

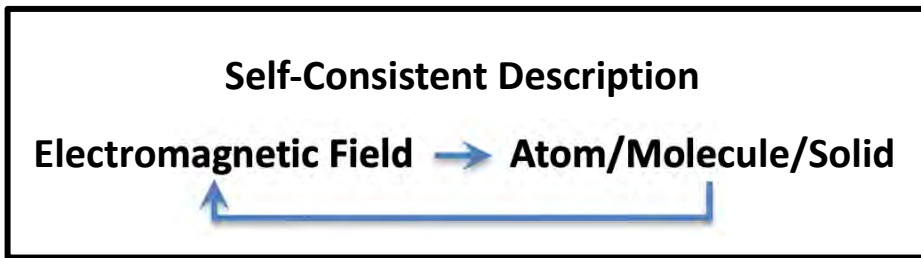
Quantum Theory of Light-Matter Interaction

Completed:

- Fully classical description of fields & Atoms

Next Step:

- Semiclassical description
 - Classical field
 - Quantum atoms



Needed: Quantum theory of atomic response
analogous to classical $\vec{p} = \alpha(\omega) \vec{E}$

Note: In QM the dipole is an Observable
Observable = Hermitian operator \hat{p}
Classical Field = C-valued vector \vec{E}

Cannot plug into Wave Eq. for classical field!

Wave Equation w/classical field & atoms

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \vec{E} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{p}}{\partial t^2}, \quad \vec{p} = N \vec{p}$$

How do we solve the mismatch?

Repeated measurements of $\vec{p}(t)$



Quantum fluctuations $\vec{p}(t) = \langle \vec{p}(t) \rangle + \Delta \vec{p}(t)$

where $\langle \vec{p}(t) \rangle = \langle \psi(t) | \hat{p} | \psi(t) \rangle$ mean
 $\Delta \hat{p}(t)$ fluctuations

Note: Given $|\psi(t=0)\rangle$ and \vec{E} , the mean $\langle \vec{p}(t) \rangle$
follows from the Schrödinger Eq.,
radiates coherently like classical $\vec{p}(t)$

$\langle \vec{p}(t) \rangle$ is a Real-valued vector (more later)
 \rightarrow we can plug it into the Wave Eq.

Quantum Theory of Light-Matter Interaction

Wave Equation w/classical field & atoms

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- Note:**
- The Equations look very similar
 - Polarizability, index of refraction, etc will be *very different* in some regimes
 - Notably, the model is no longer linear in \vec{E} and will lead to phenomena like saturation and wave mixing
 - $\Delta \hat{p}(t)$ represents quantum fluctuations driven by the empty modes of the EM field, a process also responsible for spontaneous decay.

Note: Do not identify $\langle \vec{p} \rangle$ and $\Delta \vec{p}$ with Stimulated and spontaneous emission. Those labels are not meaningful here.

Quantum Theory of Light-Matter Interaction

Wave Eq. w/classical field & quantum atoms

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \vec{E} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2}{\partial t^2} \vec{P}, \quad \vec{P} = N \langle \hat{\vec{p}} \rangle$$

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Atom-field interaction

Hamiltonian:

$$H = H_a + V_{\text{ext}}(\vec{R}, t)$$

- H_a : time-independent atomic Hamiltonian
- V_{ext} : time-dependent driving term, non necessarily a perturbation

Question: Time evolution of the atomic system?
Is there a steady state?

Schrödinger Eq.:

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = H |\psi(t)\rangle$$

Expand in basis $\{|\varphi_n\rangle\}$ of eigenstates of H_a

$$|\psi(t)\rangle = \sum_n a_n(t) |\varphi_n\rangle, \quad H_a |\varphi_n\rangle = E_n |\varphi_n\rangle$$

Quantum Theory of Light-Matter Interaction

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Plug into S. E. \rightarrow

$$i\hbar \sum_n \dot{a}_n(t) |\varphi_n\rangle = \sum_n a_n(t) [E_n + V_{\text{ext}}] |\varphi_n\rangle$$

Take scalar product w/ $|\varphi_m\rangle$ on both sides \rightarrow

$$i\hbar \sum_n \dot{a}_n(t) \langle \varphi_m | \varphi_n \rangle = \sum_n a_n(t) [E_n \langle \varphi_m | \varphi_n \rangle] + \overbrace{\langle \varphi_m | V_{\text{ext}} | \varphi_n \rangle}^{V_{mn}}$$

On vector-matrix form this can be written

$$i\hbar \dot{\underline{a}} = \underline{H}_a \underline{a} + \underline{V} \underline{a}$$

\leftarrow S.E. in $\{|\varphi_n\rangle\}$ rep.
Still exact!

