Quantum Theory of Light-Matter Interaction

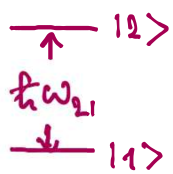
Atom-Light Interaction: 2-Level Approximation

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 $|\varphi_n\rangle \to |\varphi_m\rangle$ and far off resonance with all others.

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

State space $\mathcal{D}im(\mathcal{E}) = 2, \ \{|1\rangle, |2\rangle\}$



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

Schröd. eq.

$$i\hbar \dot{a}_1 = E_1 a_1 + V_{11} a_1 + V_{12} a_2$$

$$i\hbar \dot{a}_2 = E_2 a_2 + V_{21} a_1 + V_{22} a_2$$

Interaction

$$V_{12}(t) = -\vec{\mu}_{12} \cdot \tfrac{1}{2}(\hat{\varepsilon} E_0 e^{-i\omega t} + c.c.)$$

$$V_{21}(t) = -\vec{\mu}_{21} \cdot \tfrac{1}{2}(\hat{\varepsilon} E_0 e^{-i\omega t} + c.c.)$$

Parity selection rule

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

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

$\Rightarrow$ **Eigenstates** $\varphi(\vec{r}) = \pm\, \varphi(-\vec{r}) = \varphi(-[-\vec{r}])$

"+" for even parity
"-" for odd parity

two reflections equals the identity

The dipole $\hat{\vec{\mu}}$ is a vector operator $\Rightarrow$ transforms like a vector when $\vec{r} \to -\vec{r}$

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

$\vec{\mu}_{nm} = \int d^3r\, \varphi_n^*(\vec{r})\, \vec{\mu}\, \varphi_m(\vec{r}) \neq 0$ only when

φ_n and φ_m have <u>opposite parity</u>

Parity rule: No dipole moment in energy eigenstate!

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

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We <u>define</u>

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

$$\left.\begin{array}{l} \chi_{12} = \vec{\mu}_{12}\cdot\hat{\varepsilon}E_0/\hbar \\ \chi_{21} = \vec{\mu}_{21}\cdot\hat{\varepsilon}E_0/\hbar \end{array}\right\}$$

interaction energy is $\hbar\chi$

χ **Rabi frequency**

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

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

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

Atom-Light Interaction: 2-Level Approximation

We define

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

$$\chi_{12} = \vec{\mu}_{12} \cdot \hat{\varepsilon} E_0 / \hbar$$

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

$$i\dot{a}_1 = -\frac{1}{2}\left(\chi_{12} e^{-i\omega t} + \chi_{21}^* e^{i\omega t}\right) a_2$$

$$i\dot{a}_2 = \omega_{21} a_2 - \frac{1}{2}\left(\chi_{21} e^{-i\omega t} + \chi_{12}^* e^{i\omega t}\right) a_1$$

Switch to rotating frame (slow variables)

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

$$i\dot{C}_1(t) = -\frac{1}{2}\left(\chi_{12} e^{-i2\omega t} + \chi_{21}^*\right) C_2(t)$$

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

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

$$i\dot{C}_1(t) = -\frac{1}{2} \chi_{21}^* C_2(t)$$

$$i\dot{C}_2(t) = \Delta C_2(t) - \frac{1}{2} \chi_{21} C_1(t)$$

$\Delta = \omega_{21} - \omega$ (detuning)

Exactly Solvable !

Atom-Light Interaction: 2-Level Approximation

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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

$$i\dot{c}_1(t) = -\frac{1}{2} \chi_{21}^* c_2(t)$$

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Exactly Solvable !

To simplify, make a global phase choice such that

$\chi_{21} = \vec{\mu}_{21} \cdot \hat{\varepsilon} E_0 / \hbar = \chi$ **is real (not required)**

Simplest 2-level equations

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

$$i\dot{C}_2(t) = \Delta C_2(t) - \frac{1}{2} \chi C_1(t)$$

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.** $\quad \Delta$: **Detuning**

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

Atom-Light Interaction: 2-Level Approximation

To simplify, make a global phase choice such that

$\chi_{21} = \vec{\mu}_{21} \cdot \hat{\varepsilon} E_0 / \hbar = \chi$ is real (not required)

Simplest 2-level equations

$$\boxed{\begin{aligned} i\dot{c}_1(t) &= -\frac{1}{2}\chi c_2(t) \\ i\dot{c}_2(t) &= \Delta c_2(t) - \frac{1}{2}\chi c_1(t) \end{aligned}}$$

