MANCHESTER 1824

The University of Manchester

Introduction to Lasers II

Jon Billowes

LA³NET School

Introduction to Lasers II

- Introduction to atomic physics for laser spectroscopy,
- Natural linewidth and line broadening
- Isotope shifts and nuclear properties
- Hyperfine structures and nuclear moments
- Laser spectroscopy techniques for radioacitve atoms
- Laser frequency-doubling with non-linear crystals
- Magneto-optical atom traps

(Ionization schemes – will be covered by Bruce Marsh tomorrow)

Energy scales and units used

Wavelength, λ: SI unit = m [or μm, nm or Angström, 1 Å = 10^{-10} m]

λ is *dependent* on the (refractive index of the) medium in which the wave travels

Frequency, ν: SI unit = Hz (i.e., cycles s^{-1}) [or MHz = 10^6 Hz , GHz = 10^9 Hz]

frequency is *independent* of the medium

Energy, E: SI unit = J,

BUT : It is hard to measure energy directly. Spectra are recorded as line intensities as a function of frequency or wavelength.

The conversion to energy *appears* simple: **$E = h\nu = hc/\lambda$**

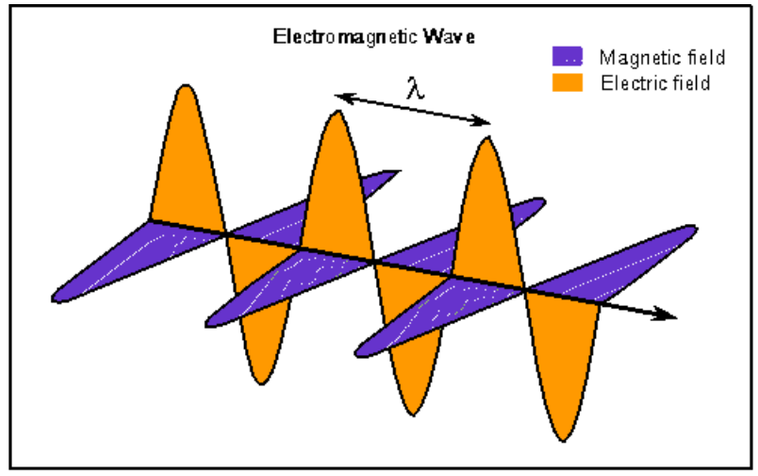
But *h* is only known to 8 significant figures. Hence, it is convenient to introduce

Wavenumber, a *property* defined as reciprocal of the vacuum wavelength: and whose units are universally quoted as **cm^{-1}** (*n.b.* not m^{-1})

$$\bar{\nu} = \frac{1}{\lambda_{vac}}$$

Wavenumber is directly proportional to **energy**, $E = hc\bar{\nu}$ and thus we commonly quote "energies" in units of cm^{-1}.

Properties of Photons



Light is electromagnetic radiation. It is composed of individual “photons” which carry an energy proportional to their frequency

Properties of the photon:

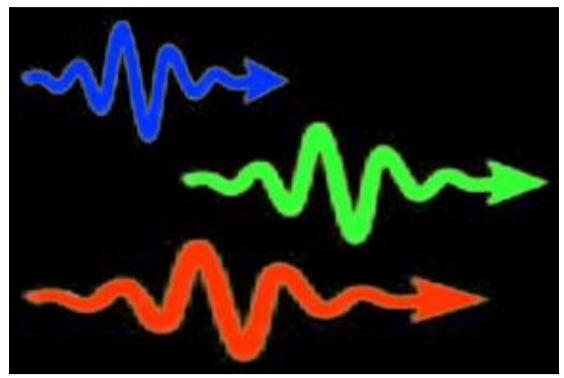
Wavelength: λ Frequency: ν Speed of light: $c = 2.998 \times 10^8$ m/sec

Relation: $c = \lambda\nu$

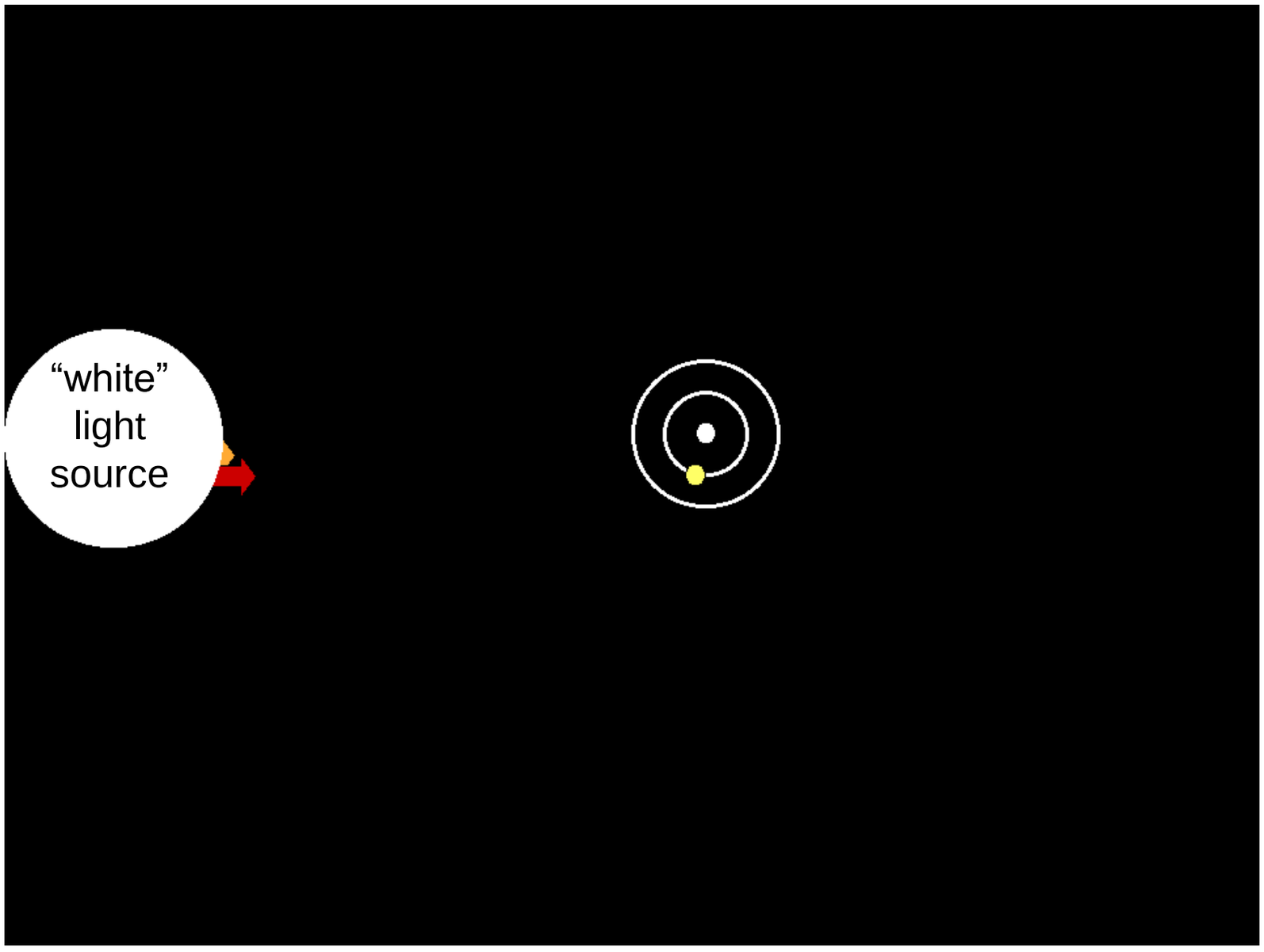
Energy: $E = h\nu$ (and alternatively $E = hc/\lambda$)

Momentum $P = \hbar k$ where $k = 2\pi/\lambda$

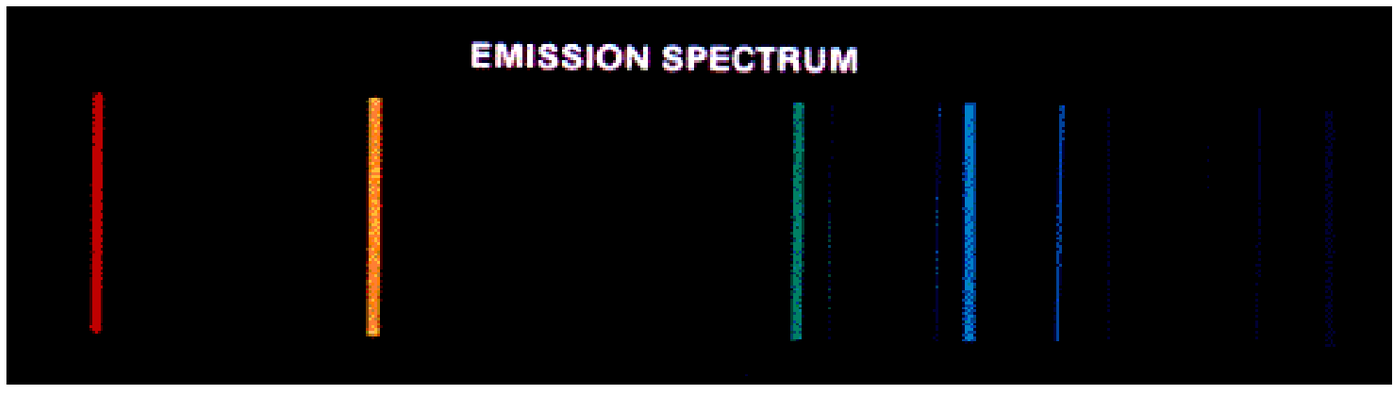
Spin = +1 (σ+) or -1 (σ-) in direction of propagation

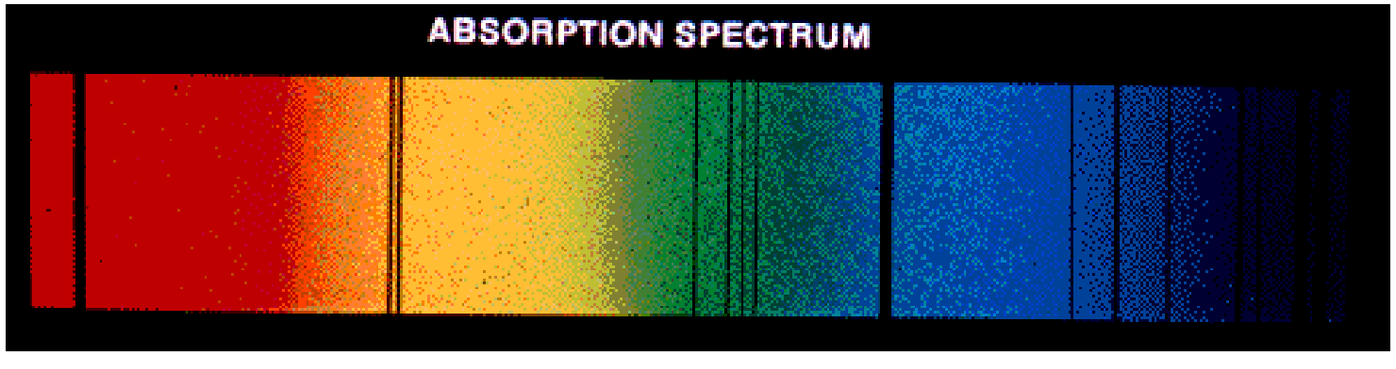


Absorption and emission spectra

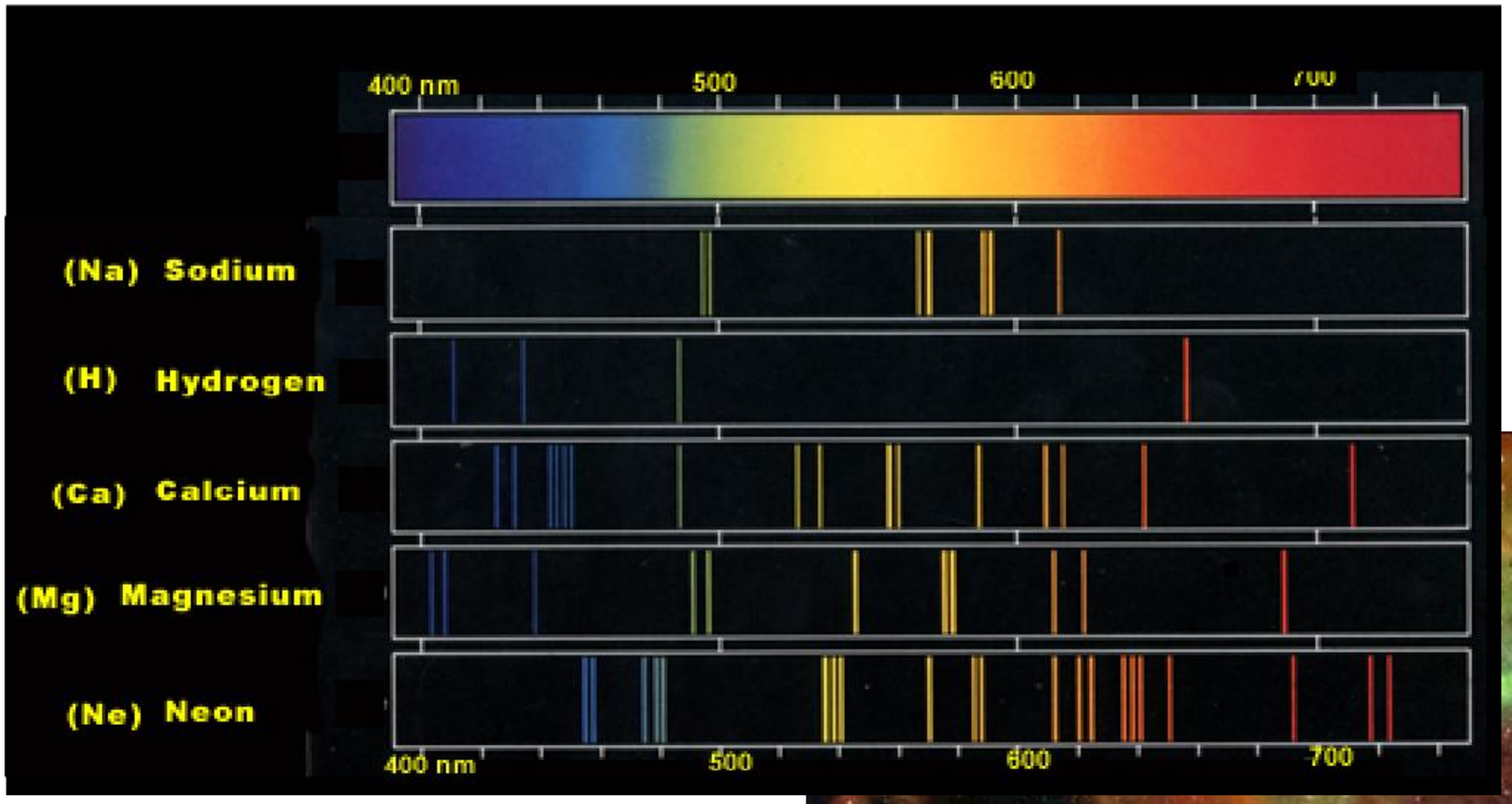


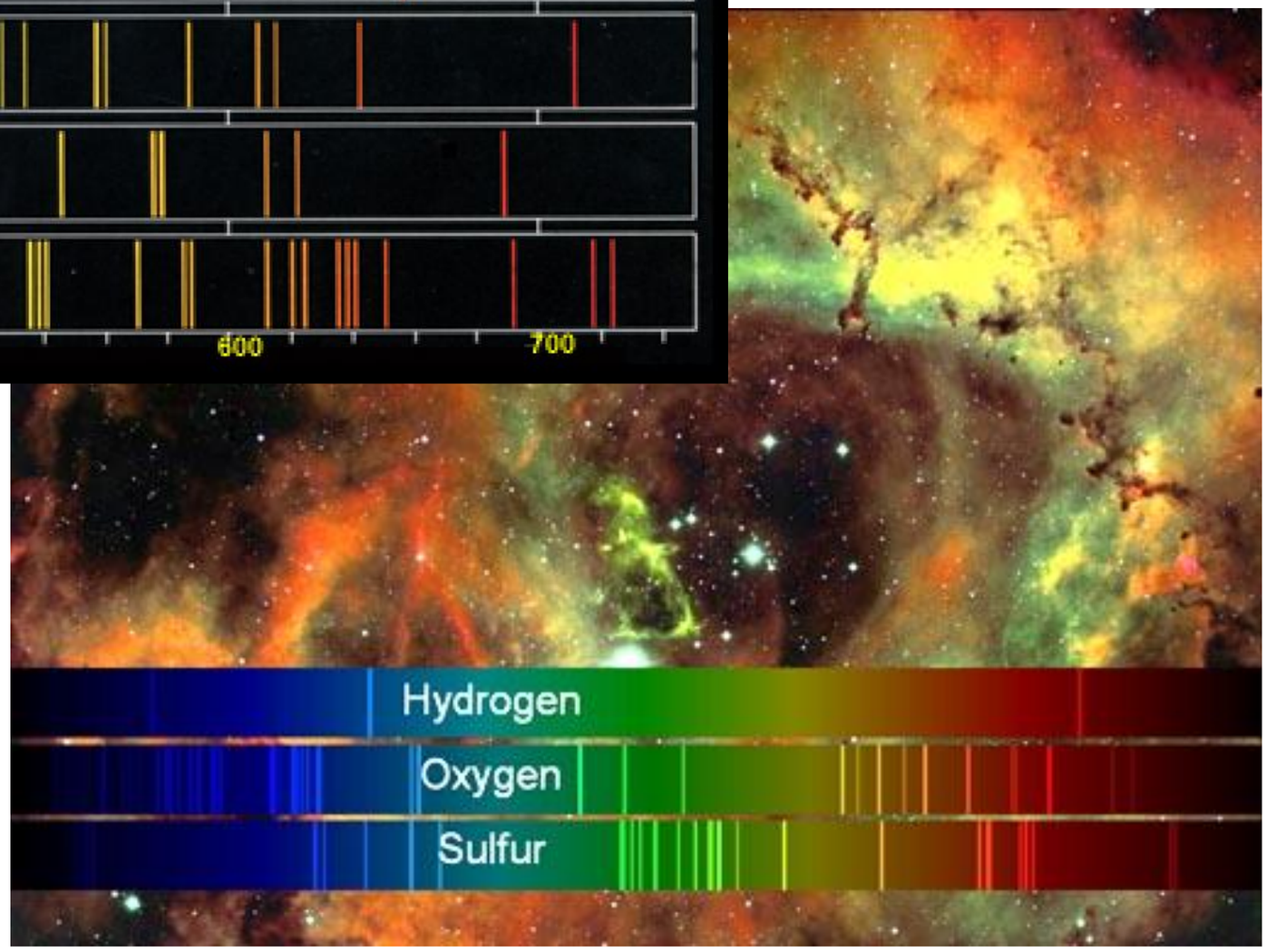
Examples of emission and absorption spectra in the visible region for vapours of two different elements:



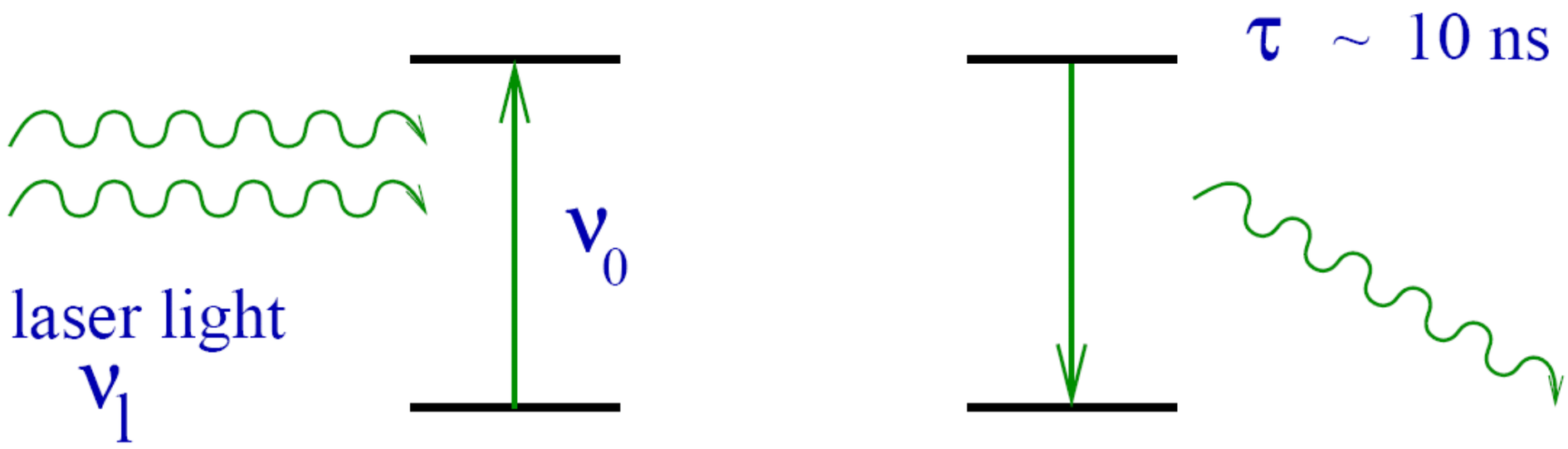


Emission spectra for light elements





Resonant absorption, spontaneous emission



Resonant absorption ($\nu_l = \nu_0$) Spontaneous emission

$$\sigma = \frac{3\lambda^2}{2\pi}$$ (much larger than size of atom)

