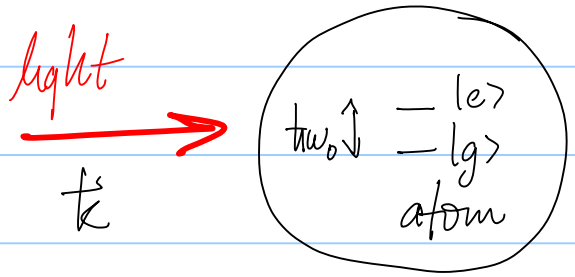
PHY 5514

Semiclassical – quantum atom, classical light

Note Title 3/2/2005

Try to improve our model!



There is much, much physics here!

$$\vec{E} = E_1 \hat{\varepsilon} \cos(kz - \omega t + \varphi) = \tfrac{1}{2} E_1 \hat{\varepsilon} \left[e^{i(\vec{k}\cdot\vec{r} - \omega t + \varphi)} + e^{-i(\vec{k}\cdot\vec{r} - \omega t + \varphi)} \right]$$

What is the interaction?

$$H = H_0 + H_I$$

$$H_I = -\vec{p}\cdot\vec{E}$$ electric dipole!

$$= e\frac{E_1}{2}\vec{r}\cdot\hat{\varepsilon}\left[e^{i(\vec{k}\cdot\vec{r} - \omega t + \varphi)} + e^{-i(\vec{k}\cdot\vec{r} - \omega t + \varphi)} \right]$$

Let's take our H_0 as the Hamiltonian for an atom in free space, w/ eigenkets $|g\rangle$ and $|e\rangle$ and $E_g = 0$, $E_e = \hbar\omega_0$.

(we'll make life more complicated shortly)

Also, right now: $e^{i\vec{k}\cdot\vec{r}} = 1 + i\vec{k}\cdot\vec{r} + \ldots \simeq 1$ since the electron is very localized compared with the wavelength of light

(we'll relax this shortly too!)

So, in the interaction picture:

$$H_I' = e^{iH_0 t/\hbar} H_I e^{-iH_0 t/\hbar}$$

$$= \frac{e}{2} e^{iH_0 t/\hbar} \left[e^{-i(\omega t - \varphi)} + e^{i(\omega t - \varphi)} \right] \vec{r}\cdot\hat{\varepsilon}\, e^{-iH_0 t/\hbar}$$

How do we evaluate $\vec{r} \cdot \hat{\varepsilon}$?

- for a complicated atom, this is tricky — more soon
- we can learn a lot by considering a hydrogen atom with a spinless electron and nucleus
- we'll need matrix elements such as

$$\langle e | \vec{r} \cdot \hat{\varepsilon} | g \rangle$$

typically we write $\langle e | \vec{r} \cdot \hat{\varepsilon} | g \rangle = D_{12} I_{ang}$

I_{ang}: angular integral (selection rules)

D_{12}: dipole matrix element

so, for our example:

$$D_{12} = \int_0^\infty R_{n_2,\ell_2}\, r\, R_{n_1,\ell_1}\, r^2 dr$$

$$I_{ang} = \int d\Omega\, Y^*_{\ell_2 m_2}\, \hat{r} \cdot \hat{\varepsilon}\, Y_{\ell_1 m_1}$$

let's rewrite $\vec{r}\cdot\hat{\varepsilon}$

(this is all consistent with spherical tensor notes)

we need:

(1) $\hat{r} = \sin\theta\cos\varphi\,\hat{x} + \sin\theta\sin\varphi\,\hat{y} + \cos\theta\,\hat{z}$

(2) $\sin\theta\cos\varphi = \sqrt{\frac{2\pi}{3}}\left(Y_{1,-1} - Y_{1,1}\right)$

$\sin\theta\sin\varphi = \sqrt{\frac{2\pi}{3}}\left(Y_{1,-1} + Y_{1,1}\right)$

$\cos\theta = \sqrt{\frac{4\pi}{3}}\,Y_{1,0}$

(3) $\hat{\varepsilon} = \varepsilon_-\left(\frac{\hat{x}-i\hat{y}}{\sqrt{2}}\right) + \varepsilon_0\hat{z} + \varepsilon_+\left(-\frac{\hat{x}+i\hat{y}}{\sqrt{2}}\right)$

atomic conventions: σ_- π σ_+

taking this dot product in cartesian coordinates (i.e., keeping things simple)

$$\hat{r}\cdot\hat{\varepsilon} = \varepsilon_- Y_{1,-1} + \varepsilon_0 Y_{1,0} + \varepsilon_+ Y_{1,1}$$

for $\mathcal{E}_0$, "π transitions"

$\hat{\mathcal{E}}$ along $\hat{z}$

$$I_{ang} = \int d\Omega \, Y^*_{\ell_2 m_2} \cos\theta \, Y_{\ell_1 m_1}$$

See Foot or many other sources —

$I_{ang} \neq 0$ iff $m_1 - m_2 = 0$, $\boxed{\Delta m = 0}$

for $\mathcal{E}_\pm$, "σ transitions"

$\hat{\mathcal{E}} \perp \hat{z}$

$$I_{ang} = \int d\Omega \, Y^*_{\ell_2 m_2} \sin\theta \, e^{\pm i\varphi} \, Y_{\ell_1 m_1}$$

for $I_{ang} \neq 0$, require $m_1 - m_2 \pm 1 = 0$

$\boxed{\Delta m = \pm 1}$

Using various recursion relations, these integrals can be evaluated (Arfken p 753)

There is an additional selection rule that you find if you do these integrals — the "parity selection rule"

This can be motivated physically —

$\hat{H}$ commutes with the parity operator, so I_{ang} should not change under parity:

$$\hat{P} Y_{lm} = (-1)^l Y_{lm}$$

← just a property of spherical harmonics

under parity, $I_{ang} \to I_{ang} (-1)^{l_2 + l_1 + 1}$,

which requires

$$l_2 + l_1 + 1 = 2n \qquad n = 0, 1, 2, \ldots$$

The spherical harmonics Y_{1m} have angular momentum $l = 1$, and so we require $l_2 + l_1 = \pm 1$, per usual angular momentum addition rules.

