

Atom-Light Interaction: Multi-Level Atoms

Begin 02-13-2025

General ED Selection Rules

$$\Delta L = \pm 1 \quad \vec{L}: \text{total e orbital A. M.}$$

$$\Delta F = 0, \pm 1 \quad \vec{F}: \text{total orbital + spin A. M.}$$

$$\Delta m_F = q = 0, \pm 1 \quad q: \text{polarization of EM field}$$

Clebsch-Gordan coefficients ($E_{F', m_{F'}} > E_{F, m_F}$)

$$\langle F', m_{F'} | V | F, m_F \rangle \propto \langle 1, q; F, m_F | F', m_{F'} \rangle$$

$$\langle F, m_F | V | F', m_{F'} \rangle \propto \langle 1, -q; F', m_{F'} | F, m_F \rangle$$

Hydrogen atom

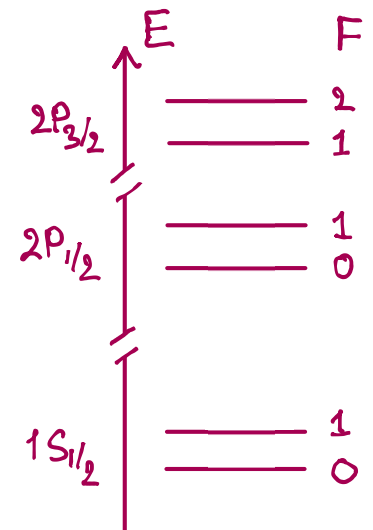
1S - 2S : forbidden 1S - 2P : allowed

Total spin: $\vec{F} = \vec{J} + \vec{I}$, $\vec{J} = \vec{L} + \vec{S}$

nuclear
 orbital
 electron spin

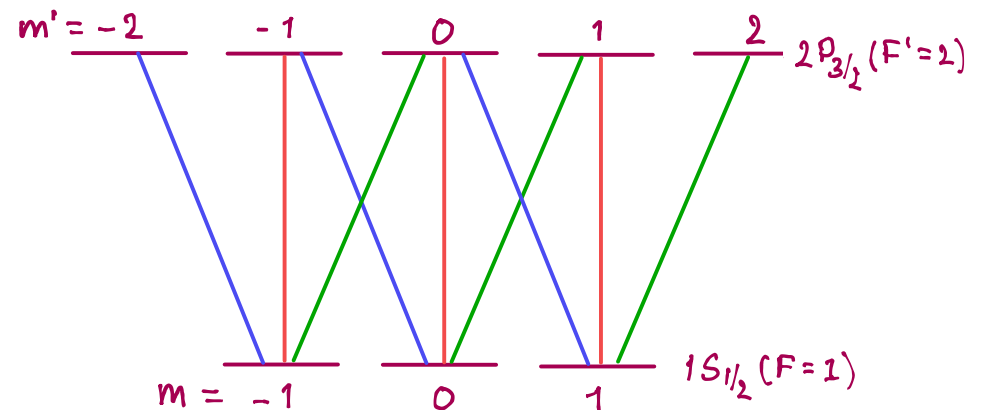
1S State:
 $J = 1/2, F = 0, 1$

2P State:
 $J = 1/2, F = 0, 1$
 $J = 3/2, F = 1, 2$



Level diagram for transitions

$1S_{1/2} (F=1) \rightarrow 2P_{3/2} (F=2)$

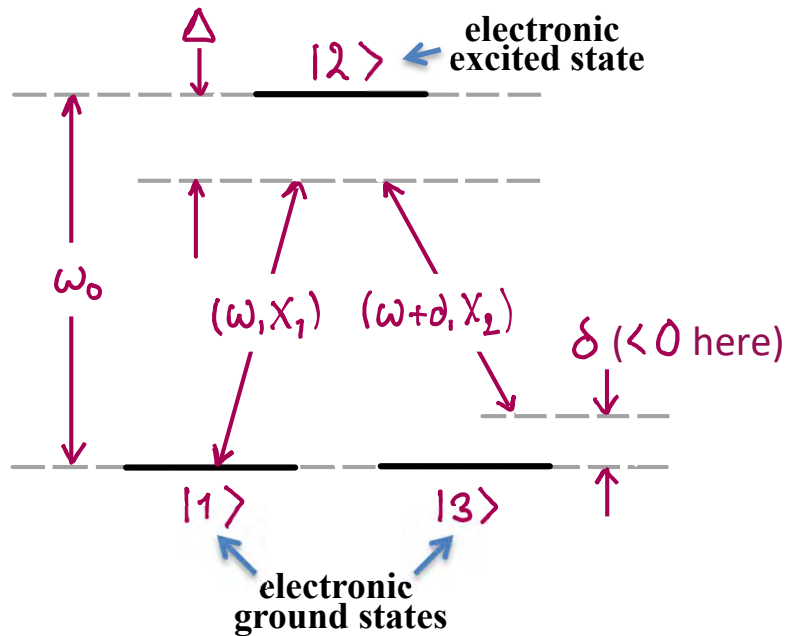


Polarization: $| \quad q=0 \quad / \quad q=1 \quad \backslash \quad q=-1$

Raman Coupling in 3-level Atoms

Raman Coupling in 3-Level Systems

Consider an atom with this 3-level structure



For simplicity we set $E_1 = E_3$ (no loss of generality)

Fields $\left\{ \begin{array}{l} \text{at } \omega, \text{ coupling } |1\rangle, |2\rangle \text{ w/Rabi freq. } \chi_1 \\ \text{at } \omega + \delta, \text{ coupling } |3\rangle, |2\rangle \text{ w/Rabi freq. } \chi_2 \end{array} \right.$

The Hamiltonian for this system is (χ_1, χ_2 real)