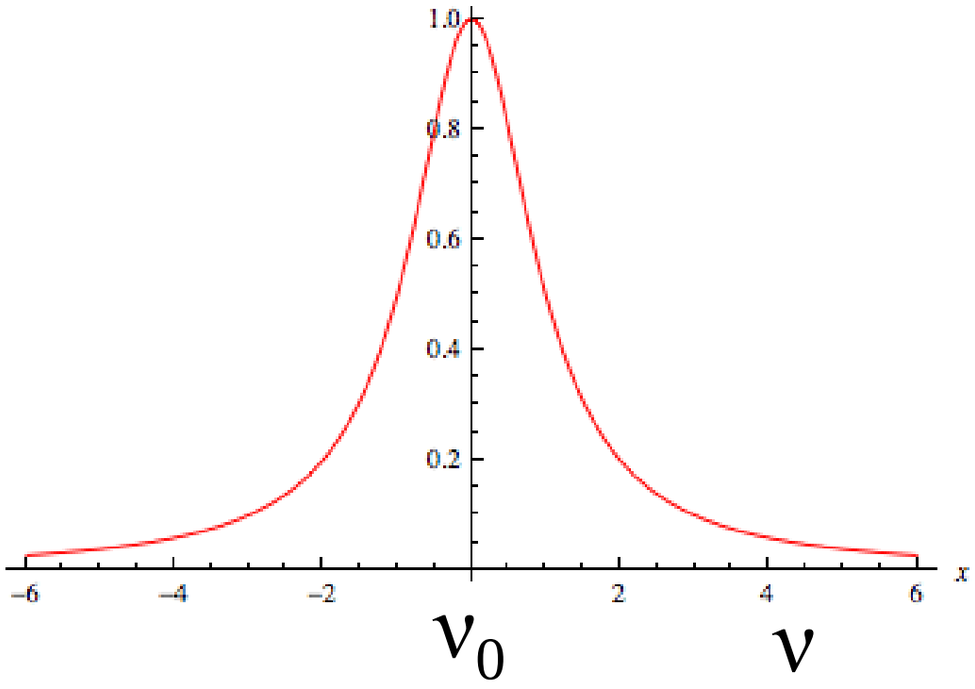
Natural linewidth $\Delta\nu = 1/2\pi\tau$ (Heisenberg uncert.)

~ 16 MHz

single–mode CW laser bandwidth < 1MHz

Natural linewidth has Lorentzian shape:

$L(\nu)$

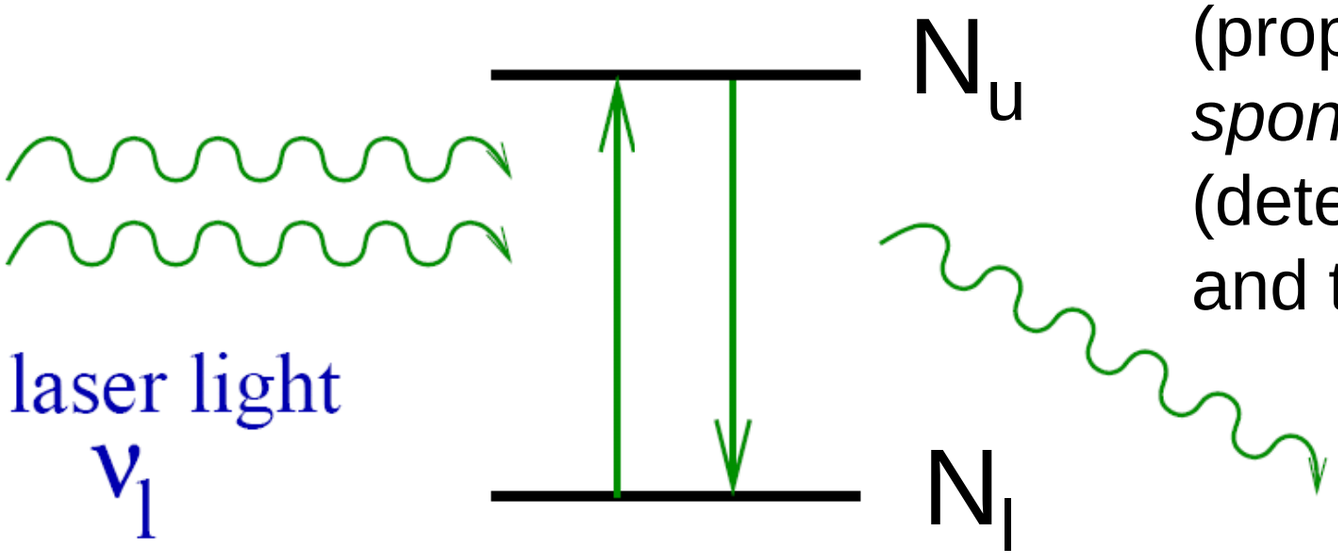


$$L(\nu) = const \times \left\{ \frac{1}{1 + [4\pi\tau(\nu - \nu_0)]^2} \right\}$$

$$FWHM = {}^{1}/_{2\pi\tau}$$

Line broadening

Power Broadening

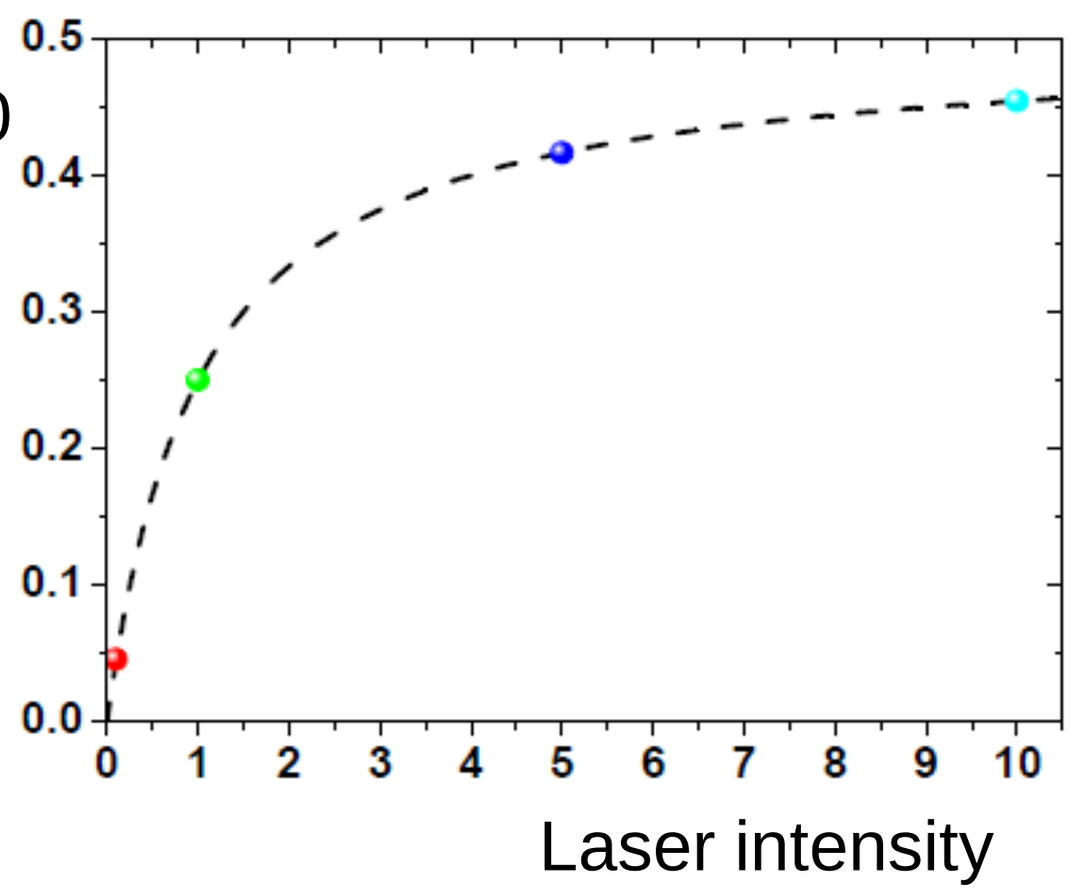


The population of an excited state is determined by the *excitation rate* (proportional to laser intensity), the *spontaneous decay* rate (determined by the natural lifetime) and the *stimulated emission* rate.

Atoms in sample:

$$N_0 = N_l + N_u$$

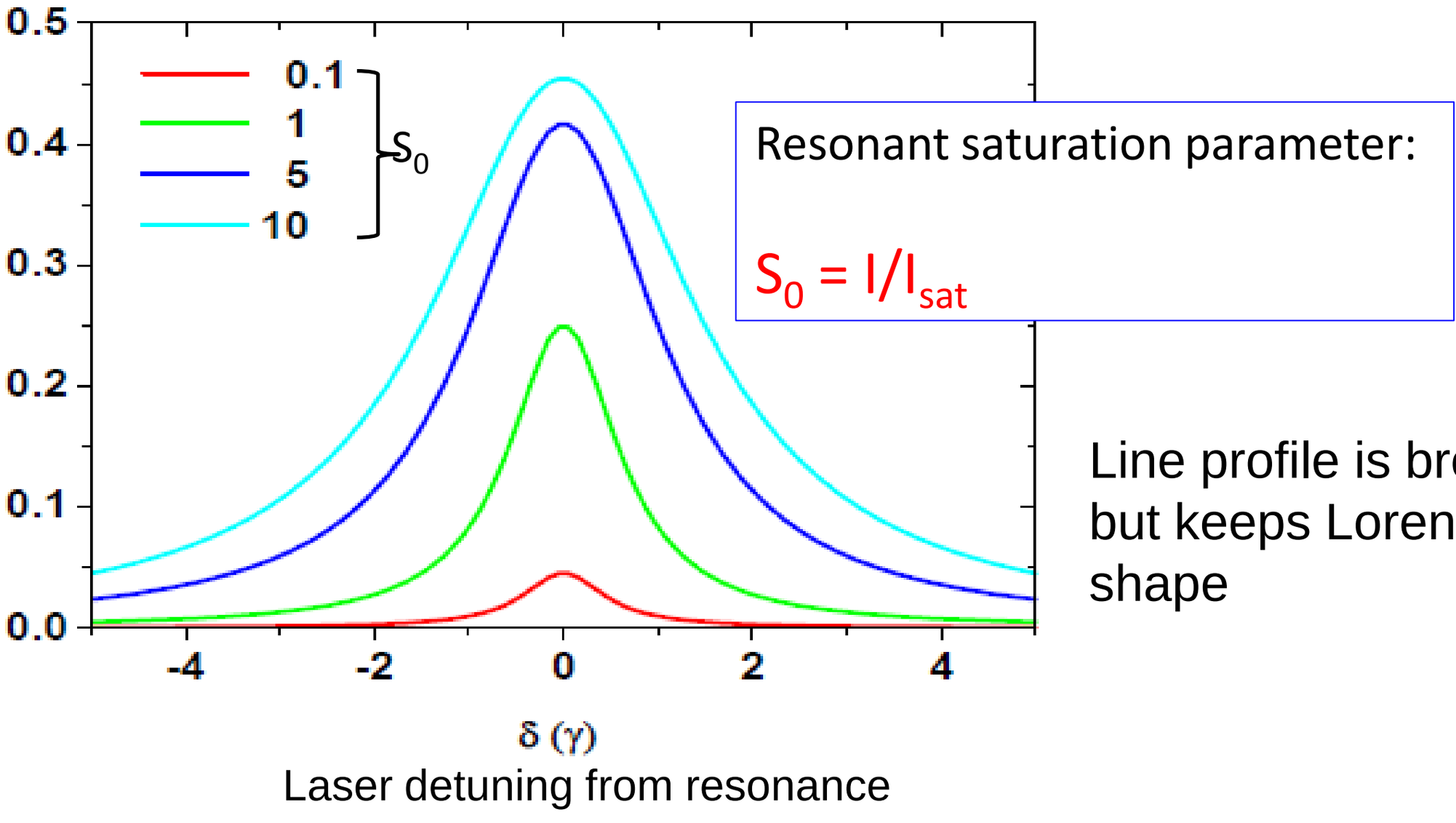
As laser intensity is increased, the fraction in the upper atomic state saturates to 50% - this limits the spontaneous decay rate



Power Broadening

The saturation intensity I_{sat} is that for which the stimulated emission rate is equal to the spontaneous emission rate

$$I_{sat} = \frac{\pi h c}{3\lambda^3 \tau}$$



Laser detuning from resonance

Resonant saturation parameter:

$S_0 = I/I_{sat}$

Line profile is broadened but keeps Lorentzian shape

Main problem in laser spectroscopy:

Doppler broadening (inhomogeneous)

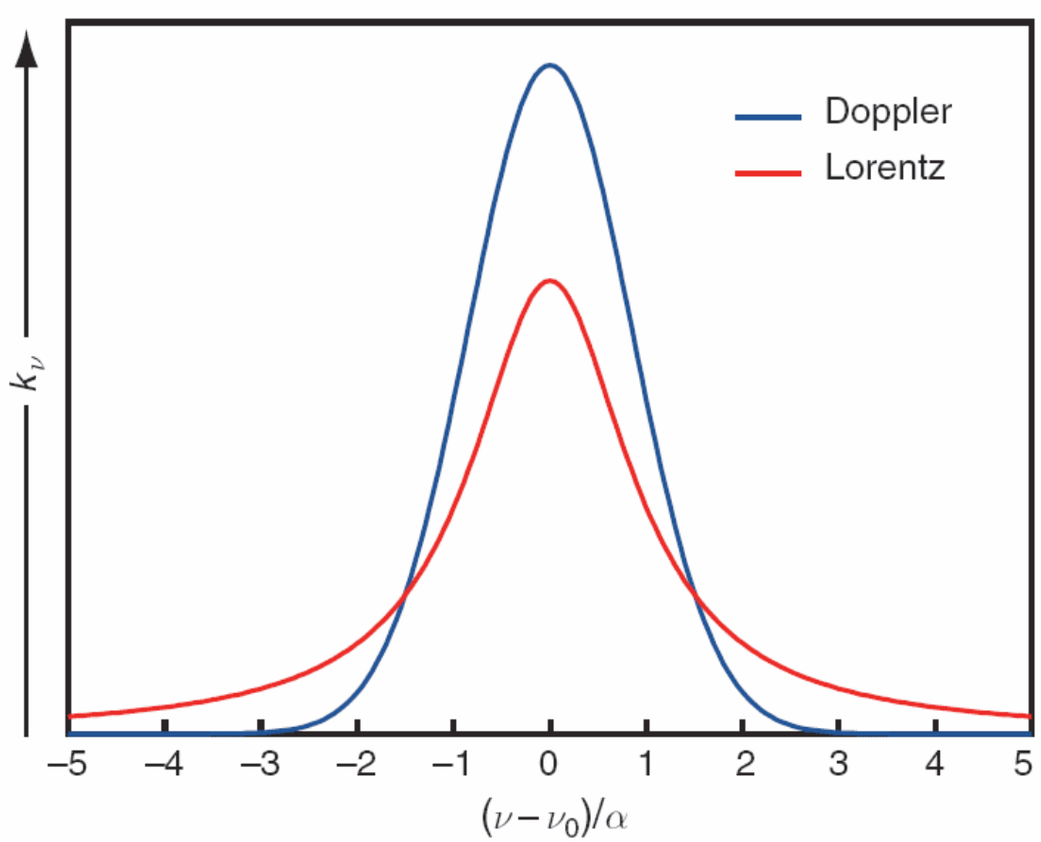
atomic vapour: Maxwell–Boltzmann velocity distribution

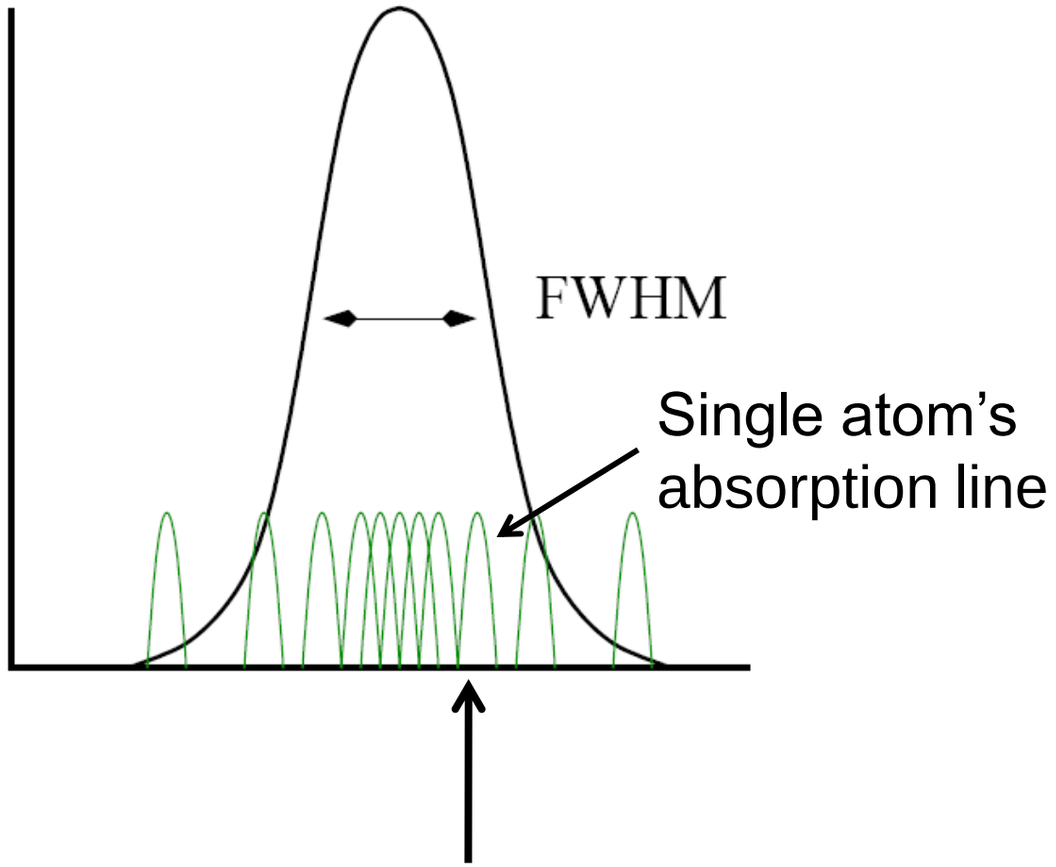
Resonance of atom shifted to $\nu_0 (1 + v_x / c)$

Velocity distribution (Gaussian) :

$$P (v_x) = \exp (-m v_x^2 / 2 k T)$$

$$\text{FWHM} = \sqrt{8kT \ln 2 / mc^2} \sim 1 \text{ GHz}$$





Fixed frequency laser only excites a fraction of atoms in sample

Pressure broadening (or density broadening)

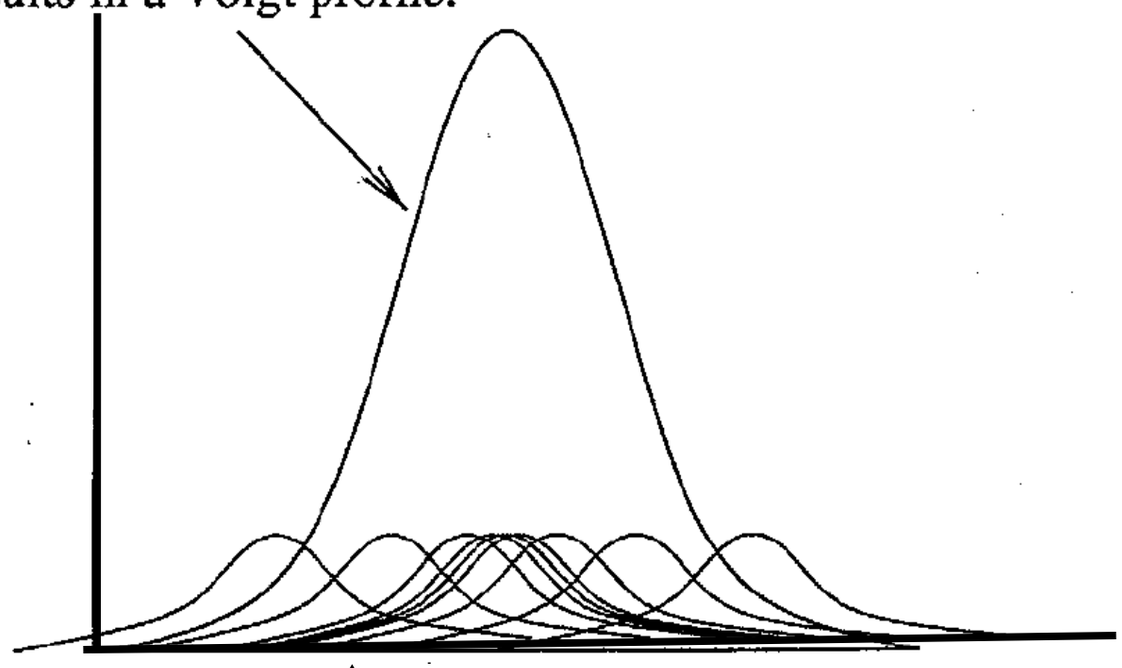
Atomic collisions in vapours or buffer gases interrupt the phase of the wavefunction - this broadens the energy of the state according to the Heisenberg uncertainty principle:

$$\Delta\nu = 1 / (2\pi \cdot \text{collision time})$$

$$\sim \text{typically } 10 \text{ MHz / Torr of gas}$$

(In addition there is a pressure shift of the transition)

This is homogeneous broadening, giving a Lorentzian lineshape. The convolution with Doppler broadening results in a Voigt profile.