All of these selection rules are straightforward if you evaluate I_{any} directly (see Arfken p753):

$$I_{any} = \delta_{m_2 m_1} \left[\delta_{l_2 l_1 + 1} \sqrt{\frac{(l_1 - m_1 + 1)(l_1 + m_1 + 1)}{(2l_1 + 1)(2l_1 + 3)}} + \delta_{l_2 l_1 - 1} \sqrt{\frac{(l_1 - m_1)(l_1 + m_1)}{(2l_1 - 1)(2l_1 + 1)}} \right] \varepsilon_0$$

π

$$\delta_{m_2 m_1 \pm 1} \left[\mp \delta_{l_2 l_1 + 1} \sqrt{\frac{(l_1 \pm m_1 + 1)(l_1 \pm m_1 + 2)}{(2l_1 + 1)(2l_1 + 3)}} \pm \delta_{l_2 l_1 - 1} \sqrt{\frac{(l_1 \mp m_1)(l_1 \mp m_1 - 1)}{(2l_1 - 1)(2l_1 + 1)}} \right] \varepsilon_\pm$$

$\sigma_\pm$

- keep things simple for now with the time-dependent problem →

say we only have $\mathcal{E}_+$, and therefore write

$$\hat{\mathcal{E}} \cdot \vec{r} = \langle e|r_+|g\rangle |e\rangle\langle g| + \langle g|r_+|e\rangle |g\rangle\langle e|$$

or:

$$E_1 r_+ = \langle r\rangle \left\{ \mathcal{E}_1 |e\rangle\langle g| + \mathcal{E}_1^* |g\rangle\langle e| \right\}$$

where $\langle r\rangle = \langle e|r_+|g\rangle$

Then: $H_I' = \langle r \rangle \frac{e}{2} \mathcal{E}_+ e^{i H_0 t/\hbar} \left(E_1 |e\rangle\langle g| + E_1^* |g\rangle\langle e| \right) \cdot e^{-i H_0 t/\hbar}$

$$\cdot \left[e^{-i(\omega t - \varphi)} + e^{i(\omega t - \varphi)} \right]$$

$$= \langle r \rangle \frac{e}{2} \mathcal{E}_+ \left[e^{i\omega_0 t} E_1 |e\rangle\langle g| + e^{-i\omega_0 t} E_1^* |g\rangle\langle e| \right]$$

$$\left[e^{-i(\omega t - \varphi)} + e^{i(\omega t - \varphi)} \right]$$

look familiar? it's a coupled two-level system!

$$H_I' = \frac{e}{2} \mathcal{E}_+ \langle r_+ \rangle \left[e^{i\omega_0 t} \sigma_+ E_1 + e^{-i\omega_0 t} \sigma_- E_1^* \right]$$

$$\left[e^{-i(\omega t - \varphi)} + e^{i(\omega t - \varphi)} \right]$$

We can make the RWA:

$$H_I' \simeq \frac{e}{2} \mathcal{E}_+ \langle r_+ \rangle \left[e^{-i(\delta t - \varphi)} \sigma_+ E_1 + e^{i(\delta t - \varphi)} \sigma_- E_1^* \right]$$

where $\delta = \omega - \omega_0$

and $\sigma_+ = |e\rangle\langle g|$ and $\sigma_- = |g\rangle\langle e|$

write $|\psi\rangle = c_g|g\rangle + c_e|e\rangle$

Schrödiger's equation:

$$i\hbar \frac{\partial}{\partial t}|\psi\rangle = H_I'|\psi\rangle$$

*we're using the interaction picture, but we're not using time-dependent PT — this is an exact solution!

$$i\hbar \dot{c}_g = \frac{\hbar\Omega^*}{2} e^{i(\delta t+\varphi)} c_e$$
$$i\hbar \dot{c}_e = \frac{\hbar\Omega}{2} e^{-i(\delta t+\varphi)} c_g$$

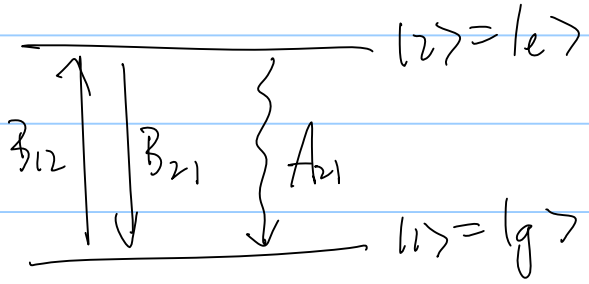
where $\hbar\Omega = eE_1\langle r_{+-}\rangle$
(this is Foot's, Metcalf's, etc. convention)

We already know what happens! Rabi oscillations.

