Atom-Light Interaction: Multi-Level Atoms

General ED Selection Rules

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

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

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

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

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

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

Hydrogen atom

$1S - 2S$: forbidden $\qquad$ $1S - 2P$: allowed

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

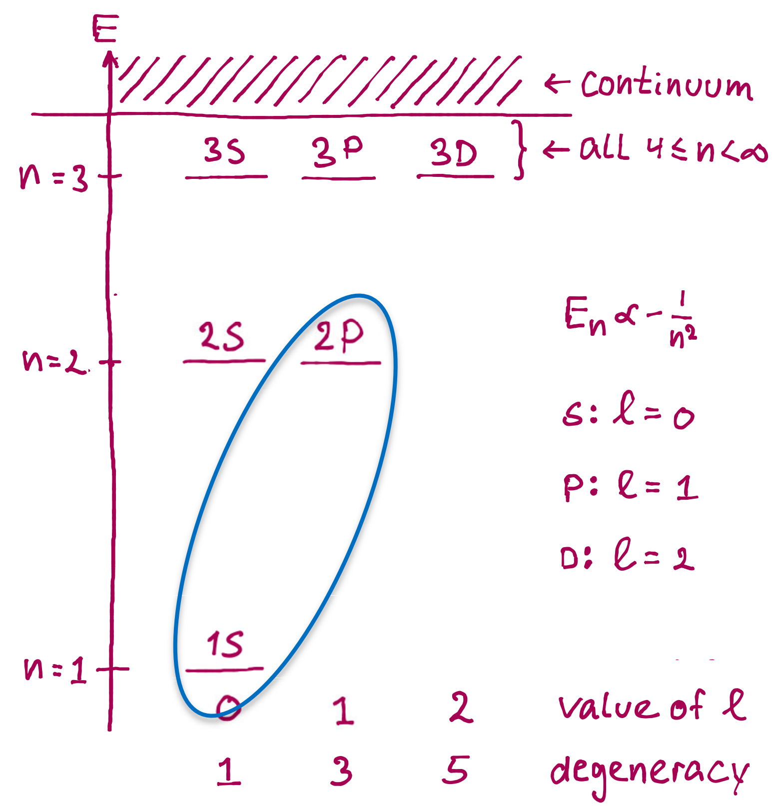
nuclear $\quad$ orbital $\quad$ electron spin

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 $\qquad$ $\vec{R}$: center-of-mass



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nuclear orbital electron spin

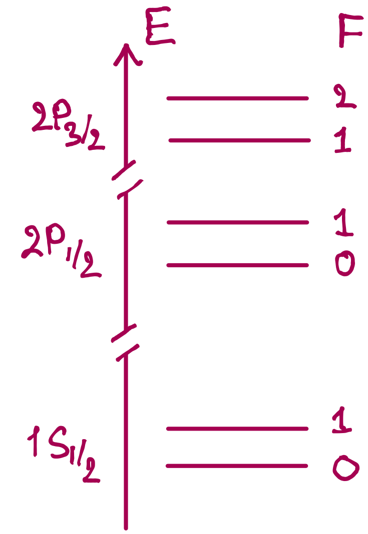
$1S$ State:

$J = 1/2, \ F = 0, 1$

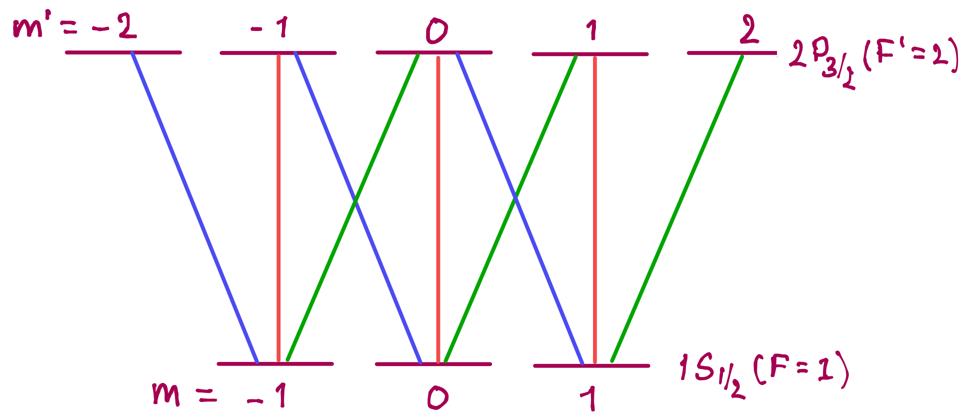
$2P$ State:

$J = 1/2, \ F = 0, 1$

$J' = 3/2, \ F = 1, 2$



Level diagram for transitions $1S_{1/2}(F=1) \rightarrow 2P_{3/2}(F=2)$



Polarization: $q = 0$ $q = 1$ $q = -1$

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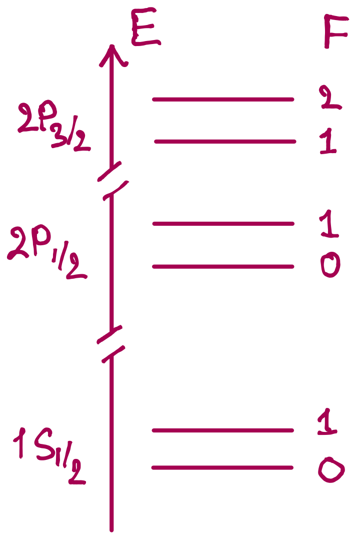

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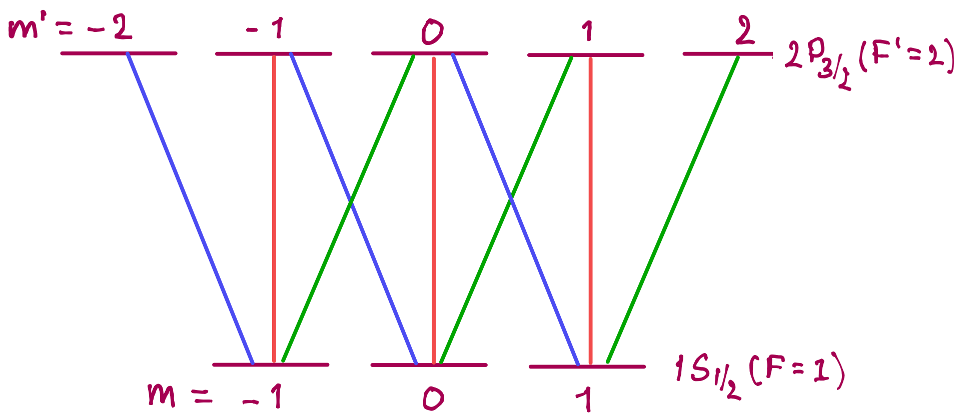

Polarization: | $q = 0$ / $q = 1$ \ $q = -1$

Note: When the field polarization is pure linear or circular the levels are coupled in pairs, and the oscillator strengths depend on the Clebsch-Gordan coefficients

Demo: Clebsch-Gordan Coefficients and Oscillator Strengths from Mathematica

- <u>Dense or hot gases:</u> Collisions redistribute Atoms between *m*-levels on very short time scales and the gas looks like a gas of 2-level atoms w/an effective coupling strength. If the dipole is oriented at random with the field, Then $\langle \vec{\eta} \cdot \vec{\mathcal{E}}_q \rangle_{angles} \sim \frac{1}{3} |\langle \vec{\eta} \rangle|$.
  The same is true for <u>unpolarized light</u>

- <u>Short interaction time:</u> If the atoms are "unpolarized"(random *m*-level populations) and the interaction too brief to change this, the atoms behave as an ensemble with different oscillator strengths

- <u>Optical pumping:</u> In dilute gases without collisions, atoms can be "pumped" into a single, pure state, e. g., $1S_{1/2}(F=1, m_F=1)$.
  If driven with $\vec{\mathcal{E}}_q = 1$ polarization this will realize a true 2-level system, as $2P_{3/2}(F'=2, m_F'=2)$ can only decay back to $1S_{1/2}(F=1, m_F=1)$

- If more than one frequency or polarization is Present, one can often drive Raman transitions

Raman Coupling in 3-Level Systems

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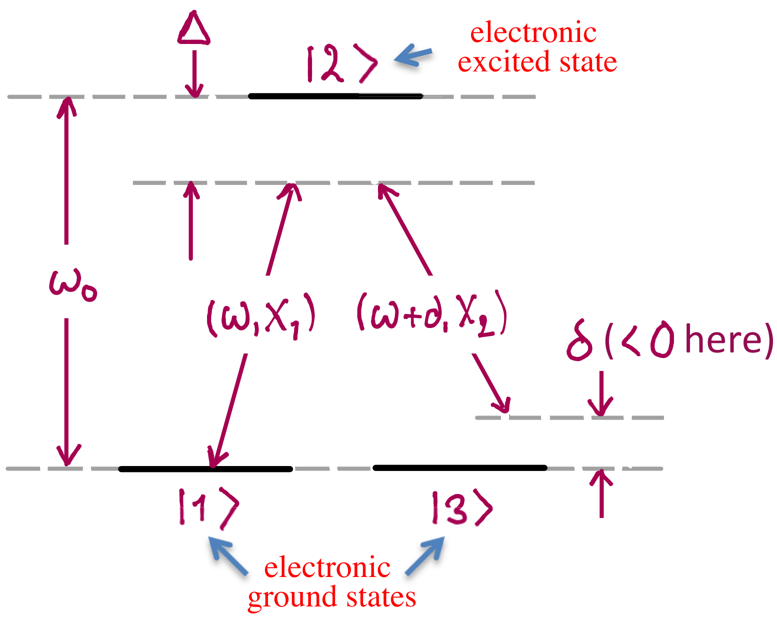
Consider an atom with this 3-level structure



