# Atom-Light Interaction: Multi-Level Atoms

The matrix element = overlap integral

$$V_{21} = \langle n'\ell' m' | V_{ext} | n\ell m\rangle$$

$$\propto \int_{\mathbb{R}^3} d^3r\, \varphi^*_{n'\ell' m'}(\vec r)\, r\left(Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t}\right)\varphi_{n\ell m}(\vec r)$$

where $\varphi_{n\ell m}(\vec r) = R_{n\ell}(r)\, Y_\ell^m(\theta,\varphi)$

$$V_{21} = \langle n'\ell' m' | V_{ext} | n\ell m\rangle$$

$$= \underbrace{\mathcal{R}}_{\text{radial integral}} \times \underbrace{\int_{4\pi} d\Omega\, (Y_{\ell'}^{m'})^* \left(Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t}\right) Y_\ell^m}_{\text{angular integral}}$$

Thus, to within a constant factor

$$V_{21} = \langle \ell' m' | Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t} | \ell m\rangle = V_{12}^*$$

And thus in the RWA we get

$$V_{21} \propto \langle \ell' m' | Y_1^q e^{-i\omega t} | \ell m\rangle$$

$$V_{12} \propto \langle \ell m | (-1)^q Y_1^{-q} e^{i\omega t} | \ell' m'\rangle$$

dropping factor $(-1)^q$

$$V_{21} \propto \int d\Omega\, (Y_{\ell'}^{m'})^* Y_1^q Y_\ell^m \;\propto\; \langle 1, q; \ell m | \ell' m'\rangle$$

$$V_{12} \propto \int d\Omega\, (Y_{\ell}^{m})^* Y_1^{-q} Y_{\ell'}^{m'} \;\propto\; \langle 1, -q; \ell' m' | \ell m\rangle$$

Clebsch-Gordan coefficients

**Next:** We can understand this as conservation of angular momentum when a photon is absorbed or emitted

**Selection Rules for Electric Dipole Transitions**

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$$\boxed{\begin{aligned} V_{21} &\propto \langle \ell' m' | Y_1^q e^{-i\omega t} | \ell m \rangle \\ V_{12} &\propto \langle \ell m | (-1)^q Y_1^{-q} e^{i\omega t} | \ell' m' \rangle \end{aligned}}$$

$$V_{21} \propto \int d\Omega \, (Y_{\ell'}^{m'})^* Y_1^q Y_\ell^m \propto \langle 1, q; \ell m | \ell' m' \rangle$$

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**Next:** We can understand this as conservation of angular momentum when a photon is absorbed or emitted

**Selection Rules for Electric Dipole Transitions**

**Revisit: Addition of Angular Momenta**

Let $\vec{J} = \vec{J}_1 + \vec{J}_2 \Rightarrow$ eigenstates $\begin{cases} |j_1 m_1\rangle \\ |j_2 m_2\rangle \\ |j m\rangle \end{cases}$

We can write $|jm\rangle$ in the basis $|j_1 m_1\rangle |j_2 m_2\rangle$

$$|jm\rangle = \underbrace{\sum_{m_1, m_2} |j_1 m_1; j_2 m_2\rangle\langle j_1 m_1; j_2 m_2|}_{\text{identity}} |jm\rangle$$

$$= \sum_{m_1 m_2} \langle j_1 m_1; j_2 m_2 | jm \rangle \, |j_1 m_1; j_2 m_2\rangle$$

Clebsch-Gordan coefficients

CG's are non-zero when
(Conservation of Angular Momentum)

$$\boxed{\begin{gathered} |j_1 - j_2| \le j \le j_1 + j_2 \\ m_1 + m_2 = m \end{gathered}}$$

Going back to the matrix element, $V_{21} \neq 0$ where

$|1q\rangle$ combined w/ $|\ell m\rangle$ is consistent w/ $|\ell' m'\rangle$

"photon" AM — ground state AM — excited state AM

# Atom-Light Interaction: Multi-Level Atoms

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The corresponding Selection Rules are

$$\ell' - \ell = 0, \pm 1 \;,\; m' - m = q \;,\; q = 0, \pm 1$$

Combining this with the Parity Rule we get

## Electric Dipole Selection Rules

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**Remarkably:**

(✱) These selection rules generalize to complex many – electron atoms, and after we include both electron and nuclear spins in the theory.