(we can drive rotations, etc)

Let's add in spontaneous emission ad-hoc, even though we can derive the optical Bloch equations that follow using quantum treatment of light field (we'll do that later).

Interestingly, Einstein's A & B coefficients argument is another way to get to the OBEs with minimal assumptions about the quantum nature of light.



$A_{21} = \Gamma = 1/\tau$ lifetime of $|e\rangle$

Einstein's argument: $A_{21} = \frac{\hbar\omega^3}{\pi^2 c^3} B_{21}$ $\quad B_{12} = B_{21}$

let's convert to Foot's notation:

$$\Omega = \frac{e}{\hbar}\langle g|\vec{r}\cdot\vec{E}_0|e\rangle = e/\hbar\langle 1|\vec{r}\cdot\vec{E}_0|2\rangle$$

$$B_{12} = \frac{\pi e^2 |D_{12}|^2}{3\epsilon_0 \hbar^2} \qquad \vec{D}_{12} = \langle 1|\vec{r}|2\rangle \approx \text{few } a_0 \text{ for alkalis}$$

thus assumes all possible polarizations.

so $\boxed{A_{12} = \frac{g_1}{g_2}\frac{e^2\omega^3}{3\pi\epsilon_0\hbar c^3}|D_{12}|^2}$ after some math

Amazing! Spontaneous emission rate depends only on ω^3 and $|D_{12}|^2$ — note ω dependence different than classical result!

We put this by hand into the OBEs:

$$\dot{c}_e = \ldots - \Gamma/2\, c_e$$

Note: Einstein's argument really about $|\dot{c}_e|^2$, but, if we put this into ρ_{ee} then we get an unphysical result (the coherences do not decay)

This is like a T_1 and T_2 process combined...

Foot uses a standard "rotating frame" (it's not) operator transformation:

$$\tilde{c}_g = c_g e^{-i\delta t/2} \qquad \tilde{c}_e = e^{i\delta t/2} c_e$$

$$\tilde{\rho}_{gg} = \rho_{gg} \quad \tilde{\rho}_{ee} = \rho_{ee} \quad \tilde{\rho}_{ge} = e^{-i\delta t} \rho_{ge} \quad \tilde{\rho}_{eg} = e^{i\delta t} \rho_{eg}$$

Then:

$$\begin{cases} \dot{u} = \delta v - \Gamma/2\, u \\ \dot{v} = -\delta u + \Omega w - \Gamma/2\, v \\ \dot{w} = -\Omega v - \Gamma(w-1) \end{cases} \quad \text{or} \quad \begin{cases} \dot{\rho}_{gg} = \Gamma \rho_{ee} + i/2 \left(\Omega^* \tilde{\rho}_{eg} - \Omega \tilde{\rho}_{ge}\right) \\ \dot{\rho}_{ee} = -\Gamma \rho_{ee} + i/2 \left(\Omega \tilde{\rho}_{ge} - \Omega^* \tilde{\rho}_{eg}\right) \\ \dot{\tilde{\rho}}_{ge} = -\left(\Gamma/2 + i\delta\right) \tilde{\rho}_{ge} + i/2\, \Omega^* \left(\rho_{ee} - \rho_{gg}\right) \\ \dot{\tilde{\rho}}_{eg} = -\left(\Gamma/2 - i\delta\right) \tilde{\rho}_{eg} + i/2\, \Omega \left(\rho_{gg} - \rho_{ee}\right) \end{cases}$$

Let's look at the long-time / equilibrium behavior:
(set all time derivatives to zero and solve)

$$\begin{pmatrix} u \\ v \\ w \end{pmatrix} = \frac{1}{\delta^2 + \Omega^2/2 + \Gamma^2/4} \begin{pmatrix} \Omega\delta \\ \Omega\Gamma/2 \\ \delta^2 + \Gamma^2/4 \end{pmatrix}$$

$$\rho_{22} = \frac{1-w}{2}$$

define: $s = \frac{2\Omega^2}{\Gamma^2} = \frac{I}{I_{sat}}$ $\qquad I_{sat} = \frac{hc\pi}{3\lambda^3\tau}$

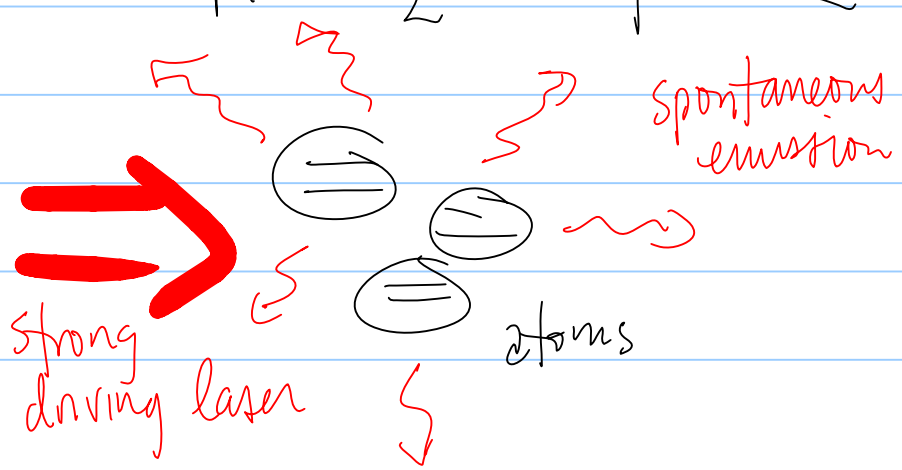
(using $I = \epsilon_0 c \frac{|E_0|^2}{2}$)

