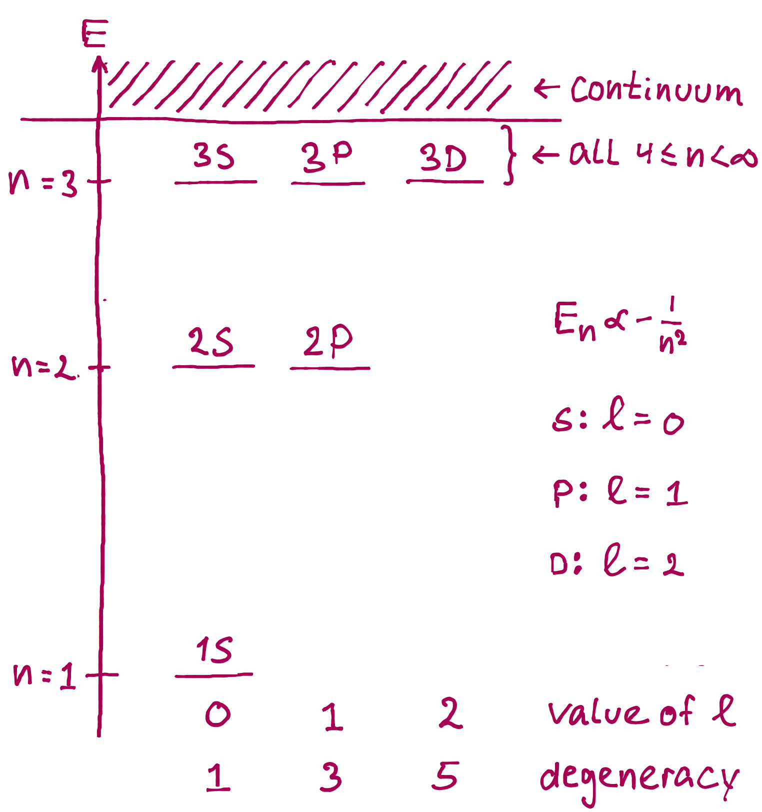
Atom-Light Interaction: Multi-Level Atoms

Starting point – the Hydrogen atom

$$\boxed{\begin{array}{l} H_a = \dfrac{p^2}{2m} - \dfrac{1}{4\pi\varepsilon_0}\dfrac{e^2}{|\vec{r}|} \\[2ex] V_{ext}(\vec{r},\vec{R},t) = -e\vec{r}\cdot\vec{E}(\vec{R},t) \end{array}}$$

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



Note: Frequencies for transitions $n \to n'$, $n'' \to n'''$ are <u>very</u> 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 <u>Selection Rules</u>

Interaction matrix element

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

Wavefunction parity is even/odd depending on ℓ

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

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

This is the Parity Selection Rule !

Atom-Light Interaction: Multi-Level Atoms

And thus in the RWA we get (use $(Y_\ell^m)^* = (-1)^m Y_\ell^{-m}$)

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

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

dropping the factor $(-1)^q$

$$\boxed{\begin{aligned} V_{21} &\propto \int d\Omega \, (Y_{\ell'}^{m'})^* Y_1^q Y_\ell^m \propto \langle 1, q; \ell m | \ell' m' \rangle \\ V_{12} &\propto \int d\Omega \, (Y_\ell^m)^* Y_1^{-q} Y_{\ell'}^{m'} \propto \langle 1, -q; \ell' m' | \ell m \rangle \end{aligned}}$$

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

Reminder: **Addition of Angular Momenta**

Let $\vec{J} = \vec{J}_1 + \vec{J}_2$ ⇨ 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 $|j_1 m_1\rangle |j_2 m_2\rangle$

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

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

Clebsch-Gordan coefficients

CG's are non-zero when $\boxed{\begin{gathered} |j_1 - j_2| \le j \le j_1 + j_2 \\ m_1 + m_2 = m \end{gathered}}$

