Light-Matter Interaction

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Light-matter interactions occur when charged particles accelerate in a time-dependent electric field. An accelerating charge particle generates light, and conversely, electric fields cause forces on the charged particles. In most scenarios, the magnetic field does not directly interact with matter since it is easier to have charges and electric dipoles than magnetic dipoles.

Time-dependent charges can be described by a charge density $\rho(\mathbf{r}, t)$. Dipoles and currents are used more often to describe light-matter interaction. Polarization $\mathbf{P}$ (dipole) and currents density $\mathbf{J}$ have the relations

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0, \tag{0.1}$$

$$\mathbf{J} = \frac{\partial \mathbf{P}}{\partial t}. \tag{0.2}$$

1 Hamiltonian

1.1 Interaction Hamiltonian

According to classical mechanics, a charged particle has the Hamiltonian (SI units)

$$\mathcal{H} = \frac{(\mathbf{p} - q\mathbf{A})^2}{2m} + q\Phi(\mathbf{r}, t), \tag{1.1}$$

where q is the charge of the particle, not the position. $\Phi(\mathbf{r}, t)$ is the electric potential. In the case of an electron, $q = -e$, we have

$$\mathcal{H} = \frac{(\mathbf{p} + e\mathbf{A})^2}{2m} - e\Phi(\mathbf{r}, t). \tag{1.2}$$

We can decompose it into $\mathcal{H}_0$ and $\mathcal{H}_I$,

$$\mathcal{H}_0 = \frac{p^2}{2m}, \tag{1.3}$$

$$\mathcal{H}_I = \frac{e(\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p})}{2m} + \frac{e^2 A^2}{2m} - e\Phi. \tag{1.4}$$

Typically, the term $\frac{e^2A^2}{2m}$ is dropped since the momentum of field $e\mathbf{A}$ is usually small than the electron's momentum $\mathbf{p}$ [1] Since the momentum $\mathbf{p}$ is a differential operator, $\mathbf{p} \cdot \mathbf{A}$ is not equal to $\mathbf{A} \cdot \mathbf{p}$. The vector potential $\mathbf{A}$ and Coulomb's potential Φ are not unique. The Maxwell's equations are invariant under the gauge transformations

$$\mathbf{A}' = \mathbf{A} + \nabla\lambda(\mathbf{r}, t), \tag{1.5}$$

$$\Phi' = \Phi - \frac{\partial \lambda(\mathbf{r}, t)}{\partial t}. \tag{1.6}$$

[1] Well, this is a sloppy argument. In electromagnetism, the source of the vector potential $\mathbf{A}$ is current, so $\mathbf{A}$ is proportional to $\frac{v}{c}$, where v is the electron speed and c is the light speed. The term $\frac{e^2A^2}{2m}$ is proportional to $\frac{v^2}{c^2}$, which is typically small. .

The fields are given by

$$\mathbf{B} = \nabla \times \mathbf{A}, \tag{1.7}$$

$$\mathbf{E} = -\nabla\Phi - \frac{\partial \mathbf{A}}{\partial t}. \tag{1.8}$$

To simplify the Hamiltonian, the Coulomb gauge ($\nabla \cdot \mathbf{A}$) with the condition $\Phi = 0$[2] is used most of the time. In the the Coulomb gauge, using $\mathbf{p} = -i\hbar\nabla$, we have

$$\mathbf{p} \cdot \mathbf{A} = \mathbf{p} \cdot \mathbf{A}$$

The interaction Hamiltonian becomes

$$\mathcal{H}_I = \frac{e(\mathbf{A} \cdot \mathbf{p})}{m} \tag{1.9}$$

$$= -\int dv \mathbf{A} \cdot \mathbf{J} \tag{1.10}$$

where we use $\int dv\mathbf{J} = \frac{-e\mathbf{p}}{m}$.

The interaction Hamiltonian is now related to current and vector potential. It is possible to replace current and vector potential by dipoles and electric field. We use the Göppert-Mayer gauge,

$$\lambda = -(\mathbf{r} - \mathbf{r}_0) \cdot \mathbf{A}(\mathbf{r}_0). \tag{1.11}$$

Using this gauge and Eq. (1.6), we have

$$\mathbf{A}' = \mathbf{A}(\mathbf{r}) - \mathbf{A}(\mathbf{r}_0), \tag{1.12}$$

$$-e\Phi' = e(\mathbf{r} - \mathbf{r}_0) \cdot \mathbf{E}(\mathbf{r}_0) \equiv -\mathbf{d} \cdot \mathbf{E}, \tag{1.13}$$

where $\mathbf{d} = -e(\mathbf{r} - \mathbf{r}_0)$ is the dipole operator since $\mathbf{r}$ is the position operator. The so-called dipole approximation is when $\mathbf{A}(\mathbf{r})$ is almost a constant, i.e., $\mathbf{A}(\mathbf{r}) \simeq \mathbf{A}(\mathbf{r}_0)$. In this approximation, the new vector potential $\mathbf{A}'(\mathbf{r}_0)$ vanishes. This approximation is valid if the field changes gradually over the range of the charge distributions. For example, the charge distribution of an atom is about 0.1 nm, and the electric field of visible light is almost constant over the atom since the wavelengths range from 400 to 700 nm. The interaction Hamiltonian becomes

$$\mathcal{H}_I = -\mathbf{E} \cdot \mathbf{d} \tag{1.14}$$

Although we did not define the field operator $\mathbf{A}$, it can be obtained by the relation of the electric filed operator and the vector potential operator

$$\boldsymbol{\mathcal{E}} = -\frac{\partial}{\partial t}\boldsymbol{\mathcal{A}} \tag{1.15}$$

$$= i\omega\boldsymbol{\mathcal{A}} \tag{1.16}$$

$$\mathbf{A} = \left(\frac{\boldsymbol{\mathcal{E}}a - \boldsymbol{\mathcal{E}}^* a^\dagger}{2i\omega}\right). \tag{1.17}$$

[2] In the region without charges $\nabla \cdot \mathbf{E} = 0$, we can define $\mathbf{E} = -\nabla\Phi$. Using the gauge transformation $\lambda = \int \Phi dt$, we can eliminate Φ and make $\nabla \cdot \mathbf{A} = 0$.