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \chi_1(t) & 0 \\ \chi_1(t) & \omega_0 & \chi_2(t) \\ 0 & \chi_2(t) & 0 \end{pmatrix}$$

where

$$\chi_1(t) = \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t})$$

$$\chi_2(t) = \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t})$$

Setting $|2(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle$
we get a S.E.

$$\dot{a}_1 = -i \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t}) a_2$$

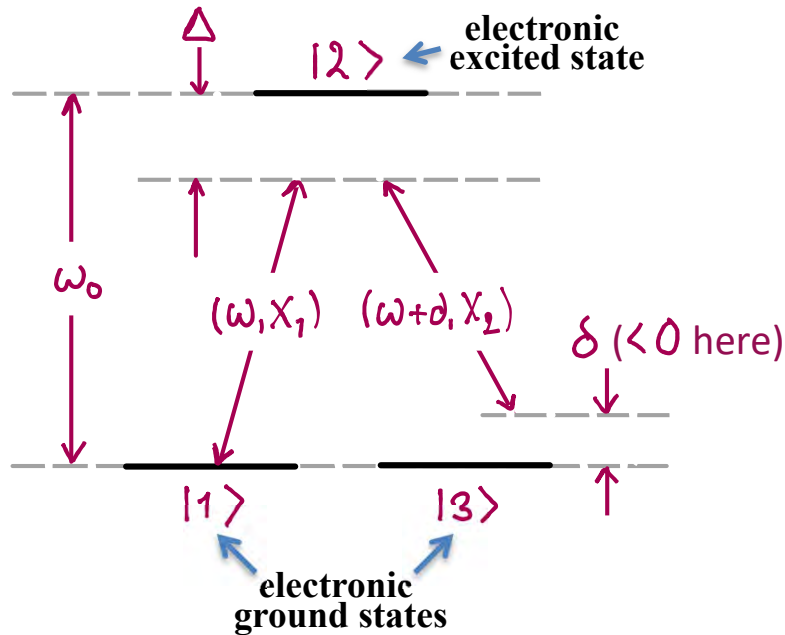
$$\dot{a}_2 = -i \omega_0 a_2 - i \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t}) a_1 - i \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t}) a_3$$

$$\dot{a}_3 = -i \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t}) a_2$$

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Rotating Wave Approximation.

Let $a_1 = b_1, a_2 = b_2 e^{-i\omega t}, a_3 = b_3 e^{i\delta t}$

Plug into in S.E.

$$\begin{aligned} \dot{b}_1 &= -i \frac{\chi_1}{2} (1 + e^{-i2\omega t}) b_2 \\ \dot{b}_2 &= -i(\omega_0 - \omega) b_2 - i \frac{\chi_1}{2} (e^{i2\omega t} + 1) b_1 \\ &\quad - i \frac{\chi_2}{2} (e^{i2(\omega + \delta)t} + 1) b_3 \\ \dot{b}_3 &= -i\delta b_3 - i \frac{\chi_2}{2} (1 + e^{-i2(\omega + \delta)t}) b_2 \end{aligned}$$

Drop non-resonant terms, set $\omega_0 - \omega = \Delta$

$$\begin{aligned} \dot{b}_1 &= -i \frac{\chi_1}{2} b_2 \\ \dot{b}_2 &= -i\Delta b_2 - i \frac{\chi_1}{2} b_1 - i \frac{\chi_2}{2} b_3 \\ \dot{b}_3 &= -i\delta b_3 - i \frac{\chi_2}{2} b_2 \end{aligned}$$