A fixed frequency laser can excite all the atoms in the velocity distribution

MANCHESTER 1824

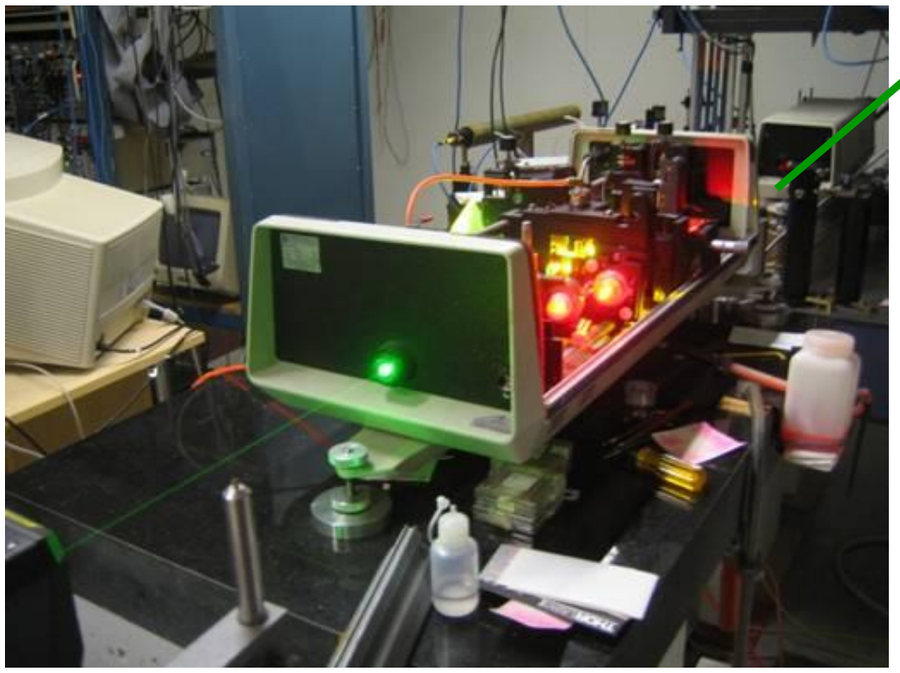
The University of Manchester

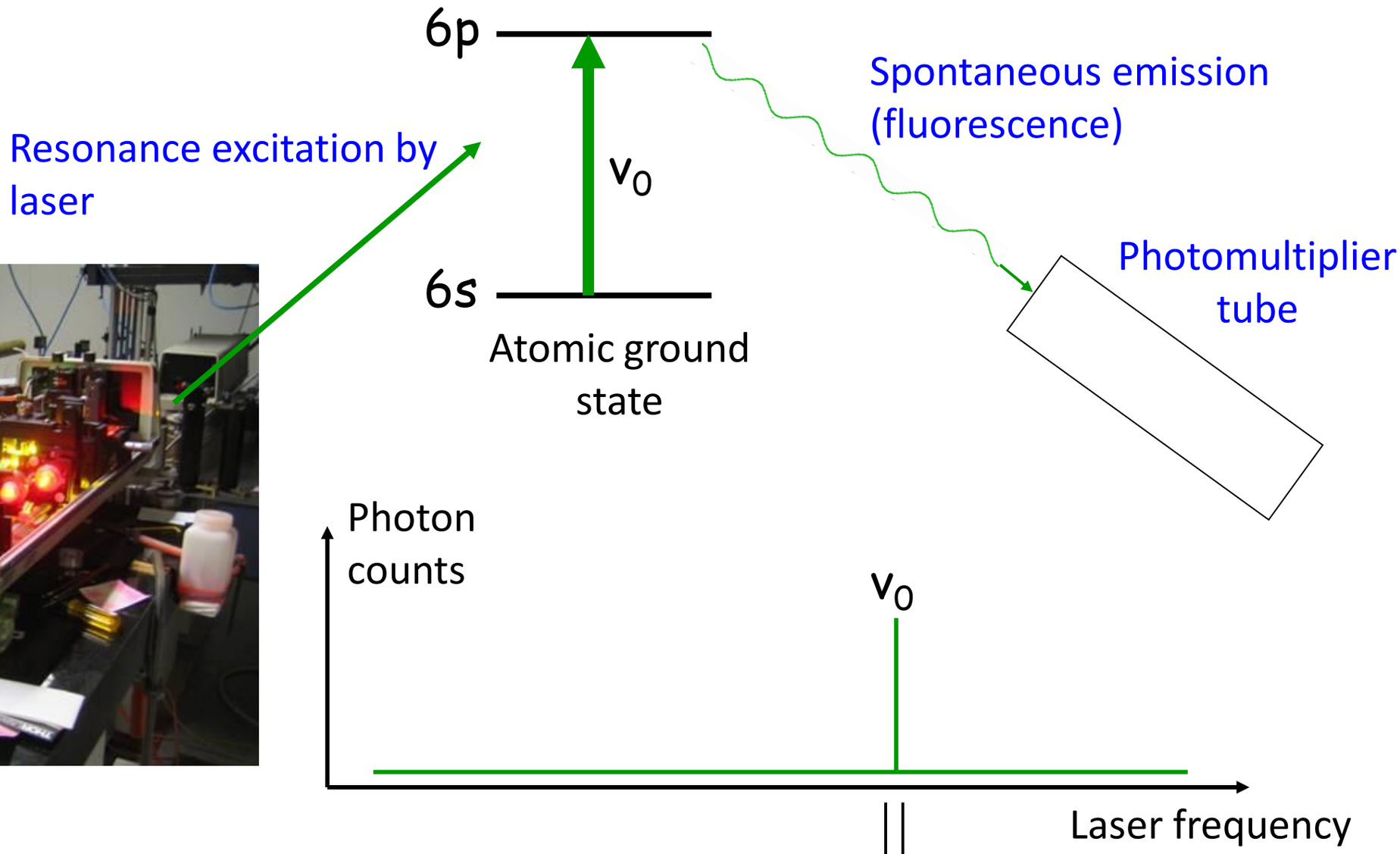
Isotope shifts and nuclear properties

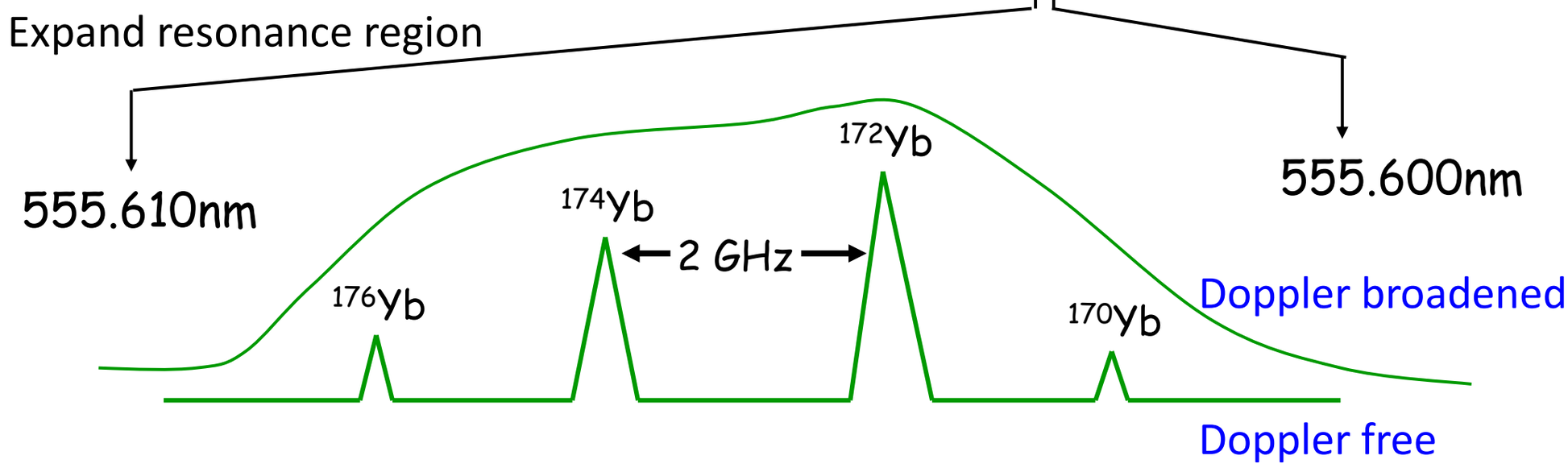
MANCHESTER 1824

The University of Manchester

Laser-fluorescence of an atomic vapour (of ytterbium)





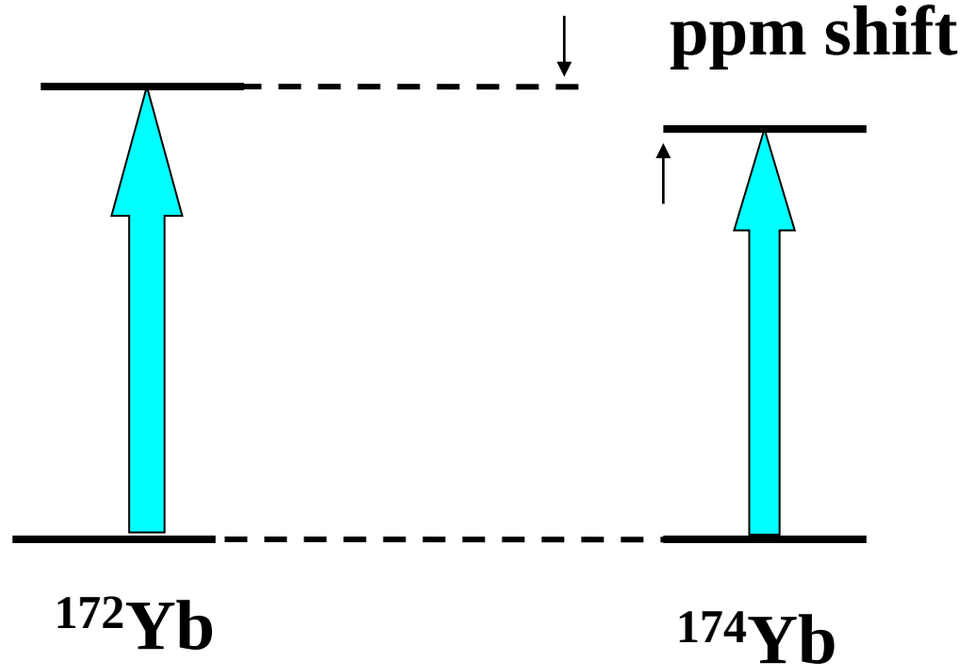


Isotope shift of an atomic transition

Shift has two components

• change in reduced mass of electron (*Mass Shift*)

• change in size of the nucleus (*Volume Shift*)



The *Volume Shift* is directly proportional to the change in nuclear mean square charge radius, $\delta\langle r^2\rangle$. Analysis of the shift provides information on the nuclear size and shape – even for short-lived (radioactive nuclei):

$$\delta\langle r^2\rangle = \delta\langle r^2\rangle_{\mathrm{sph}} + \langle r^2\rangle_{\mathrm{sph}} \frac{5}{4\pi} \delta\langle \beta_2^2\rangle$$

volume deformation

But, first, this requires the mass shift to be evaluated...

Mass Shift

Kinetic energy (nucleus + electrons) $$T = \frac{P_n^2}{2M_n} + \sum_i \frac{p_i^2}{2m_e}$$

But in centre of mass frame $$\mathbf{P_n} = -\sum_{\mathbf{i}} \mathbf{p_i}$$

Thus nucleus kinetic energy is $$T_{nuc} = \frac{1}{2M_n}\sum_i \mathbf{p_i^2} + \frac{1}{2M_n}\sum_{i\neq j}(\mathbf{p_i}\cdot\mathbf{p_j})$$

Energy change between two isotopes A, A’

(m_u = atomic mass unit)

$$\delta T_{nuc} = \frac{1}{2m_u}\left(\frac{A'-A}{AA'}\right)\left(\sum_i(\mathbf{p_i})^2 + \sum_{i>j}(2\mathbf{p_i}\cdot\mathbf{p_j})\right)$$

“normal” “specific”

$$\delta\nu_{MS}^{A,A'} = (N+S)\left(\frac{A-A'}{AA'}\right).$$

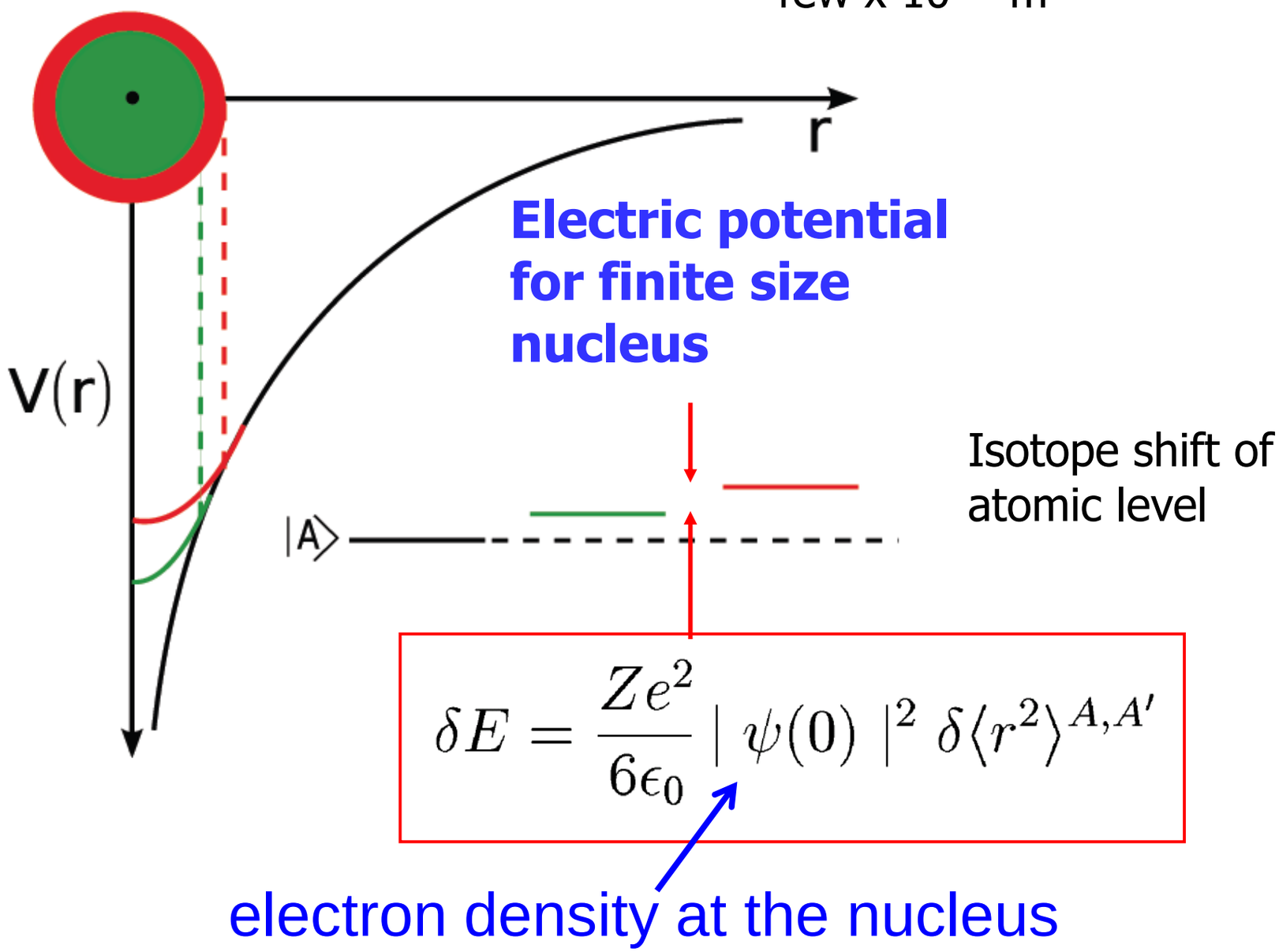
$$N = \frac{m_e}{m_u}\nu_0$$

S must be evaluated by experiment or calculation – often difficult to do well

The volume shift

Nuclear radius:
few x 10^{-15} m

Electron radius:
few x 10^{-10} m

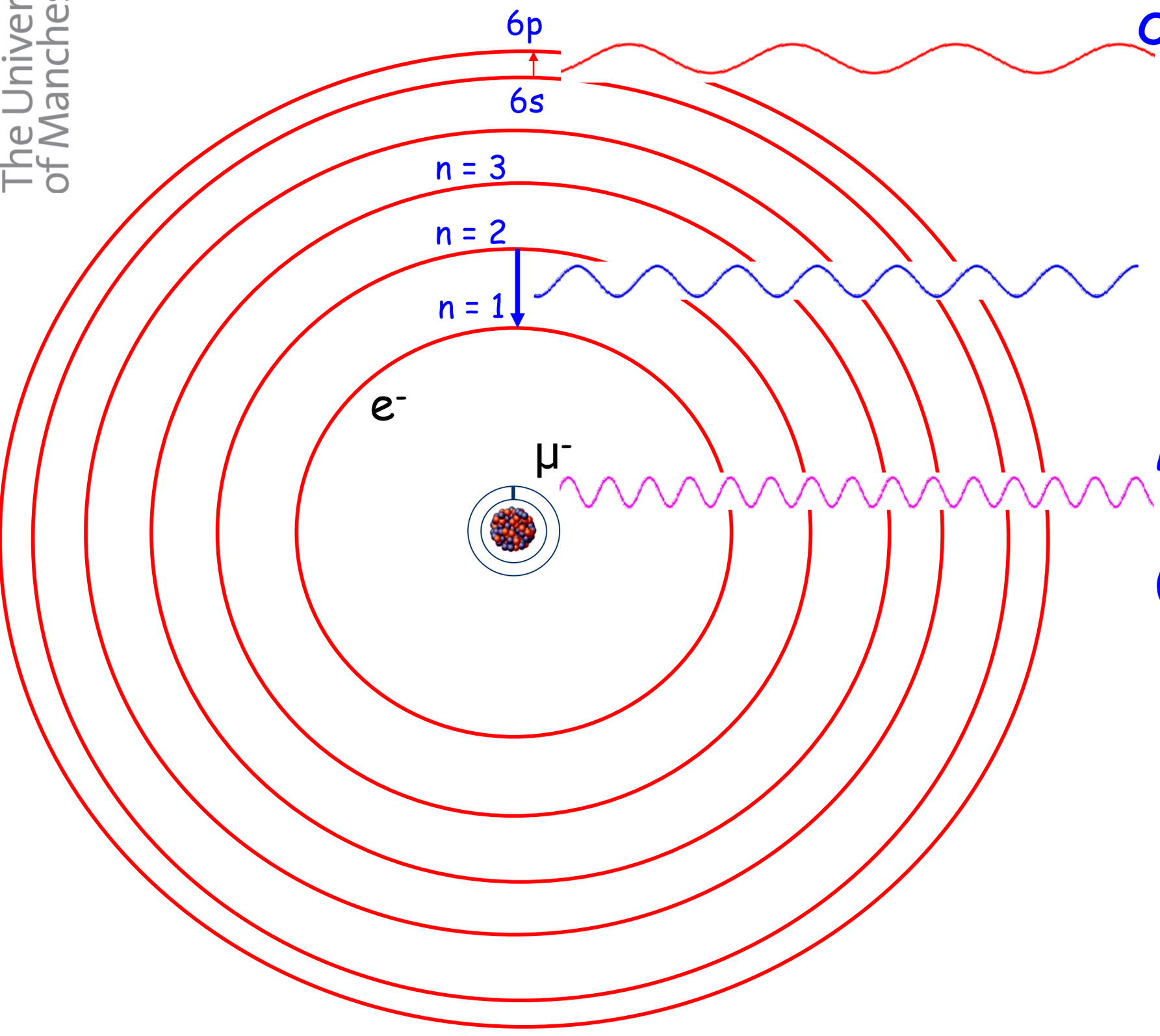


$$\delta E = \frac{Ze^2}{6\epsilon_0} \mid \psi(0) \mid^2 \delta\langle r^2 \rangle^{A,A'}$$

electron density at the nucleus

Thus volume shift = $F\ \delta\langle r^2 \rangle^{A'A}$ where F is an atomic factor which must be evaluated

