

Combining laser light with synchrotron radiation

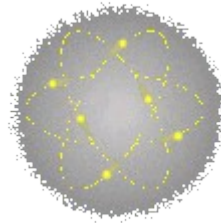
Part 5

Optical pumping

Interaction of Atoms with visible Light

Green light wavelength 5000\AA

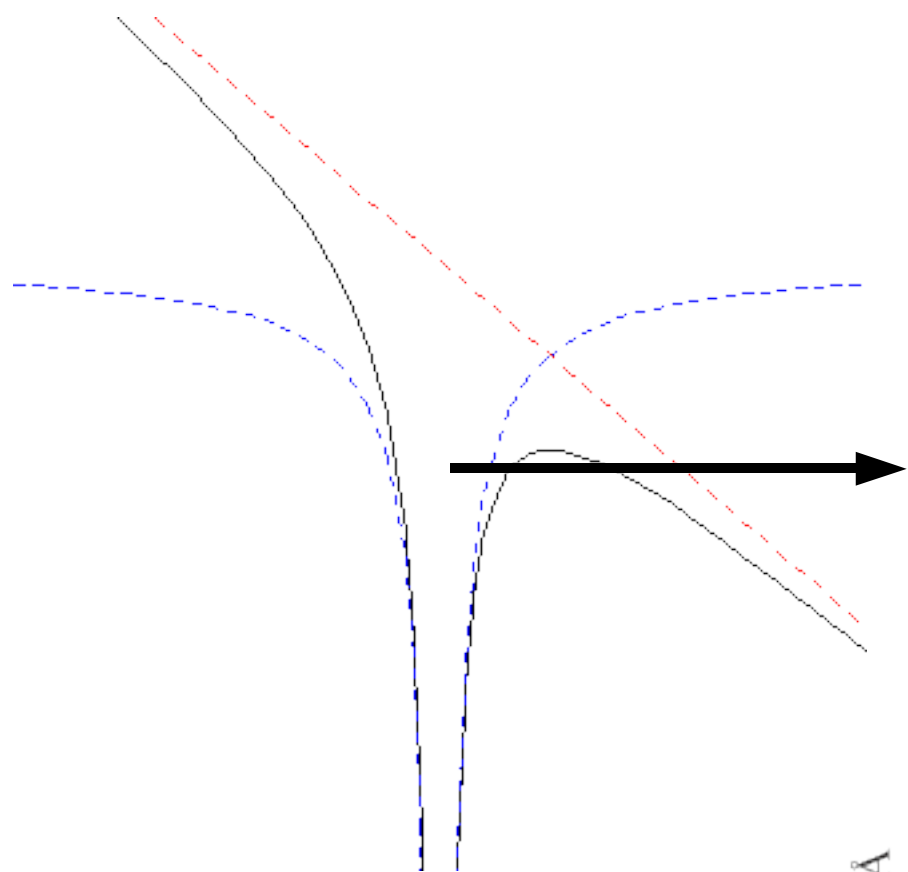
$\sim 1\text{\AA}$



The wavelength is much larger than the atom

The light electric field is weak with respect to the coulomb force

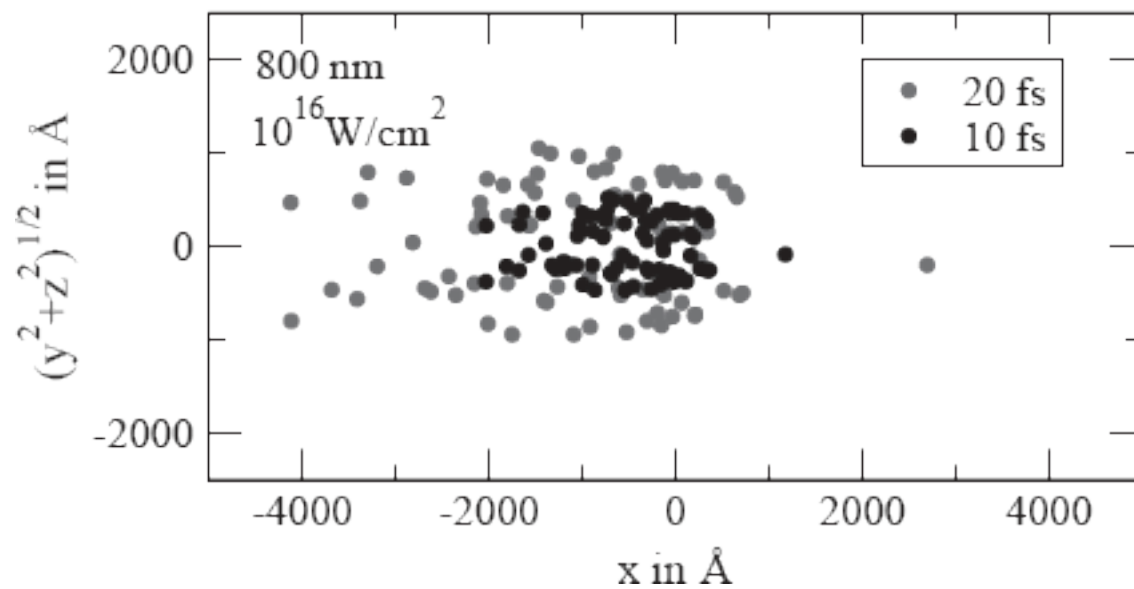
Strong electric field



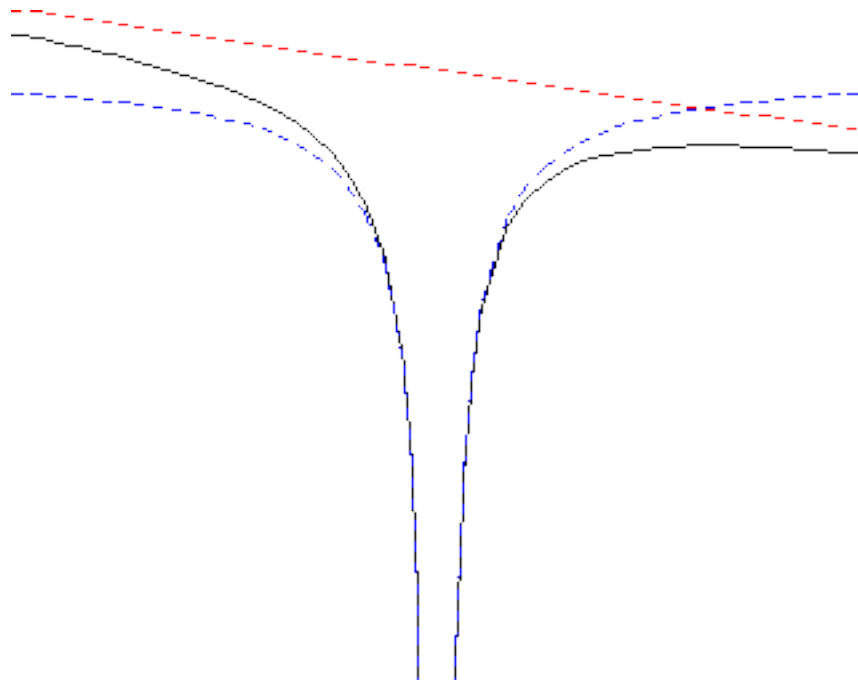
Tunnel
ionization

Classical Simulations

Xe₁₃ 78 free electrons



Weak electric field



The light field is only a weak perturbation of the coulomb field

Perturbation theory → Part 1 of this lecture

Oscillator Model

Hooke's Law:

If a material body is slightly deformed, the restoring force is linear to the deformation.