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Rotating Wave Approximation.

$$\text{Let } a_1 = b_1, \quad a_2 = b_2 e^{-i\omega t}, \quad a_3 = b_3 e^{i\delta t}$$

Plug into in S.E.

$$\dot{b}_1 = -i \frac{\chi_1}{2} (1 + e^{-i2\omega t}) b_2$$

$$\dot{b}_2 = -i(\omega_0 - \omega) b_2 - i \frac{\chi_1}{2} (e^{i2\omega t} + 1) b_1 - i \frac{\chi_2}{2} (e^{i2(\omega + \delta)t} + 1) b_3$$

$$\dot{b}_3 = -i\delta b_3 - i \frac{\chi_2}{2} (1 + e^{-i2(\omega + \delta)t}) b_2$$

Drop non-resonant terms, set $\omega_0 - \omega = \Delta$

$$\dot{b}_1 = -i \frac{\chi_1}{2} b_2$$

$$\dot{b}_2 = -i\Delta b_2 - i \frac{\chi_1}{2} b_1 - i \frac{\chi_2}{2} b_3$$

$$\dot{b}_3 = -i\delta b_3 - i \frac{\chi_2}{2} b_2$$

This S.E. has no explicit time dependence
Easy to solve numerically...

Now assume that $b_2(t=0) = 0$ → the atom is in the electronic ground state at $t=0$ when the fields turn on.

→ we can solve eq. for $b_2(t)$:

$$\dot{b}_2(t) = -i\Delta b_2 - i g(t), \quad g(t) = \left(\frac{\chi_1}{2} b_1 + \frac{\chi_2}{2} b_3 \right)$$

$$\begin{aligned} b_2(t) &= -e^{-i\Delta t} \int_0^t e^{i\Delta t'} g(t') dt' \quad \leftarrow (A) \\ &= -e^{-i\Delta t} \left(\underbrace{\left[\frac{1}{\Delta} e^{i\Delta t'} g(t') \right]_0^t}_{(B)} - \int_0^t \frac{1}{\Delta} e^{i\Delta t'} \dot{g}(t') dt' \right) \end{aligned}$$

Reminder: Integration by parts

$$\int_a^b f(x) g(x) dx = \left[F(x) g(x) \right]_a^b - \int_a^b F(x) g'(x) dx$$

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(B)

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Now consider the relative magnitude of (A) & (B)

(1) Let Rabi freqs be of the same order, $\chi_1 \sim \chi_2 \sim \chi$

(2) b_1, b_3 are at most ~ 1 \rightarrow $g(t)$ in (A) is $\sim \chi$

(3) In (B), the part $\frac{1}{\Delta} \dot{g}(t) = \frac{\chi}{\Delta} (\dot{b}_1 + \dot{b}_3)$

Where \dot{b}_1, \dot{b}_3 are $\sim \chi b_2$ and $b_2 \sim \frac{\chi}{\Delta}$ from Rabi solutions

$$\begin{aligned} \dot{b}_1 &= -i \frac{\chi_1}{2} b_2 \\ \dot{b}_2 &= -i\Delta b_2 - i \frac{\chi_1}{2} b_1 - i \frac{\chi_2}{2} b_3 \\ \dot{b}_3 &= -i\Delta b_3 - i \frac{\chi_2}{2} b_2 \end{aligned}$$

$$\begin{aligned} c_1(t) &= \left(\cos \frac{\Omega t}{2} + i \frac{\Delta}{\Omega} \sin \frac{\Omega t}{2} \right) e^{-i\Delta t/2} \\ c_2(t) &= \left(i \frac{\chi}{\Omega} \sin \frac{\Omega t}{2} \right) e^{-i\Delta t/2} \end{aligned}$$

Rabi Solutions

$$\Omega \equiv \sqrt{\chi^2 + \Delta^2}$$

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 We can ignore (B) when $\Delta^2 \gg \chi^2$



$$b_2(t) \approx -\frac{1}{\Delta} g(t) + \frac{1}{\Delta} e^{-i\Delta t} g(0)$$

$$= -\left[\frac{\chi_1}{2\Delta} b_1(t) + \frac{\chi_2}{2\Delta} b_3(t) \right]$$

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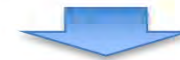
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(5) Finally, the last term $\propto \frac{e^{-i\Delta t}}{\Delta}$ can be ignored because it averages to zero on the timescale on which b_1, b_3 evolve.