Atom-Light Interaction: 2-Level Approximation

General observation:

- Atoms and molecules often behave as if they have a single, dominant transition frequency
- We expect this when the freq. of the driving is resonant with one transition $| \phi_n \rangle \rightarrow | \phi_m \rangle$ and far off resonance with all others.

Interaction

$$V_{ext} = -\hat{\vec{\mu}} \cdot \vec{E}(t)$$

State space

$$\text{Dim}(\mathcal{E}) = 2, \{ |1\rangle, |2\rangle \}$$

$$\begin{array}{c} \xrightarrow{|2\rangle} \\ \hline \hbar\omega_{21} \\ \xleftarrow{|1\rangle} \end{array}$$

State vector

$$|\psi(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle$$

Schröd. eq.

$$\begin{aligned} i\hbar\dot{a}_1 &= E_1 a_1 + V_{11} a_1 + V_{12} a_2 \\ i\hbar\dot{a}_2 &= E_2 a_2 + V_{21} a_1 + V_{22} a_2 \end{aligned}$$

Interaction

$$\begin{aligned} V_{12}(t) &= -\vec{\mu}_{12} \cdot \frac{1}{2}(\dot{\vec{E}}_0 e^{-i\omega t} + \text{c.c.}) \\ V_{21}(t) &= -\vec{\mu}_{21} \cdot \frac{1}{2}(\dot{\vec{E}}_0 e^{-i\omega t} + \text{c.c.}) \end{aligned}$$

Parity selection rule

Definition: $\vec{r} \rightarrow -\vec{r}$ is a reflection through the origin

Atomic Hamiltonian $H \propto \frac{1}{r} \Rightarrow H(\vec{r}) = H(-\vec{r})$

\Rightarrow Eigenstates $\psi(\vec{r}) = \pm \psi(-\vec{r}) = \psi(-[-\vec{r}])$

"+" for even parity two reflections equals the identity
"-" for odd parity

The dipole $\hat{\vec{\mu}}$ is a vector operator \Rightarrow

transforms like a vector when $\vec{r} \rightarrow -\vec{r}$

Thus $\hat{\vec{\mu}}(\vec{r}) = e^{\vec{r}} = -\hat{\vec{\mu}}(-\vec{r})$ and

$$\hat{\mu}_{nm} = \int d^3r \psi_n^*(\vec{r}) \hat{\vec{\mu}} \psi_m(\vec{r}) \neq 0 \quad \text{only when}$$

ψ_n and ψ_m have opposite parity

Parity rule:

No dipole moment in energy eigenstate!

$$\begin{aligned} \hat{\mu}_{12} &= \langle 1 | \hat{\vec{\mu}} | 2 \rangle, \quad \hat{\mu}_{21} = \hat{\mu}_{12}^* \\ \hat{\mu}_{11} &= \hat{\mu}_{22} = 0 \Rightarrow V_{11} = V_{22} = 0 \end{aligned}$$

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We define

$$\begin{aligned} \omega_{21} &= \frac{E_2 - E_1}{\hbar}, & E_1 &= 0 \\ \chi_{12} &= \vec{p}_{12} \cdot \hat{\vec{E}} E_0 / \hbar \\ \chi_{21} &= \vec{p}_{21} \cdot \hat{\vec{E}} E_0 / \hbar \end{aligned} \left. \vphantom{\begin{aligned} \omega_{21} \\ \chi_{12} \\ \chi_{21} \end{aligned}} \right\} \begin{array}{l} \text{interaction energy is } \hbar \chi \\ \chi \text{ Rabi frequency} \end{array}$$

