. For simplicity, we assume $E_1 = E_2$, and we obtain:

$$(33) \qquad \frac{4|\Omega|^2}{E_1^{\,2}\Gamma} = \left(\frac{1}{\Delta^2} + \frac{1}{\left(\Delta+\omega_{21}\right)^2} - \frac{2}{\Delta\left(\Delta+\omega_{21}\right)}\right)\left(\frac{1}{\Delta^2} + \frac{1}{\left(\Delta+\omega_{21}\right)^2}\right)^{-1}$$

Equation (33) is maximized at $\Delta = -\omega_{21}/2$. This improves the Rabi frequency by a factor of two over the case where $b = 0$.

Equation (23) is a system of two coupled linear first order differential equations. We take the detuning $\delta \to 0$ and solve with the initial conditions $c_{g1}(0) = 1$, $c_{g2}(0) = 0$. Some Rabi flopping curves in the presence of spontaneous decay are shown in figure 4.

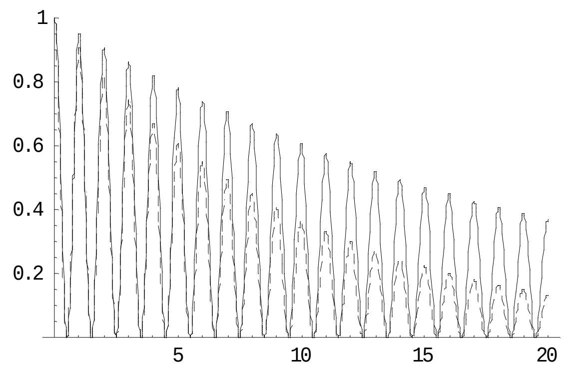


Figure 5. Evolution of $|c_{e1}(t)|^2$ in the presence of spontaneous emission. $\Omega = \pi$ in both curves. Solid curve: Γ = 0.05. Dashed curve: Γ = 0.1.

Conclusion

We have shown that two excited states coupled to a common ground state by the vacuum can produce an effective coupling between the excited states themselves. This coupling causes the system originally prepared in one of the excited states to evolve into dark state where spontaneous decay is somewhat inhibited. Spontaneous emission can either be enhanced or suppressed by a proper choice of initial conditions.

We then explored spontaneous emission during stimulated Raman transitions for a system consisting of two excited states and two ground states. We showed that the two ground states evolve according to a non-hermitian effective Hamiltonian by adiabatically eliminating the excited states. The imaginary parts of the diagonal elements of this two-by-two matrix lead to decay of the amplitudes of the ground states into states with non-zero photons in the vacuum. We then analyzed in which circumstances spontaneous emission can be suppressed during a Raman transition. We found that spontaneous decay cannot be suppressed without also reducing the Rabi frequency for the two ground states. We then maximized the Rabi frequency to spontaneous emission rate ratio by choosing the orientation of the atomic dipole moments and the detuning of the laser fields from the excited states. We found that the ratio of Rabi frequency to spontaneous emission rate can be improved by a factor of two because of the effective coupling of the excited states by the vacuum. Unfortunately, we were unable to suppress spontaneous decay below its regular value (excited states uncoupled from each other via the vacuum).

Appendix

A1. Calculation of the Spontaneous Emission Rate

A2. Calculation of the $e^{i\phi_d} \cos\theta_d$ Term in Part 1

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