Note:

The ground state amplitudes evolve slowly because $\chi_1/\Delta, \chi_2/\Delta \ll 1$, while the excited state amplitude evolves fast and adiabatically follows the instantaneous values of b_1, b_3

Plug the solution for $b_2(t)$ into the eqs. for b_1, b_3

$$\begin{aligned} \dot{b}_1(t) &= i \frac{\chi_1^2}{4\Delta} b_1(t) + i \frac{\chi_1 \chi_2}{4\Delta} b_3(t) \\ \dot{b}_3(t) &= -i \left(\delta - \frac{\chi_2^2}{4\Delta} \right) b_3(t) + i \frac{\chi_1 \chi_2}{4\Delta} b_1(t) \end{aligned}$$


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$$\begin{aligned} b_2(t) &\approx -\frac{1}{\Delta} g(t) + \frac{1}{\Delta} e^{-i\Delta t} g(0) \\ &= -\left[\frac{\chi_1}{2\Delta} b_1(t) + \frac{\chi_2}{2\Delta} b_3(t) \right] \\ &\quad + e^{-i\Delta t} \left[\frac{\chi_1}{2\Delta} b_1(0) + \frac{\chi_2}{2\Delta} b_3(0) \right] \end{aligned}$$

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Plug the solution for $b_2(t)$ into the eqs. for b_1, b_3



$$\begin{aligned} \dot{b}_1(t) &= i \frac{X_1^2}{4\Delta} b_1(t) + i \frac{X_1 X_2}{4\Delta} b_3(t) \\ \dot{b}_3(t) &= -i \left(\delta - \frac{X_2^2}{4\Delta} \right) b_3(t) + i \frac{X_1 X_2}{4\Delta} b_1(t) \end{aligned}$$

We simplify by making a final change of variables

$$C_1(t) = b_1(t) e^{-i \frac{X_1^2}{4\Delta} t}, \quad C_3(t) = b_3(t) e^{-i \frac{X_1^2}{4\Delta} t}$$



$$\begin{aligned} \dot{C}_1(t) &= i \frac{X_1 X_2}{4\Delta} C_3(t) \\ \dot{C}_3(t) &= -i \left(\delta + \frac{X_1^2 - X_2^2}{4\Delta} \right) C_3(t) + i \frac{X_1 X_2}{4\Delta} C_1(t) \end{aligned}$$

These are two-level equations!

Physical Discussion: We have an effective 2-level atom with effective Rabi Frequency and detuning.

$$\chi_{\text{eff}} = \frac{X_1 X_2}{2\Delta}, \quad \delta_{\text{eff}} = \delta + \frac{X_1^2 - X_2^2}{4\Delta}$$

Note that $\chi_{\text{eff}} \sim X^2/\Delta$ while the excited state population $P_2 \sim X^2/\Delta^2$. This means that for large X, Δ we can have large χ_{eff} and no opportunity for spontaneous decay.



Coherent Rabi oscillations and long lived superposition states

Raman Coupling in 3-level Atoms

We simplify by making a final change of variables

$$c_1(t) = b_1(t) e^{-i \frac{\chi_1^2}{4\Delta} t}, \quad c_3(t) = b_3(t) e^{-i \frac{\chi_1^2}{4\Delta} t}$$



$$\dot{c}_1(t) = i \frac{\chi_1 \chi_2}{4\Delta} c_3(t)$$

These are two-level equations!

$$\dot{c}_3(t) = -i \left(\delta + \frac{\chi_1^2 - \chi_2^2}{4\Delta} \right) c_3(t) + i \frac{\chi_1 \chi_2}{4\Delta} c_1(t)$$

Physical Discussion: We have an effective 2-level atom with effective Rabi Frequency and detuning.

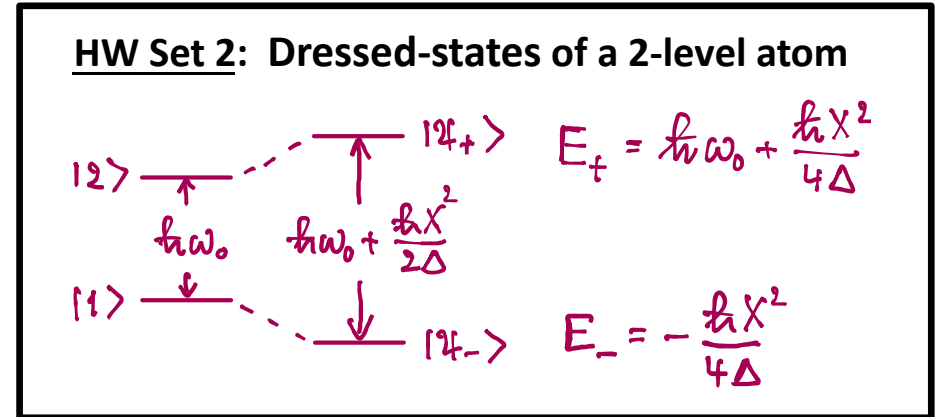
$$\chi_{\text{eff}} = \frac{\chi_1 \chi_2}{2\Delta}, \quad \delta_{\text{eff}} = \delta + \frac{\chi_1^2 - \chi_2^2}{4\Delta}$$

Note that $\chi_{\text{eff}} \sim \chi^2/\Delta$ while the excited state population $P_2 \sim \chi^2/\Delta^2$. This means that for large χ, Δ we can have large χ_{eff} and no opportunity for spontaneous decay.



