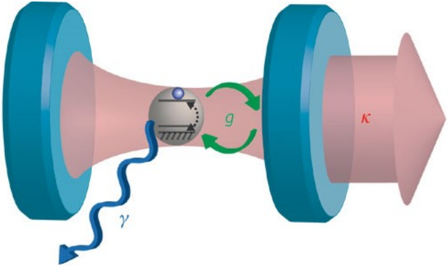
Quantum Light-Matter Interactions

QIC 895: Theory of Quantum Optics



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Outline

Background Review

Jaynes-Cummings Model

- Vacuum Rabi Oscillations, Collapse & Revival

Spontaneous Emission

- Wigner-Weisskopf Theory

Photoelectric Effect

- Semi-Classical vs. Quantized EM Field Predictions

Summary

Semi-classical L-M Interactions

Semi-classically, an electron in an EM field evolves as

$$i\hbar\frac{\partial\psi}{\partial t} = \Big[-\frac{\hbar^2}{2m}\Big(\nabla - \frac{ie}{\hbar}\vec{A}(\vec{r},t)\Big)^2 + \hat{V}(r)\Big]\psi$$

- $\vec{A}$ is the magnetic potential in the Coulomb (radiation) gauge
- V is the atomic binding potential

In the dipole approximation, we define $\varphi(\vec{r},t) = \exp(ie\vec{A}\cdot\vec{r}/\hbar)\psi(\vec{r},t)$ and find

$$i\hbar\frac{\partial\varphi}{\partial t} = \Big[\underbrace{\frac{\hat{p}^2}{2m} + \hat{V}(r)}_{H_A} \underbrace{-e\hat{\vec{r}}\cdot\vec{E}(\vec{r}_0,t)}_{H_{\text{int}}}\Big]\varphi$$

where $\vec{r}_0$ is the location of the nucleus.

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Quantum L-M Interactions

We'd like to treat the EM field quantum mechanically. This will require two changes:

1. $|\psi\rangle$ will need to represent the joint atom-field state
2. We'll need a Hamiltonian that acts on $\mathcal{H}_A \otimes \mathcal{H}_F$

Generically, we want $H = H_A + H_F + H_{\text{int}}$:

- We know $H_F = \sum_k \hbar\nu_k \left(a_k^\dagger a_k + \frac{1}{2}\right)$
- Let's use semi-classical $H_{\text{int}} = -e\vec{r} \cdot \vec{E}$ and promote $\vec{E} \to \hat{\vec{E}}$

For an atom at the origin:

$$\hat{\vec{E}}(t) = \sum_k \vec{\epsilon_k} \mathcal{E}_k \hat{a}_k e^{-i\nu_k t} + \text{h.c.}$$

In the Schrödinger picture we'll use $\hat{\vec{E}} := \hat{\vec{E}}(0)$ in place of $\hat{\vec{E}}(t)$.

Quantum L-M Interactions (2)

In analogy to our semi-classical analysis, we'll focus on an electron transition that is (nearly) resonant with some mode:

- Consider two electron energy eigenstates $|g\rangle$ and $|e\rangle$ with energy gap $\hbar\omega$
- In the $\{|g\rangle, |e\rangle\}$ subspace

$$H_A = \frac{\hbar\omega}{2}|e\rangle\langle e| - \frac{\hbar\omega}{2}|g\rangle\langle g| = \frac{\hbar\omega}{2}\sigma_z$$

  up to a multiple of I.

- As in the semi-classical picture, we take the dipole operator

$$e\hat{\vec{r}} \propto \underbrace{|e\rangle\langle g|}_{\sigma_+} + \underbrace{|g\rangle\langle e|}_{\sigma_-} = \sigma_x$$

Quantum L-M Interactions (3)

Putting it all together, we get the multi-mode Rabi Hamiltonian:

$$H = \underbrace{\sum_k \hbar\nu_k a_k^\dagger a_k}_{H_F} + \underbrace{\frac{\hbar\omega}{2}\sigma_z}_{H_A} + \underbrace{\hbar\sum_k g_k(\sigma_+ + \sigma_-)(a_k + a_k^\dagger)}_{H_{\text{int}} = -e\hat{\vec{r}}\cdot\hat{\vec{E}}}$$

From here we're going to make two more approximations:

1. Single-mode approximation
2. Rotating wave approximation

These will get us to the well-known Jaynes-Cummings model.