Note: $\left\{ \begin{array}{l} \chi_{12}^* = \vec{p}_{21} \cdot (\hat{\vec{E}} E_0 / \hbar)^* \neq \chi_{21} \\ \chi_{21}^* = \vec{p}_{12} \cdot (\hat{\vec{E}} E_0 / \hbar)^* \neq \chi_{12} \end{array} \right.$

Plug into $i\hbar \dot{\underline{a}} = \underline{H}_a \underline{a} + \underline{V} \underline{a}$ (S. E.) to get

$$\begin{aligned} i\dot{a}_1 &= -\frac{1}{2} (\chi_{12} e^{-i\omega t} + \chi_{21}^* e^{i\omega t}) a_2 \\ i\dot{a}_2 &= \omega_{21} a_2 - \frac{1}{2} (\chi_{21} e^{-i\omega t} + \chi_{12}^* e^{i\omega t}) a_1 \end{aligned}$$

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$$\omega_{21} = \frac{E_2 - E_1}{\hbar}, \quad E_1 = 0$$

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Switch to rotating frame (slow variables)

$$C_1(t) = a_1(t), \quad C_2(t) = a_2(t) e^{i\omega t}$$



$$\begin{aligned} i\dot{C}_1(t) &= -\frac{1}{2} (\chi_{12} e^{-i2\omega t} + \chi_{21}^*) C_2(t) \\ i\dot{C}_2(t) &= (\omega_{21} - \omega) C_2(t) - \frac{1}{2} (\chi_{21} + \chi_{12}^* e^{i2\omega t}) C_1(t) \end{aligned}$$

Rotating Wave Approximation (RWA)

Very important, equivalent to resonant approximation

Terms $\propto e^{\pm i2\omega t}$ average to zero on time scale for variations in C_1, C_2



$$\begin{aligned} i\dot{C}_1(t) &= -\frac{1}{2} \chi_{21}^* C_2(t) \\ i\dot{C}_2(t) &= \Delta C_2(t) - \frac{1}{2} \chi_{21} C_1(t) \end{aligned} \quad \begin{array}{l} \Delta = \omega_{21} - \omega \\ \text{(detuning)} \end{array}$$

Exactly Solvable !

Atom-Light Interaction: 2-Level Approximation

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Exactly Solvable !

To simplify, make a global phase choice such that

$$\chi_{21} = \vec{p}_{21} \cdot \hat{\epsilon} E_0 / \hbar = \chi \quad \text{is real (not required)}$$



Simplest 2-level equations

$$i\dot{C}_1(t) = -\frac{1}{2} \chi C_2(t)$$

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Rabi Solutions for $C_1(0) = 1, C_2(0) = 0$

$$C_1(t) = \left(\cos \frac{\Omega t}{2} + i \frac{\Delta}{\Omega} \sin \frac{\Omega t}{2} \right) e^{-i\Delta t/2}$$

$$C_2(t) = \left(i \frac{\chi}{\Omega} \sin \frac{\Omega t}{2} \right) e^{-i\Delta t/2}$$

χ : Rabi freq. Δ : Detuning

$\Omega \equiv \sqrt{\chi^2 + \Delta^2}$: Generalized Rabi freq.

Atom-Light Interaction: 2-Level Approximation

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Note: The Rabi Solutions give us the entire state, in the lab (a's) and rotating (c's) frames