then: $\rho_{22} = \frac{1}{2} \frac{s}{1 + s + 4\delta^2/\Gamma^2}$

rate of spontaneous emission $= \Gamma\rho_{22} = R$

first, notice that in the $s \gg 1$ limit and $\delta = 0$,

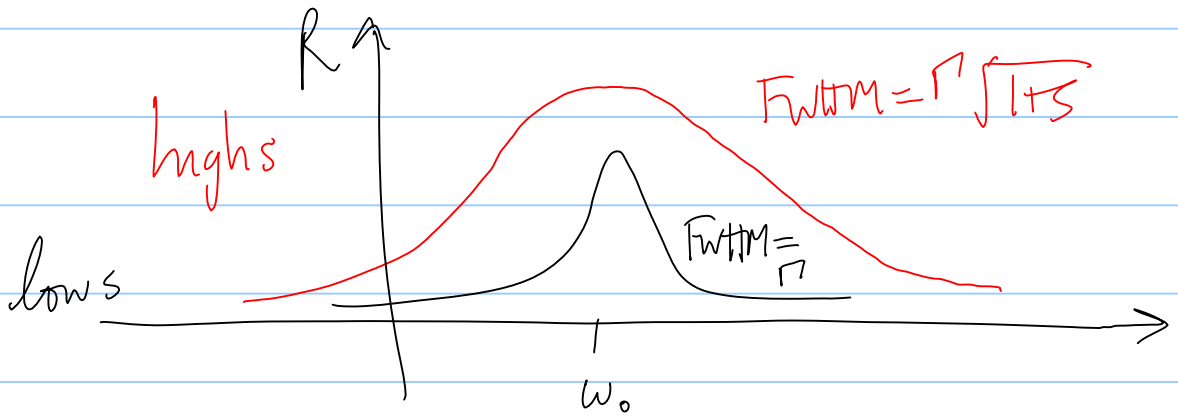
$R \to \Gamma/2$ and $\rho_{22} \to 1/2$



The atoms spend at most half of their time in the excited state

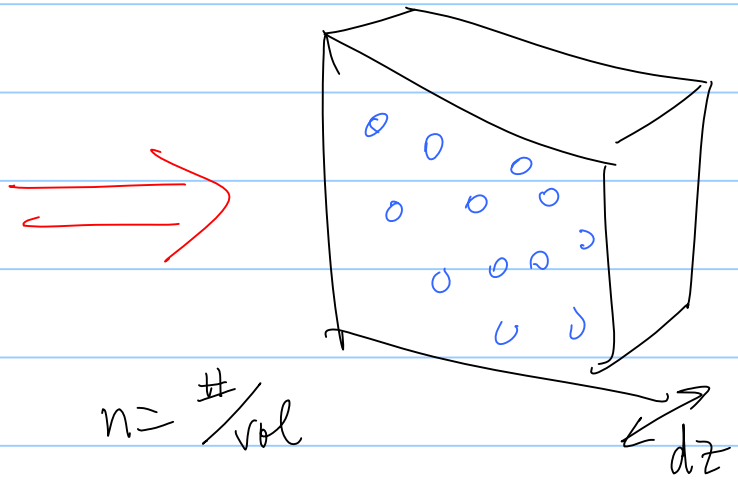
Power broadening

$$R = \frac{\Gamma}{2} s \frac{\Gamma^2/4}{\Gamma^2/4 (1+s) + (\omega - \omega_0)^2}$$



physics: near resonance, very saturated — fluorescence reduced; away from resonance — not much change

What about absorption?



atoms / area = $n\,dz$

$n\,\sigma\,dz$ = fraction of surface covered by atoms

$\frac{dI}{I} = -n\sigma dz$ fraction of light absorbed

$\frac{dI}{dz} = -n\sigma I$ this defines the cross section σ

using steady state conservation of energy:

n_1: density of atoms in state 1

$(n_1 - n_2)\sigma I = n_2 \Gamma \hbar\omega$

net rate of energy in — rate of energy out

using $w = \frac{n_2 - n_1}{n}$ $\rho_{22} = n_2/n$ $n = n_1 + n_2$

$$\sigma = \frac{3\lambda^2}{2\pi} \frac{1}{1 + 4\delta^2/\Gamma^2}$$

on resonance, $\sigma = \frac{3\lambda^2}{2\pi} \simeq \lambda^2/2$

that's a big cross-section for something so small!

Note! There is some averaging here to be careful about.

Our definition of B_{12} has a factor of $1/3$ for average over polarization.

Everyone handles this differently. The right way to deal with this is to solve the OBEs for a real, multilevel atom, using the exact dipole matrix elements.