<u>Rabi Solutions</u> for $c_1(0) = 1$, $c_2(0) = 0$

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χ: Rabi freq. Δ: Detuning

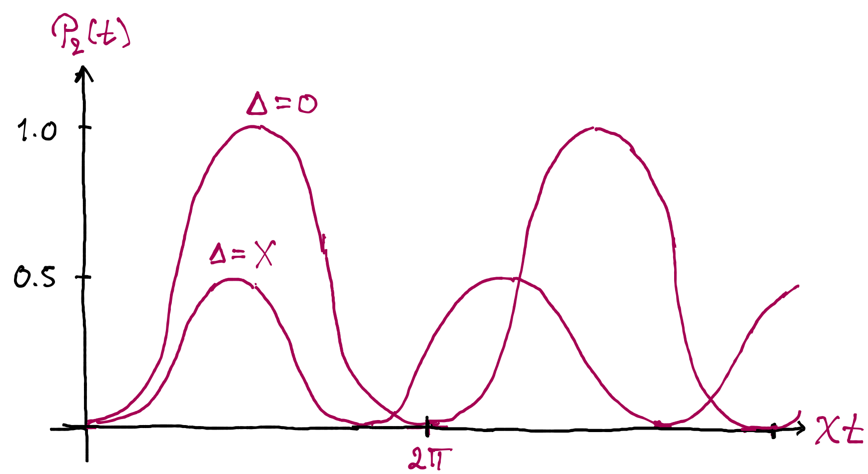
$\Omega \equiv \sqrt{\chi^2 + \Delta^2}$: Generalized Rabi freq.

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 $|1\rangle$, $|2\rangle$:

$$\boxed{\begin{aligned} P_1(t) &= |c_1(t)|^2 = \frac{1}{2}\left(1 + \frac{\Delta^2}{\Omega^2}\right) + \frac{1}{2}\frac{\chi^2}{\Omega^2}\cos\Omega t \\ P_2(t) &= |c_2(t)|^2 = \frac{1}{2}\frac{\chi^2}{\Omega^2}\left[1 - \cos\Omega t\right] \end{aligned}}$$



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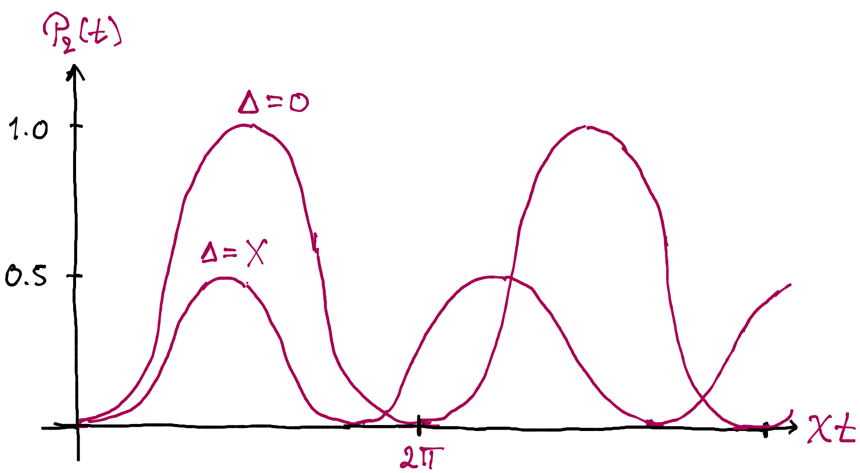


Note: All 2-level systems are isomorphic

- 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

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

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 $\vec{\varepsilon}E_0$ is real-valued and $V_{12} = V_{21} = V$ ⇒ The eqs for the *a* 's are

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

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

With this we have

$$\frac{d}{dt}a_1^* a_2 = \left(\dot{a}_1^* a_2 + a_1^* \dot{a}_2\right)$$

$$= -i\underbrace{\frac{E_2 - E_1}{\hbar}}_{\omega_0} a_1^* a_2 - i\frac{V}{\hbar}\left(|a_1|^2 - |a_2|^2\right)$$

Differentiating again gives us

$$\frac{d^2}{dt^2}\left(a_1^* a_2\right) = -\omega_0^2 a_1^* a_2 - \frac{\omega_0 V}{\hbar}\left(|a_1|^2 - |a_2|^2\right)$$

$$-i\hbar\frac{d}{dt}\left[\frac{V}{\hbar}\left(|a_1|^2 - |a_2|^2\right)\right]$$

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

This gives us

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

$$= \frac{2\omega_0}{\hbar}\vec{p}_{12}\left(\vec{p}_{12}\cdot\vec{E}\right)\left(|a_1|^2 - |a_2|^2\right)$$