If the electron shell of an atom is slightly deformed, the restoring force is linear to the deformation.

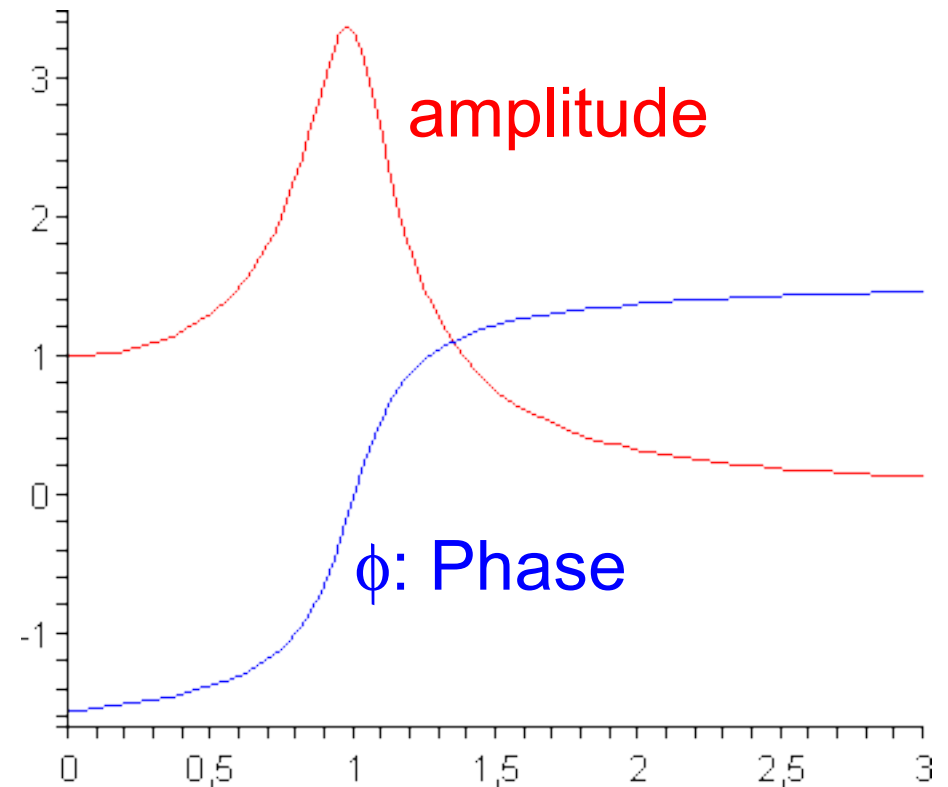
A linear restoring force leads to harmonic oscillations.

Driven harmonic oscillator

$$m \frac{d^2 x}{dt^2} + r \frac{dx}{dt} + kx = F_0 \cos(\omega t)$$

$$x(t) = \frac{F_0}{\omega \sqrt{r^2 + (\omega m - k/\omega)^2}} \sin(\omega t - \phi)$$

$$\phi = \arctan\left(\frac{\omega m - k/\omega}{r}\right)$$



Atomic wave functions

Two energy eigenstates of an atomic system:

$$\Psi_1(\vec{r}, t) = u_1(\vec{r}) \exp\left[-i(E_1/\hbar)t\right]$$

$$\Psi_2(\vec{r}, t) = u_2(\vec{r}) \exp\left[-i(E_2/\hbar)t\right]$$

Superposition of eigenstates is a solution of the time dependent Schrödinger-equation:

$$\Psi = a_1 \Psi_1 + a_2 \Psi_2$$

with $|a_1|^2 + |a_2|^2 = 1$

Atomic Susceptibility

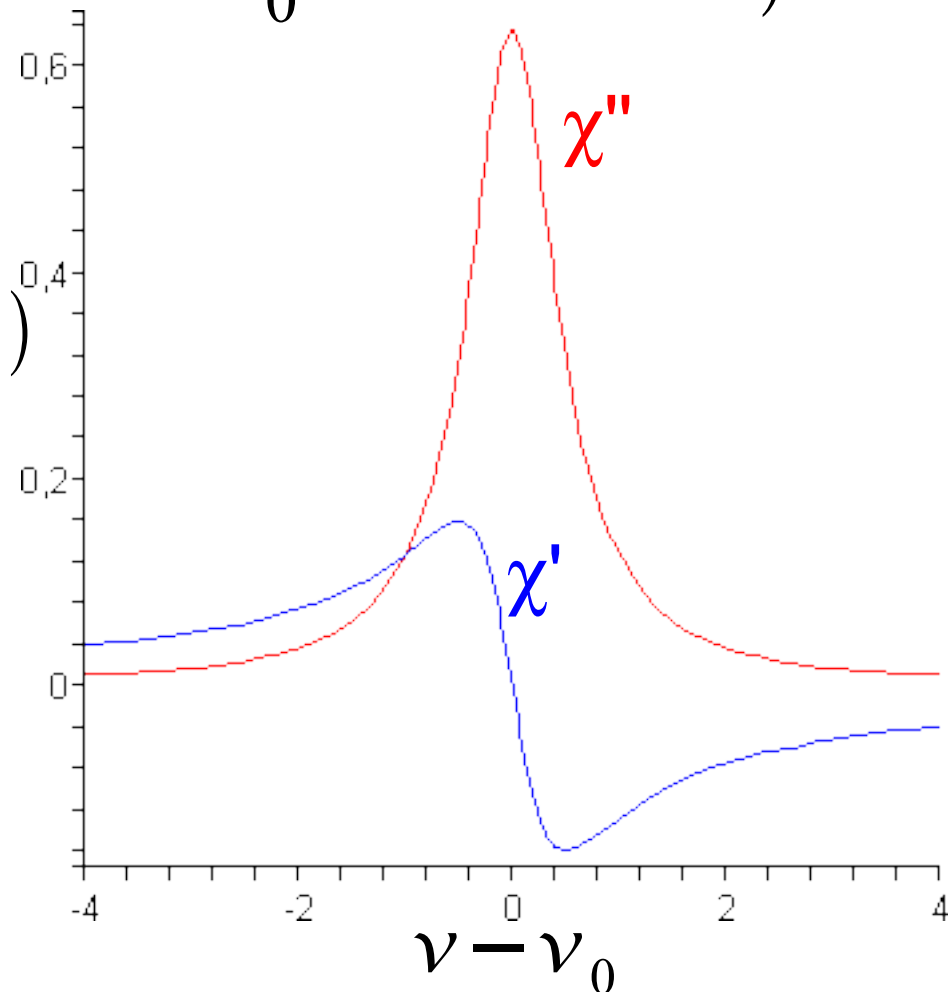
$$\vec{E}(t) = \vec{E}_0 \cos \omega t$$