Isotope shifts in atomic transitions



Optical transitions (3 eV)

Shift ~ 10^{-6} eV

K X-rays (50 keV)

Shift ~ 0.1 eV

Muonic X-rays (1 MeV)

Shift ~ 5,000 eV
(Theory allows absolute size measurement)

Approximate magnitudes for ΔA = 2

Isotope shift = (normal + specific) mass shift + volume shift

$$F\,\delta\langle r^2\rangle^{A'A}$$

Element	Transition	Normal	Specific	Volume	Doppler Broadening
$_{2}$He	2s – 3p	35000 MHz	8000 MHz	-1.4 MHz	3300 MHz
$_{11}$Na	3s – 3p	550 MHz	200 MHz	-10 MHz	1400 MHz
$_{70}$Yb	6s – 6p	20 MHz	‹ 20 MHz	-1500 MHz	500 MHz

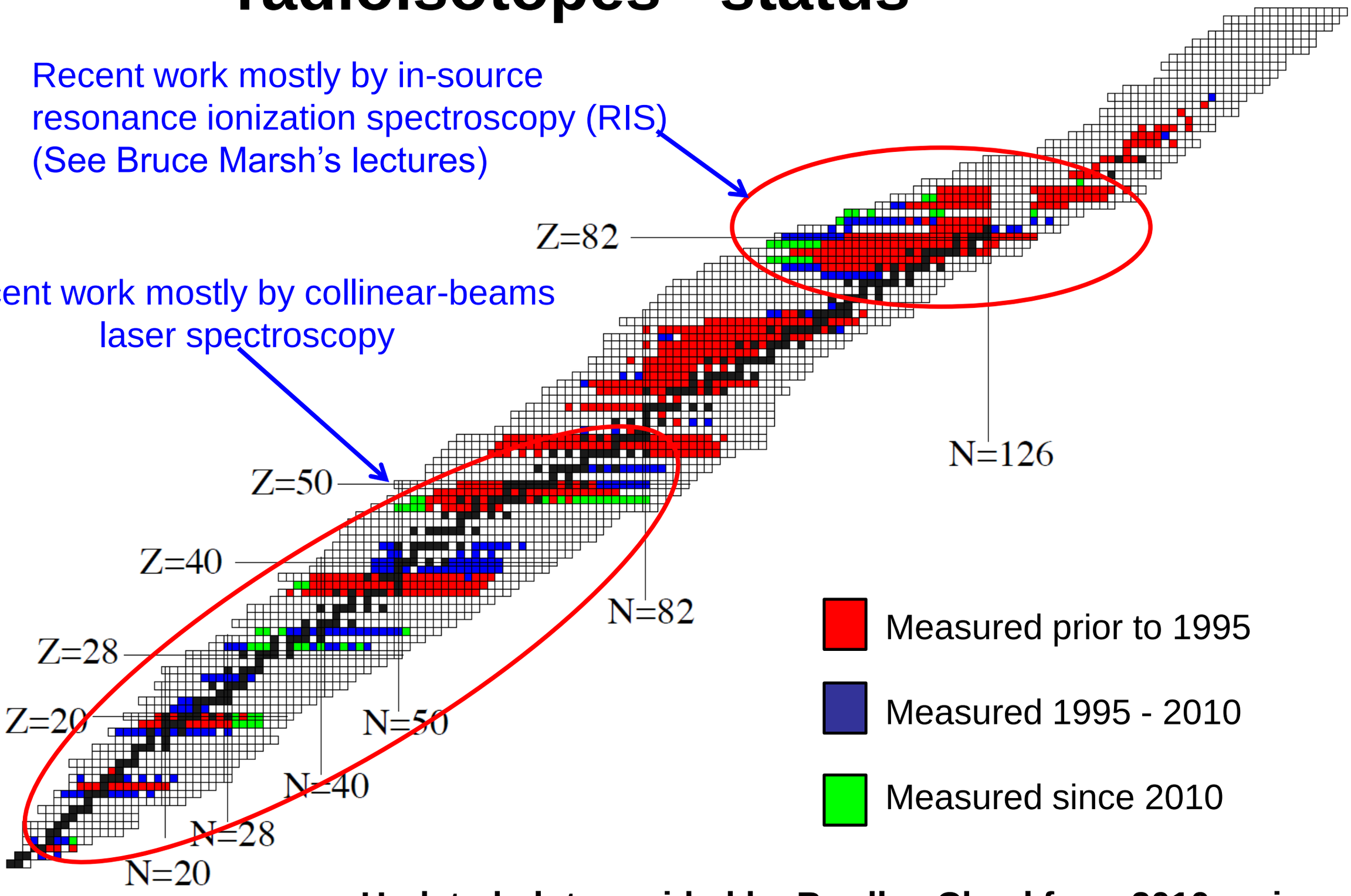
Heavy elements: useful results can be obtained from Doppler-broadened spectra

Light and medium mass elements: Doppler-free methods of laser spectroscopy

Laser spectroscopy of radioisotopes - status

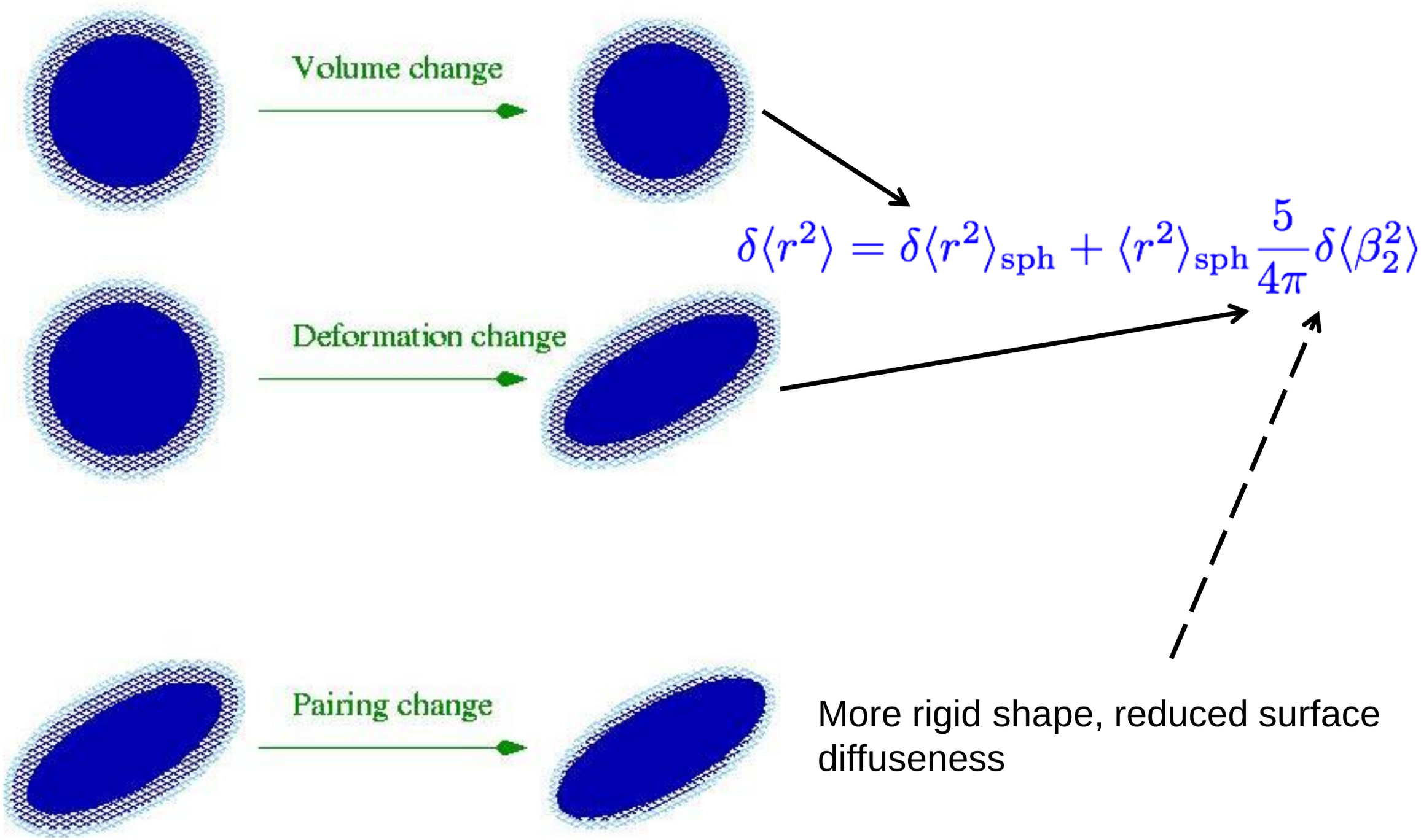
Recent work mostly by in-source resonance ionization spectroscopy (RIS)
(See Bruce Marsh's lectures)

Recent work mostly by collinear-beams laser spectroscopy

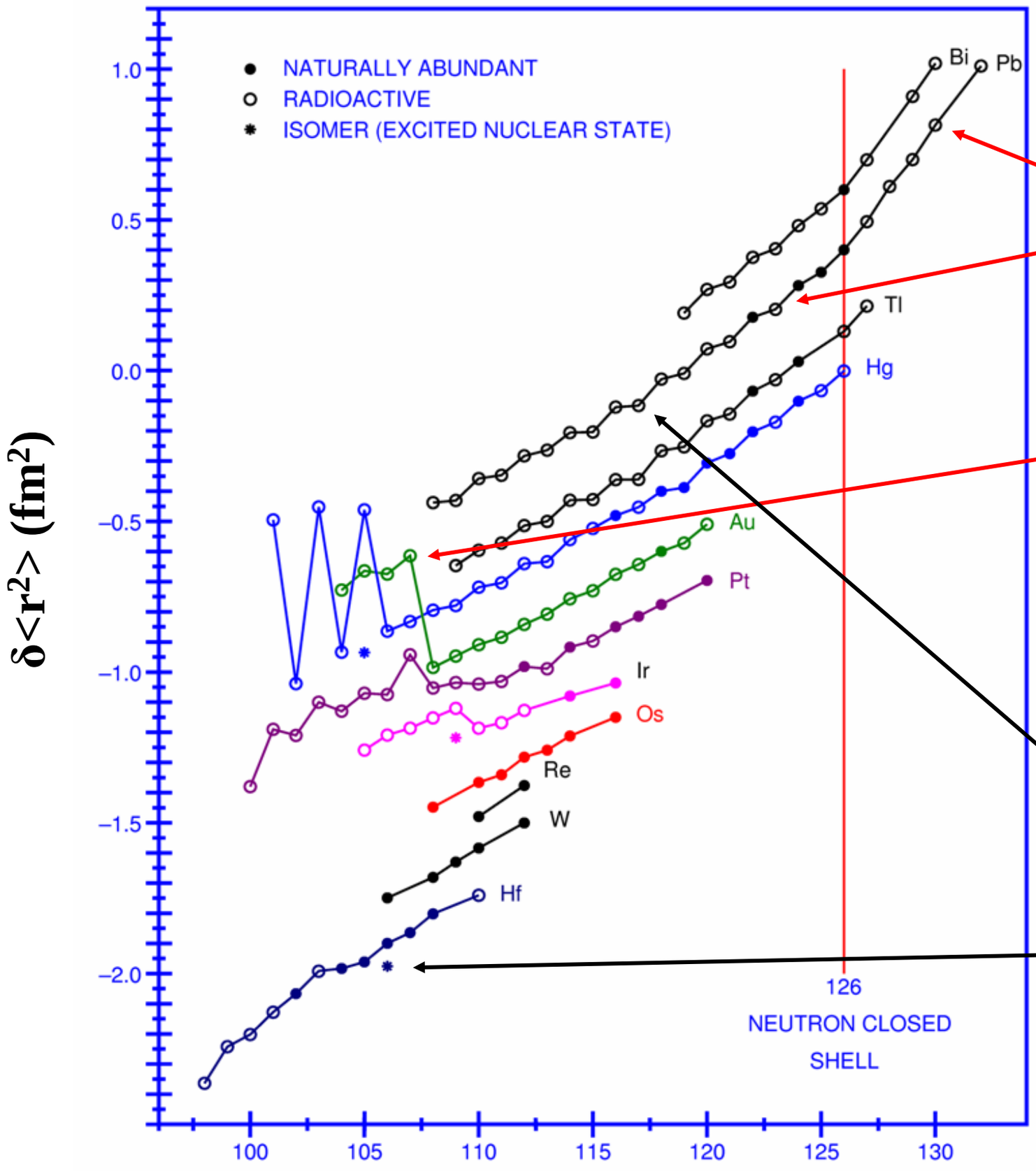


Updated plot provided by Bradley Cheal from 2010 review
(B. Cheal & K.T. Flanagan J. Phys. G 37 (2010) 113101)

Factors controlling the nuclear mean square charge radius



Mean square charge radii



Volume

Deformation

Pairing effects?

Odd-even staggering

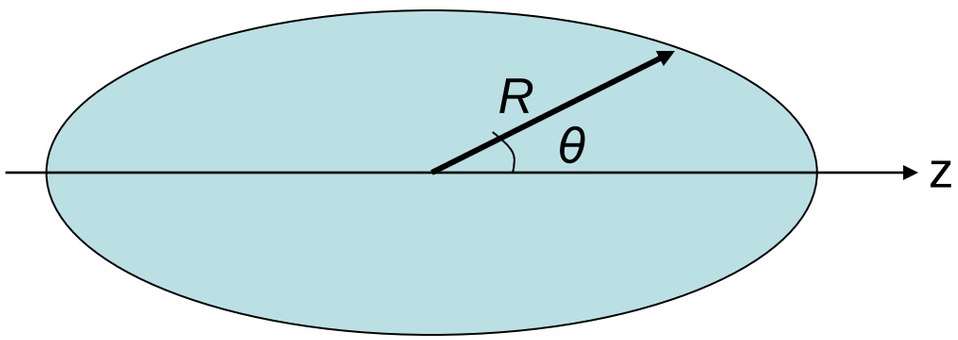
Isomer shifts

Example: deformation in zirconium isotopes (JYFL)

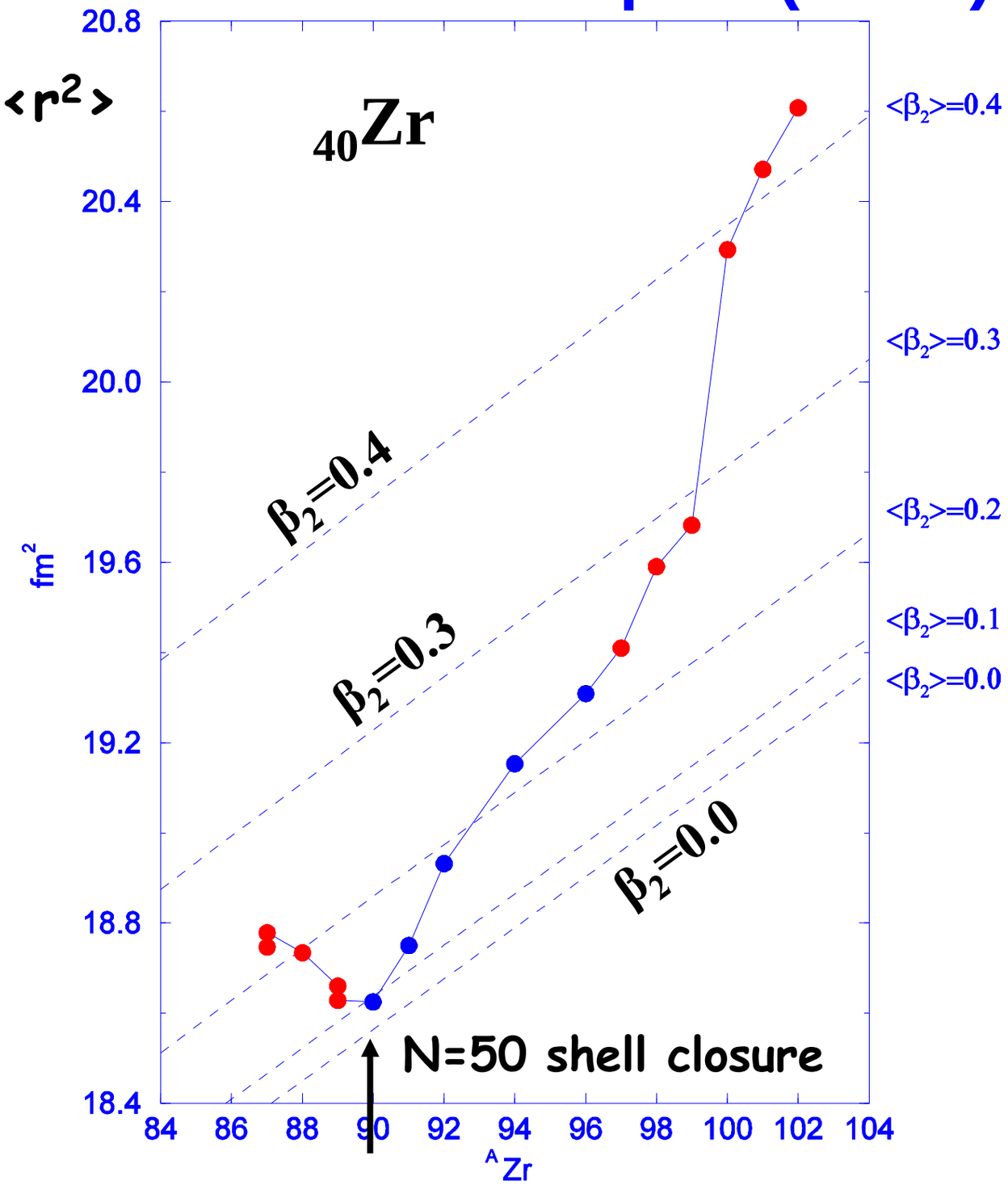
Quadrupole deformation parameter

$$\langle r^2 \rangle = \langle r^2 \rangle_0 \left(1 + \frac{5}{4\pi}(\langle \beta_2^2 \rangle + ...)\right)$$

Radius of spherical nucleus of same volume



$$R = R_0 \, (\, 1 + \beta_2 \, Y_{2,0} \, (\theta, \varphi))$$



MANCHESTER 1824

The University of Manchester

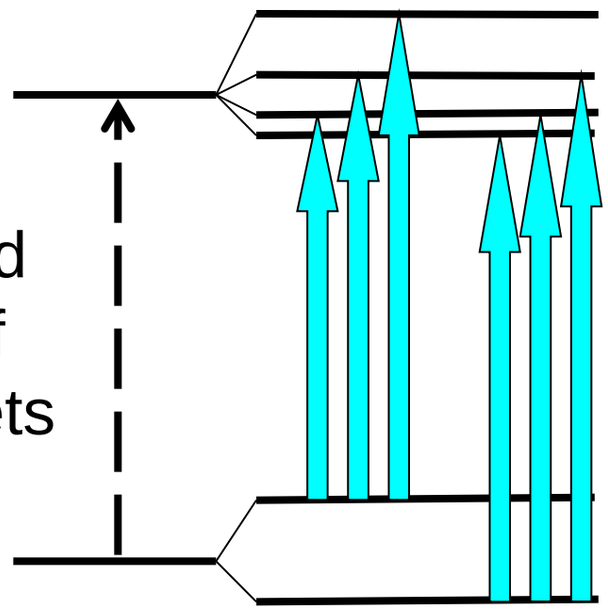
Hyperfine structure and nuclear moments

Hyperfine structure of an atomic transition

The nuclear magnetic dipole and electric quadrupole moments cause small perturbations to the energy of an atomic level, producing a hyperfine structure.