1.2 Total Hamiltonian

The total Hamiltonian of the light-matter is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I + \mathcal{H}_F. \tag{1.18}$$

where

$$\mathcal{H}_F = \sum_m \int dv \left(\frac{\epsilon(\mathbf{r}) E_m^2(\mathbf{r})}{2} + \frac{B_m^2(\mathbf{r})}{2\mu(\mathbf{r})} \right) \tag{1.19}$$

$$= \sum_m \hbar\omega_m \left(a_m^\dagger a_m + \frac{1}{2} \right). \tag{1.20}$$

The Hamiltonian of matter $\mathcal{H}_0$ is not necessary in the form of a free particle. In general, $\mathcal{H}_0$ describes a N-level system,

$$\mathcal{H}_0 = \sum_n E_n |E_n\rangle\langle E_n|. \tag{1.21}$$

The simplest case is a two-level system (TLS)

$$\mathcal{H}_{TLS} = \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix}. \tag{1.22}$$

The interaction Hamiltonian for a two-level system is

$$\mathcal{H}_I = \begin{pmatrix} \langle E_c| - \mathbf{E}\cdot\mathbf{d}|E_c\rangle & \langle E_c| - \mathbf{E}\cdot\mathbf{d}|E_v\rangle \\ \langle E_v| - \mathbf{E}\cdot\mathbf{d}|E_c\rangle & \langle E_v| - \mathbf{E}\cdot\mathbf{d}|E_v\rangle \end{pmatrix} \tag{1.23}$$

$$= -\mathbf{E}\cdot \begin{pmatrix} \mathbf{d}_{cc} & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & \mathbf{d}_{vv} \end{pmatrix}, \tag{1.24}$$

where the dipole matrix element is $\mathbf{d}_{nn'} = \langle E_n|\mathbf{d}|E_{n'}\rangle$. In many cases, the diagonal elements of dipole matrices vanishes since the charge densities of the eigenfunctions are typically symmetric.

2 Classical Fields and Quantum Matter

We consider that the matter is described by a N-level system and treat the electric field $\mathbf{E}(\mathbf{r}, t)$ as a number. The Hamiltonian is

$$\mathcal{H} = \sum_n E_n |E_n\rangle\langle E_n| - \mathbf{E}\cdot\mathbf{d}. \tag{2.1}$$

In the case of a TLS system, the Hamiltonian is

$$\mathcal{H} = \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \mathbf{E}\cdot \begin{pmatrix} 0 & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & 0 \end{pmatrix}, \tag{2.2}$$

where we assume the diagonal elements of the dipole matrix are zeros. To solve the dynamics, we start with the interaction picture where the state is

$$|\psi\rangle = C_c(t) e^{-i\omega_c t}|E_c\rangle + C_v(t) e^{-i\omega_v t}|E_v\rangle. \tag{2.3}$$

It is clear that without an external field $\mathbf{E}$, the coefficients $C_c(t)$ and $C_v(t)$ are constant in time. Plugging Eq. (2.3) in the Schrödinger equation, we obtain

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}C_c\\C_v\end{pmatrix} = -\mathbf{E}\cdot\begin{pmatrix}0 & \mathbf{d}_{cv}e^{i(\omega_c-\omega_v)t}\\ \mathbf{d}_{vc}e^{i(\omega_v-\omega_c)t} & 0\end{pmatrix}\begin{pmatrix}C_c\\C_v\end{pmatrix}. \tag{2.4}$$

The dipole matrix elements in the interaction picture oscillate rapidly in time. The electric field $\mathbf{E} = \boldsymbol{\mathcal{E}}_\omega e^{-i\omega t} + \boldsymbol{\mathcal{E}}^*_\omega e^{i\omega t}$ needs to have a frequency $\omega \simeq (\omega_c - \omega_v)$ in order to create transition. We write

$$\omega = \omega_{cv} + \Delta, \tag{2.5}$$

where $\omega_{cv} = \omega_c - \omega_v$ and Δ is the detuning.

2.1 Rabi Model

Let the external field $\mathbf{E} = \mathbf{E}_0\cos\omega t = \mathbf{E}_0\left(\frac{e^{-i\omega t}+e^{i\omega t}}{2}\right)$. The equation of the coefficients is

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}C_c\\C_v\end{pmatrix} = \begin{pmatrix}0 & \frac{V_0}{2}\left[e^{-i\Delta t}+e^{i(2\omega_{cv}+\Delta)t}\right]\\ \frac{V_0^*}{2}\left[e^{i\Delta t}+e^{-i(2\omega_{cv}+\Delta)t}\right] & 0\end{pmatrix}\begin{pmatrix}C_c\\C_v\end{pmatrix}. \tag{2.6}$$

where

$$V_0 = -\mathbf{E}_0\cdot\mathbf{d}_{cv}. \tag{2.7}$$

The equation needs to be solved numerically. The rotating-wave approximation (RWA), where the high-frequency terms are dropped, is often used. Under the RWA, the equation reads

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}C_c\\C_v\end{pmatrix} = \begin{pmatrix}0 & \frac{V_0}{2}e^{-i\Delta t}\\ \frac{V_0^*}{2}e^{i\Delta t} & 0\end{pmatrix}\begin{pmatrix}C_c\\C_v\end{pmatrix}. \tag{2.8}$$

Eliminating the variable C_v, we obtain the second-order differential equation

$$\ddot{C}_c + i\Delta\dot{C}_c + \frac{|V_0|^2}{4\hbar^2}C_c = 0. \tag{2.9}$$

The general solution is

$$C_c(t) = A_+e^{i\lambda_+t} + A_-e^{i\lambda_-t} \tag{2.10}$$

with

$$\lambda_\pm = \Delta \pm \sqrt{\Delta^2 + \frac{|V_0|^2}{\hbar^2}} \equiv \Delta \pm \Omega_R. \tag{2.11}$$

The Rabi frequency $\Omega_R = \sqrt{\Delta^2 + \frac{|V_0|^2}{\hbar^2}}$. If initially $C_v(0) = 1$ and $C_c(0) = 1$, the solution is

$$C_c = e^{i\frac{\Delta t}{2}}\frac{iV_0}{\hbar\Omega_R}\sin\frac{\Omega_R t}{2}, \tag{2.12}$$

$$C_v = e^{i\frac{\Delta t}{2}}\left[\cos\frac{\Omega_R t}{2} - i\frac{\Delta}{\Omega_R}\sin\frac{\Omega_R t}{2}\right]. \tag{2.13}$$