A reasonable approach from Foot:

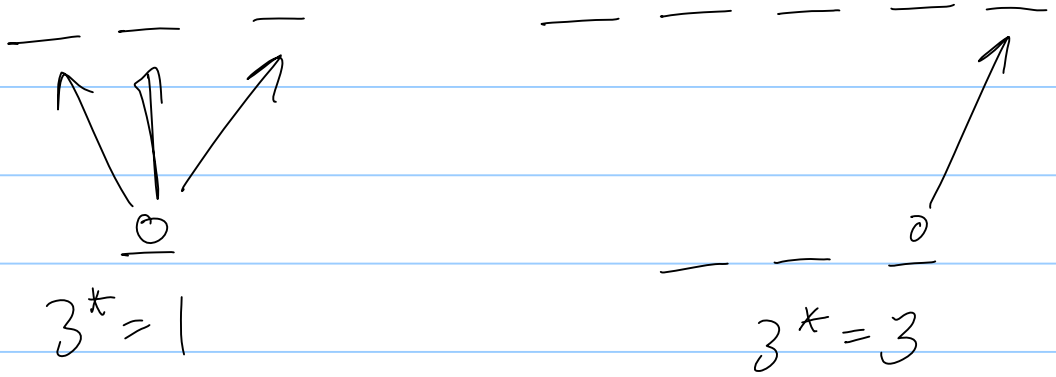
$$\sigma = \underbrace{\frac{g_2}{g_1}}_{\text{degeneracy factors from } A_{21}} \frac{\pi^2 c^2}{\omega_0^2} A_{21} \underbrace{\frac{1}{2\pi} \frac{\Gamma}{\delta^2 + \Gamma^2/4}}_{\text{lineshape function}}$$

→ good for low excited state populations

A lot of people put averages into I_{sat}:

$$I_{sat} = \frac{hc\pi}{3^* \lambda^3 \tau} \qquad 3^* = 1 \Rightarrow 3$$

unpolarized light or atoms: $3^* = 1$
polarized light and atoms: $3^* = 3$



for alkali atoms, $I_{sat} \sim 5\text{-}6\ mW/cm^2$

So, what is the right way to calculate dipole matrix elements?

We want: $\langle F' m_F' | \vec{r} | F m_F \rangle$

This is not a fun matrix element to calculate!

procedure:
1. expand to $|m_J, m_I\rangle$ basis
2. then expand to $|m_\ell, m_s, m_I\rangle$ basis
3. then calculate the matrix element

result:

$$\langle F' m_F' | r_q | F m_F \rangle = (-1)^{1+L'+S+J+J'+I-m_F'} \langle \alpha' L' \| r \| \alpha L \rangle \cdot \sqrt{(2J+1)(2J'+1)(2F+1)(2F'+1)} \cdot \begin{Bmatrix} L' & J' & S \\ J & L & 1 \end{Bmatrix} \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix} \cdot \begin{pmatrix} F & 1 & F' \\ m_F & q & -m_F' \end{pmatrix}$$

reduced matrix element

6-j symbols

selection rules come from this 3-j symbol

the reduced matrix element is:

$$R = \int_0^\infty r^2 dr\, R_{n'\ell'}\, r\, R_{n\ell}$$

and must, in general, be determined experimentally (e.g., from σ)!

Mathematica knows 3-j and 6-j symbols:

http://mathworld.wolfram.com/Wigner6j-Symbol.html

http://mathworld.wolfram.com/Wigner3j-Symbol.html

A standard reference on this is Elementary Theory of Angular Momentum by Rose:

http://www.amazon.com/Elementary-Theory-Angular-Momentum-M-E/dp/0486684806

selection rules: * the polarization in the atomic basis "matters" (relative to a quantization magnetic field)

$m_F' = q + m_F$

$\Delta l = \pm 1$ (remember "parity rule")

$\Delta F = \pm 1, 0$; $\Delta F = 0$ and $\Delta m_F = 0$ not allowed

No one in their right mind calculates this directly!

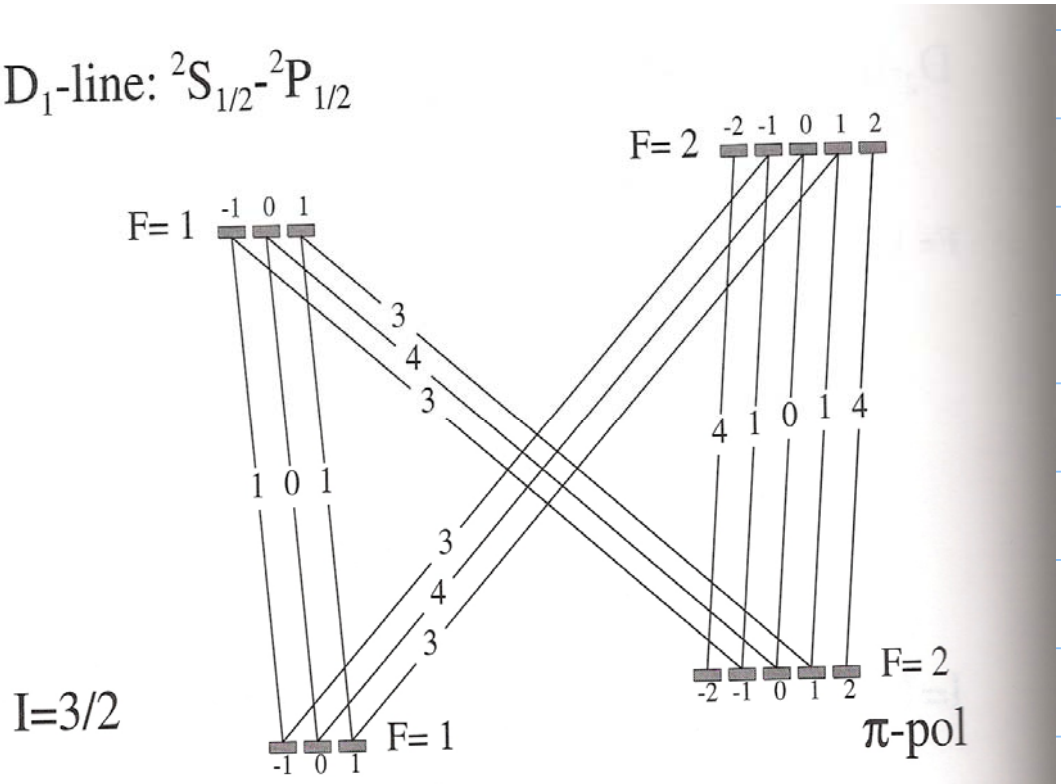
Talk to your favorite atomic physicist to get a chart!