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

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

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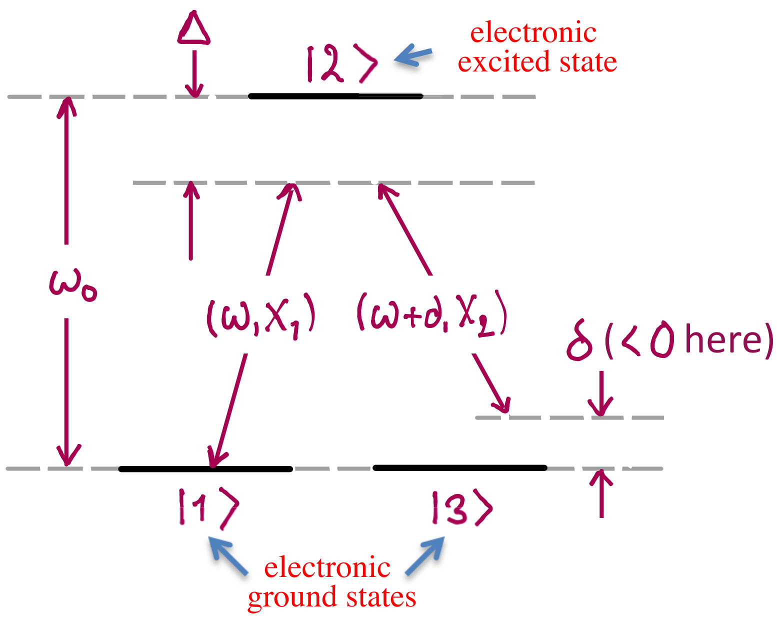

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The Hamiltonian for this system is (χ_1, χ_2 real)

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

where

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

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

Setting $|\psi(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle$

we get a Schrödinger Equation

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

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

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$$H = \hbar \begin{pmatrix} 0 & X_1(t) & 0 \\ X_1(t) & \omega_0 & X_2(t) \\ 0 & X_2(t) & 0 \end{pmatrix}$$

where

$$X_1(t) = \frac{\chi_1}{2}\left(e^{i\omega t} + e^{-i\omega t}\right)$$

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Rotating Wave Approximation.

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.

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

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

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

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

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

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\dot{b}_1 &= -i\frac{\chi_1}{2}\left(1+e^{-i2\omega t}\right) b_2 \\
\dot{b}_2 &= -i(\omega_0-\omega) b_2 - i\frac{\chi_1}{2}\left(e^{i2\omega t}+1\right) b_1 \\
&\quad - i\frac{\chi_2}{2}\left(e^{i2(\omega+\delta)t}+1\right) b_3 \\
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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 $b_2(t)$ **:**

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

$$b_2(t) = -e^{-i\Delta t}\int_0^t i e^{i\Delta t'} g(t')\,dt' \quad \leftarrow \quad (A)$$

$$= -e^{-i\Delta t}\left(\left[\frac{1}{\Delta} e^{i\Delta t'} g(t')\right]_0^t - \underbrace{\int_0^t \frac{1}{\Delta} e^{i\Delta t'} \dot g(t')\,dt'}_{(B)}\right)$$

Math result:

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

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Next, we 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 ⇨ $g(t)$ 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

(4) Therefore $\frac{1}{\Delta}\dot{g}(t) \sim \frac{\chi^3}{\Delta^2}$ and $\frac{(B)}{(A)} = \frac{1}{\Delta}\frac{\dot{g}(t)}{g(t)} \sim \frac{\chi^2}{\Delta^2}$

⇨ 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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(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

(4) Therefore $\frac{1}{\Delta}\dot{g}(t) \sim \frac{\chi^3}{\Delta^2}$ and $\frac{(B)}{(A)} = \frac{1}{\Delta}\frac{\dot{g}(t)}{g(t)} \sim \frac{\chi^2}{\Delta^2}$

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

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

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

$$+ e^{-i\Delta t}\left[\frac{\chi_1}{2\Delta}b_1(0) + \frac{\chi_2}{2\Delta}b_3(0)\right]$$

(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

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

Raman Coupling in 3-Level Systems

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