It can be checked that $|C_c|^2 + |C_v|^2 = 1$. The population of the excited state is

$$P_c(t) = |C_c(t)|^2 = \frac{|V_0|^2 \sin^2 \frac{\Omega_R t}{2}}{\hbar^2 \Omega_R^2} \tag{2.14}$$

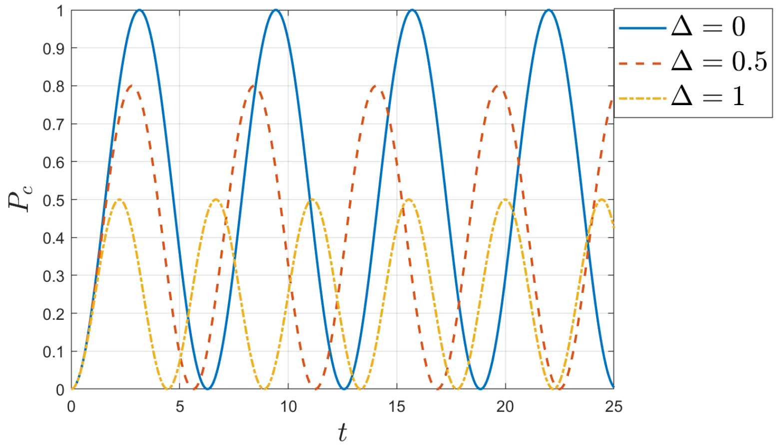

Figure 1: Population of the excited state as a function of time with $\frac{V_0}{\hbar} = 1$.

2.2 Fermi's Golden Rule

If the external field is small, we can obtain from Eq. (2.12) [3]

$$P_c(t) = |C_c|^2 = \frac{|V_0|^2 \sin^2 \frac{\Delta t}{2}}{\hbar^2 \Delta^2}. \tag{2.15}$$

[3] the formal method to obtain this result is the the perturbation method (for example, see Chapter 5 of Ref. [1])

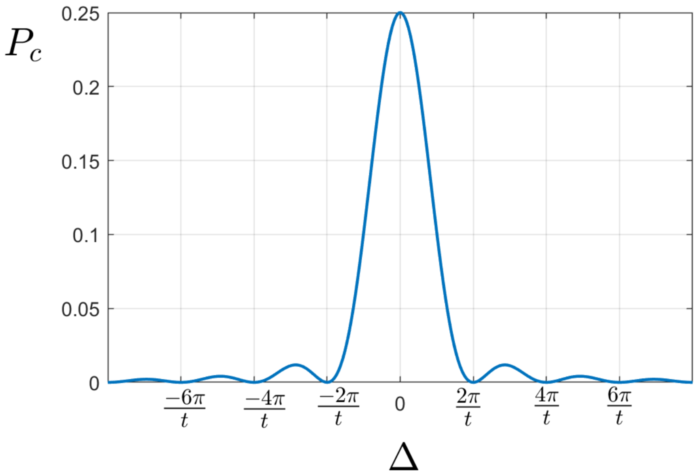

Figure 2: The transition probability $P_c(t)$ at a momentum t. When t is large, the function is approximately a delta function.

When t is large, the fraction is approximately a delta function

$$\frac{\sin^2 \frac{\Delta t}{2}}{\Delta^2} \simeq \frac{\pi t}{2} \delta(\Delta). \tag{2.16}$$

The transition rate $W_{v \to c}$ is

$$W_{v \to c} = \frac{P_c(t)}{t} = \frac{\pi}{2} \frac{|V_0|^2}{\hbar^2} \delta(\omega - \omega_{cv}) \tag{2.17}$$

$$= \frac{\pi}{2} \frac{|\mathbf{E}_0 \cdot \mathbf{d}_{cv}|^2}{\hbar^2} \delta(\omega - \omega_{cv}) \tag{2.18}$$

$$= \frac{\pi}{2} \frac{|\langle c|\mathbf{H}_I|v\rangle|^2}{\hbar^2} \delta(\omega - \omega_{cv}), \tag{2.19}$$

which is the famous Fermi's Golden rule. The unit of $\delta(\omega - \omega_{cv})$ is one over frequency. The delta function $\delta(\omega - \omega_{cv})$ is interpreted as the density of states. Since we consider only a two-level system, there is only one final state for $\omega_{cv} - d\omega/2 < \omega < \omega_{cv} + d\omega/2$. If instead, we consider there are many states between $\omega_{cv} - d\omega/2$ and $\omega_{cv} + d\omega/2$, we will use the the density of states $\rho(\omega)$, defined by

$$\rho(\omega) = \frac{dN}{d\omega}, \tag{2.20}$$

where N is the number of states between $\omega_{cv} - d\omega/2$ and $\omega_{cv} + d\omega/2$. In this case, Fermi's Golden rule becomes

$$W = \frac{\pi}{2} \frac{|\langle c|\mathbf{H}_I|v\rangle|^2}{\hbar^2} \rho(\omega), \tag{2.21}$$

or, in terms of energies,

$$W = \frac{\pi}{2} \frac{|\langle c|\mathbf{H}_I|v\rangle|^2}{\hbar} \rho(E), \tag{2.22}$$

where $\rho(E)dE$ is the number of states for E between $E_{cv} - dE/2$ and $E_{cv} + dE/2$.

Using the Fermi Golden rule, one can derive the famous rate of spontaneous emission in a vacuum,

$$W_{\mathrm{sp}} = \frac{\omega^3 |d_{cv}|^2}{3\pi\epsilon_0 \hbar c^3}. \tag{2.23}$$

Note 1: Fermi's Golden Rule

- Fermi's golden rules are valid in the perturbation regime ($|\langle c|\mathbf{H}_I|v\rangle|$ is small compared to E_{cv})
- Fermi's golden rules describe the incoherent excitation. The excitation events are independent, and the final state is almost empty. These conditions are not true for the Rabi oscillation (coherent excitation).
- The rate is proportional to the square of the transition dipole element $|\langle c|\mathbf{d}|v\rangle|^2$
- The rate is proportional to the density of the final states.