Notes on charts:

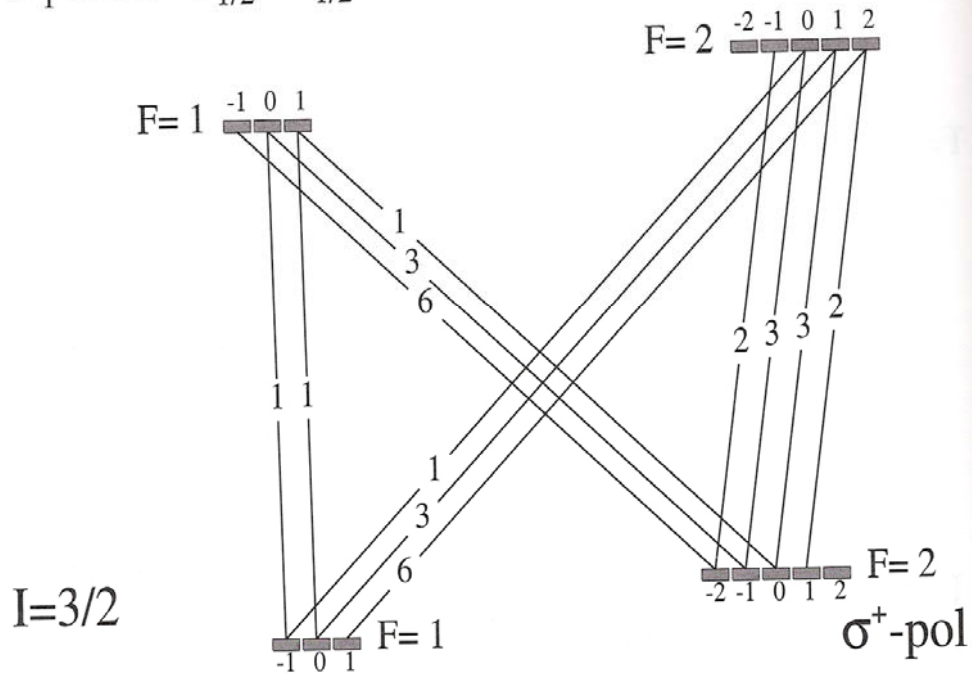
- everyone has their own conventions
- most people leave out signs — do this at your own peril!

Lesson — be sure that you know what someone means!