Interaction Picture Refresher

If the Schrödinger Hamiltonian has the form $H_0 + H_1$ then:

- States transform as
$$|\psi_I(t)\rangle = e^{iH_0 t/\hbar}|\psi_S(t)\rangle$$
- Operators transform as
$$A_I(t) = e^{iH_0 t/\hbar} A_S e^{-iH_0 t/\hbar}$$

N.b. Both states and operators have time dependence in this picture!

States evolve according to

$$i\hbar \frac{d}{dt}|\psi_I(t)\rangle = H_{1,I}(t)|\psi_I(t)\rangle$$

Rotating Wave Approximation

If we move into the interaction picture (rotating frame) with $H_0 = H_A + H_F$ we find:

$$H_{\text{int},I} = \hbar g \big(\underbrace{a\sigma_- e^{-i(\omega+\nu)t} + a^\dagger \sigma_+ e^{i(\omega+\nu)t}}_{\text{High Frequency}} + \underbrace{a\sigma_+ e^{i(\omega-\nu)t} + a^\dagger \sigma_- e^{-i(\omega-\nu)t}}_{\text{Low Frequency}} \big)$$

- Low freq. terms: $|g\rangle|n\rangle \to |e\rangle|n-1\rangle$ "conserve energy"
  $|e\rangle|n\rangle \to |g\rangle|n+1\rangle$
- High freq. terms: $|g\rangle|n\rangle \to |e\rangle|n+1\rangle$ no classical analogue
  $|e\rangle|n\rangle \to |g\rangle|n-1\rangle$

Rotating Wave Approximation (2)

The solution to the interaction picture SE will involve $\int_0^t H_{\text{int},I}(\tau) d\tau$.

This will give terms of the form $\int_0^t e^{i(\omega \pm \nu)\tau} d\tau$. In general:

$$\int_0^t e^{i\xi\tau} d\tau = -\frac{i}{\xi}\left(e^{i\xi t} - 1\right)$$

- Low freq. terms ($\xi = \omega - \nu$ small):

$$\int_0^t e^{i(\omega-\nu)\tau} d\tau = -\frac{i}{\xi}\Big(1 + i\xi t + O(\xi^2) - 1\Big) = t + O(\xi)$$

- High freq. terms ($\xi = \omega + \nu$ big):

$$\int_0^t e^{i(\omega+\nu)\tau} = -\underbrace{\frac{i}{\omega+\nu}}_{\text{small}}\underbrace{\left(e^{i(\omega+\nu)t} - 1\right)}_{\text{bounded}}$$

Jaynes-Cummings Model

Discarding the high frequency (counter-rotating) terms and returning to the Schrödinger picture we find

$$H = \hbar\nu a^\dagger a + \frac{\hbar\omega}{2}\sigma_z + \hbar g\big(a\sigma_+ + a^\dagger\sigma_-\big)$$

This Hamiltonian can be diagonalized exactly:

$$E_{\pm,n} = \hbar\nu\big(n + \frac{1}{2}\big) \pm \frac{\hbar}{2}\sqrt{\Delta^2 + 4g^2(n+1)}$$

$$|n, \pm\rangle = a_{n,\pm}|e\rangle|n\rangle + b_{n,\pm}|g\rangle|n+1\rangle$$

where $\Delta := \nu - \omega$ is the detuning.

Jaynes-Cummings Model (2)

Scully writes a general state in the uncoupled energy eigenbasis:

$$|\psi(t)\rangle = \sum_n \Big(\alpha_n(t)|g\rangle|n\rangle + \beta_n(t)|e\rangle|n\rangle\Big)$$

Plugging into the Schrödinger equation, we get

$$\dot{\beta}_n(t) = -ig\sqrt{n+1}e^{i\Delta t}\alpha_{n+1}(t)$$

$$\dot{\alpha}_{n+1}(t) = -ig\sqrt{n+1}e^{-i\Delta t}\beta_n(t)$$

The probability of finding n photons in the cavity is $P(n) = |\alpha_n(t)|^2 + |\beta_n(t)|^2$, which depends non-trivially on t (see animation).

Vacuum Rabi Oscillations

Consider $W(t) = -\langle \sigma_z \rangle$, the "atomic inversion".