2.3 Density Matrix Approach

Consider a classical light interacted with an ensemble of the same two-level systems. We need to use the density matrix

$$\rho = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} \tag{2.24}$$

and use the quantum Liouville's equation to obtain

$$\frac{d\rho_{11}}{dt} = i\Omega_R(\rho_{12} - \rho_{21}), \tag{2.25}$$

$$\frac{d\rho_{22}}{dt} = -i\Omega_R(\rho_{12} - \rho_{21}), \tag{2.26}$$

$$\frac{d\rho_{12}}{dt} = i\Omega_R(\rho_{11} - \rho_{22}), \tag{2.27}$$

$$\frac{d\rho_{21}}{dt} = -i\Omega_R(\rho_{11} - \rho_{22}), \tag{2.28}$$

where ρ_{11} and ρ_{22} describe probabilities, and ρ_{12} and ρ_{21} describe coherence. Further simplifications give

$$\frac{d^2\rho_{11}}{dt^2} = -2\Omega_R^2(\rho_{11} - \rho_{22}) \tag{2.29}$$

$$= -2\Omega_R^2(2\rho_{11} - 1). \tag{2.30}$$

3 Classical Matter and Quantum Fields

Currents and charges are treated as classical numbers. Time-dependent charges and currents are not independent variables. They are related by the continuity equation.

This assumption is adequate when currents come from a lot of electrons and the quantum fluctuations are ignored. The typical problem is how a current source $\mathbf{I}(\mathbf{r},t)$ interacts with photons. The current is a control and macroscopic parameter which can be treated classically as a number. Thus, currents are given functions, and the problem is to solve the filed Hamiltonian.

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_F + \mathcal{H}_I && (3.1)\\ &= \sum_m \hbar\omega_m a_m^\dagger a_m - \sum_m \mathbf{E}_m \cdot \mathbf{d} && (3.2)\\ &= \sum_m \hbar\omega_m a_m^\dagger a_m - \sum_m \left(\frac{\mathcal{E}_m a + \mathcal{E}_m^* a^\dagger}{2}\right)\cdot \mathbf{d}, && (3.3)\end{aligned}$$

The above interaction Hamiltonian has the dipole instead of a current. Dynamically, dipoles and currents are related. Let the current be $\mathbf{I}(\mathbf{r},t) = \mathbf{I}(\mathbf{r})_0 e^{-i\omega t}$. The current is related to the current density $\mathbf{J}$ by

$$\mathbf{J}(\mathbf{r},t) = \frac{\mathbf{I}(\mathbf{r},t)}{da_\perp}. \tag{3.4}$$

From this relation, we can find the current density $\mathbf{J}(\mathbf{r},t) = \mathbf{J}_0(\mathbf{r})e^{-i\omega t}$. Now we can use the interaction Hamiltonian in terms of $\mathbf{J}$ and $\mathbf{A}$. Considering a single mode and $\omega_m = \omega$, the Hamiltonian becomes

$$\mathcal{H} = \hbar\omega a^\dagger a - \int dv \mathbf{A}\cdot\mathbf{J}. \tag{3.5}$$

3.1 Generation of Coherent States

We are going to show a coherent state $|\alpha\rangle$ can be generated by a harmonic oscillating current density

$$\mathbf{J} = \frac{\mathbf{J}_0(\mathbf{r})e^{-i\omega t} + \mathbf{J}_0^*(\mathbf{r})e^{i\omega t}}{2} \tag{3.6}$$

This current density oscillating with the frequency ω can excite photons of the same frequency. The total Hamiltonian is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I, \tag{3.7}$$

with the photon Hamiltonian $\mathcal{H}_0 = \hbar\omega a^\dagger a$ and $\mathcal{H}_I = -\int dv \mathbf{A}\cdot\mathbf{J}$. Using Eq. (1.17) and the RWA, the interaction Hamiltonian becomes

$$\mathcal{H}_I = \left(V_0 a + V_0^* a^\dagger\right), \tag{3.8}$$

where

$$V_0(t) = \frac{e^{i\omega t}\int dv \mathcal{E}_\omega^*(\mathbf{r})\cdot\mathbf{J}_0(\mathbf{r})}{4i\omega}. \tag{3.9}$$

Now, the term $V_0(t)$ is time-dependent. We can use the interaction picture to remove the time dependence. In the interaction picture[4], the interaction Hamiltonian becomes[5]

$$\tilde{\mathcal{H}}_I = \left(V_I a + V_I^* a^\dagger \right), \tag{3.10}$$

where the interaction potential becomes time-independent and reads

$$V_I = \frac{\int dv \boldsymbol{\mathcal{E}}_\omega^*(\mathbf{r}) \cdot \mathbf{J}_0(\mathbf{r})}{4i\omega}. \tag{3.11}$$

The evolution of a state is given by

$$|\psi(t)\rangle_I = \hat{\mathcal{T}}[e^{-i\int \frac{\tilde{\mathcal{H}}_I(t)}{\hbar} dt}]|\psi(0)\rangle_I \tag{3.12}$$

where $\hat{\mathcal{T}}[]$ denotes the time-ordering[6]. In this case, the interaction Hamiltonian in the interaction picture is time-independent,

$$|\psi(t)\rangle_I = e^{-i\frac{\tilde{\mathcal{H}}_I(t)}{\hbar}t}|\psi(0)\rangle_I \tag{3.13}$$

$$= e^{\alpha^* a - \alpha a^\dagger}|\psi(0)\rangle_I, \tag{3.14}$$

where

$$\alpha = i\frac{V_I^*}{\hbar}t. \tag{3.15}$$

Equation (3.14) is indeed the displacement operator. If the initial state is the ground state $|0\rangle$, the final state is a coherent state,

$$|\psi(t)\rangle_I = e^{\alpha^* a - \alpha a^\dagger}|0\rangle \tag{3.16}$$

$$= |\alpha\rangle. \tag{3.17}$$

One interesting observation is that $|\alpha| \sim t$ and the photon number $n \sim t^2$ grows quadratically.