$$\vec{P}(t) = \vec{E}_0 (\epsilon_0 \chi' \cos \omega t + \epsilon_0 \chi'' \sin \omega t)$$

$$\chi''(\nu) \propto \Delta N g(\nu)$$

$$\chi'(\nu) \propto \Delta N (\nu_0 - \nu) g(\nu)$$

$$g(\nu) = \frac{\Delta \nu / 2\pi}{(\nu - \nu_0)^2 + \left(\frac{\Delta \nu}{2}\right)^2}$$



In phase response

$$\chi'(\nu) \propto \Delta N (\nu_0 - \nu) g(\nu)$$

$$\epsilon = (1 + \chi') \epsilon_0$$

Dispersive signal.

Influences the speed of light far from the resonance

No energy transfer

Out of phase response

$$\chi''(\nu) \propto \Delta N g(\nu)$$

Absorption signal

Lorentzian line shape

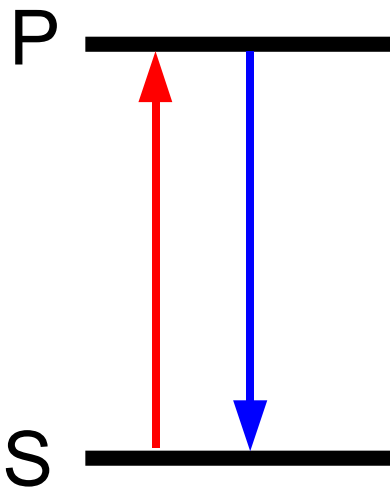
Proportional to transition probability

Both signals saturate if $\Delta N \rightarrow 0$

Optical pumping

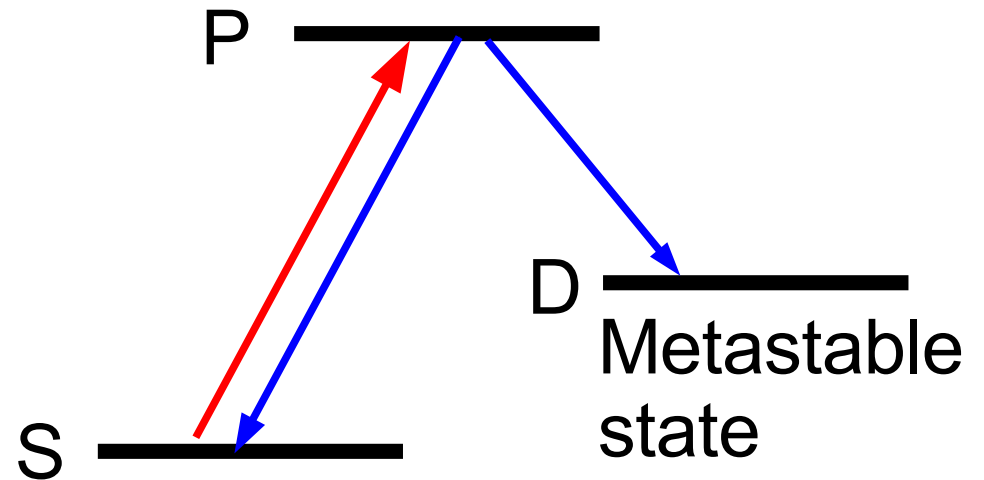
Excitation of atoms into different states

Alkali-Atoms



Two level system

Alkaline earths

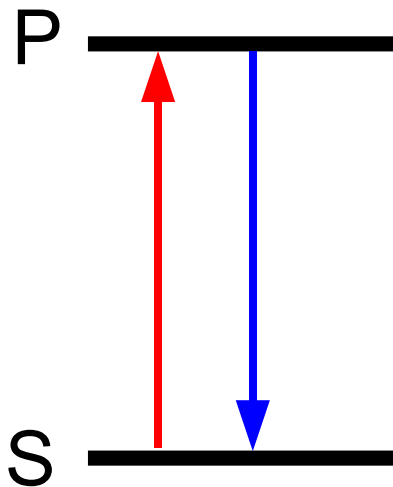


Λ -Type three level system

Two Level Systems

Alkali Metals

Two fine structure levels in the upper state, but no allowed transition between them.



Na: $3s \rightarrow 3p_{3/2}$: 589.0 nm, $A=6.2 \cdot 10^7 \text{ s}^{-1}$

$3s \rightarrow 3p_{1/2}$: 589.6 nm, $A=6.1 \cdot 10^7 \text{ s}^{-1}$

K: $4s \rightarrow 4p_{3/2}$: 766.5 nm, $A=3.8 \cdot 10^7 \text{ s}^{-1}$

$4s \rightarrow 4p_{1/2}$: 769.9 nm, $A=3.7 \cdot 10^7 \text{ s}^{-1}$

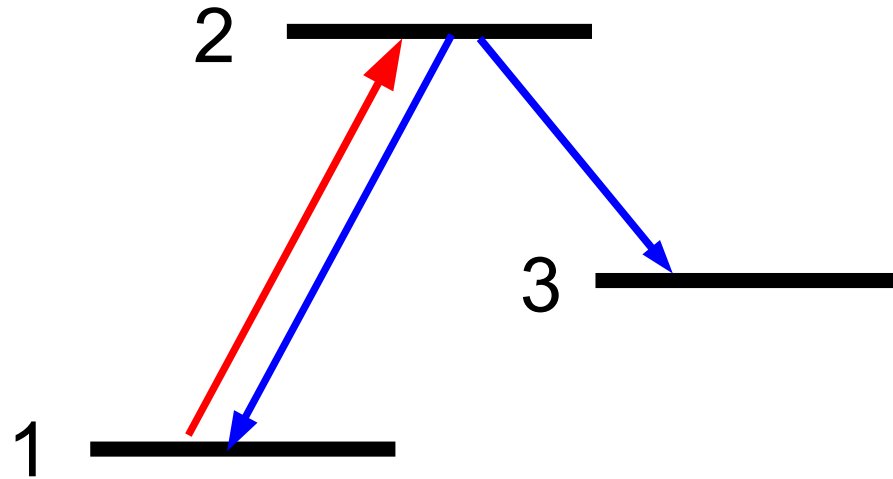
Rb: $5s \rightarrow 5p_{3/2}$: 780.0 nm, $A=3.7 \cdot 10^7 \text{ s}^{-1}$