The magnitude of this structure is comparable to the (ppm) size of the isotope shifts.

Isotope shift found using centroids of hyperfine multiplets



^{177}Hf

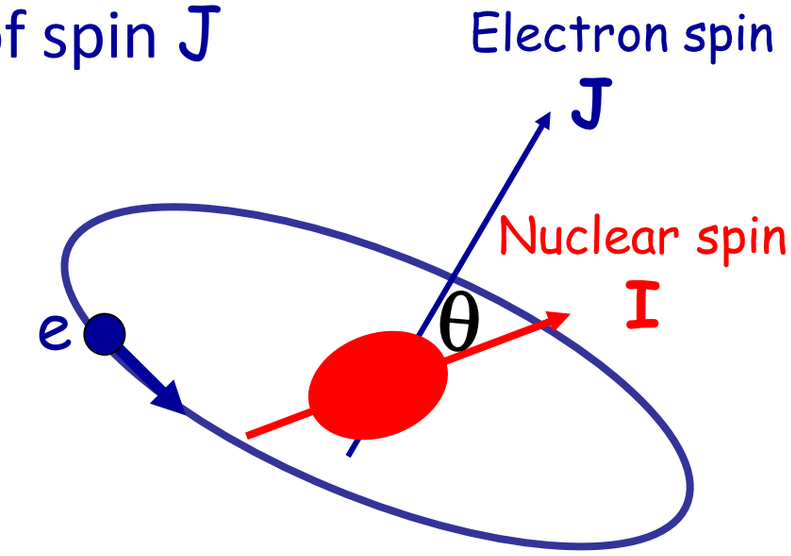
μ = magnetic dipole moment

Q_s = spectroscopic quadrupole moment (projection of intrinsic moment Q_0 on the quantization axis)

Hyperfine Interactions in free atoms

Hyperfine interaction = the interaction of nuclear magnetic and electric moments with electromagnetic fields.

We will consider the effect on an atomic orbit of spin **J**

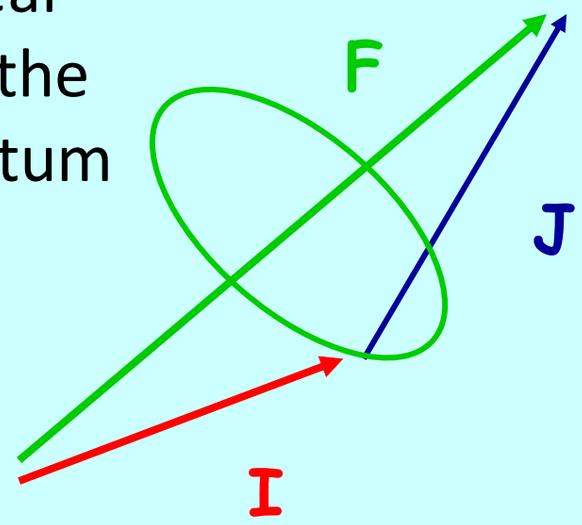


The atomic and nuclear spins couple to form the total angular momentum

F = **I** + **J**

Each state **J** has several **F**-states:

$$|I - J| \leq F \leq I + J$$



The interaction energy depends on the angle ϑ thus for the same **I** and **J**, the different **F**-states are at slightly different energies:

Magnetic dipole interaction

$$E = -\boldsymbol{\mu} \cdot \mathbf{B_e} = -\mu B_e \cos\theta$$

Electric quadrupole interaction

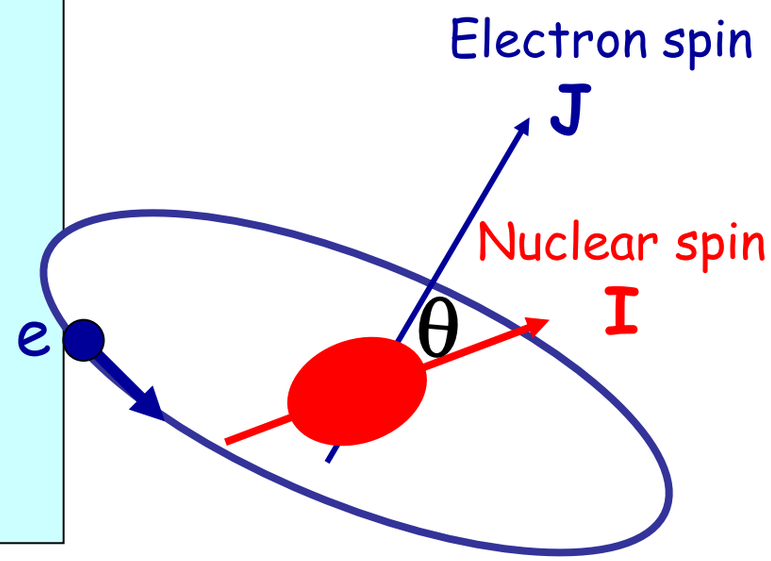
$$E = \tfrac{1}{4}\, e\, Q_0\, V_{JJ}\, P_2(\cos\theta)$$

Magnetic dipole interaction

$$E = -\boldsymbol{\mu} \cdot \mathbf{B}_e = -\mu B_e \cos\theta$$

Since $\boldsymbol{\mu} = g\mathbf{I}\mu_N$ and $\mathbf{B}_e = -(\frac{B_e}{J})\mathbf{J}$

then interaction Hamiltonian can be expressed as $H_m = (\frac{gB_e\mu_N}{J})\mathbf{I}.\mathbf{J} = A\mathbf{I}.\mathbf{J}$

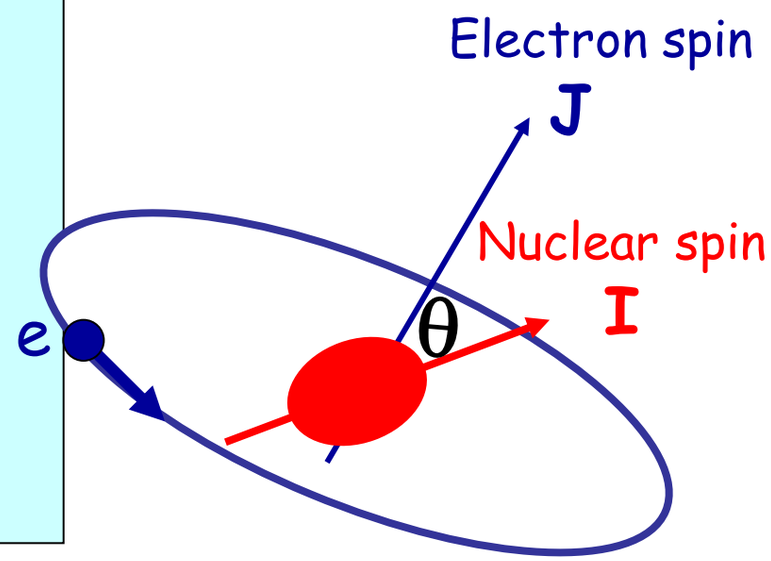


Magnetic dipole interaction

$$E = -\boldsymbol{\mu} \cdot \mathbf{B_e} = -\mu B_e \cos\theta$$

Since $\boldsymbol{\mu} = g\mathbf{I}\mu_N$ and $\mathbf{B}_e = -(\frac{B_e}{J})\mathbf{J}$

then interaction Hamiltonian can be expressed as $H_m = (\frac{gB_e\mu_N}{J})\mathbf{I}.\mathbf{J} = A\mathbf{I}.\mathbf{J}$



The different energy shifts of the different F-states are then $\Delta E = \langle IJF \mid H_m \mid IJF \rangle = A\langle \mathbf{I}.\mathbf{J} \rangle$

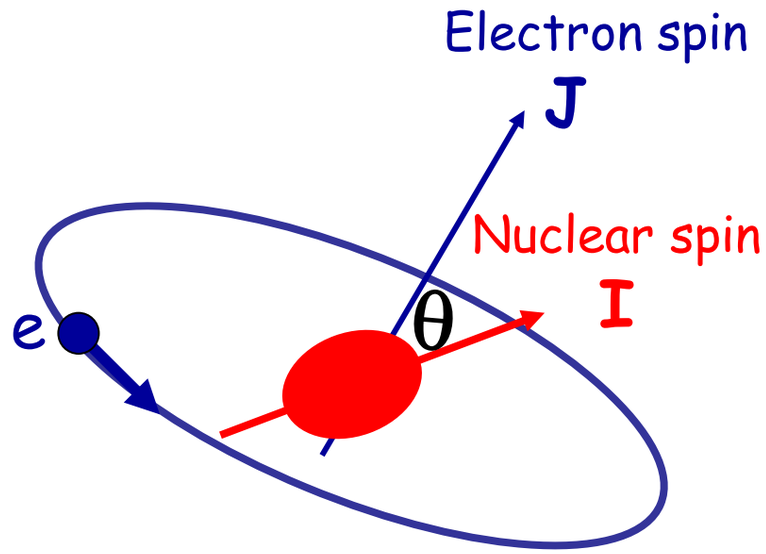
where $\langle \mathbf{I}.\mathbf{J} \rangle = \frac{1}{2}\langle F^2 - I^2 - J^2 \rangle = \frac{1}{2}[F(F+1) - I(I+1) - J(J+1)]$

B_e, the magnetic field at the nucleus produced by the atomic electrons can be calibrated by measuring the energy shifts for a isotope of known magnetic moment.

Electric quadrupole interaction

$$E = \tfrac{1}{4}\, e\, Q_0\, V_{JJ}\, P_2(\cos\theta)$$ [compare $E = -\boldsymbol{\mu}.\mathbf{B} = -\mu\, B\, P_1(\cos\theta)$]

Electric field gradient along J-direction due to atomic electrons.



Energy shifts of the F-states are then

$$\Delta E_Q = \frac{B}{4}\frac{\frac{3}{2}C(C+1) - 2I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$

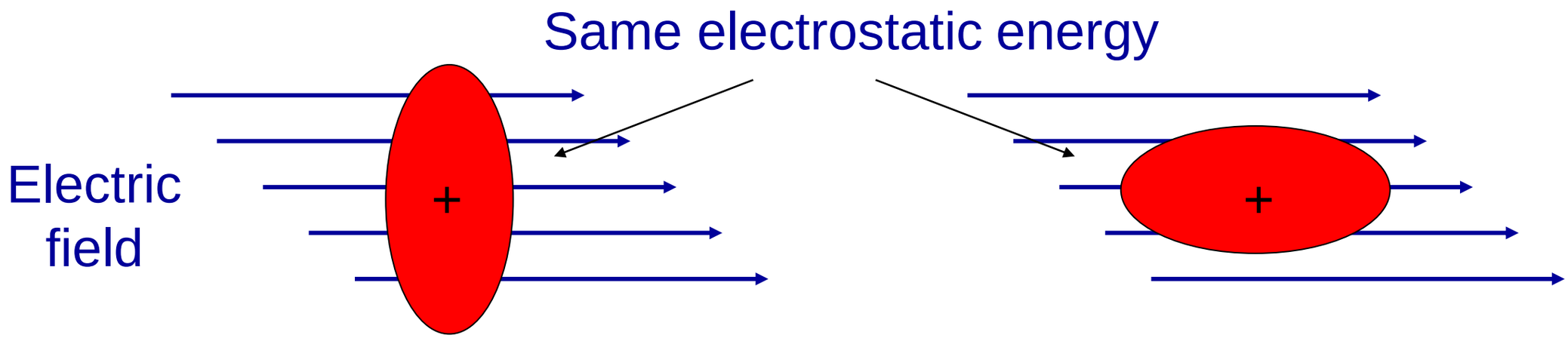
where $C = [F(F+1) - I(I+1) - J(J+1)]$

$$B = eQ_s\langle\frac{\partial^2 V}{\partial Z^2}\rangle = eQ_s V_{JJ}$$ is the hyperfine factor measured by experiment.

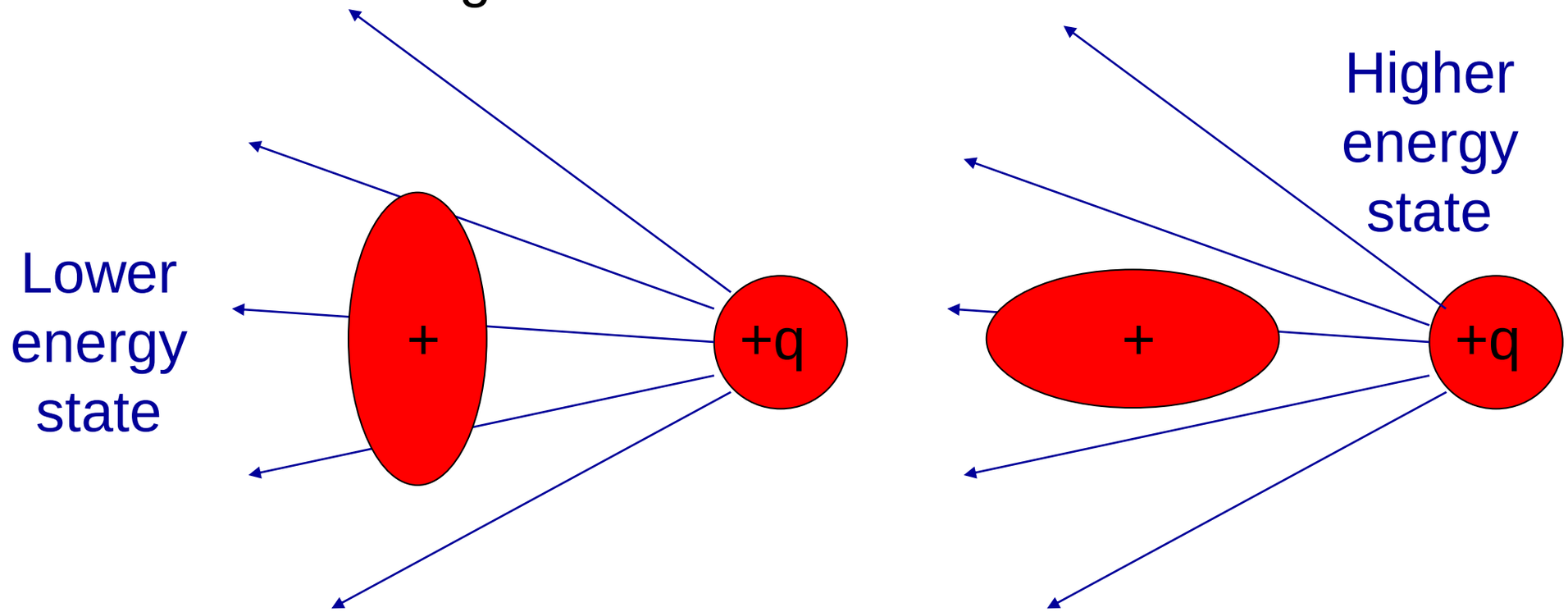
The electric field gradient V_{JJ} may be calibrated with an isotope with know Q_s

Electric quadrupole interaction

In a uniform electric field the energy of an electric quadrupole moment is independent of angle and therefore there is no quadrupole interaction

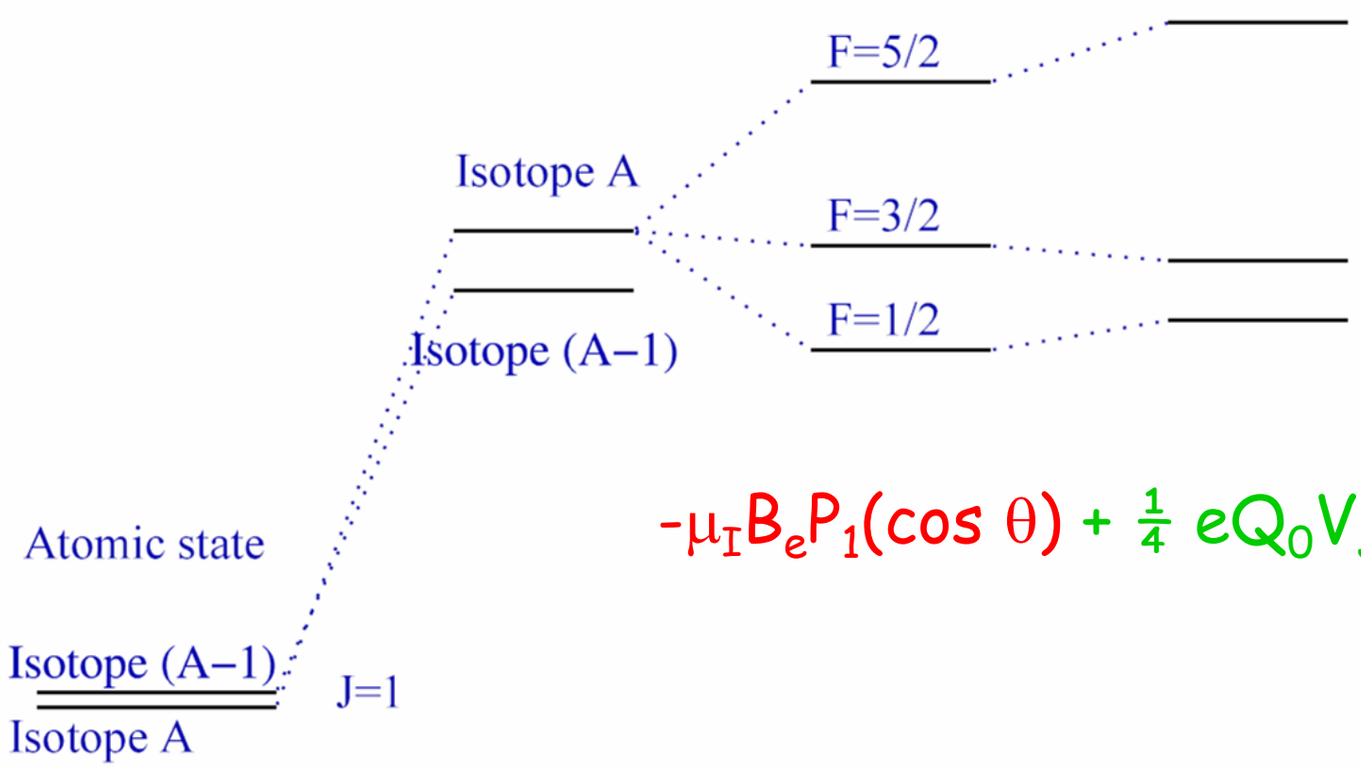


There is an angle-dependence in an electric field gradient and thus different F-state have different energies