(✱) From a physics perspective, this reflects the conservation of angular momentum in rotationally invariant systems, and therefore transitions that do not conserve angular momentum are forbidden

(✱) To find the Clebsch-Gordan coefficients for different transitions we would need to use the Wigner-Eckart theorem, the proof of which is beyond this course.

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## General ED Selection Rules

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

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

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

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

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

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

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**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 orbital electron spin

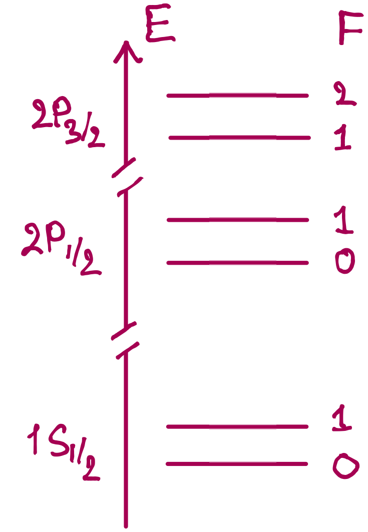
$1S$ State:

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

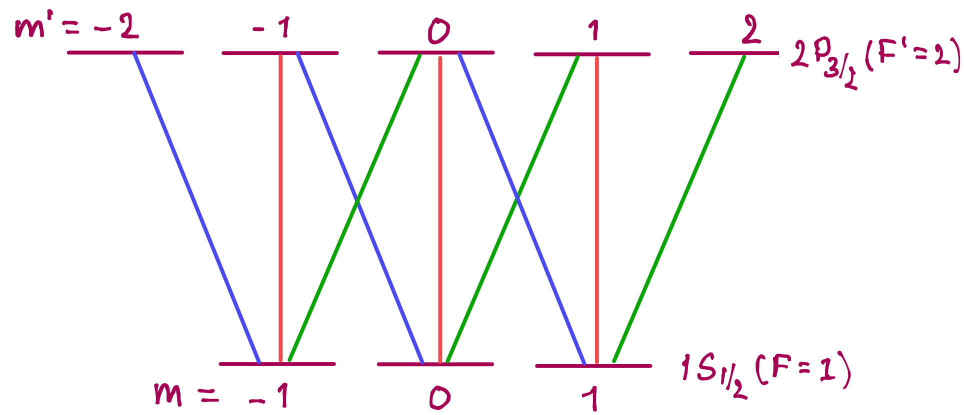
$2P$ State:

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

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




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




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

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# Atom-Light Interaction: Multi-Level Atoms

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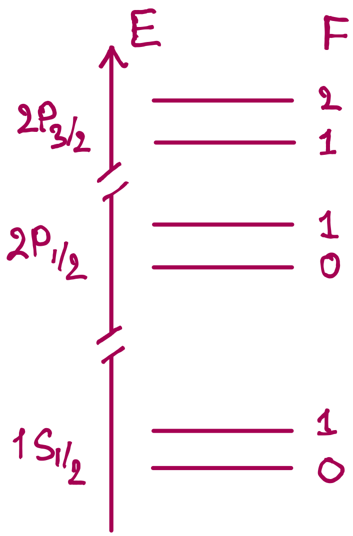


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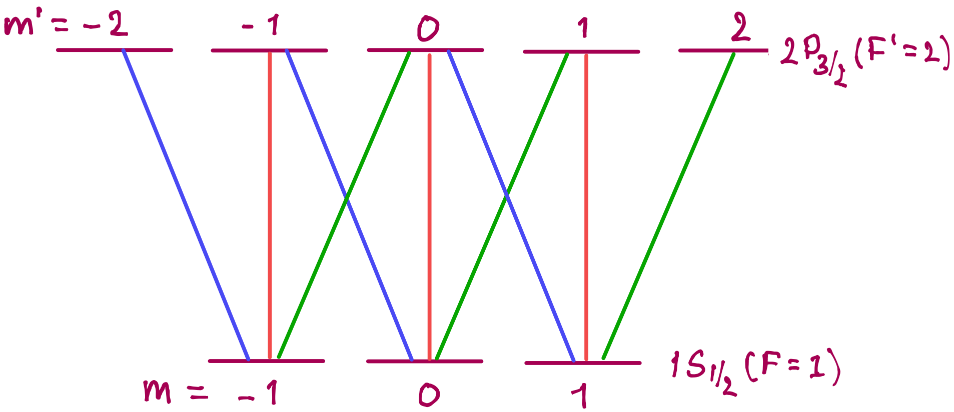