4 Fully Quantum Approach

When both electrons and fields are quantized, the Hamiltonian includes three parts: photons, electrons, and interactions. The Hamiltonian is

$$\mathcal{H} = \mathcal{H}_F + \mathcal{H}_e + \mathcal{H}_I \tag{4.1}$$

$$= \sum_m \hbar\omega_m a_m^\dagger a_m + \sum_n E_n |E_n\rangle\langle E_n| - \mathbf{E}\cdot\mathbf{d}. \tag{4.2}$$

[4]Rotating with the $\mathcal{H}_0$.
[5]To avoid confusion, we use $\tilde{\mathcal{H}}_I$ to denote the interaction Hamiltonian in the interaction picture.
[6]Time-ordering is necessary if H_I is time-dependent and $[H_I(t_1), H_I(t_2)] \neq 0$

It should be noted that both the field **E** and the dipole **d** are operators. The electric field operator is

$$\mathbf{E} = \sum_m \frac{\mathcal{E}_m a_m + \mathcal{E}_m^* a_m^\dagger}{2}, \tag{4.3}$$

and the dipole matrix operator in the energy basis is

$$\begin{pmatrix} \mathbf{d}_{11} & \mathbf{d}_{12} & \cdots \\ \mathbf{d}_{21} & \mathbf{d}_{22} & \\ \vdots & & \ddots \end{pmatrix}, \tag{4.4}$$

with $\mathbf{d}_{nn'} = \langle E_n|\mathbf{d}|E_{n'}\rangle$ and $\mathbf{d} = q\mathbf{r} = -e\mathbf{r}$.

The Hilbert space of the Hamiltonian includes both the photon and electron parts. The total space is indeed the tensor direct product of each space,

$$|\psi\rangle = |\text{photon}\rangle \otimes |\text{electron}\rangle. \tag{4.5}$$

The dimension of the total space is the product of the dimension of each space. In this definition, the photonic operators such as a and $a^\dagger$ will only be applied on the photonic ket $|\text{photon}\rangle$, and the electronic operators such as **d** will only be applied on the electronic ket $|\text{electron}\rangle$.

$$\langle\psi|\mathcal{H}_F|\psi\rangle = \langle\text{photon}|\mathcal{H}_F|\text{photon}\rangle \otimes \langle\text{electron}|\text{electron}\rangle = \langle\text{photon}|\mathcal{H}_F|\text{photon}\rangle \otimes \mathbb{1}_e, \tag{4.6}$$

$$\langle\psi|\mathcal{H}_e|\psi\rangle = \langle\text{photon}|\text{photon}\rangle \otimes \langle\text{electron}|\mathcal{H}_|\text{electron}\rangle = \mathbb{1}_F \otimes \langle\text{photon}|\mathcal{H}_e|\text{photon}\rangle, \tag{4.7}$$

$$\langle\psi|\mathbf{E}\cdot\mathbf{d}|\psi\rangle = \langle\text{photon}|\mathbf{E}|\text{photon}\rangle \cdot \langle\text{electron}|\mathbf{d}|\text{electron}\rangle. \tag{4.8}$$

For example, we can write the photonic ket in a number basis and the electron ket in the energy basis,

$$|\text{photon}\rangle = \sum_n C_n|n\rangle, \tag{4.9}$$

$$|\text{electron}\rangle = \sum_m D_m|E_m\rangle. \tag{4.10}$$

Now, all the possible states of the total space can be written as

$$|\psi\rangle = \left(\sum_n C_n|n\rangle\right) \otimes \left(\sum_m D_m|E_m\rangle\right). \tag{4.11}$$

In principle, the dimension of the total space is infinite since the dimension of the number state is infinite. In practical computation, we will truncate the photon number so that the maximum number is finite, say n_m. The photon basis vectors now include $|0\rangle$, $|1\rangle$,... , $|n_m\rangle$, so the dimension of the photonic part is m. If now we consider a two-level system of electrons, the dimension of the total space is $m \times 2$. All the basis vectors of the total space are $|0\rangle|E_c\rangle$, $|1\rangle|E_c\rangle$,... , $|n_m\rangle|E_c\rangle$, and $|0\rangle|E_v\rangle$, $|1\rangle|E_v\rangle$,... , $|n_m\rangle|E_v\rangle$.

4.1 Two-Level System and Single-Mode Photons

The Hamiltonian is

$$\mathcal{H} = \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \mathbf{E}\cdot\mathbf{d}. \tag{4.12}$$

where the electric field operator is

$$\mathbf{E} = \frac{\boldsymbol{\mathcal{E}}_\omega a + \boldsymbol{\mathcal{E}}_\omega^* a^\dagger}{2}, \tag{4.13}$$

and the dipole matrix operator is

$$\begin{pmatrix} 0 & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & 0 \end{pmatrix}, \tag{4.14}$$

where we assume that the diagonal terms vanish. The transition rate from $|n\rangle|E_c\rangle$ to $|n+1\rangle|E_v\rangle$ is obtained by

$$W_{\text{emission}} = \frac{\pi}{2}\frac{|\langle n+1|\langle E_v|\mathbf{H}_I|n\rangle|E_c\rangle|^2}{\hbar^2}\delta(\omega-\omega_{cv}) \tag{4.15}$$

$$= \frac{(n+1)\pi}{2}\frac{|\boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}|^2}{\hbar^2}\delta(\omega-\omega_{cv}). \tag{4.16}$$

An interesting result occurs when $n = 0$. The emission is not zero when $n = 0$. This is the phenomenon of "spontaneous emission". When $n > 0$, it corresponds to the stimulated emission. The transition rate from $|n\rangle|E_v\rangle$ to $|n-1\rangle|E_c\rangle$ is obtained by

$$W_{\text{absorption}} = \frac{\pi}{2}\frac{|\langle n-1|\langle E_c|\mathbf{H}_I|n\rangle|E_v\rangle|^2}{\hbar^2}\delta(\omega-\omega_{cv}) \tag{4.17}$$

$$= \frac{n\pi}{2}\frac{|\boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}|^2}{\hbar^2}\delta(\omega-\omega_{cv}). \tag{4.18}$$

4.2 Jaynes-Cummings Model

The TLS and single-mode photon Hamiltonian can be further simplified with the RWA,

The Hamiltonian is

$$\mathcal{H} = \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \frac{1}{2}\begin{pmatrix} 0 & \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}a + \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{cv}a^\dagger \\ \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{vc}a + \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}a^\dagger & 0 \end{pmatrix} \tag{4.19}$$

$$\simeq \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \frac{1}{2}\begin{pmatrix} 0 & \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}a \\ \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}a & 0 \end{pmatrix} \tag{4.20}$$

$$= \hbar\omega a^\dagger a + \frac{E_c+E_v}{2} + \frac{\hbar\omega_{cv}}{2}\sigma_z + \hbar\left(\lambda\sigma_+ a + \lambda^*\sigma_- a^\dagger\right) \tag{4.21}$$

where

$$\lambda = \frac{-\boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}}{2\hbar}. \tag{4.22}$$