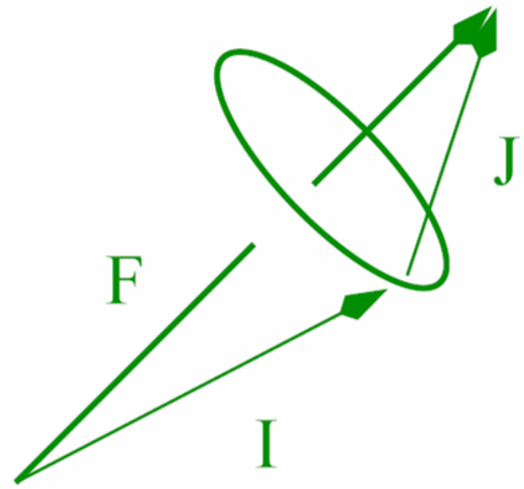


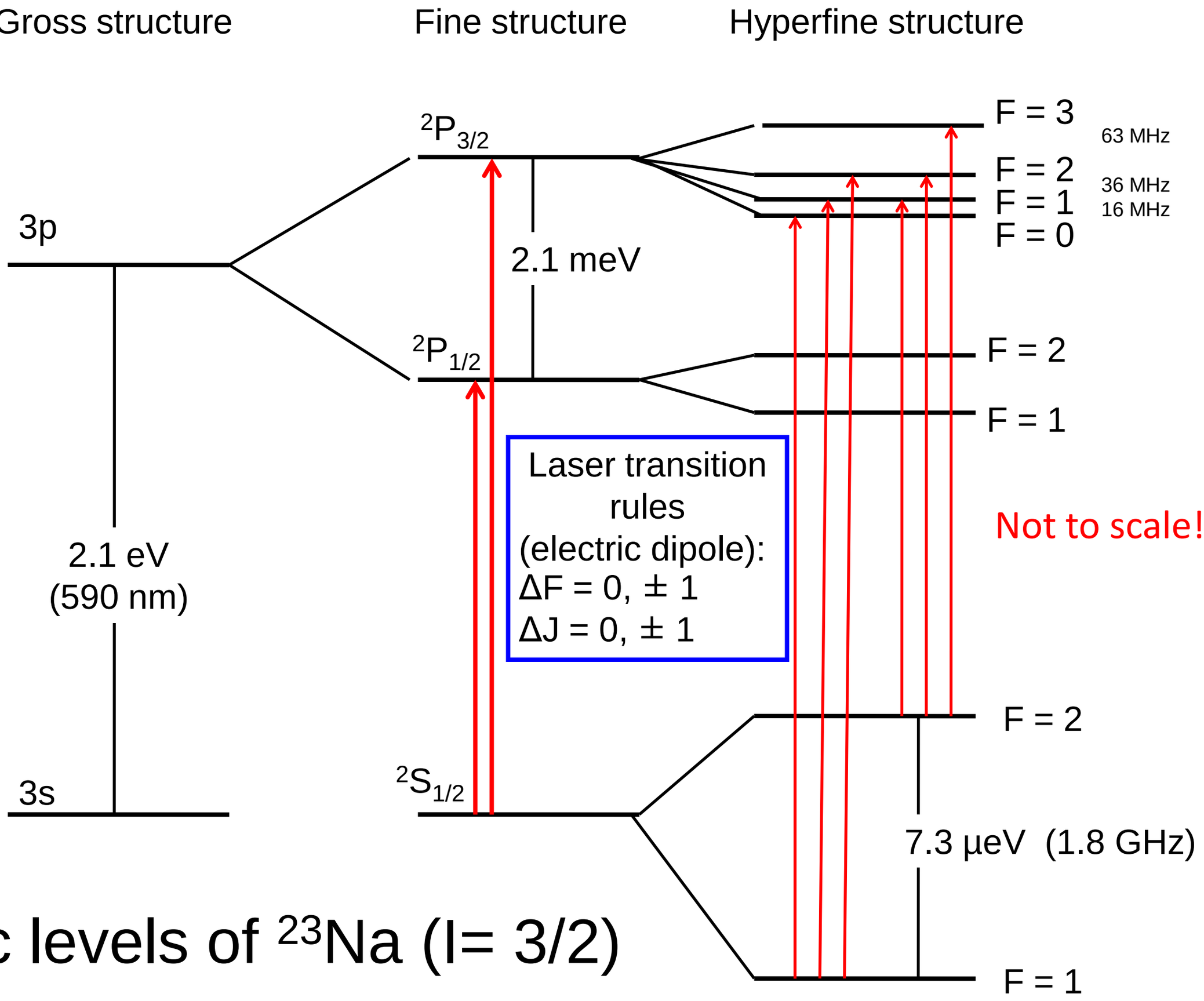
Summary of atomic structure (example uses J=1 and I = 3/2)

Point nucleus + Finite size of nucleus + Magnetic dipole + Electric quadrupole + higher multipoles (too small to consider in laser measurements)



$-\mu_I B_e P_1(\cos\theta) + \frac{1}{4} eQ_0 V_{JJ} P_2(\cos\theta)$ + ...





Atomic levels of ^{23}Na (I= 3/2)

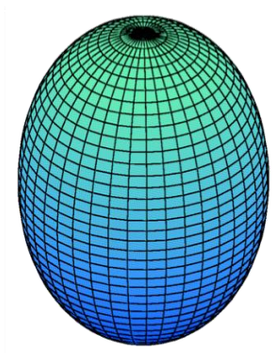
MANCHESTER 1824

The University of Manchester

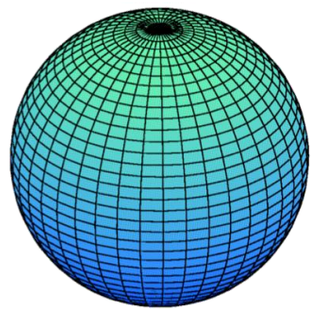
Laser spectroscopy of radioactive atoms

Laser spectroscopy of optical transitions in radioactive atoms can:

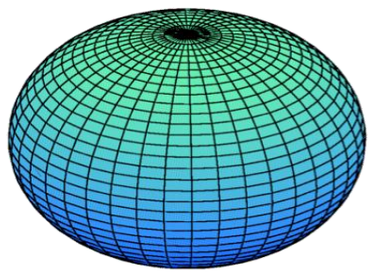
- Confirm existence of an exotic nuclide or isomer
- Determine the valence proton and neutron orbitals and configurations (via the nuclear spin and magnetic moment)
- Determine the size and shape (static and dynamic) of the nucleus
- Provide pure nuclide sample for low-background nuclear spectroscopy (to measure half-lives, decay modes, etc)



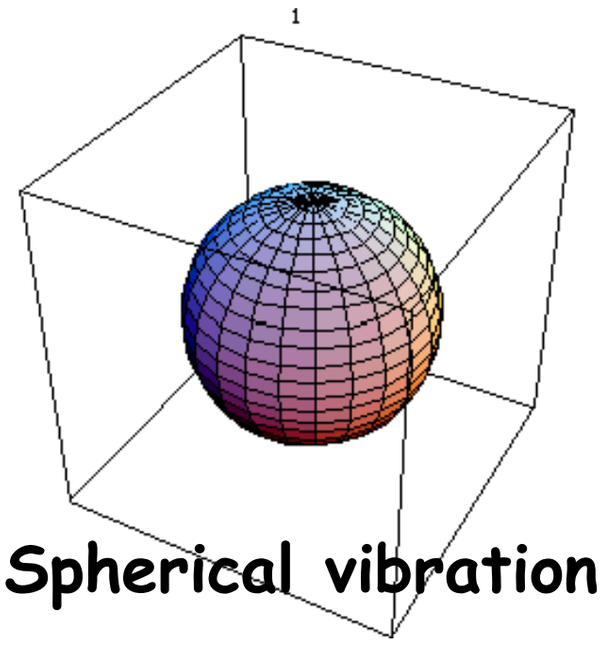
Prolate vibration



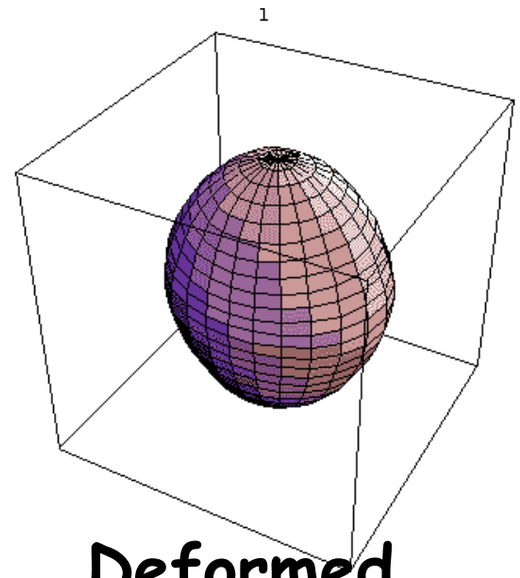
Spherical



Oblate



Spherical vibration

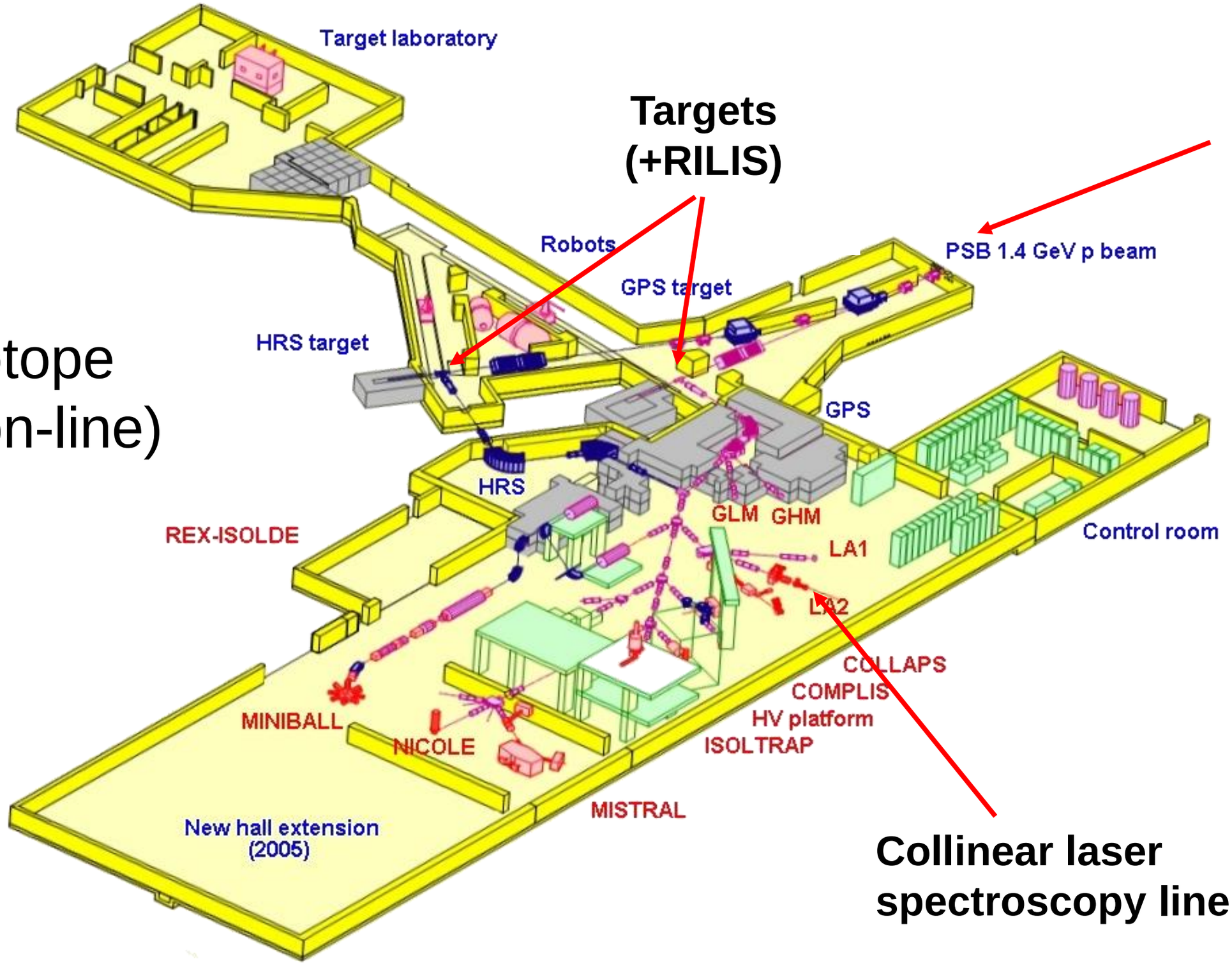


Deformed

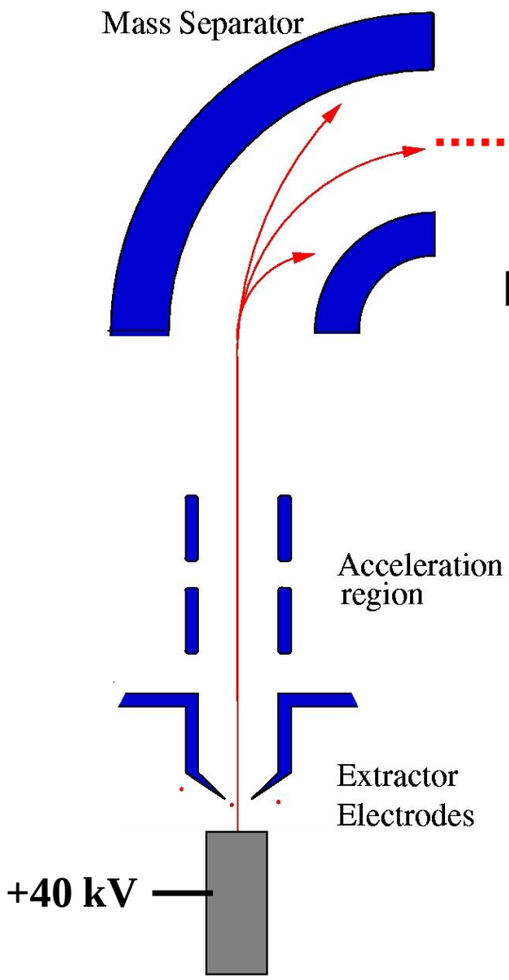
THE CERN Site

Collinear-beams laser spectroscopy at ISOLDE (suppressed Doppler-broadening)

(ISOL = Isotope separator on-line)



Standard method: Collinear-beams laser spectroscopy



Ion source
(1+ ions)

T = 2,500 K

Fast ion beam (40 keV)

Beam flux ~ 10^4 ions/sec

Laser beam

1 mW

Principle behind the collinear beams method

Adding a fixed energy to each ion in the sample

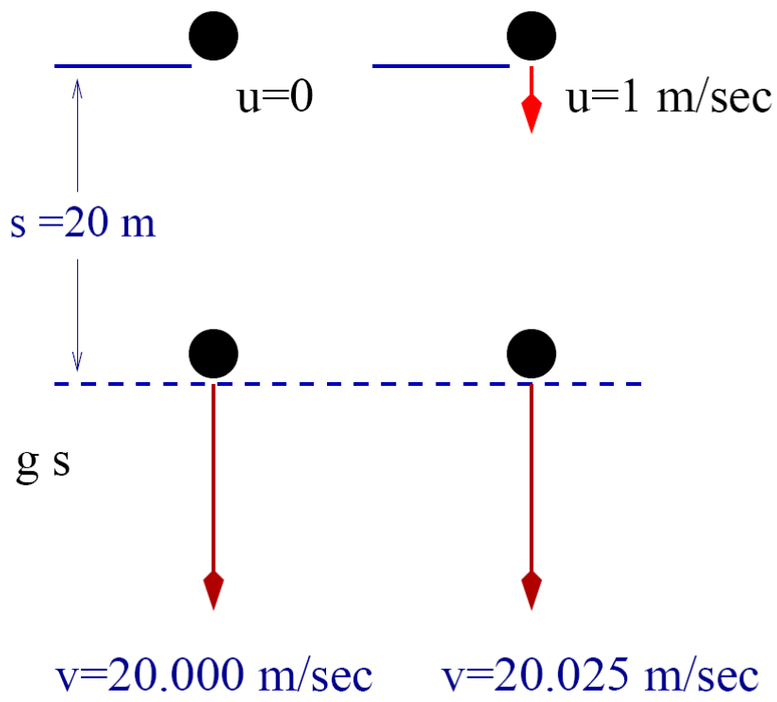
→ increases the velocity

but

reduces the velocity spread

An example:

Dropping a particle through a fixed distance



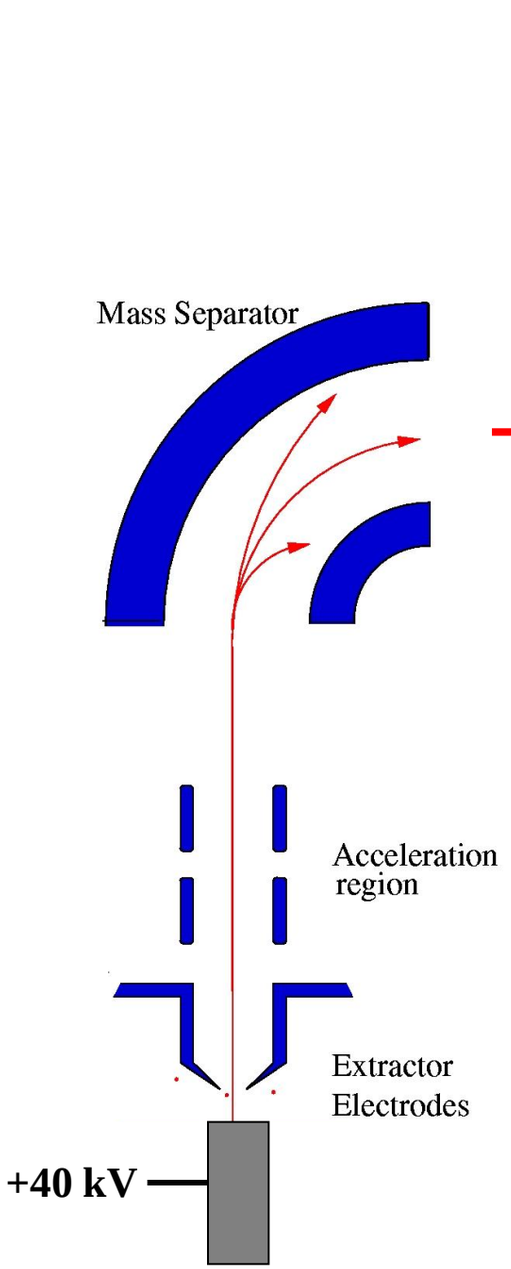
Count rates for low-flux ion beams

Signal (laser on resonance) =
1 photon detected per 1,000 ions in beam

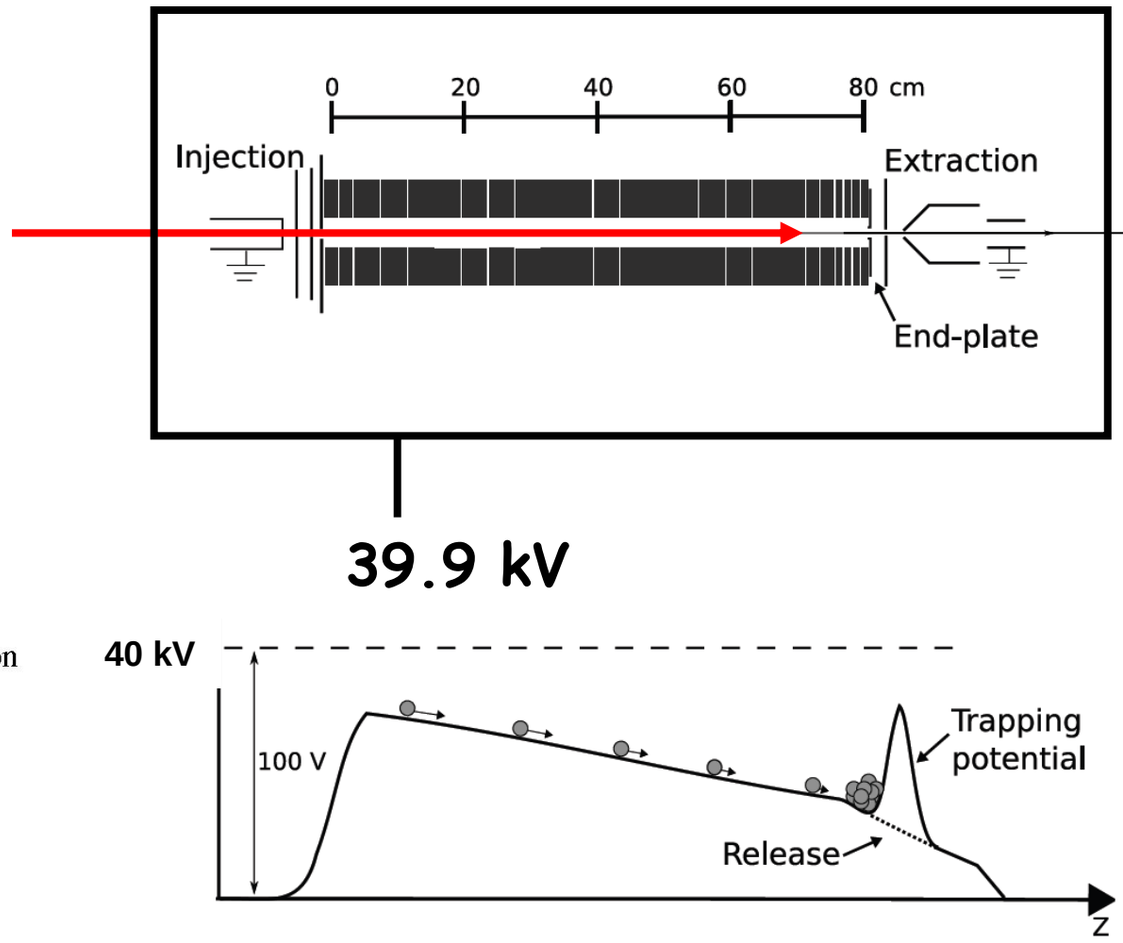
Background (laser light scatter) = 200 photons / second

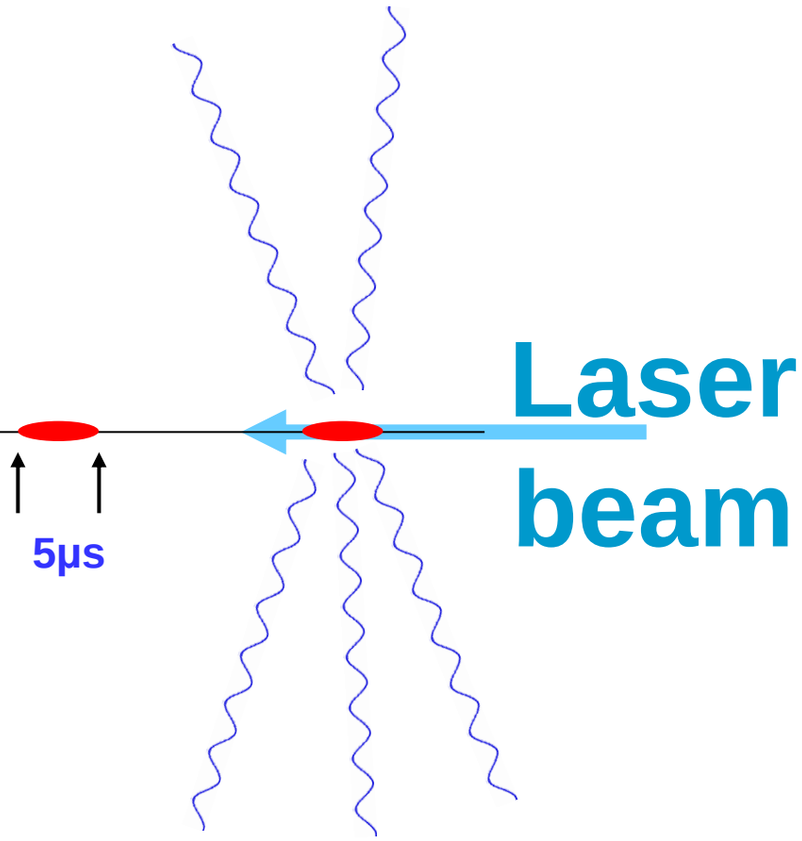
Low-flux beams (10,000 ions / sec): background must be suppressed to see signal.

