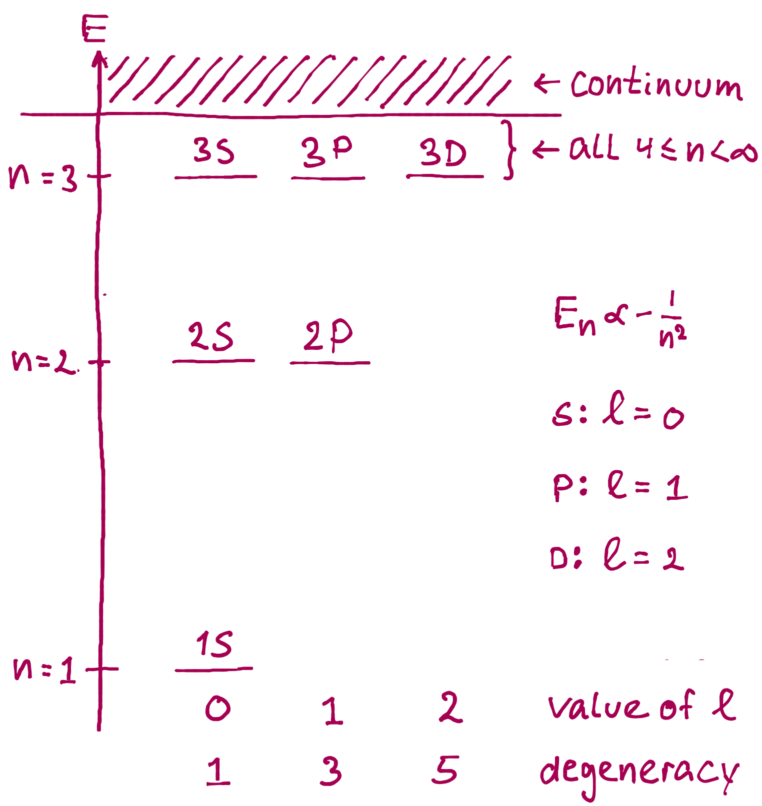
Atom-Light Interaction: Multi-Level Atoms

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 !

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

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Next: We will find selection rules that derive from the <u>angular symmetry</u> of the matrix element

We need to develop the proper math language ⇒ spherical basis vectors and harmonics

Consider an arbitrary set of orthonormal basis vectors $\vec\varepsilon_i, \vec\varepsilon_j, \vec\varepsilon_k$. We can always write

$$\vec r = (\vec r \cdot \vec\varepsilon_i)\vec\varepsilon_i + (\vec r \cdot \vec\varepsilon_j)\vec\varepsilon_j + (\vec r \cdot \vec\varepsilon_k)\vec\varepsilon_k$$

Cartesian basis: (real-valued)

$$\vec\varepsilon_i = \vec\varepsilon_x \qquad \vec\varepsilon_j = \vec\varepsilon_y \qquad \vec\varepsilon_k = \vec\varepsilon_z$$

Spherical basis: (complex-valued)

$$\begin{cases} \vec\varepsilon_i = \vec\varepsilon_1 = -\frac{1}{\sqrt2}(\vec\varepsilon_x + i\vec\varepsilon_y) \\ \vec\varepsilon_j = \vec\varepsilon_{-1} = \frac{1}{\sqrt2}(\vec\varepsilon_x - i\vec\varepsilon_y) \\ \vec\varepsilon_k = \vec\varepsilon_0 = \vec\varepsilon_z \end{cases}$$

Reminder: Scalar products of complex vectors

Dirac notation	Regular notation
$\{\,|a\rangle + i|b\rangle, |c\rangle\}$	$(\vec a + i\vec b)\cdot\vec c$
$= (\langle a| - i\langle b|)\,|c\rangle$	$= \vec a\cdot\vec c - i\,\vec b\cdot\vec c$
$= \langle a|c\rangle - i\langle b|c\rangle$	(anti-linear in 1st factor)

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Scalar Products in the spherical basis

Homework: prove the relations

$$\boxed{\vec{\varepsilon}_q^{\,*} = (-1)^q \vec{\varepsilon}_q, \quad \vec{\varepsilon}_{q'} \cdot \vec{\varepsilon}_q = \delta_{qq'}, \quad \vec{\varepsilon}_{q'} \cdot \vec{\varepsilon}_q^{\,*} = (-1)^q \delta_{-q'q}}$$

Next: Rewrite $\vec{r}\cdot\vec{\varepsilon}_q$ in polar coordinates

$$\vec{r}\cdot\vec{\varepsilon}_x = x = r\sin\theta\cos\varphi$$
$$\vec{r}\cdot\vec{\varepsilon}_y = y = r\sin\theta\sin\varphi$$
$$\vec{r}\cdot\vec{\varepsilon}_z = z = r\cos\theta$$