$5s \rightarrow 5p_{1/2}$: 794.8 nm, $A=3.4 \cdot 10^7 \text{ s}^{-1}$

Cs: $6s \rightarrow 6p_{3/2}$: 852.1 nm, $A=3.3 \cdot 10^7 \text{ s}^{-1}$

$6s \rightarrow 6p_{1/2}$: 894.3 nm, $A=2.9 \cdot 10^7 \text{ s}^{-1}$

Λ -Type Three level System

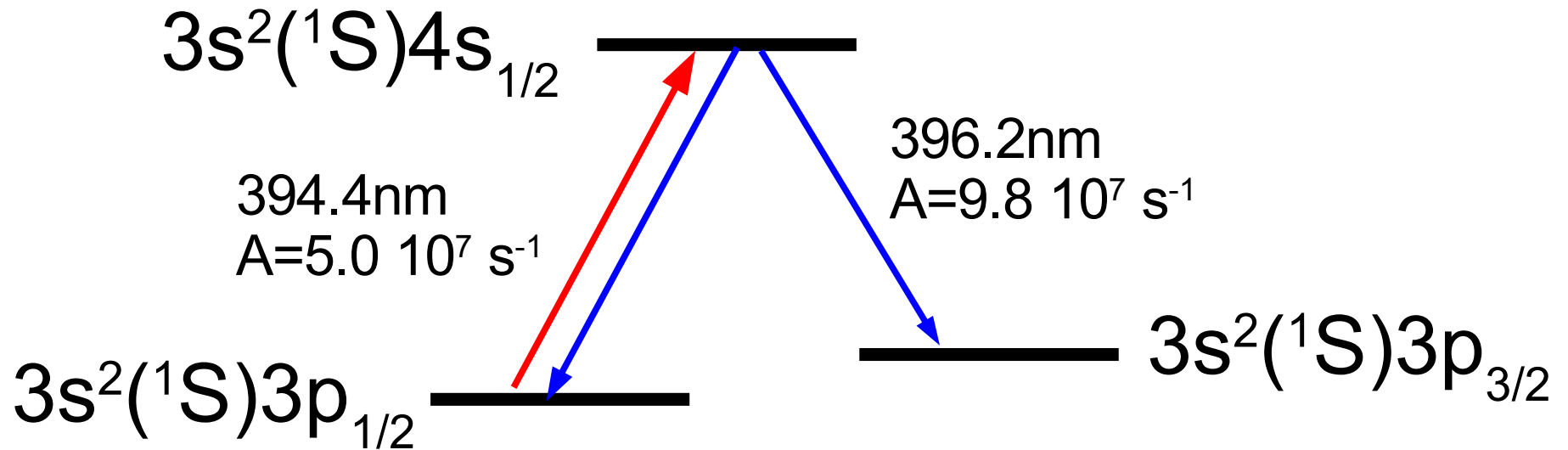


$$\frac{dN_3}{dt} = N_2 A_{23}$$

$$\frac{dN_2}{dt} = +(N_1 - N_2) B_{12} \rho - N_2 A_{21} - N_2 A_{23}$$

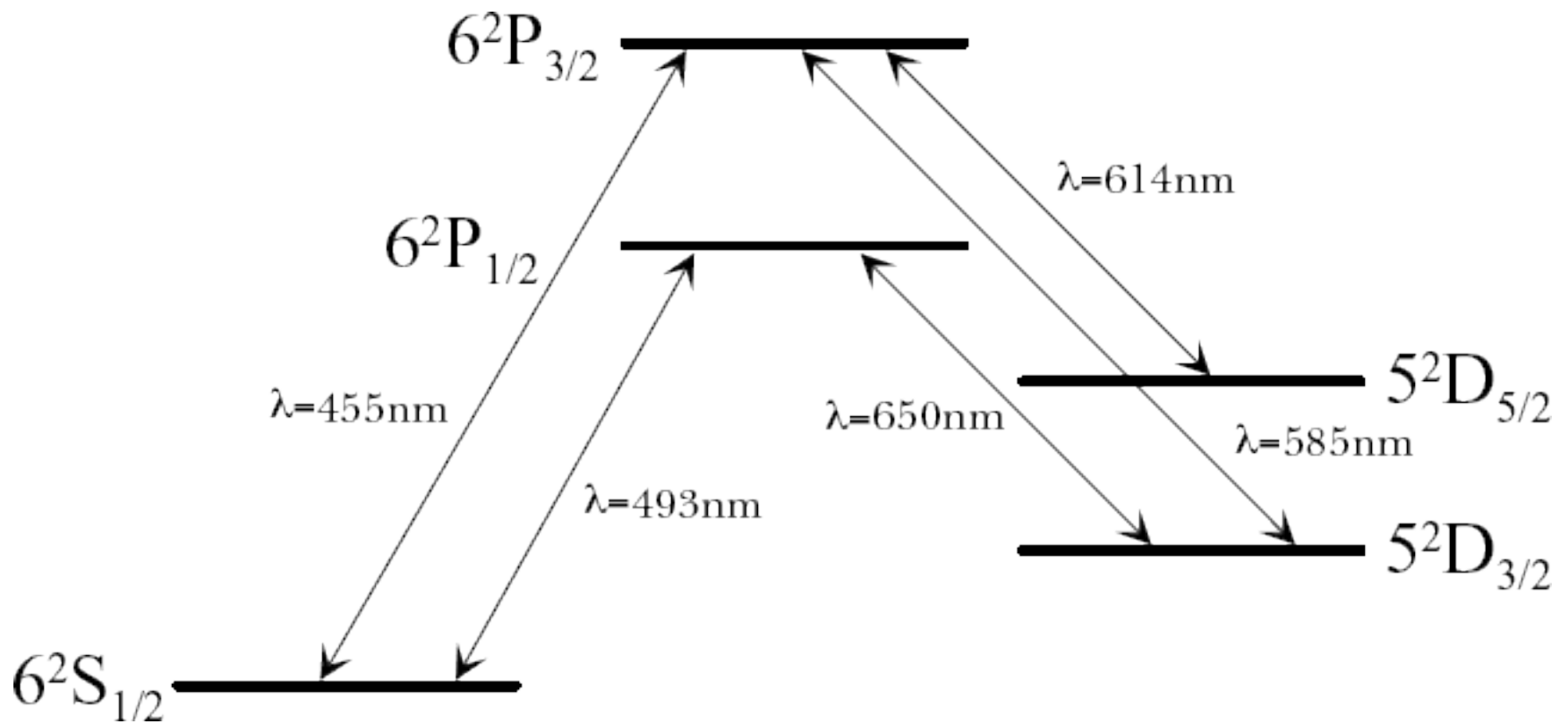
$$\frac{dN_1}{dt} = -(N_1 - N_2) B_{12} \rho + N_2 A_{21}$$