The average energy $\frac{E_c+E_v}{2}$ is only a constant, so as irrelevant to dynamics. In most cases, it is possible to make λ real by choosing the phase of $\mathbf{d}_{cv}$. The JaynesCummings Model is then obtained as

$$\mathcal{H}_{JC} = \hbar\omega a^\dagger a + \frac{\hbar\omega_{cv}}{2}\sigma_z + \hbar\lambda\left(\sigma_+ a + \sigma_- a^\dagger\right). \tag{4.23}$$

We have used the Pauli matrices

$$\sigma_z = |E_c\rangle\langle E_c| - |E_v\rangle\langle E_v| = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{4.24}$$

$$\sigma_+ = |E_c\rangle\langle E_v| = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \tag{4.25}$$

$$\sigma_- = |E_v\rangle\langle E_c| = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{4.26}$$

The electron number operator is an identity,

$$N_e = |E_c\rangle\langle E_c| + |E_v\rangle\langle E_v|, \tag{4.27}$$

and the excitation number operator is

$$N_{ex} = |E_c\rangle\langle E_c| + a^\dagger a. \tag{4.28}$$

These numbers are conservative since the commutators vanish

$$[H, N_e] = 0, \tag{4.29}$$

$$[H, N_{ex}] = 0. \tag{4.30}$$

Exercise 1: Excitation Number

Show Eq. (4.38).

The Jaynes-Cummings Model is then obtained as

$$\mathcal{H}_{JC} = \hbar\omega a^\dagger a + \frac{\hbar\omega_{cv}}{2}\sigma_z + \hbar\lambda\left(\sigma_+ a + \sigma_- a^\dagger\right). \tag{4.31}$$

We have used the Pauli matrices

$$\sigma_z = |E_c\rangle\langle E_c| - |E_v\rangle\langle E_v| = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{4.32}$$

$$\sigma_+ = |E_c\rangle\langle E_v| = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \tag{4.33}$$

$$\sigma_- = |E_v\rangle\langle E_c| = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{4.34}$$

The electron number operator is an identity,

$$N_e = |E_c\rangle\langle E_c| + |E_v\rangle\langle E_v|, \tag{4.35}$$

and the excitation number operator is

$$N_{ex} = |E_c\rangle\langle E_c| + a^\dagger a. \tag{4.36}$$

These numbers are conservative since the commutators vanish

$$[\mathcal{H}, N_e] = 0, \tag{4.37}$$
$$[\mathcal{H}, N_{ex}] = 0, \tag{4.38}$$

which mean that the total Hamiltonian can be **block-diagonalized**, and in each block, the excitation number and the electron number are the same. The basis kets are

$$|n\rangle \otimes |E_m\rangle \equiv |n\rangle|E_m\rangle \tag{4.39}$$

where $E_m = E_c$ or E_v and $n = 0,\ 1,\ ,2,\ 3,...$ It seems that if we want to use the number states as the basis, the dimension of the Hamiltonian would be infinite. This is true, but the Hamiltonian can be block-diagonalized. **Because the excitation number is conserved, only the states with the same excitation number are coupled**. Within each block, the excitation number is the same. Eventually, one finds that each block is just a 2 by 2 matrix. This is because the state $|E_c\rangle|n\rangle$ is only coupled to $|E_v\rangle|n+1\rangle$. The problem is then solved using a two-dimensional Hamiltonian since each block is independent.

The Hamiltonian is decomposed as

$$\mathcal{H}_{JC} = \mathcal{H}_N + \mathcal{H}_D \tag{4.40}$$
$$\mathcal{H}_N = \hbar\omega N_{ex} - \hbar\frac{\omega}{2}N_e, \tag{4.41}$$
$$\mathcal{H}_D = -\frac{\hbar\Delta}{2}\sigma_z + \hbar\lambda\left(\sigma_+ a + \sigma_- a^\dagger\right). \tag{4.42}$$

with $\omega = \omega_{cv} + \Delta$. The two Hamiltonians $\mathcal{H}_N$ and $\mathcal{H}_D$ commute with each other,

$$[\mathcal{H}_N, \mathcal{H}_D] = 0, \tag{4.43}$$

which means the two Hamiltonians are decoupled, so

$$e^{-i\frac{\mathcal{H}_N+\mathcal{H}_D}{\hbar}t} = e^{-i\frac{\mathcal{H}_N}{\hbar}t}e^{-i\frac{\mathcal{H}_D}{\hbar}t} = e^{-i\frac{\mathcal{H}_D}{\hbar}t}e^{-i\frac{\mathcal{H}_N}{\hbar}t}. \tag{4.44}$$

In the basis by Eq. (4.39), the Hamiltonian $\mathcal{H}_N$ is indeed diagonal, which means that as time increases, $\mathcal{H}_N$ only adds the phase in each basis vector but does not cause the transitions between the basis kets. The physical reason is that the Hamiltonian $\mathcal{H}_N$ describes the conservative numbers so that it is irrelevant to dynamics. Therefore, the dynamics is given by $\mathcal{H}_D$. We can use the interaction picture where $\mathcal{H}_0 = \mathcal{H}_D$ so that the dynamics is given by

$$i\hbar\frac{\partial}{\partial t}|\psi\rangle_I = \mathcal{H}_D|\psi\rangle_I. \tag{4.45}$$

The ket here is in the interaction picture. Because of being block-diagonalized, the dimension of $|\psi\rangle_I$ is effectively 2.