Compare to the Spherical Harmonics

$$Y_1^0(\theta,\varphi) = \sqrt{\frac{3}{4\pi}}\cos\theta, \quad Y_1^{\pm 1}(\theta,\varphi) = \pm\sqrt{\frac{3}{8\pi}}\sin\theta\, e^{\pm i\varphi}$$

$$\left.\begin{aligned} x &= -r\sqrt{\frac{2\pi}{3}}\left(Y_1^1 - Y_1^{-1}\right) \\ y &= ir\sqrt{\frac{2\pi}{3}}\left(Y_1^1 + Y_1^{-1}\right) \\ z &= r\sqrt{\frac{4\pi}{3}}\,Y_1^0 \end{aligned}\right\} \Rightarrow \boxed{\vec{r}\cdot\vec{\varepsilon}_q = r\sqrt{\frac{4\pi}{3}}\,Y_1^q}$$

to verify, plug in $\vec{\varepsilon}_q$ in terms of $\vec{\varepsilon}_x, \vec{\varepsilon}_y, \vec{\varepsilon}_z$

This finally gives us $\vec{\varepsilon}_q$ in the spherical basis:

$$\boxed{\vec{r} = \sum_{q=0,\pm 1} (\vec{r}\cdot\vec{\varepsilon}_q)\vec{\varepsilon}_q = r\sqrt{\frac{4\pi}{3}}\sum_{q=0,\pm 1} Y_1^q\,\vec{\varepsilon}_q}$$

End math preamble

Back to the Matrix Elements

First:

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

electric dipole interaction

$$\vec{E}(t) = \frac{1}{2}E_0\left(\vec{\varepsilon}_q e^{-i\omega t} + \vec{\varepsilon}_q^{\,*} e^{i\omega t}\right)$$
$$= \frac{1}{2}E_0\left(\vec{\varepsilon}_q e^{-i\omega t} + (-1)^q \vec{\varepsilon}_q e^{i\omega t}\right)$$

electric field polarization $\vec{\varepsilon}_q$

$$V_{ext} = -\sqrt{\pi/3}\,eE_0 r\left(\sum_{q'} Y_1^{q'}\,\underbrace{\vec{\varepsilon}_{q'}\Big)\cdot\Big(\vec{\varepsilon}_q}_{\delta_{q'q}} e^{-i\omega t} + (-1)^q \vec{\varepsilon}_{-q} e^{i\omega t}\right)$$

$\delta_{q'(-q)}$

$$\boxed{V_{ext} \propto r\left(Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t}\right)}$$

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$$\underbrace{\vec{\mathcal{E}}_{q'}\cdot\vec{\mathcal{E}}_q}_{\delta_{q'q}} \qquad \overbrace{\vec{\mathcal{E}}_{q'}\cdot\vec{\mathcal{E}}_{-q}}^{\delta_{q'(-q)}}$$

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with $\underbrace{\vec{\varepsilon}_{q'}\cdot\vec{\varepsilon}_q}_{\delta_{q'q}}$ and $\overbrace{\vec{\varepsilon}_{q'}\cdot\vec{\varepsilon}_{-q}}^{\delta_{q'(-q)}}$

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$$\boxed{V_{ext} \propto r\left(Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t}\right)}$$

The matrix element = overlap integral of the form

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

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

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

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

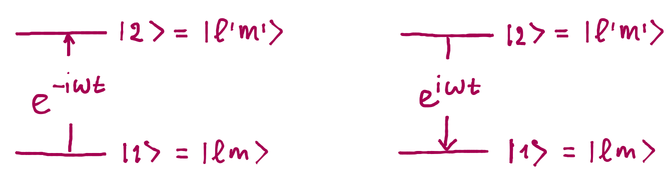
$$= \mathcal{R} \times \underbrace{\int_{4\pi} d\Omega\,\left(Y_{\ell'}^{m'}\right)^*\left(Y_1^q e^{-i\omega t} + (-1)^q Y_1^{-q} e^{i\omega t}\right)Y_\ell^m}_{\text{angular integral}}$$

radial integral

Thus, to within a constant factor

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

From the RWA, we know the resonant terms are



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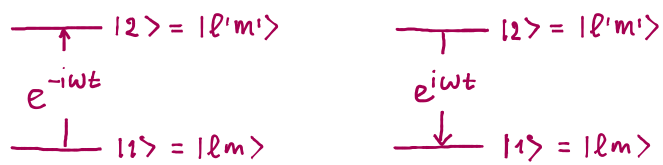

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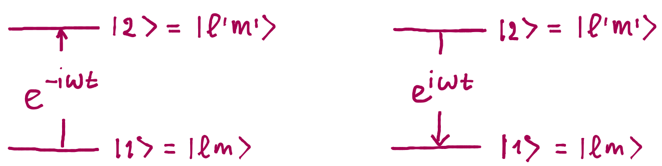

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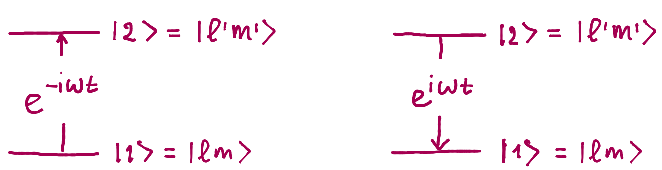

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dropping the factor $(-1)^q$

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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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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 $|j_1 m_1\rangle |j_2 m_2\rangle$

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

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

Clebsch-Gordan coefficients

CG's are non-zero when

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

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

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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

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