$\vec{p}_{12} = \langle 1 | \hat{\vec{p}} | 2 \rangle$: dipole matrix element

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

Pick linear polarization so $\vec{\varepsilon}$ is real-valued.
Pick quantization axis along $\vec{\varepsilon}$ ⇒ $\vec{p}_{12} = p_{12}\vec{\varepsilon}$

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

Atom-Light Interaction: 2-Level Approximation

This gives us

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

$$= \frac{2\omega_0}{\hbar}\vec{r}_{12}\left(\vec{r}_{12}\cdot\vec{E}\right)\left(|a_1|^2-|a_2|^2\right)$$

$\vec{r}_{12} = \langle 1|\hat{\vec{r}}|2\rangle$: **dipole matrix element**

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

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

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

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 $|a_1|^2 \sim 1$, $|a_2|^2 \sim 0$

This is the case for $\Delta \gg \chi$
Or when $\chi \ll \Gamma$

Limit of weak Excitation !

Decay rate of $|2\rangle$

Oscillator Strength

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

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

Like the classical equation, but with modified polarizability !

Atom-Light Interaction: Multi-Level Atoms

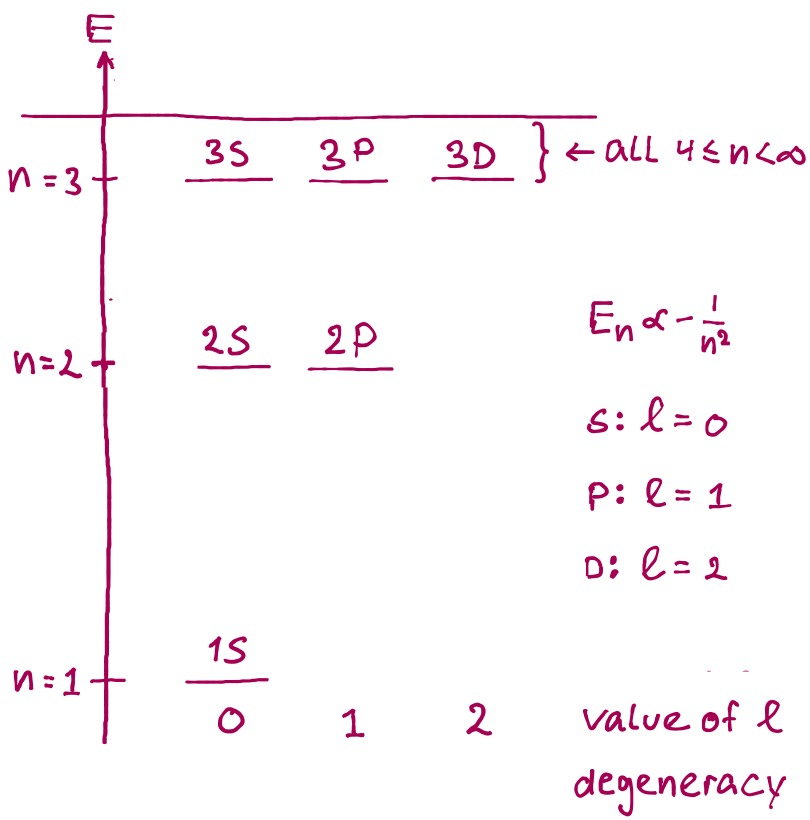
Atom-Light Interaction: Multi-Level Atoms

Starting point – the Hydrogen atom

$$H_a = \frac{p^2}{2m} - \frac{1}{4\pi\varepsilon_0}\frac{e^2}{|\vec{r}|}$$

$$V_{ext}(\vec{r},\vec{R},t) = -e\vec{r}\cdot\vec{E}(\vec{R},t)$$

$\vec{r}$: relative $\quad \vec{R}$: center-of-mass



Note: Frequencies for transitions $n \to n'$, $n'' \to n'''$ are very different ➡ near-resonant approx. with a single transition frequency $\omega \sim \omega_0$

Levels $|n\ell\rangle$ are generally degenerate with respect to the quantum number m (*), so we cannot isolate a 2-level system only through its transition frequency.

We must therefore consider Selection Rules

Interaction matrix element

$$\langle n'\ell' m' | V_{ext} | n\ell m\rangle \propto \int_{-\infty}^{\infty} dr^3\, \varphi^*_{n'\ell' m'}(\vec{r})\, \vec{r}\, \varphi_{n\ell m}(\vec{r})$$

Wavefunction parity is even/odd depending on ℓ

$$\varphi_{n\ell m}(\vec{r}) = (-1)^{\ell}\, \varphi_{n\ell m}(-\vec{r})$$

➡ $\langle\, | V |\, \rangle$ can be non-zero only if $(\ell - \ell')$ is odd.

This is the Parity Selection Rule !

(*) This is not strictly true due to spin-orbit coupling.