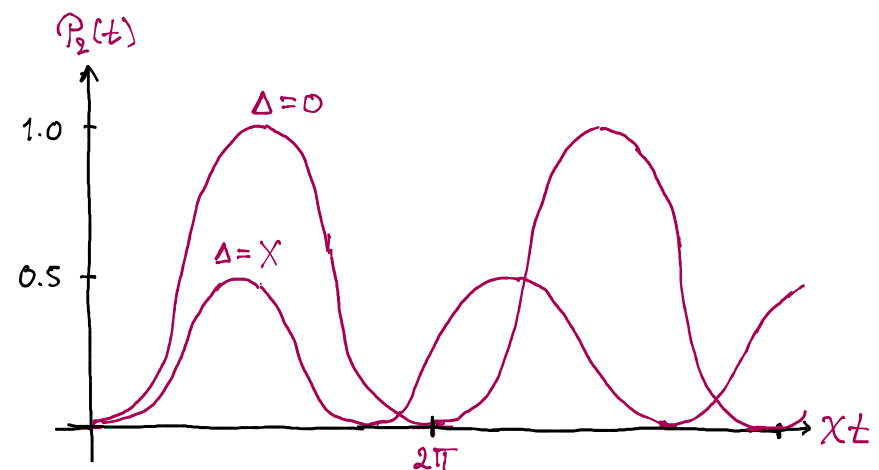


We have maximum information about the system and can make any predictions allowed by QM

Probabilities of finding the atom in $|1\rangle, |2\rangle$:

$$P_1(t) = |C_1(t)|^2 = \frac{1}{2} \left(1 + \frac{\Delta^2}{\Omega^2} \right) + \frac{1}{2} \frac{\chi^2}{\Omega^2} \cos \Omega t$$

$$P_2(t) = |C_2(t)|^2 = \frac{1}{2} \frac{\chi^2}{\Omega^2} (1 - \cos \Omega t)$$



Atom-Light Interaction: 2-Level Approximation

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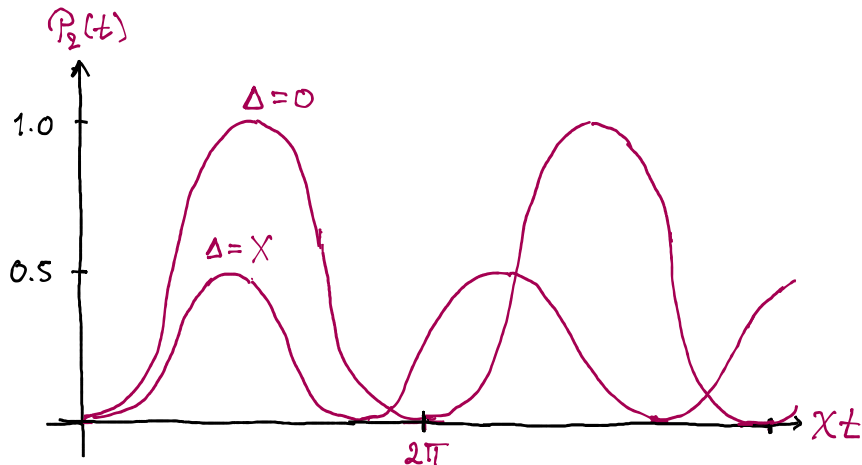


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Note: All 2-level systems are isomorphic

- Equivalent Observables
- Equivalent Phenomena
- The Rabi problem was first solved in ESR and NMR, for spin-1/2 particles with a magnetic moment $\vec{\mu}$ driven by a magnetic field \vec{B} with interaction $H = \vec{\mu} \cdot \vec{B}$
- 2-level systems are now often called qubits

Dressed States

The 2-level eqs. in the RWA look like a S.E. with

$$H_{RWA} = \hbar \begin{pmatrix} 0 & \frac{1}{2}\chi \\ -\frac{1}{2}\chi & \Delta \end{pmatrix}$$

The eigenstates of H_{RWA} are called Dressed States

The DS are stationary only in the Rotating Frame. In the Lab Frame (Schrödinger Picture) they are periodic, oscillating w/frequency ω

Atom-Light Interaction: 2-Level Approximation

We pick linear polarization so $\vec{\hat{\epsilon}} \cdot \vec{E}_0$ is real-valued

and $V_{12} = V_{21} = V \Rightarrow$ The eqs for the a 's are

$$\hbar \dot{a}_1^* = i(E_1 a_1^* + V a_2^*)$$

$$\hbar \dot{a}_2 = -i(E_2 a_2 + V a_1)$$

With this we have

$$\begin{aligned} \frac{d}{dt} a_1^* a_2 &= (\dot{a}_1^* a_2 + a_1^* \dot{a}_2) \\ &= -i \underbrace{\frac{E_2 - E_1}{\hbar}}_{\omega_0} a_1^* a_2 - i \frac{V}{\hbar} (|a_1|^2 - |a_2|^2) \end{aligned}$$