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**Note:** When the field polarization is pure linear or circular the levels are coupled in pairs, and the oscillator strengths depend on the Clebsch-Gordan coefficients

**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 $\langle \vec{\eta} \cdot \vec{\varepsilon}_q \rangle_{angles} \sim \frac{1}{3} |\langle \vec{\eta} \rangle|$
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 $\vec{\varepsilon}_q = 1$ 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

# Atom-Light Interaction: Multi-Level Atoms

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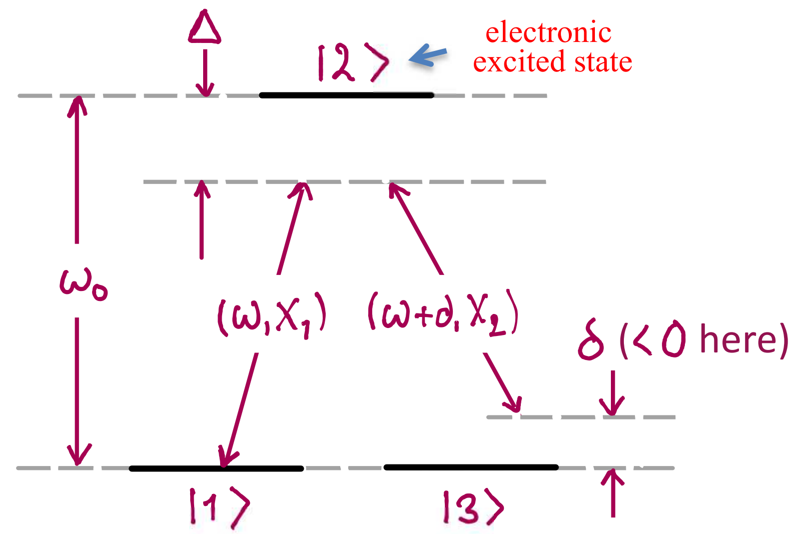
## Raman Coupling in 3-Level Systems

Consider an atom with this 3-level structure




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

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

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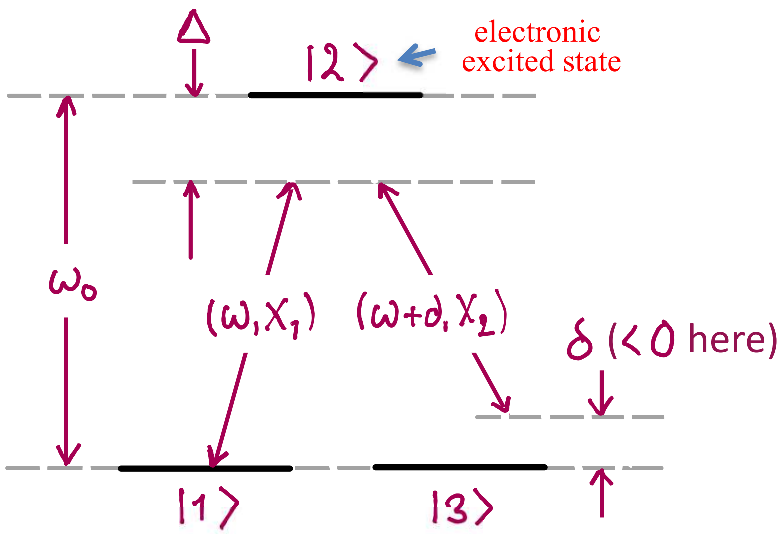


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**The Hamiltonian for this system is (** $\chi_1, \chi_2$ **real )**

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

**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 S.E.**

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

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

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

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# Raman Coupling in 3-level Atoms

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

$$i\dot c_1(t) = -\frac{1}{2}\left(\chi_{12} e^{-i2\omega t} + \chi_{21}^*\right) c_2(t)$$

$$i\dot c_2(t) = (\omega_{21}-\omega) c_2(t) - \frac{1}{2}\left(\chi_{21} + \chi_{12}^* e^{i2\omega t}\right) c_1(t)$$

$$i\dot c_1(t) = -\frac{1}{2}\chi_{21}^* c_2(t)$$

$$i\dot c_2(t) = \Delta c_2(t) - \frac{1}{2}\chi_{21} c_1(t)$$

$\Delta = \omega_{21} - \omega$

**(detuning)**

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

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

**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** $b_2(t)$ **:**

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

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

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

**Reminder: Integration by parts**

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