- Semi-classically, when an EM wave is incident on an atom, $W(t)$ oscillates (Rabi oscillations)
  - This does not happen in the classical vacuum (when $\vec{E} = \vec{B} = \vec{0}$)
- With a quantized field $W(t) = \sum_n \left[|\beta_n(t)|^2 - |\alpha_n(t)|^2 \right]$

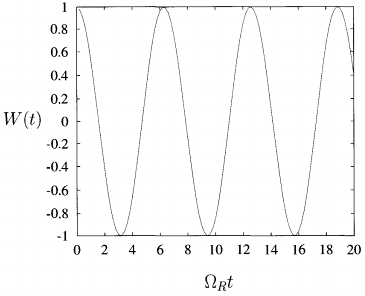
For an initial state $|e\rangle|0\rangle$:

$$W(t) = \frac{1}{\Delta^2 + 4g^2} \left[\Delta^2 + 4g^2 \cos\left(t\sqrt{\Delta^2 + 4g^2} \right) \right]$$

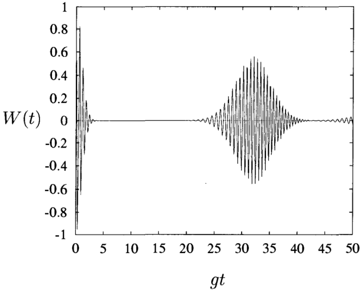
Key feature of quantum L-M interactions: Rabi oscillations occur even in the vacuum.

Collapse and Revival

Semi-classically, an EM wave induces Rabi oscillations at a single frequency



(a) Semi-classical Rabi oscillations



(b) Fully quantum Rabi oscillations

A fully quantum description predicts that the oscillations will vanish and then re-appear.

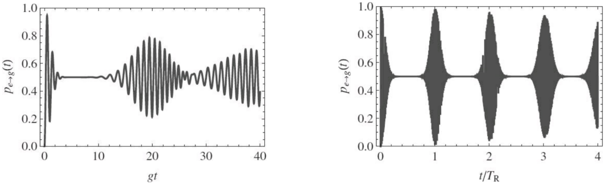
Collapse and Revival (2)

If the atom and field mode are initially uncorrelated with $\Delta = 0$:

$$W(t) = \sum_{n=0}^{\infty} |\alpha_n|^2 \cos\left(gt\sqrt{n+1}\right)$$

where $\sum_n \alpha_n |n\rangle$ is the initial field state.

- Notice that a single mode can produce oscillations at many frequencies
- These different contributions interfere to create complex collapse/revival behaviour (observed experimentally)



Collapse and Revival (3)

Scully argues that if n were continuous there would be collapse, but no revival.

Let's estimate the timescale of revival (with $\Delta = 0$):

- Each n contributes with frequency $\Omega_n := g\sqrt{n+1}$
- For a mode with photon number sharply peaked around $\bar{n}$, the frequencies near $\Omega_{\bar{n}} = g\sqrt{\bar{n}+1}$ will dominate
- How quickly do $\Omega_{\bar{n}}$ and $\Omega_{\bar{n}-1}$ move in and out of phase?

$$\big(\Omega_{\bar{n}} - \Omega_{\bar{n}-1}\big)t_r = 2\pi m \qquad m \in \mathbb{N}$$

$$\implies t_r \sim \frac{2\pi m\sqrt{\bar{n}}}{g}$$

- Revivals keep occurring periodically.

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Spontaneous Emission

- Collapse/revival observed experimentally, but only in certain settings (e.g., reflective cavities)
- A more common phenomenon is for an atom to relax (irreversibly) by emitting a photon
- To analyse this scenario, we'll need to consider multiple modes again:

$$H = \underbrace{\sum_k \hbar \nu_k a_k^\dagger a_k + \frac{\hbar\omega}{2}\sigma_z}_{H_0} + \underbrace{\hbar \sum_k g_k (\sigma_+ + \sigma_-)(a_k + a_k^\dagger)}_{H_{\text{int}}}$$

- In the interaction picture, under the RWA:

$$H_{\text{int},I} = \hbar \sum_k g_k \left(a_k \sigma_+ e^{i(\omega - \nu_k)t} + a_k^\dagger \sigma_- e^{-i(\omega-\nu_k)t} \right)$$

Spontaneous Emission (2)

If, initially, the atom is excited and the field is in the vacuum state, the general solution has the form

$$|\psi(t)\rangle = \alpha(t)|e\rangle|0\rangle + \sum_k \beta_k(t)|g\rangle|1_k\rangle$$

under the RWA. Here $|0\rangle$ is the total field vacuum, and $|1_k\rangle = a_k^\dagger|0\rangle$.