Example 1: Number State

Let the light in the number state $|n\rangle$. The two basis kets are

$$|n+1\rangle|E_v\rangle \equiv |i\rangle, \tag{4.46}$$
$$|n\rangle|E_c\rangle \equiv |f\rangle. \tag{4.47}$$

An arbitrary state in the interaction picture is

$$|\psi(t)\rangle = C_i(t)|i\rangle + C_f(t)|f\rangle. \tag{4.48}$$

Plugging this state in Eq. (4.45), we obtain

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} C_f \\ C_i \end{pmatrix} = \begin{pmatrix} -\frac{\hbar\Delta}{2} & \sqrt{n+1}\hbar\lambda \\ \sqrt{n+1}\hbar\lambda & \frac{\hbar\Delta}{2} \end{pmatrix}\begin{pmatrix} C_f \\ C_i \end{pmatrix}. \tag{4.49}$$

The eigenfrequencies are

$$\omega_\pm = \pm\sqrt{\frac{\Delta^2}{4} + (n+1)\lambda^2}. \tag{4.50}$$

and the eigenvectors (using the Bloch sphere representation) are

$$|\omega_+\rangle = \begin{pmatrix} \cos\frac{\theta}{2} \\ \sin\frac{\theta}{2} \end{pmatrix} e^{-i\omega_+ t} \tag{4.51}$$
$$|\omega_-\rangle = \begin{pmatrix} \sin\frac{\theta}{2} \\ -\cos\frac{\theta}{2} \end{pmatrix} e^{-i\omega_- t} \tag{4.52}$$

with

$$\theta = -\tan^{-1}\left(\frac{2\sqrt{n+1}\lambda}{\Delta}\right). \tag{4.53}$$

If the initial state is $C_i = 1$ and $C_f = 0$, the solution becomes

$$|\psi\rangle = \sin\frac{\theta}{2}|\omega_+\rangle - \cos\frac{\theta}{2}|\omega_-\rangle, \tag{4.54}$$
$$C_i(t) = \cos\omega_+ t + i\cos\theta\sin\omega_+ t, \tag{4.55}$$
$$C_f(t) = -i\sin\theta\sin\omega_+ t. \tag{4.56}$$

The population of the excited state $n_e = |C_f(t)|^2$ is

$$n_e = \sin^2\theta\sin^2\omega_+ t, \tag{4.57}$$
$$= \sin^2\theta\sin^2\sqrt{\frac{\Delta^2}{4} + (n+1)\lambda^2}t. \tag{4.58}$$

This is the Rabi oscillation between the states $|E_v\rangle|n+1\rangle$ and $|E_c\rangle|n\rangle$. Only when the detuning is zero, we have $\sin\theta = 1$ and the maximum excitation. The Rabi frequency is

$$\omega_+ = \sqrt{\frac{\Delta^2}{4} + (n+1)\lambda^2}. \tag{4.59}$$

The Rabi frequency depends on the number of photons. One novel case is $n = 0$ where the frequency is not zero but

$$\omega_+(n=0) = \sqrt{\frac{\Delta^2}{4} + \lambda^2}. \tag{4.60}$$

This means that there exists the Rabi oscillation even when there is no photon.[a] This is called the "vacuum Rabi oscillations".

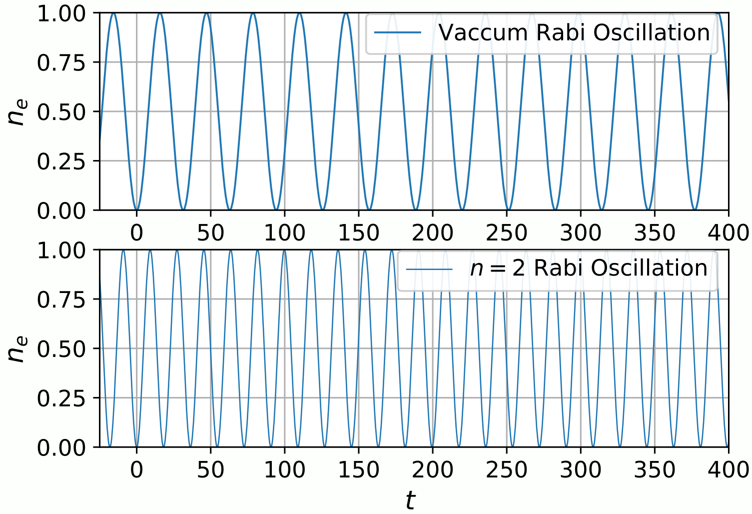

Figure 3: Rabi oscillations of the JC models for $n = 0$ and $n = 2$. The other parameters are $\Delta = 0$ and $\lambda = 0.1$

.

[a] Though, the vacuum energy is nonzero!

4.3 JC models with a Coherent State

Let us consider a more general situation where the photon state is

$$|\text{field}\rangle = \sum_{n=0}^{\infty} C_n |n\rangle, \tag{4.61}$$

and the two level system is

$$|\text{TLS}\rangle = C_c|E_c\rangle + C_v|E_v\rangle. \tag{4.62}$$

The total state is

$$|\psi\rangle = |\text{field}\rangle \otimes |\text{TLS}\rangle. \tag{4.63}$$

The solution is then (when $\Delta = 0$)

$$|\psi\rangle = \sum_n [C_c C_n \cos(\omega_{n+1} t) - i C_v C_{n+1} \sin(\omega_{n+1} t)] |n\rangle |E_c\rangle \tag{4.64}$$

$$+ \sum_n [C_v C_{n+1} \cos(\omega_{n+1} t) - i C_c C_n \sin(\omega_{n+1} t)] |n+1\rangle |E_v\rangle, \tag{4.65}$$

where

$$\omega_n = \omega_+(n). \tag{4.66}$$

Let the initial state be $C_c = 0$ and $C_v = 1$. The population of the excited state is

$$\begin{aligned} n_e = |C_c(t)|^2 &= \sum_n |C_{n+1}|^2 \sin^2 \omega_{n+1} t && (4.67) \\ &= \sum_n |C_{n+1}|^2 \left(\frac{1 - \cos 2\omega_{n+1} t}{2} \right) && (4.68) \\ &= \frac{1}{2} - \sum_n |C_{n+1}|^2 \left(\frac{\cos 2\omega_{n+1} t}{2} \right). && (4.69) \end{aligned}$$

In terms of n, we obtain

$$n_e = \frac{1}{2} - \sum_n |C_{n+1}|^2 \left(\frac{\cos 2\lambda\sqrt{n+1}\, t}{2} \right). \tag{4.70}$$