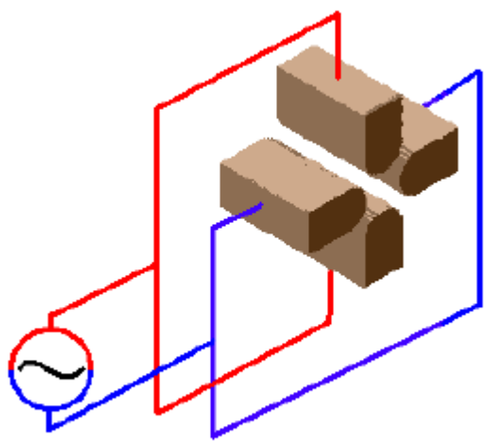
Helium-filled radio-frequency trap



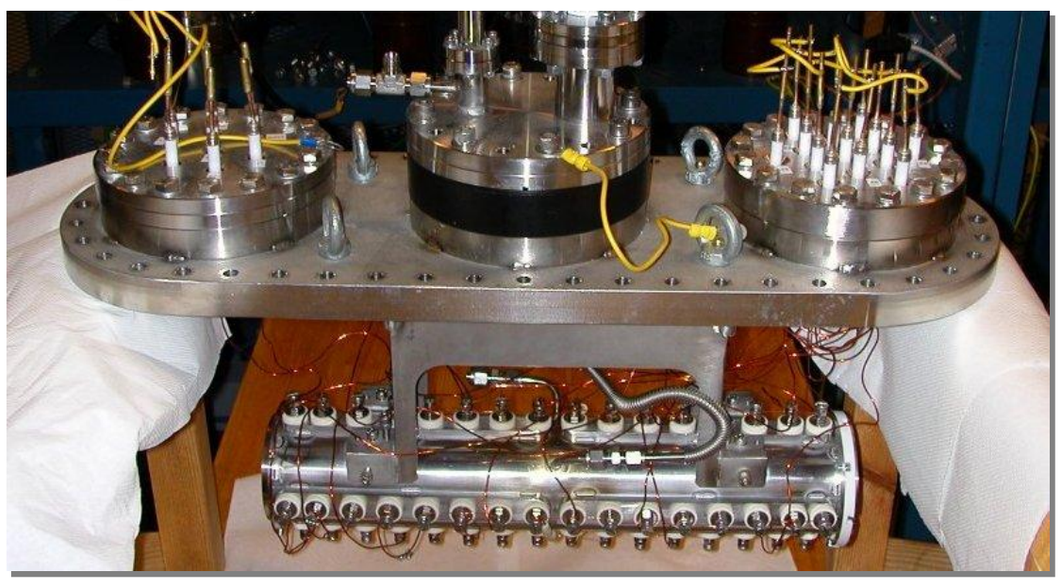
Accumulate ions for ~50 ms



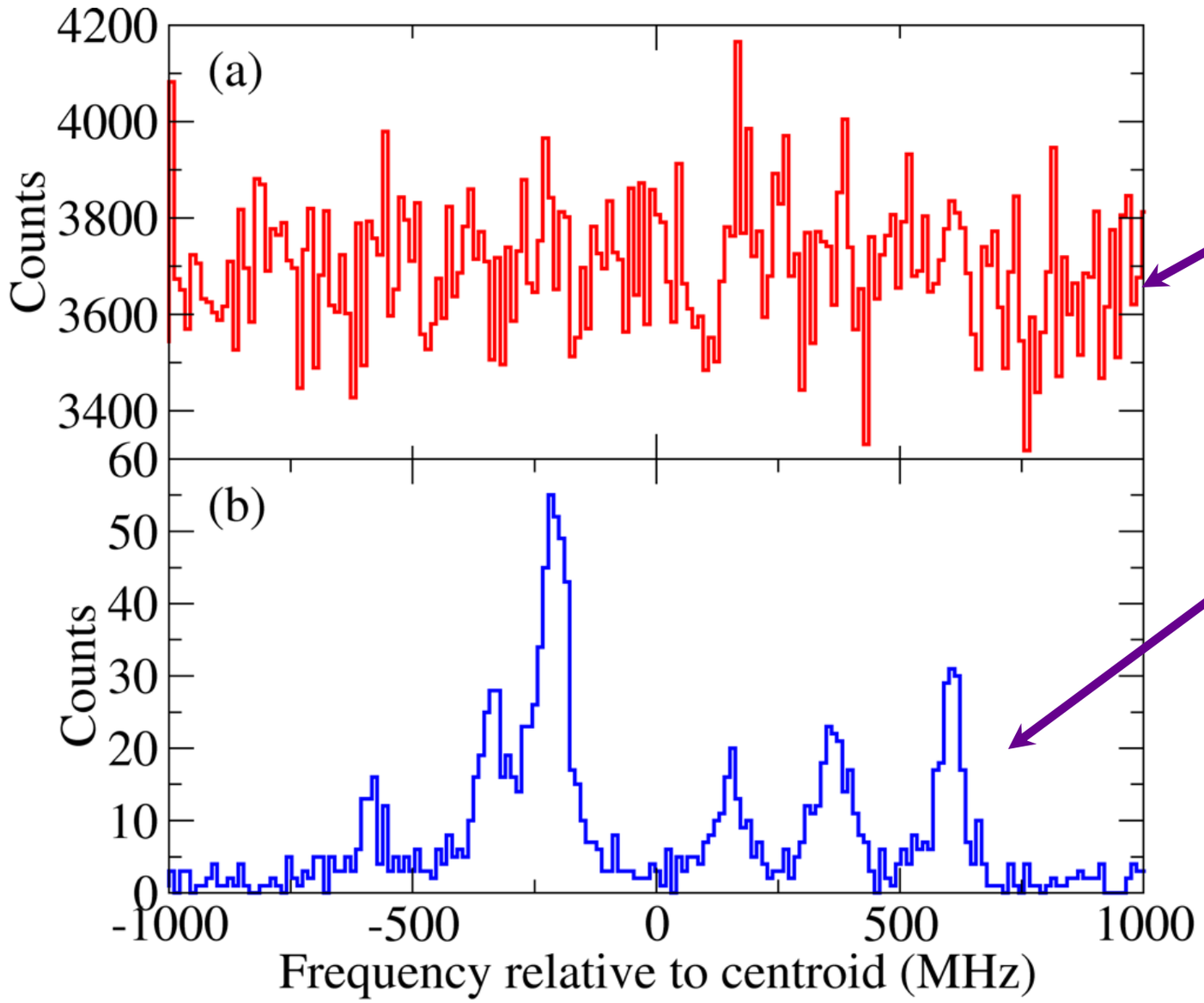




Jyväskylä ion beam cooler



Example – Photon rates for radioactive ^{76}Ga



Un-gated photons

Gated photons (time when ion bunch is in the laser interaction region)

Background suppression

50 ms / 5 μs = ~10^4

Frequency Doubling

A technique to produce narrow-bandwidth, tuneable light in the UV region (200 nm – 400 nm)

Also called Second Harmonic Generation (SHG)

Some crystals have a non-linear response (polarisability) to an applied electric field (in this case, the electric field of a laser beam)

$$\frac{P}{\epsilon_o} = \chi^{(1)}\xi + \chi^{(2)}\xi^2 + \chi^{(3)}\xi^3 + \cdots,$$

P is the electric dipole moment per unit volume. χ is the polarizability or the electric susceptibility. ξ is the electric field.

The laser electric field has a cos (ωt) time dependence.

Since $2\cos^2(\omega t) = 1 + \cos(2\omega t)$

then a component of the re-radiated light is at exactly twice the fundamental frequency.

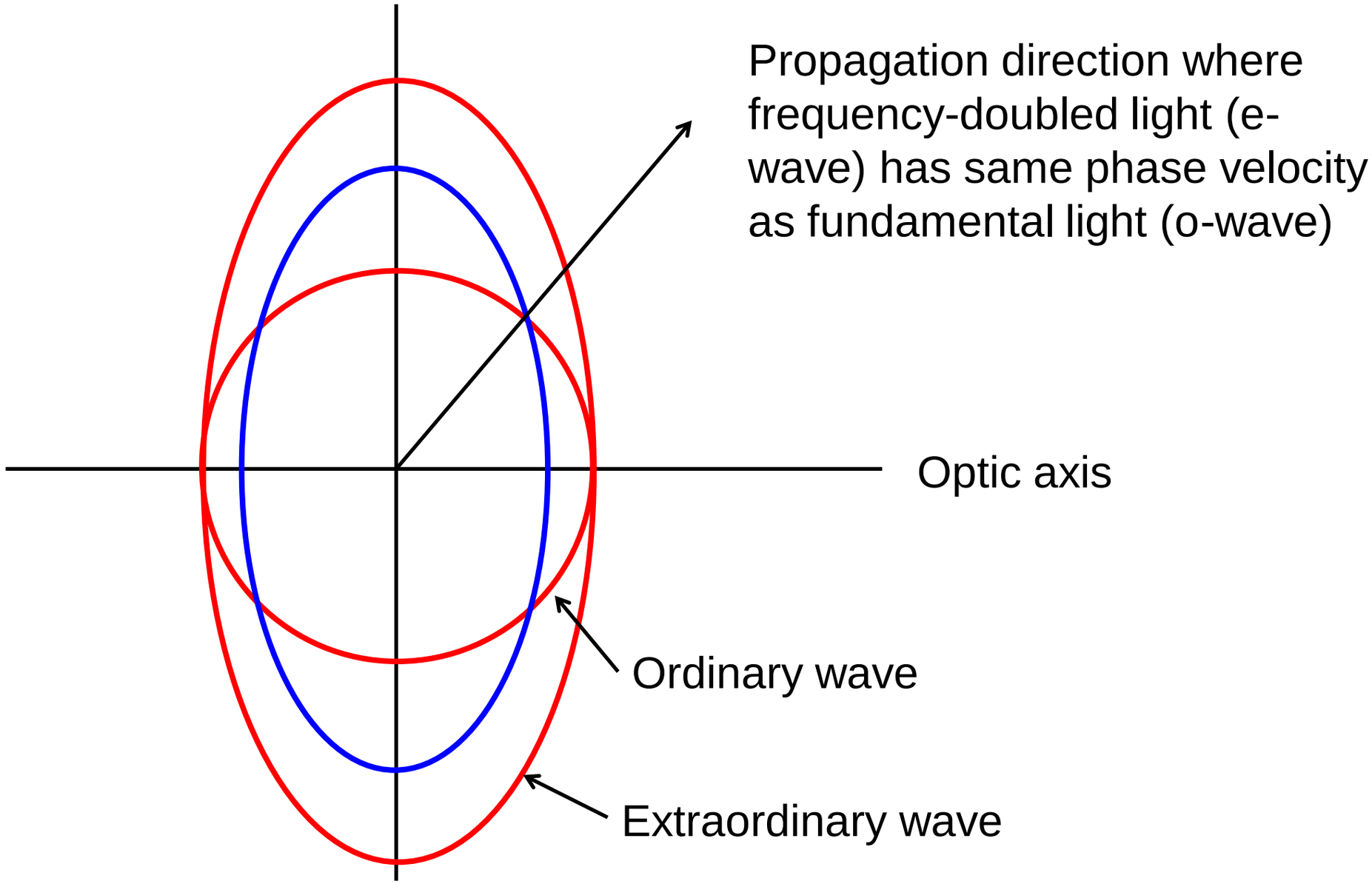
This light can sum coherently through the crystal if the frequency-doubled light has the same phase velocity as the fundamental beam.

Birefringence

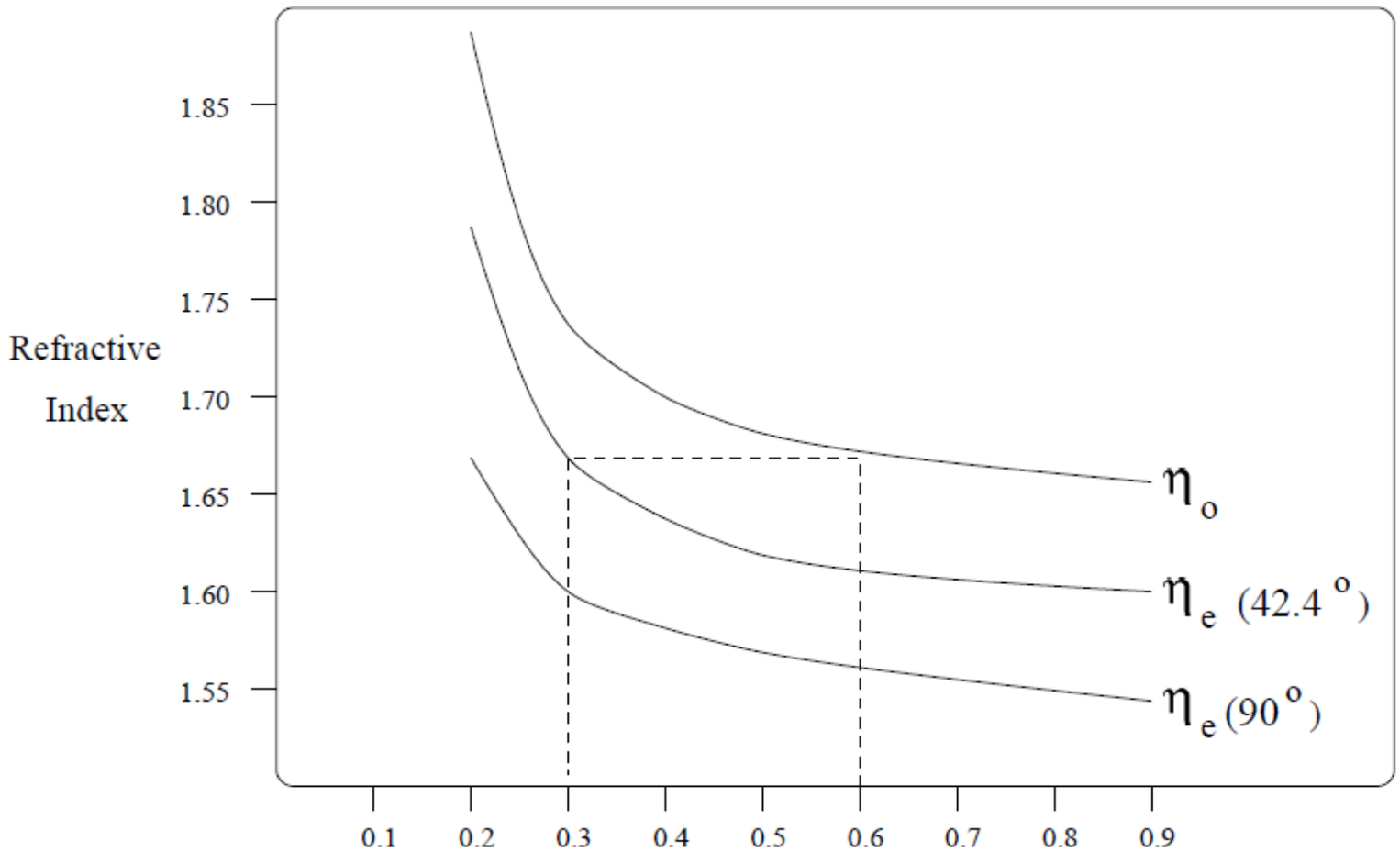
The refractive index of the material depends on the light polarization and the direction of propagation in the crystal.

Ordinary ray has its electric vector perpendicular to the optic axis and its refractive index is the same for all directions of propagation.

Extraordinary ray has a component of its electric vector along the optic axis and its refractive index varies with direction of propagation.