Differentiating again gives us

$$\begin{aligned} \frac{d^2}{dt^2} (a_1^* a_2) &= -\omega_0^2 a_1^* a_2 - \frac{\omega_0 V}{\hbar} (|a_1|^2 - |a_2|^2) \\ &\quad - i \hbar \frac{d}{dt} \left[\frac{V}{\hbar} (|a_1|^2 - |a_2|^2) \right] \end{aligned}$$

Looking at the eq. for $\langle \hat{n} \rangle$ suggests we should add the complex conjugate and multiply w $\vec{\hat{n}}_{12}$

This gives us

$$\begin{aligned} \left(\frac{d^2}{dt^2} + \omega_0^2 \right) \langle \hat{n} \rangle &= \frac{2\omega_0 \vec{\hat{n}}_{12} V}{\hbar} (|a_1|^2 - |a_2|^2) \\ &= \frac{2\omega_0}{\hbar} \vec{\hat{n}}_{12} (\vec{\hat{n}}_{12} \cdot \vec{E}) (|a_1|^2 - |a_2|^2) \\ \vec{\hat{n}}_{12} &= \langle 1 | \vec{\hat{n}} | 2 \rangle : \text{dipole matrix element} \end{aligned}$$

To wrap up, we need to know a bit about real, multilevel atoms. (We will revisit this soon)

Pick linear polarization so $\vec{\hat{\epsilon}}$ is real-valued.

Pick quantization axis along $\vec{\hat{\epsilon}} \Rightarrow \vec{\hat{n}}_{12} = \hat{\epsilon}_{12} \vec{\hat{\epsilon}}$



$$\left(\frac{d^2}{dt^2} + \omega_0^2 \right) \langle \hat{n} \rangle = \frac{2\omega_0 \hat{\epsilon}_{12}^2}{\hbar} \vec{E} (|a_1|^2 - |a_2|^2)$$

Atom-Light Interaction: 2-Level Approximation

This gives us

$$\begin{aligned} \left(\frac{d^2}{dt^2} + \omega_0^2 \right) \langle \hat{n} \rangle &= \frac{2\omega_0 \vec{r}_{12} V}{\hbar} (|a_1|^2 - |a_2|^2) \\ &= \frac{2\omega_0}{\hbar} \vec{r}_{12} (\vec{r}_{12} \cdot \vec{E}) (|a_1|^2 - |a_2|^2) \\ \vec{r}_{12} &= \langle 1 | \hat{r} | 2 \rangle : \text{dipole matrix element} \end{aligned}$$

To wrap up, we need to know a bit about real, multilevel atoms. (We will revisit this soon)

Pick linear polarization so \vec{E} is real-valued.
Pick quantization axis along $\vec{E} \Rightarrow \vec{r}_{12} = r_{12} \vec{E}$

$$\left(\frac{d^2}{dt^2} + \omega_0^2 \right) \langle \hat{n} \rangle = \frac{2\omega_0 r_{12}^2}{\hbar} \vec{E} (|a_1|^2 - |a_2|^2)$$

Compare to Classical Equation of Motion

$$\left(\frac{d^2}{dt^2} + \omega_0^2 \right) \vec{r} = \frac{e}{m} \vec{E}$$

The two eqs. have the same form if

$$\begin{aligned} |a_1|^2 &\sim 1 \\ |a_2|^2 &\sim 0 \end{aligned}$$

This is the case for $\Delta \gg \chi$
Or when $\chi \ll \Gamma$ } Limit of weak Excitation !

Decay rate of $|2\rangle$

Oscillator Strength

$$f = \frac{2m\omega_0}{\hbar e} r_{12}^2$$

$$\left(\frac{d^2}{dt^2} + \omega_0^2 \right) \langle \hat{n} \rangle = \frac{e}{m} f \vec{E}$$

Like the classical equation,
but with modified polarizability !