Conservation of Angular Momentum

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

when $|1q\rangle$ 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

The corresponding Selection Rules are

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

Combining with the Parity Rule, this gives us the

Electric Dipole Selection Rules

$$\boxed{\ell' - \ell = \pm 1, \quad m' - m = q, \quad q = 0, \pm 1}$$

End 02-08-2021 / Begin 02-10-2021

Atom-Light Interaction: Multi-Level Atoms

The corresponding Selection Rules are

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

Combining with the Parity Rule, this gives us the

Electric Dipole Selection Rules

$$\boxed{\ell' - \ell = \pm 1, \quad m' - m = q, \quad q = 0, \pm 1}$$

Remarkably

Atom-Light Interaction: Multi-Level Atoms

The corresponding Selection Rules are

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

Combining with the Parity Rule, this gives us the

Electric Dipole Selection Rules

$$\boxed{\ell' - \ell = \pm 1, \quad m' - m = q, \quad q = 0, \pm 1}$$

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.

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 $\quad (E_{F',m_F'} > E_{F,m_F})$

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

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

Atom-Light Interaction: Multi-Level Atoms

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 ($E_{F', m_F'} > E_{F, m_F}$)

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

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

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 ($E_{F', m_F'} > E_{F, m_F}$)

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

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

Atom-Light Interaction: Multi-Level Atoms

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 ($E_{F',m_F'} > E_{F,m_F}$)

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

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

Hydrogen atom

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

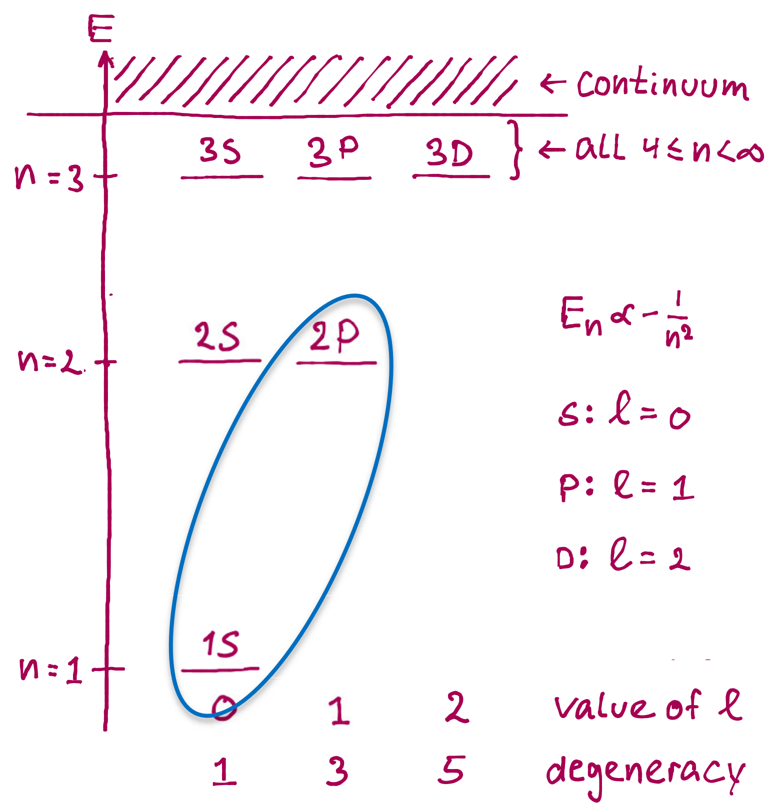
Total spin: $\vec{F} = \vec{J} + \underset{\text{nuclear}}{\vec{I}}$, $\vec{J} = \underset{\text{orbital}}{\vec{L}} + \underset{\text{electron spin}}{\vec{S}}$

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



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 ($E_{F', m_F'} > E_{F, m_F}$)

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

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

Hydrogen atom

$1S - 2S$: **forbidden** $\quad 1S - 2P$: **allowed**

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

nuclear orbital electron spin

Atom-Light Interaction: Multi-Level Atoms

General ED Selection Rules

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

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

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

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

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

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

Hydrogen atom

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

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

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

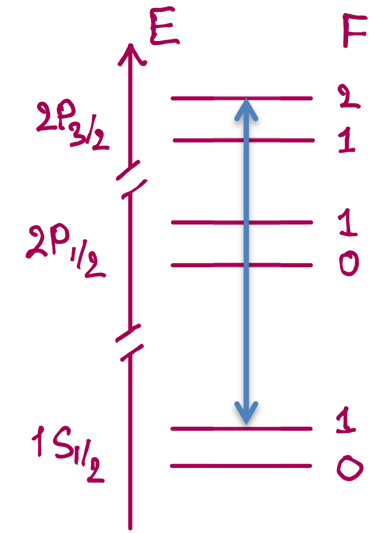
$1S$ State:

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

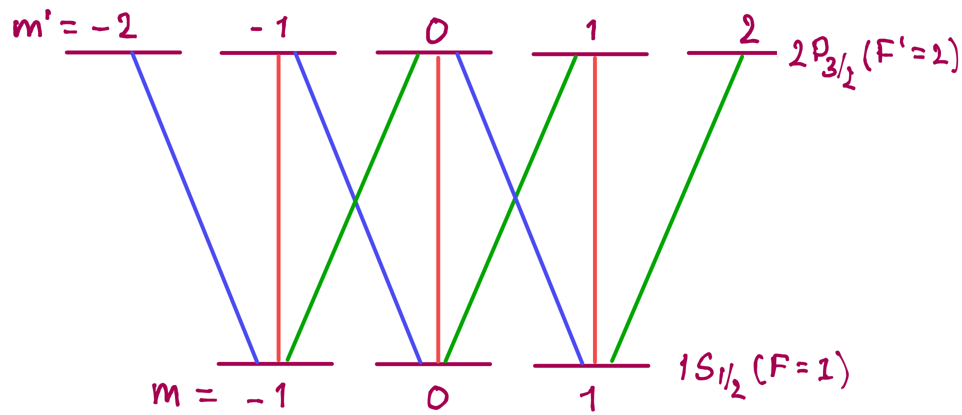
$2P$ State:

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

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



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



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

Atom-Light Interaction: Multi-Level Atoms

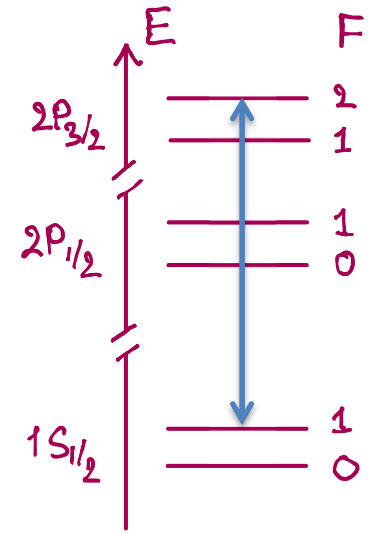
1S State:

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

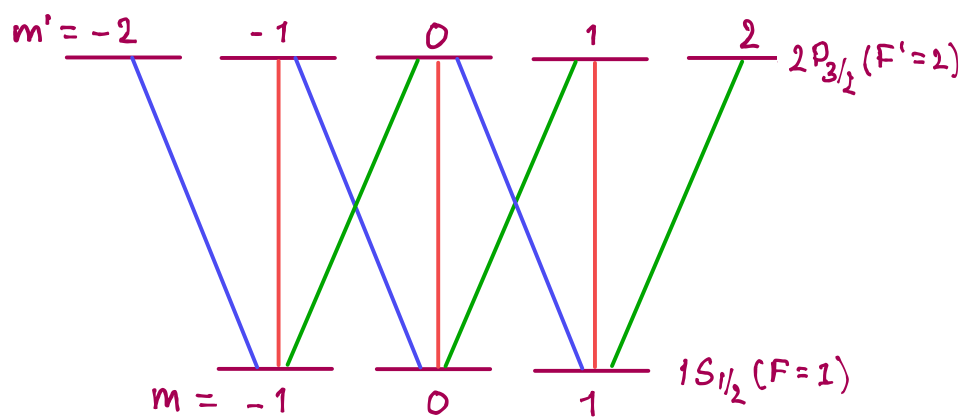
2P State:

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

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



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



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

Atom-Light Interaction: Multi-Level Atoms

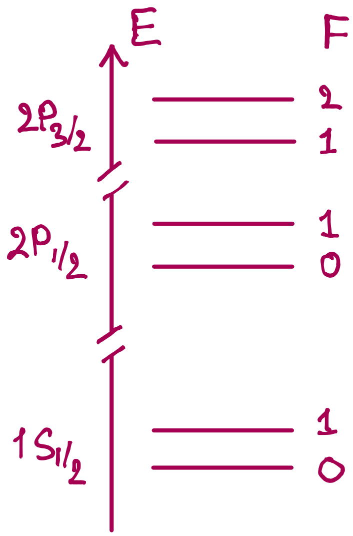
1S State:

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

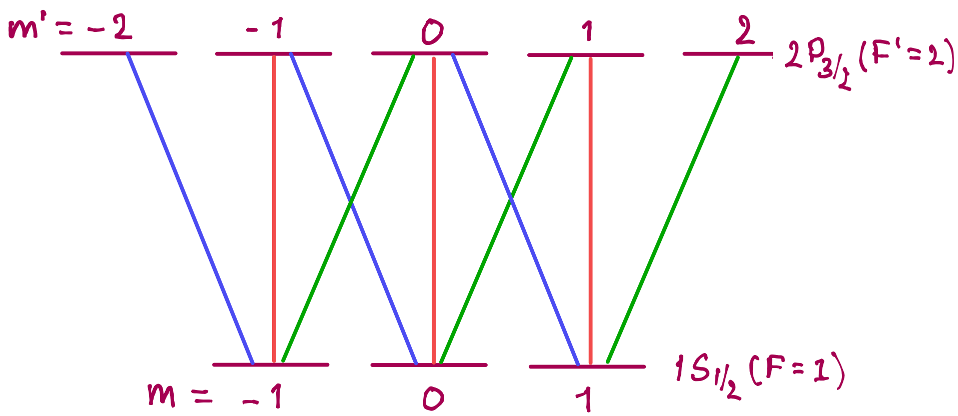
2P State:

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

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



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



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

Atom-Light Interaction: Multi-Level Atoms

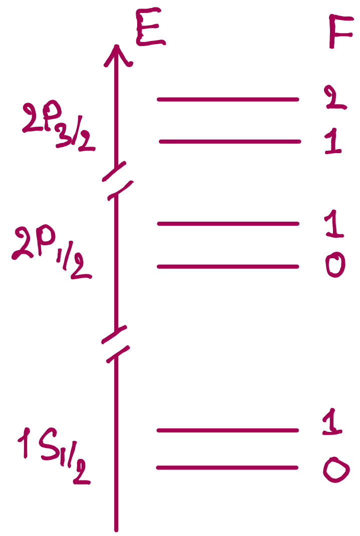
1S **State:**

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

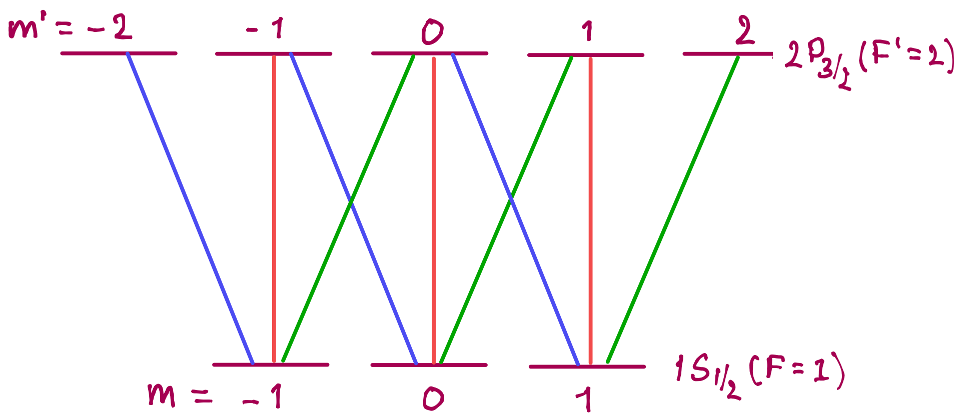
2P **State:**

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

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



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



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

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