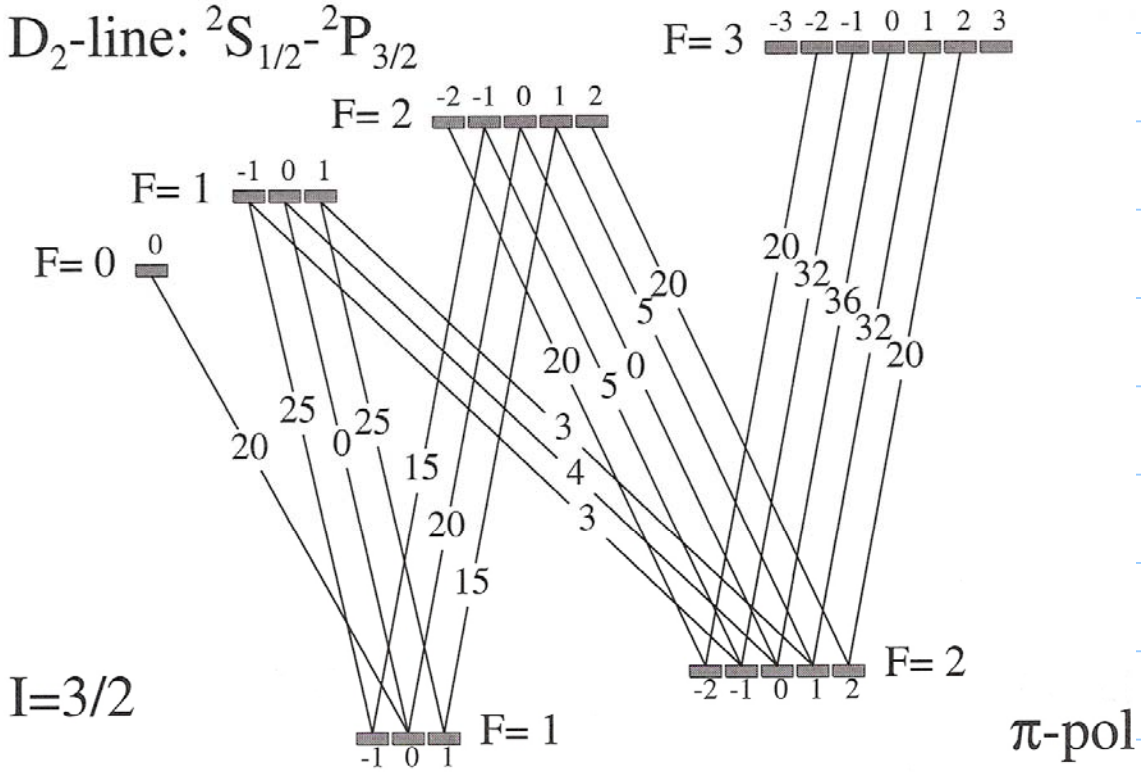
Metcalf: Laser cooling and trapping

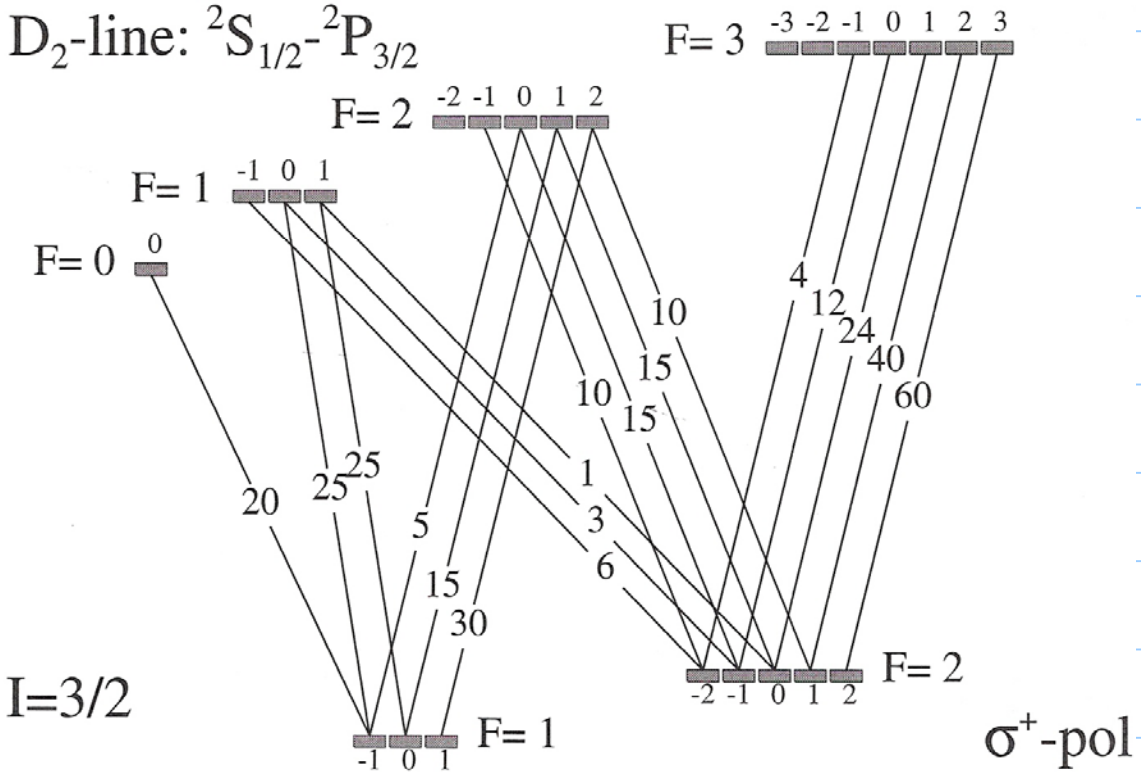


take the square root - watch normalization



Look for the note! multiply by 2 for D1:





El.	Transition			Theory		Experiment
		n_s^*	n_p^*	μ (a.u.)	Γ (MHz)	Γ (MHz)
H	1s→ 2p	1.000	2.000	0.745	99.52	99.47
He*	2s→ 2p	1.689	1.938	2.540	1.64	1.62
Li	2s→ 2p	1.589	1.959	2.352	5.93	5.92
Na	3s→ 3p	1.627	2.117	2.445	9.43	10.01
K	4s→ 4p	1.770	2.235	2.842	5.78	6.09
Rb	5s→ 5p	1.805	2.293	2.917	5.78	5.56
Cs	6s→ 6p	1.869	2.362	3.093	4.99	5.18

↑

this is $\langle l'\alpha' | r | l\alpha \rangle$

Dan Steck:

includes the signs

	$m_F = -2$	$m_F = -1$	$m_F = 0$	$m_F = 1$	$m_F = 2$
$F' = 3$	$-\sqrt{\frac{1}{6}}$	$-\sqrt{\frac{4}{15}}$	$-\sqrt{\frac{3}{10}}$	$-\sqrt{\frac{4}{15}}$	$-\sqrt{\frac{1}{6}}$
$F' = 2$	$-\sqrt{\frac{1}{6}}$	$-\sqrt{\frac{1}{24}}$	0	$\sqrt{\frac{1}{24}}$	$\sqrt{\frac{1}{6}}$
$F' = 1$		$\sqrt{\frac{1}{40}}$	$\sqrt{\frac{1}{30}}$	$\sqrt{\frac{1}{40}}$	