Λ -Type Three level Systems Aluminum

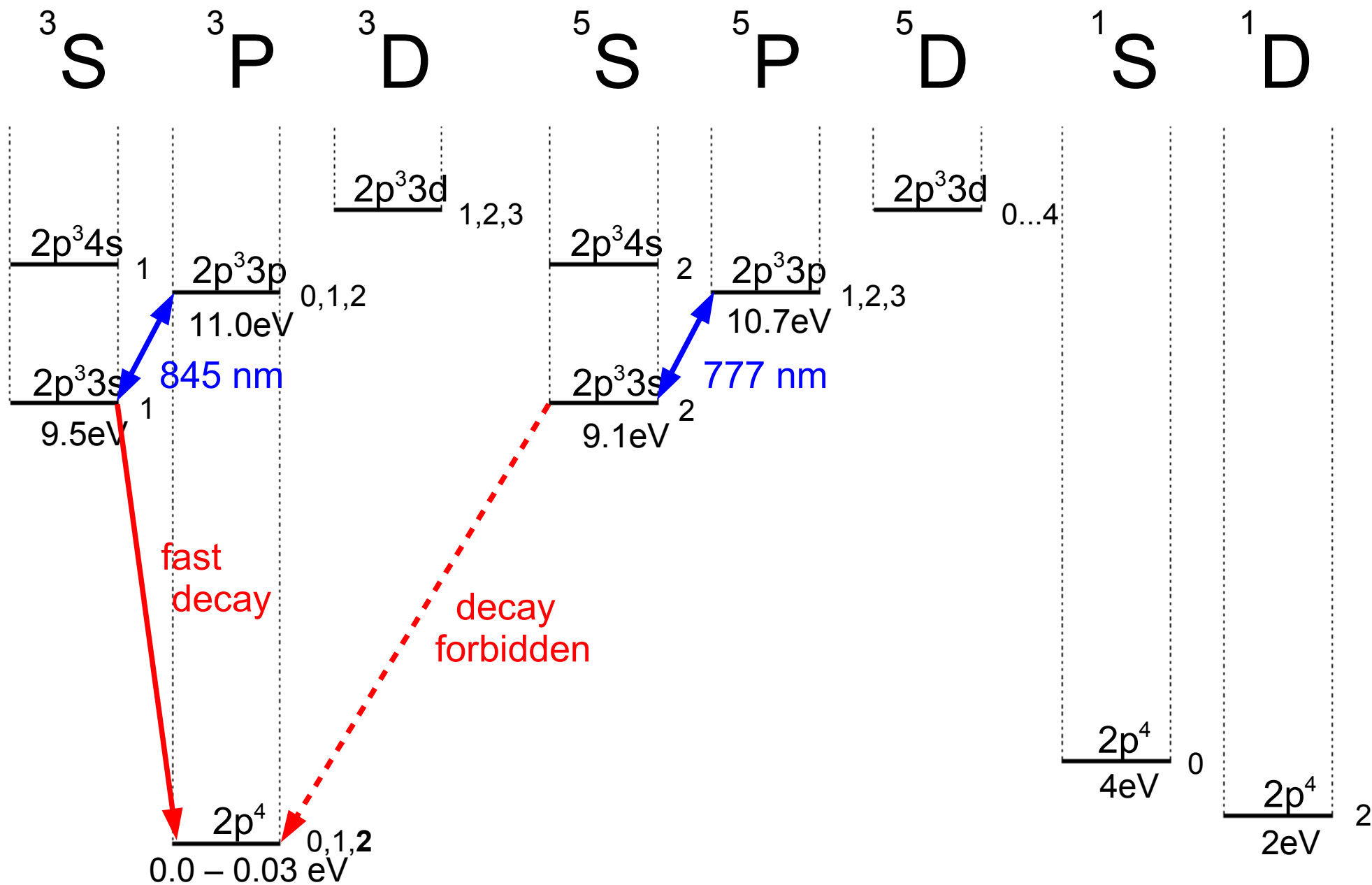


Similar systems: Ga, In

Λ -Type Three level Systems Barium Ion

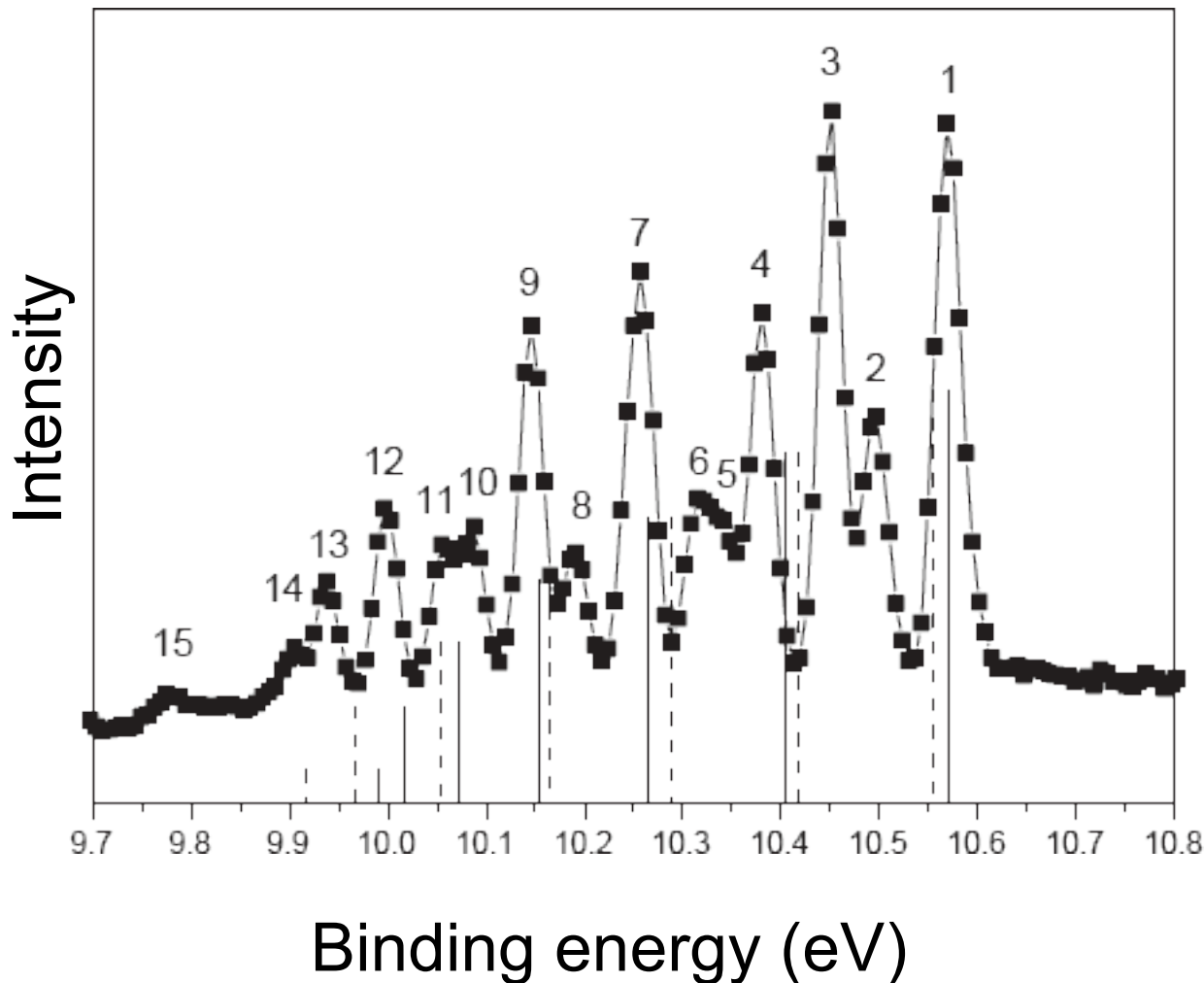


Oxigen Term Scheme



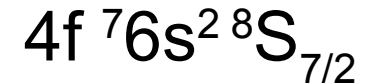
Fine Structure

With 3rd generation synchrotron sources the fine structure of photoelectron emission can be studied in great detail.

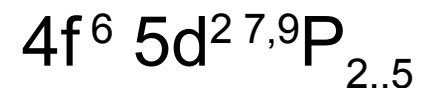
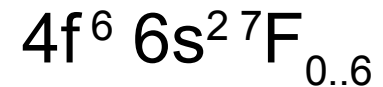


Eu 4f photoemission

Ground state:



Final states:

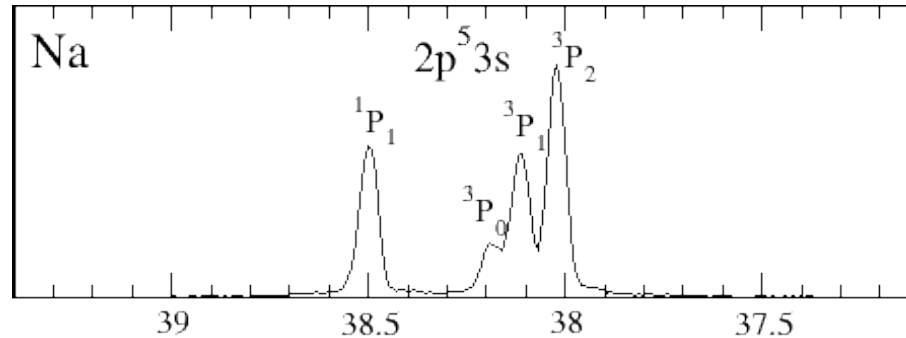


...and unclassified lines

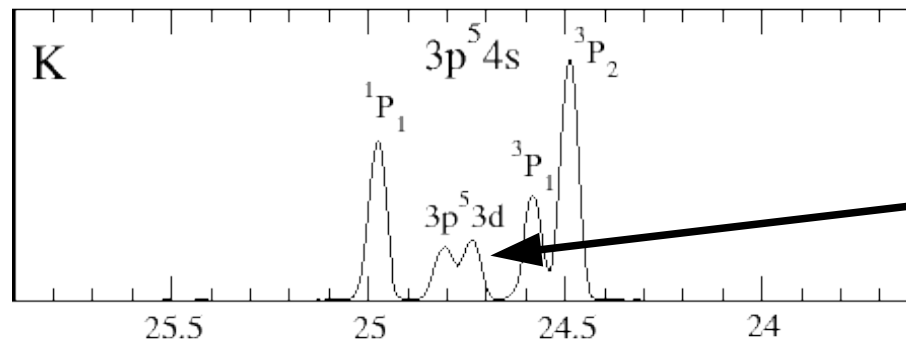
The fine structure of alkali metals

$2p^5 3s$

pure
LS-coupling

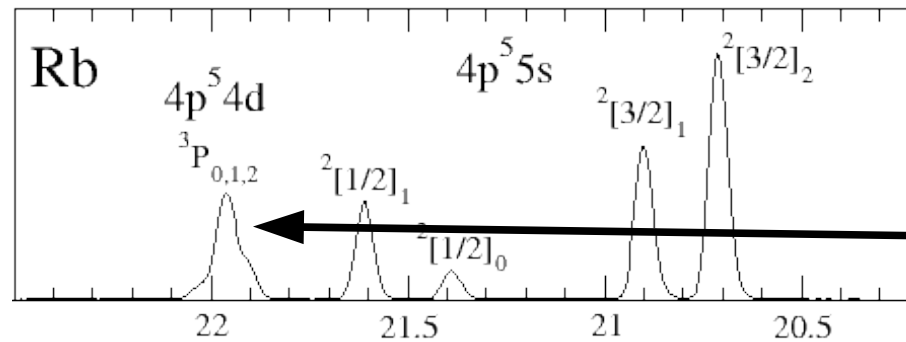


$3p^5 4s$



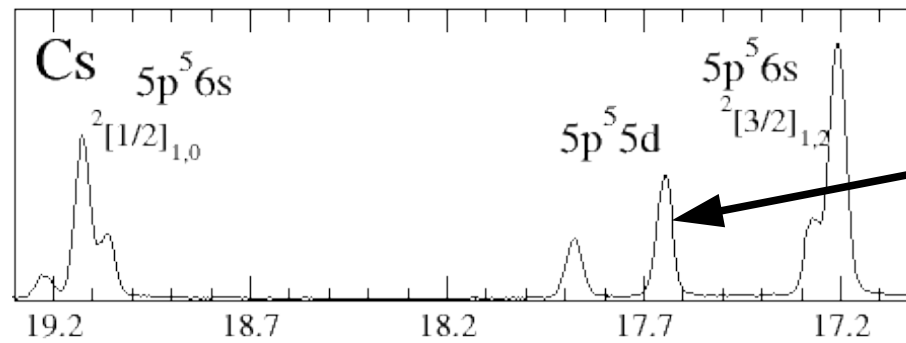
$3p^5 3d$

$4p^5 5s$