The Schrödinger equation yields

$$\dot{\alpha}(t) = -i\sum_k g_k e^{i(\omega-\nu_k)t}\beta_k(t) \tag{1}$$

$$\dot{\beta}_k(t) = -ig_k e^{-i(\omega-\nu_k)t}\alpha(t) \tag{2}$$

Substituting (2) into (1), we find

$$\dot{\alpha}(t) = -\sum_k |g_k|^2 \int_0^t dt'\, e^{-i(\omega-\nu_k)(t'-t)}\alpha(t')$$

Spontaneous Emission (3)

Approximation 1 The modes form a continuum:

$$\sum_k \to \int d^3k\, \rho(k) \qquad \text{where} \qquad \rho(k)d^3k = 2\left(\frac{L}{2\pi}\right)^3 k^2 dk\, d\phi \sin\theta d\theta$$

Substituting in the value of g_k and integrating over (ϕ, θ) we get

$$\dot{\alpha}(t) \propto \int_0^\infty d\nu_k\, \nu_k^3 \int_0^t dt'\, e^{-i(\omega-\nu_k)(t'-t)} \alpha(t')$$

Approximation 2 The integral is only important when $\omega \approx \nu_k$ (resonance), so we can replace ν_k^3 with ω^3, and integrate over $\nu_k \in \mathbb{R}$.

Spontaneous Emission (4)

Changing the order of integration, we have

$$
\begin{aligned}
\dot{\alpha}(t) &\propto -\omega^3 \int_0^t dt'\, \alpha(t') \int_{-\infty}^{\infty} d\nu_k\, e^{-i(\omega-\nu_k)(t'-t)} \\
&= -\omega^3 \int_0^t dt'\, \alpha(t')\; 2\pi\delta(t-t') \\
&= -2\pi\omega^3\alpha(t)
\end{aligned}
$$

In other words, $\frac{d\alpha}{dt} = -\frac{\Gamma}{2}\alpha(t)$ for some constant Γ, or

$$
\left|\langle e|a\rangle\right|^2 = \exp(-\Gamma t)
$$

This is a good description of energy relaxation, where $\Gamma = 1/T_1$. It is called the Weisskopf-Wigner approximation.

Spontaneous Emission (5)

What about the "emitted photon"? We can plug in $\alpha(t)$ to our ODE for $\beta_k(t)$ to get

$$\beta_k(t) = -i \int_0^t dt' g_k e^{-i(\omega - \nu_k)t' - \Gamma t'/2}$$

The frequency spectrum of the resulting radiation is given by

$$P(\nu_k) = \rho(\nu_k) \sum_{\lambda=1,2} \int d\Omega \, |\beta_k(t)|^2$$

Once the transients have vanished, we find

$$\lim_{t\to\infty} P(\nu_k) \propto \frac{1}{\Gamma^2/4 + (\omega - \nu_k)^2}$$

i.e., a Lorentzian centred around ω of width Γ (see plot).

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Fermi's Golden Rule Refresher

System evolves according to free Hamiltonian H_0, subject to perturbation $\lambda H'(t)$.

We'll be interested in the case where

$$H'(t) = Ve^{i\Omega t} + V^\dagger e^{-i\Omega t}$$

What is the probability of $\lambda H'$ inducing a transition between unperturbed energy eigenstates $|i\rangle$ and $|f\rangle$?

$$\begin{aligned} P_{|i\rangle\to|f\rangle} &= \left|\langle f|\, \mathcal{T} e^{-i\lambda \int_0^t d\tau\, H_I'(\tau)} |i\rangle\right|^2 \\ &= \left|\langle f|i\rangle\right|^2 + \lambda^2 \left| \int_0^t d\tau \langle f| e^{iH_0\tau} H'(\tau) e^{-iH_0\tau} |i\rangle\right|^2 + O(\lambda^3) \end{aligned}$$

Fermi's Golden Rule Refresher (2)

For long times t, the transitions probability $|i\rangle \to |f\rangle$ for a perturbation of the form

$$H'(t) = Ve^{i\Omega t} + V^\dagger e^{-i\Omega t}$$

is approximately

$$P_{|i\rangle\to|f\rangle} = \begin{cases} \lambda^2 \frac{2\pi t}{\hbar} \left|\langle f|V|i\rangle\right|^2 \rho(E_i + \hbar\Omega) & \text{(absorption)} \\ \lambda^2 \frac{2\pi t}{\hbar} \left|\langle f|V^\dagger|i\rangle\right|^2 \rho(E_i - \hbar\Omega) & \text{(emission)} \end{cases}$$

where $\rho(E)$ is the density of final states.

- Notice that both cases grow unbounded with t!
- Instead of P we're going to work with $\dot{P}$, which *is* bounded.