Coherent Rabi oscillations and long lived superposition states

Note also: The effective Raman detuning is shifted.



3-level system \Rightarrow ground state shifts $\frac{\chi_1^2}{4\Delta}, \frac{\chi_2^2}{4\Delta}$

\Rightarrow Differential ground state shift $\frac{\chi_1^2 - \chi_2^2}{4\Delta}$

Final note: The atomic dipole $\langle \hat{\mu} \rangle$ will have components that match the frequency and polarization of both driving fields, with amplitudes that depend on both fields.



Non-Linear wave mixing, Breakdown of superposition principle

Raman Coupling in 3-level Atoms

Note also: The effective Raman detuning is shifted.

HW Set 2: Dressed-states of a 2-level atom

$|2\rangle \xrightarrow{\hbar\omega_0} |2_+\rangle$ $E_+ = \hbar\omega_0 + \frac{\hbar X^2}{4\Delta}$
 $|1\rangle \xrightarrow{\hbar\omega_0 + \frac{\hbar X^2}{2\Delta}} |2_+\rangle$
 $|1\rangle \xrightarrow{\hbar\omega_0} |2_-\rangle$ $E_- = -\frac{\hbar X^2}{4\Delta}$
 $|1\rangle \xrightarrow{\hbar\omega_0 + \frac{\hbar X^2}{2\Delta}} |2_-\rangle$

3-level system → ground state shifts $\frac{\chi_1^2}{4\Delta}, \frac{\chi_2^2}{4\Delta}$

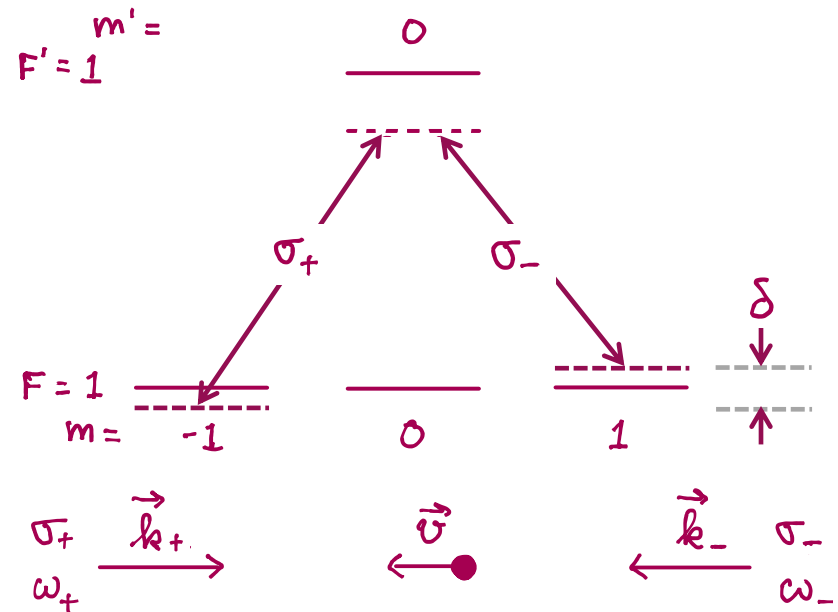
→ Differential ground state shift $\frac{\chi_1^2 - \chi_2^2}{4\Delta}$

Final note: The atomic dipole $\langle \vec{d} \rangle$ will have components that match the frequency and polarization of both driving fields, with amplitudes that depend on both fields.



**Non-Linear wave mixing,
Breakdown of superposition principle**

Example: Velocity dependent Raman Coupling



field freqs. in co-moving frame

velocity dependent Raman detuning

$$\left. \begin{aligned} \omega_+ &= \omega + k_+ v \\ \omega_- &= \omega - k_- v \end{aligned} \right\} \rightarrow \delta = 2k_+ v$$

Applications:

- Doppler velocimetry
- Raman Cooling by velocity selective momentum transfer
- What if we apply a $\pi/2$ Raman pulse?
- Atom Interferometry