$4p^5 4d$

$5p^5 6s$

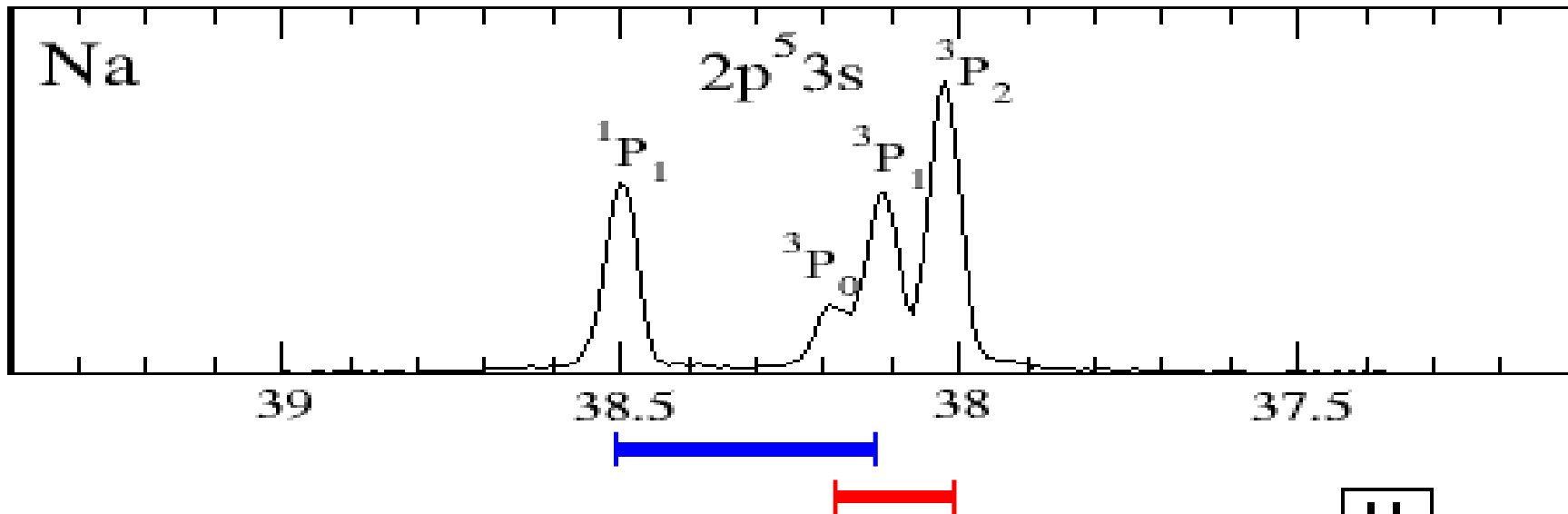


$5p^5 5d$

jj-coupling

binding energy

Na: LS coupling



Couple a 2p hole to a 3s electron:

Coupling of two spin $\frac{1}{2}$ leads to 0 and 1

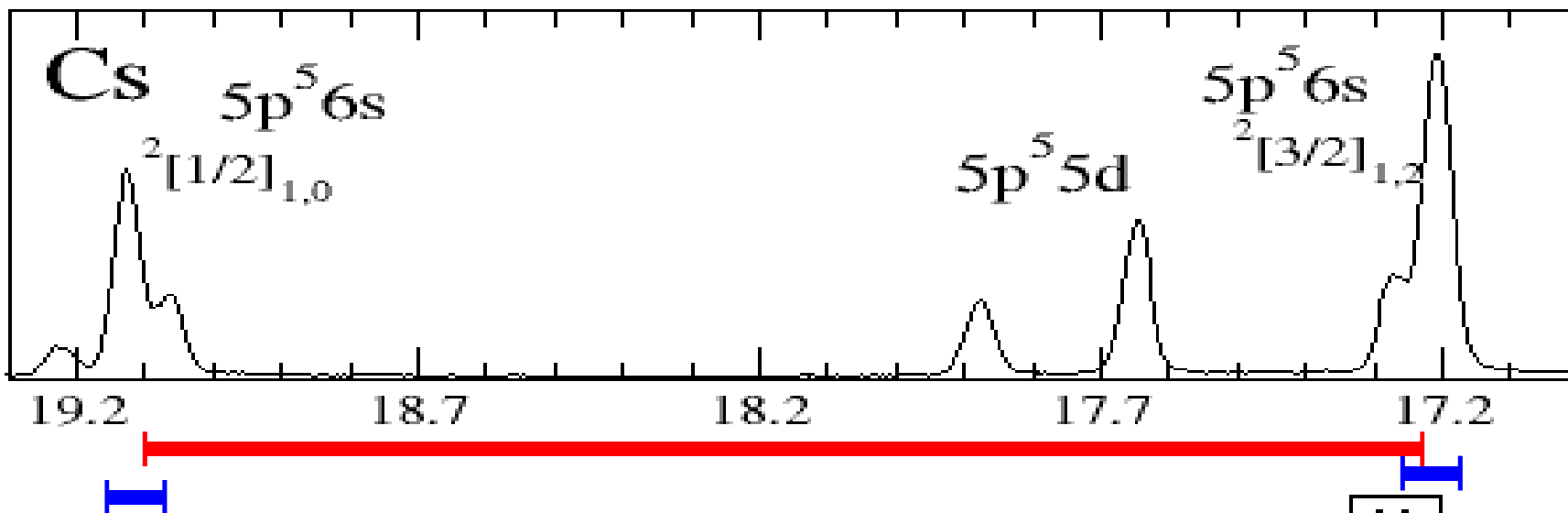
$^1P \leftrightarrow ^3P$: Coulomb exchange splitting