Figure 4 shows the populations in the cases of coherent states. Even with a coherent state, the population is not a simple harmonic oscillation as in the classical case. There are two new properties. First, the oscillation lasts for a time τ_c (the duration of the wave packet.) and **collapses**. It is shown that the time τ_c is in the limit $n \to \infty$,

$$\tau_c \simeq \frac{\sqrt{2}}{\lambda}. \tag{4.71}$$

After a rephasing time τ_{rp}, the oscillation comes back. This is called the **revival**. The time τ_{rp} is in the limit $n \to \infty$,

$$\tau_{\mathrm{rp}} \simeq \frac{4\pi|\alpha|}{\lambda}. \tag{4.72}$$

Two properties of the JC model are

- Collapsing
- Revival

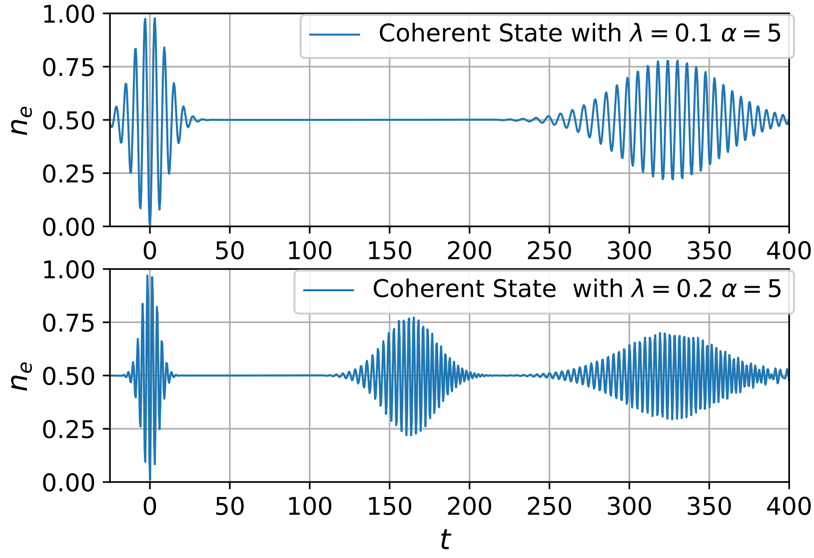

Figure 4: Rabi oscillations of the JC models for a coherent state. Collapsing and revival appear.

4.4 Dressed States

We focused on the dynamics of the JC model. Now, we discuss the eigenstates of the JC model. First, the photon energy in the vacuum is $E = n\hbar\omega$.[7] In a cavity, photons are coupled with the TLS. As a result, the photon energies are shifted. We can think that the combination of photons and the TLS leads to a new state called the "dressed state", or in the context of condensed matter physics, "polaritons". The JC Hamiltonian is block-diagonalized. Each block, denoted as $\mathcal{H}^{(n)}$, is a 2 by 2 matrix,

$$\mathcal{H}^{(n)} = n\hbar\omega + \begin{pmatrix} -\frac{\hbar\Delta}{2} & \sqrt{n+1}\hbar\lambda \\ \sqrt{n+1}\hbar\lambda & \frac{\hbar\Delta}{2} \end{pmatrix}, \tag{4.73}$$

where the matrix is spanned by the basis vectors from Eqs. (4.46) and (4.47). The eigenvalues are

$$E_{1n} = n\hbar\omega + \hbar\omega_n, \tag{4.74}$$
$$E_{2n} = n\hbar\omega - \hbar\omega_n, \tag{4.75}$$

where $\omega_n = \sqrt{\frac{\Delta^2}{4} + (n+1)\lambda^2}$ and the eigenvectors (using the Bloch sphere representation) are

$$|1n\rangle = \begin{pmatrix} \cos\frac{\theta}{2} \\ \sin\frac{\theta}{2} \end{pmatrix} e^{-i\omega_+ t} \tag{4.76}$$

$$|2n\rangle = \begin{pmatrix} \sin\frac{\theta}{2} \\ -\cos\frac{\theta}{2} \end{pmatrix} e^{-i\omega_- t} \tag{4.77}$$

[7]We drop $1/2\hbar\omega$.

with

$$\theta = -\tan^{-1}\left(\frac{2\sqrt{n+1}\lambda}{\Delta}\right). \tag{4.78}$$

The dressed photons are the eigenstates of the total system. Compared to photons in a vacuum, their frequencies shift and become non-degenerate. The splitting of dressed states is the origin of the Mollow triplet emissions.

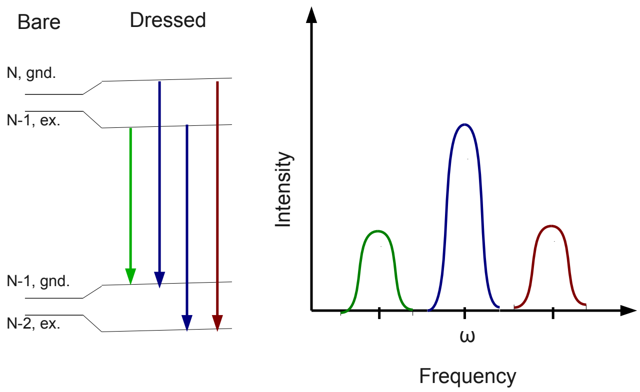

Figure 5: Mollow triplet emissions.

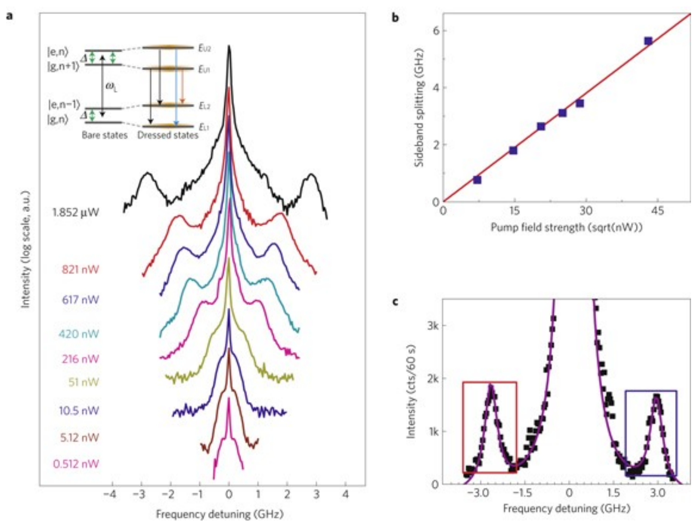

Figure 6: Experimental observation of the Mollow triplet emissions. From Nature Physics 5, 198202(2009)

References

[1] J. J. Sakurai, *Modern Quantum Mechanics*, 1994 and 2010