Photoelectric Effect (Semi-Classically)

Consider a hydrogenic atom with an electron initially in the $|n\rangle$ energy eigenstate. Suppose it is subject to a classical EM wave

$$\vec{A}(\vec{r},t) = A_0\vec{\epsilon}\left(e^{i(\vec{k}\cdot\vec{r}-\omega t)} + e^{-i(\vec{k}\cdot\vec{r}-\omega t)}\right)$$

which ejects it from the atom to a free state $|\vec{p}\rangle$.

The electron's evolution is generated by

$$H = \frac{\hat{\vec{p}}^2}{2m} - \frac{e}{m}\hat{\vec{p}}\cdot\vec{A} + \frac{e^2}{2m}\vec{A}^2 + e\hat{\phi}(\vec{r})$$

The $\vec{A}^2$ term is suppressed by the $\frac{e^2}{2m}$ factor and will not contribute to our first order transition rate. Therefore, we'll take the perturbation to be

$$\lambda H'(t) = -\frac{e}{m}\hat{\vec{p}}\cdot\vec{A} = \underbrace{-\frac{eA_0}{m}}_{\lambda}\underbrace{\left(\hat{\vec{p}}\cdot\vec{\epsilon}\right)e^{i\vec{k}\cdot\vec{r}}}_{V^\dagger}e^{-i\omega t} + \text{h.c.}$$

Photoelectric Effect (Semi-Classically, 2)

Let's look at the rate of transitions for photoabsorption $|n\rangle \to |\vec{p}\rangle$:

$$\dot{P}_{|n\rangle \to |\vec{p}\rangle} = \lambda^2 \frac{2\pi}{\hbar} \left| \langle \vec{p} | (\hat{\vec{p}} \cdot \vec{\epsilon}) e^{i\vec{k}\cdot\vec{r}} | n \rangle \right|^2 \rho(E_n + \hbar\omega)$$

For a free electron in a box of side length L, the density of states is

$$\rho(E) = \left(\frac{L}{2\pi\hbar} \right)^3 m \, ||\vec{p}|| \, d\Omega$$

where $E = \frac{\vec{p}^{\,2}}{2m}$.

- From here we could compute various cross-sections, average over final electron states etc.
- Instead though, let's set up an equivalent scenario with quantized radiation.

Photoelectric Effect (Quantized EM field)

The electron-field system evolves according to

$$H = \frac{1}{2m}\left(\hat{\vec{p}} - \frac{e\hat{\vec{A}}}{c}\right)^2 + \sum_{\vec{k},\vec{\epsilon}} \hbar\omega(a^\dagger_{\vec{k},\vec{\epsilon}} a_{\vec{k},\vec{\epsilon}} + \frac{1}{2}) + e\hat{\phi}(\vec{r})$$

where

$$\hat{\vec{A}} = \sum_{\vec{k},\vec{\epsilon}} \sqrt{\frac{2\pi\hbar}{L^3\omega_k}}\left[a_{\vec{k},\vec{\epsilon}}e^{-i\vec{k}\cdot\vec{r}} + \text{h.c.}\right]\vec{\epsilon}_{\vec{k}}$$

- Consider the initial state $|n\rangle|1_{\vec{k},\vec{\epsilon}}\rangle := |n\rangle\, a_{\vec{k},\vec{\epsilon}}|0\rangle$ and the final state $|\vec{p}\rangle|0\rangle$.
- As before, we'll omit the A^2 term as it won't contribute to our first-order transition rate.

Photoelectric Effect (Quantized EM field, 2)

After some work, Fermi's Golden Rule gives

$$\dot{P}_{|i\rangle \to |f\rangle} = \lambda^2 \frac{2\pi}{\hbar} \left| \langle f | (\hat{\vec{p}} \cdot \vec{\epsilon}) e^{i\vec{k}\cdot\vec{r}} | i \rangle \right|^2 \rho(E_f)$$

with the same λ as before.

The density of final states $\rho(E)$ is the same as in the semi-classical case.

- $\dot{P}_{|i\rangle \to |f\rangle}$ here is the same as in the semi-classical analysis.
- To first order in λ, semi-classical and fully quantum descriptions give the same transition rates, cross sections etc. for the photoelectric effect!

Summary

Present in semi-classical description:

- Rabi oscillations
- Photoelectric effect (!)

Require quantized EM field:

- *Vacuum* Rabi oscillations
- Periodic collapse and revival of Rabi oscillations
- Spontaneous emission