Coupling of the spin to orbit $P=1$ gives

$J=0,1,2$: relativistic Spin-Orbit splitting

H					
Li	Be				
Na	Mg				
K	Ca	Sc	Ti	V	
Rb	Sr	Y	Zr	N	
Cs	Ba		Hf	T	
Fr	Ra		Rf	D	
			La	Ce	P
			Ac	Th	P

Cs: jj coupling



Couple a 2p hole to a 3s electron:

Coupling of the 2p spin to it's orbit gives

$j=1/2, 3/2$: relativistic Spin-Orbit splitting

Coupling to the spin of the 6s electron

$${}^2[3/2]_1 \leftrightarrow {}^2[3/2]_2$$

H					
Li	Be				
Na	Mg				
K	Ca	Sc	Ti	V	
Rb	Sr	Y	Zr	Ni	
Cs	Ba		Hf	Ta	
Fr	Ra		Rf	D	
			La	Ce	P
			Ac	Th	P

Orientation and Alignment

Every fine structure state with $l > 0$ splits up into magnetic substates

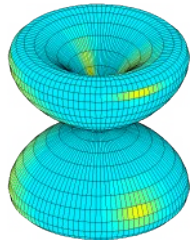
Magnetic
quantum number

d-Orbitals

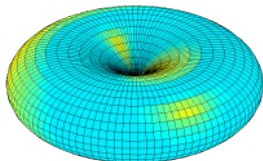
Direction in space
(alignment)



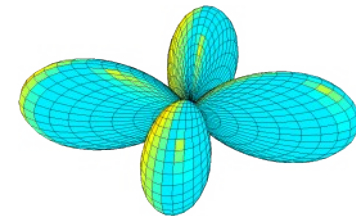
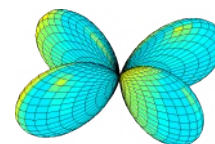
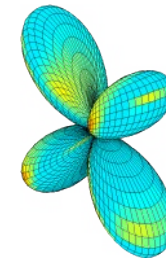
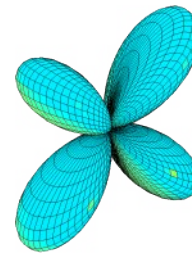
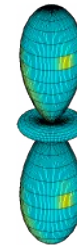
$m=0$



$m=\pm 1$

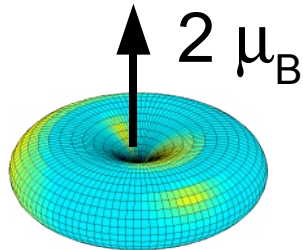


$m=\pm 2$



Orientation

The magnetic moments of the atoms show in a defined direction

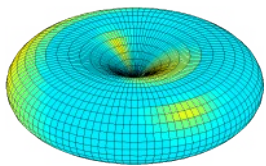


$m=2$

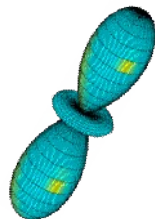
Pumped with circularly polarized light

Alignment

No magnetic field, but an axis in space is preferred or avoided



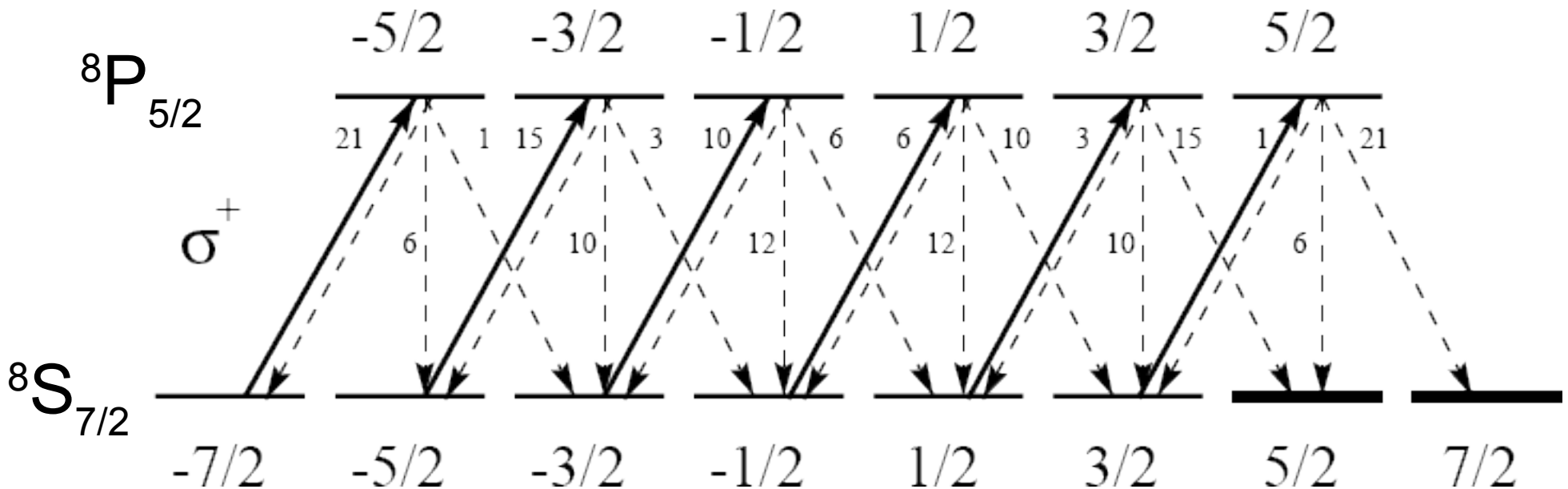
$m=2$ & $m=-2$



Pumped with linearly polarized light

Pumping into Orientation

Example: Europium $6s^2 \rightarrow 6s6p$



All atoms are pumped into the $m_j = 5/2, 7/2$ states

The final states are **spin** polarized

Multipole moments

Normalized dipole moment (orientation)

$$A_{10} = K_1(J) \sum_{m_J} m_J N_{J m_J}$$

Normalized quadrupole moment (alignment)

$$A_{20} = K_2(J) \sum_{m_J} (J(J+1) - 3m_J^2) N_{J m_J}$$

with:

Normalization constants: K_1, K_2, \dots

Occupation numbers: N_{Jm}

General definition of Multipole moments

$$\rho_{k_0 q_0}(J) = \sum_{m_J, m'_J} (-1)^{J-m'_J} (J m_J, J - m'_J | k_0 q_0) \langle J m'_J | \rho | J m_J \rangle$$

$$\rho_{00}(J) = \frac{1}{\sqrt{2J+1}} \sum_{m_J} \langle J m_J | \rho | J m_J \rangle$$

$$A_{k_0 0}(J) = \frac{\rho_{k_0 0}}{\rho_{00}}$$