D2 line,
$F = 2 \to F'$
$\hat{\varepsilon} = \hat{q}$

$$= \frac{\langle F' m_F' | r_q | F m_F \rangle}{\langle J=1/2 \| r \| J=3/2 \rangle}$$

measured: $4.227(5)$ a.u. $= 4.227(5)$ ea_0

(for D1, $\langle J=1/2 \| r \| J=1/2 \rangle = 2.992(3)$ ea_0)

from excited state lifetimes

$$\frac{\langle F m_F | r_q | F' m_F' \rangle}{\langle J \| r \| J' \rangle} = (-1)^{F'-1+m_F} \sqrt{2F+1} \begin{pmatrix} F' & 1 & F \\ m_F' & q & -m_F \end{pmatrix} \cdot (-1)^{F'+J+1+I} \sqrt{(2F'+1)(2J+1)} \begin{Bmatrix} J & J' & 1 \\ F' & F & I \end{Bmatrix}$$

and

$$\langle J \| r \| J' \rangle = \langle L \| r \| L' \rangle (-1)^{J'+L+1+S} \sqrt{(2J'+1)(2L+1)} \cdot \begin{Bmatrix} L & L' & 1 \\ J' & J & S \end{Bmatrix}$$

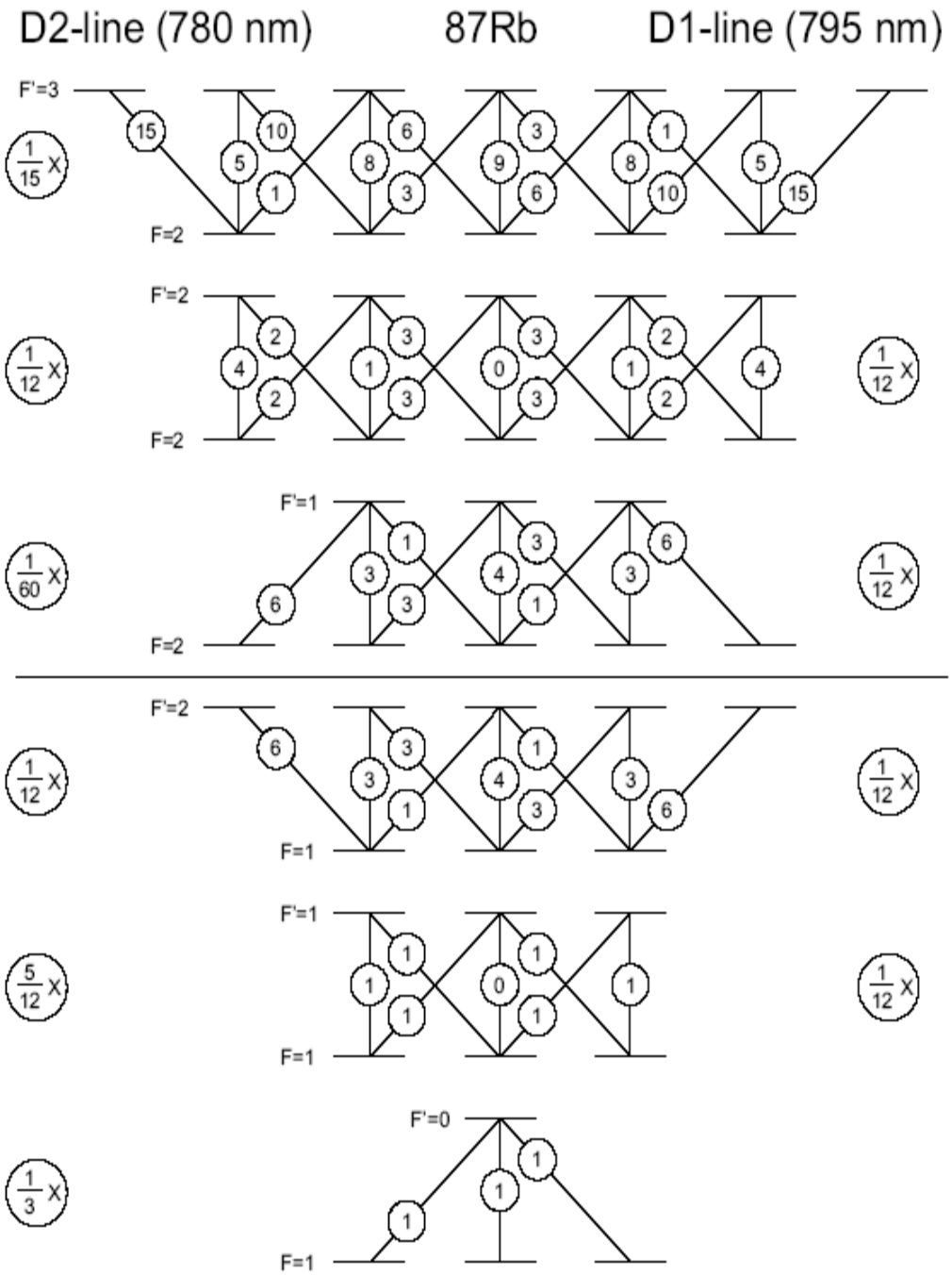
$$\frac{\langle J \| r \| J' \rangle}{\langle L \| r \| L' \rangle} = \sqrt{2} \quad \text{for D2 line}$$

$$= 1 \quad \text{for D1 line}$$

Make sure that you include these factors if you use $\langle l' \alpha' | r | l \alpha \rangle$!

From Eric Cornell / Carl Wieman's groups

No signs!



take the square root