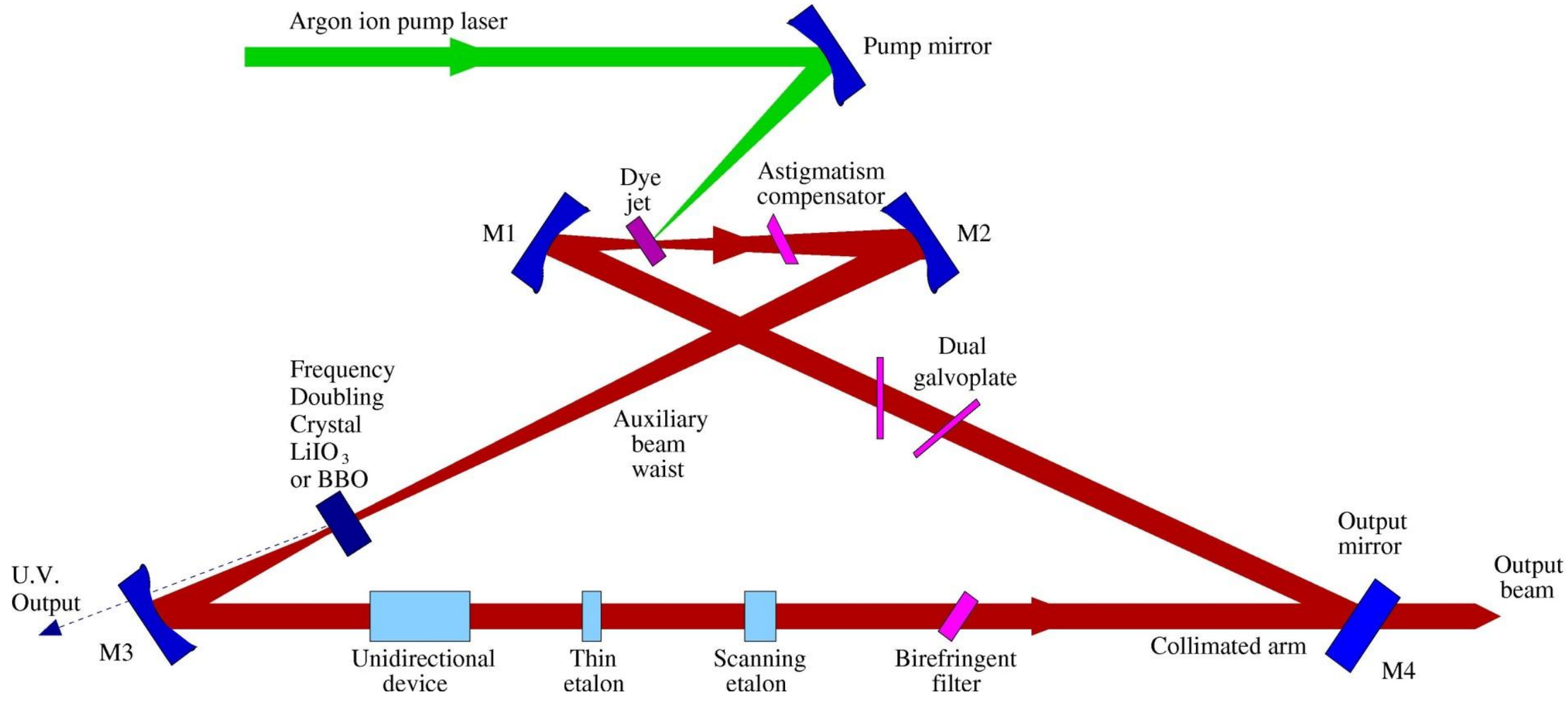


Phase velocity of light in a negative uniaxial crystal



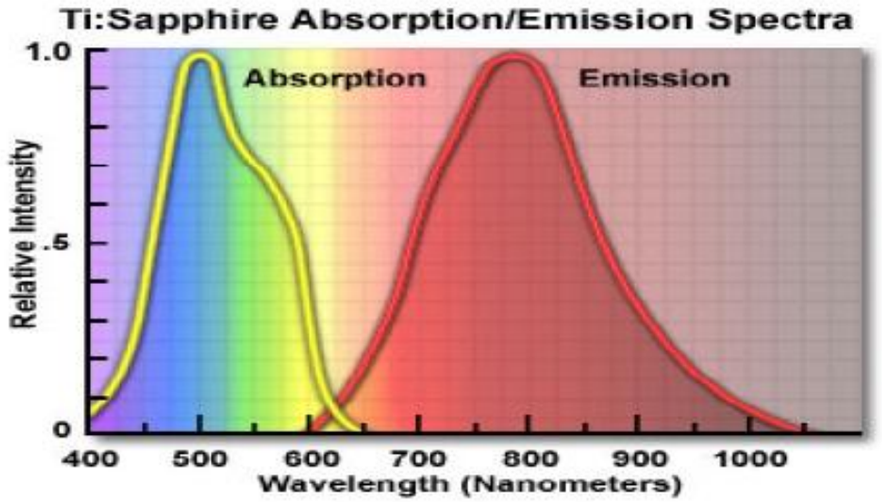
Property	α -Lithium Iodate	β -Barium Borate
Transmissivity >90%	$500 - 5000nm$	$190 - 3500nm$
Nonlinear Coefficients relative to KDP	$d_{31} = 11.9$ $d_{33} = 12.4$	$d_{11} = 5.8$ $d_{31}, d_{22} < 0.05 \times d_{11}$
d_{eff} at 600nm	~ 11	~ 3
Optical homogeneity Δn	$10^{-5} - 10^{-6} cm^{-1}$	$10^{-6} cm^{-1}$
Damage threshold	$60 MW.cm^{-2}$	$1 GW.cm^{-2}$

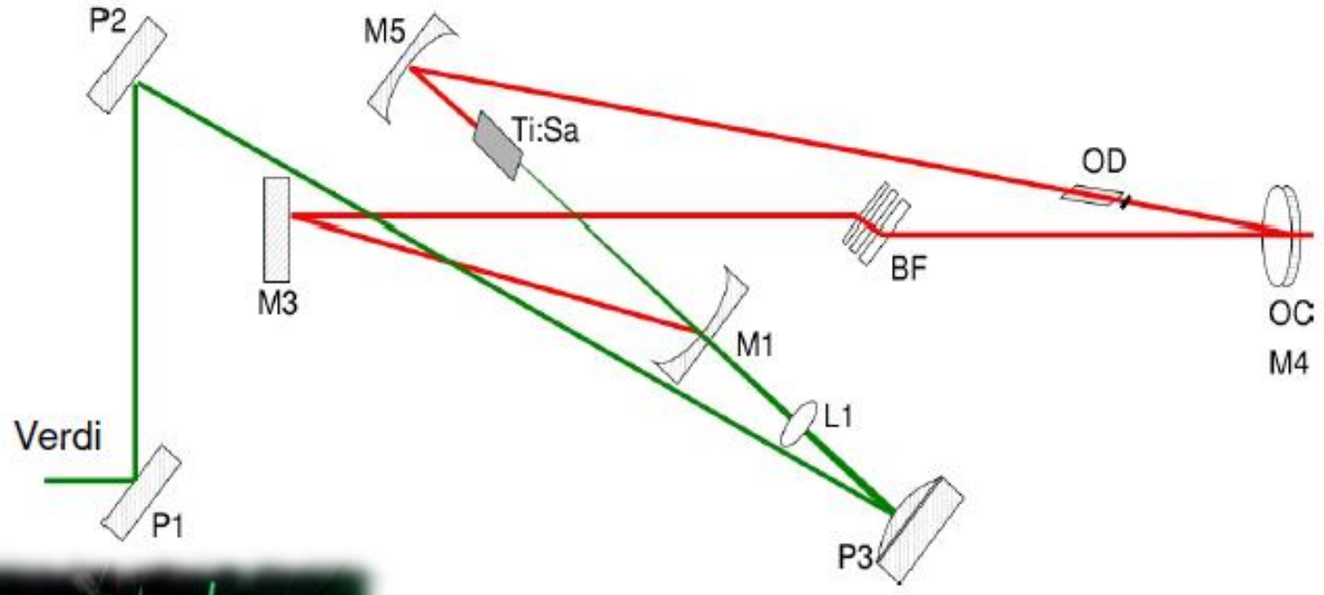
Intra-cavity frequency doubling

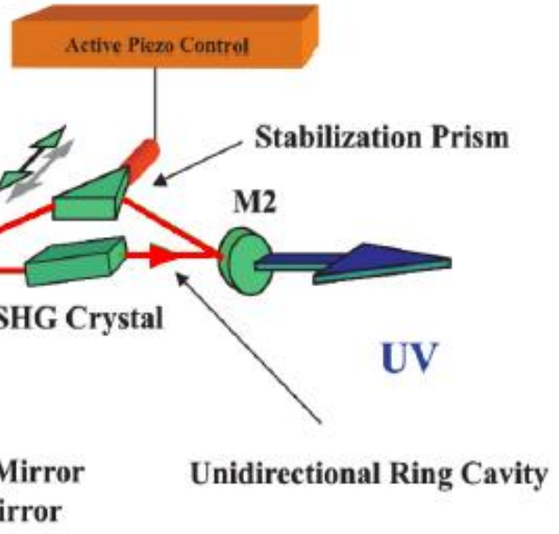


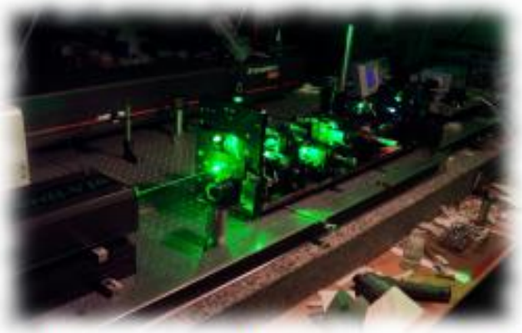
(Spectra Physics 380 single-mode ring laser cavity)

External cavity doubling







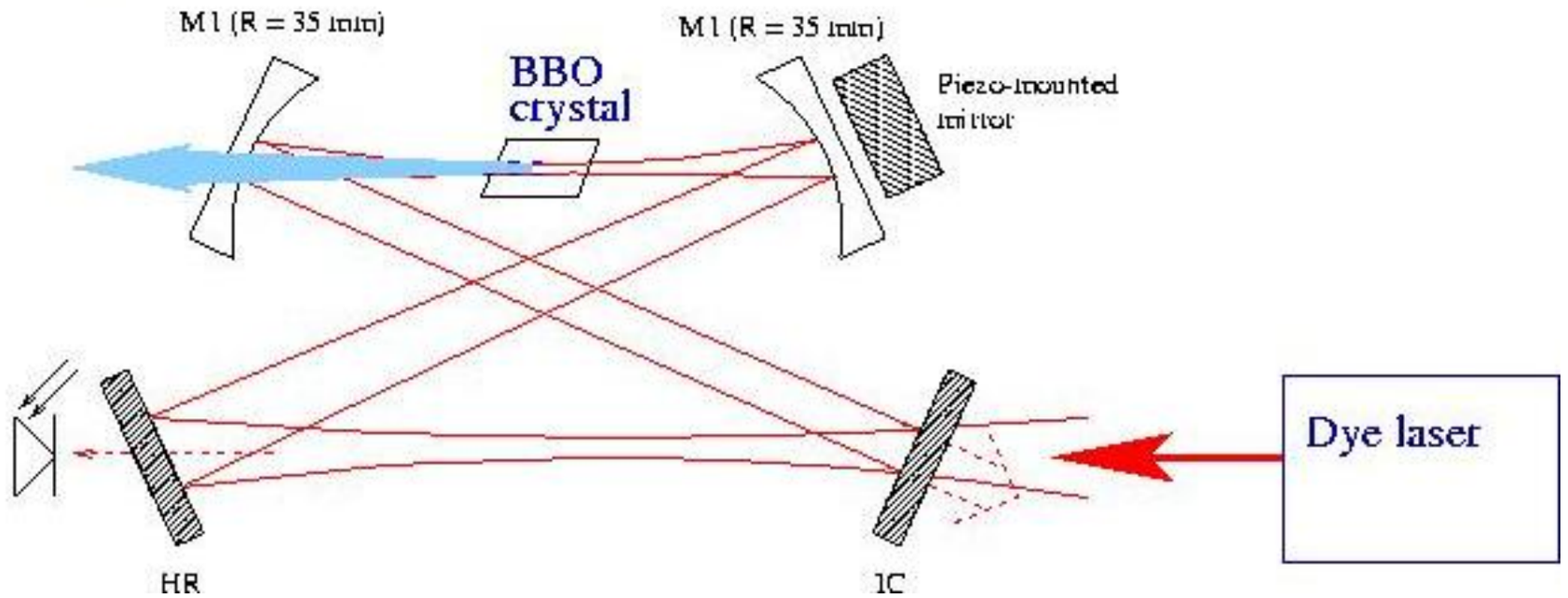


Coherent 899 (P(ω)~ 1W)

Wavetrain Delta cavity
P(2ω) ~ 0.3- 1.5 mW

- External temperature controlled etalon for short term drifts
- Wavelength was monitored with wavemeter (acc ~ 3MHz)

Four-mirror external cavity



Coherent's Monolithic Block Doubler

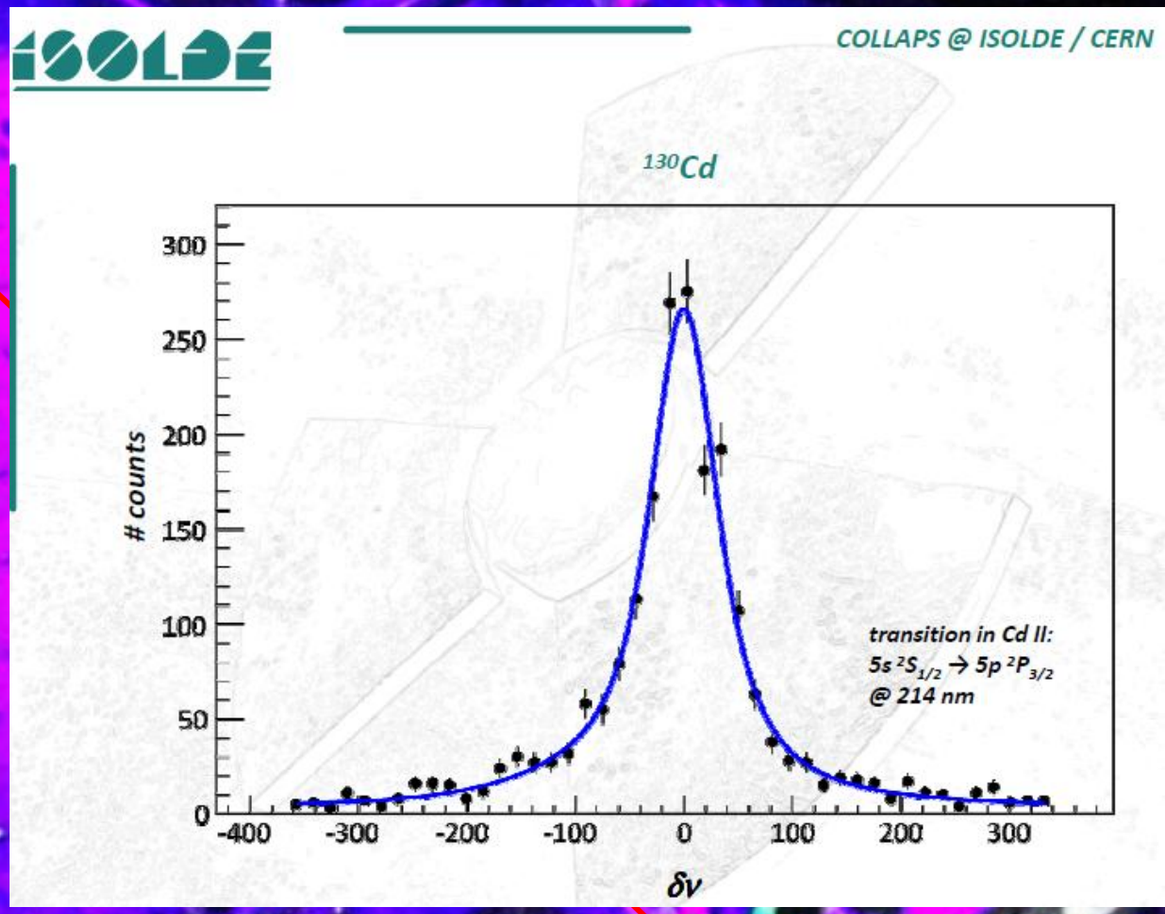
Cd isotopes – Deyan Yordanov

215 nm

430 nm

860 nm

35 mW output maintained over 5 hours



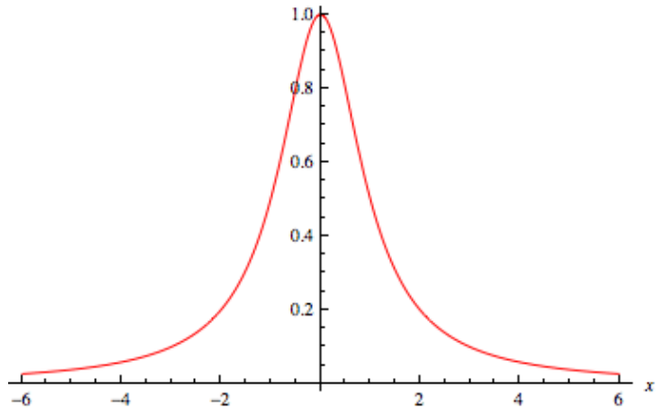
The magneto-optical trap (MOT or Zeeman trap)

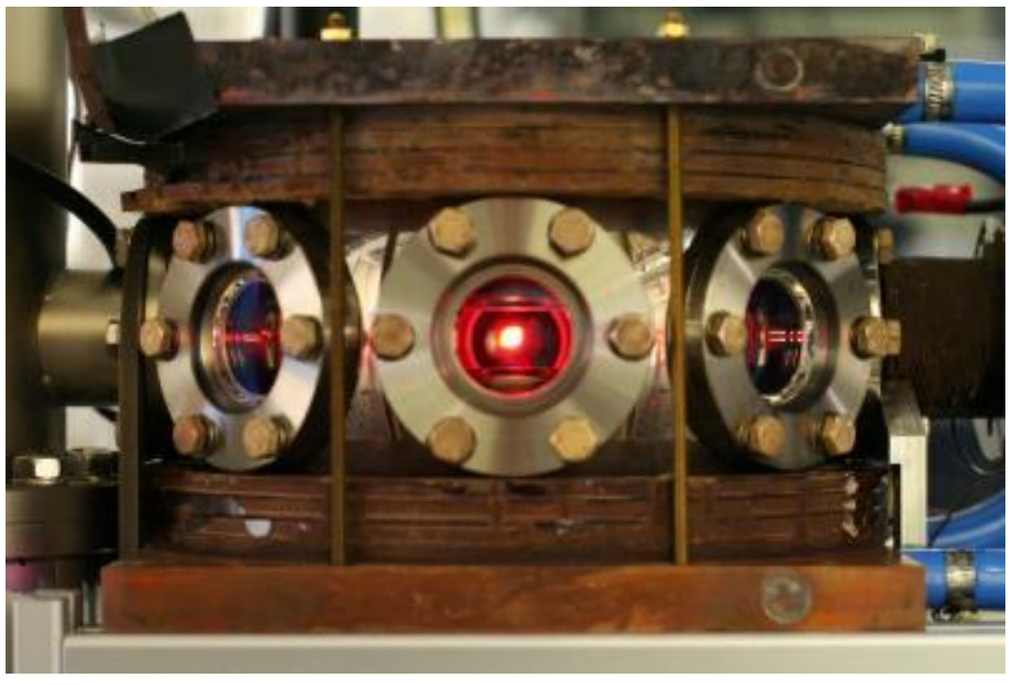
An atom trap needs:

• A velocity-dependent force – to provide damping of the atoms' motions

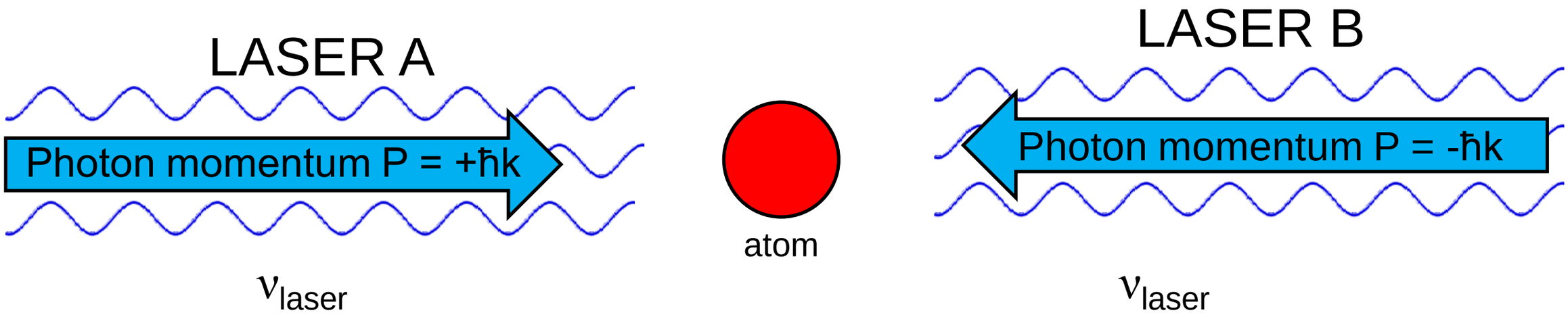
• A position-dependent force – to confine atoms

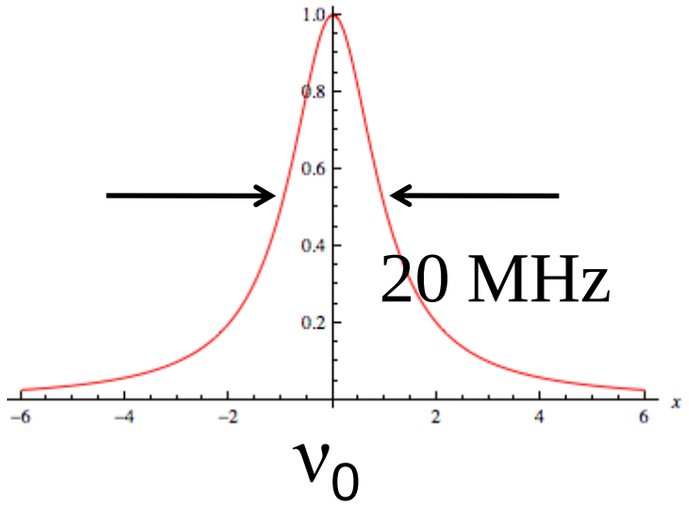
Laser can provide both





The velocity-dependent force





Tune laser slightly below the atom's resonant frequency:

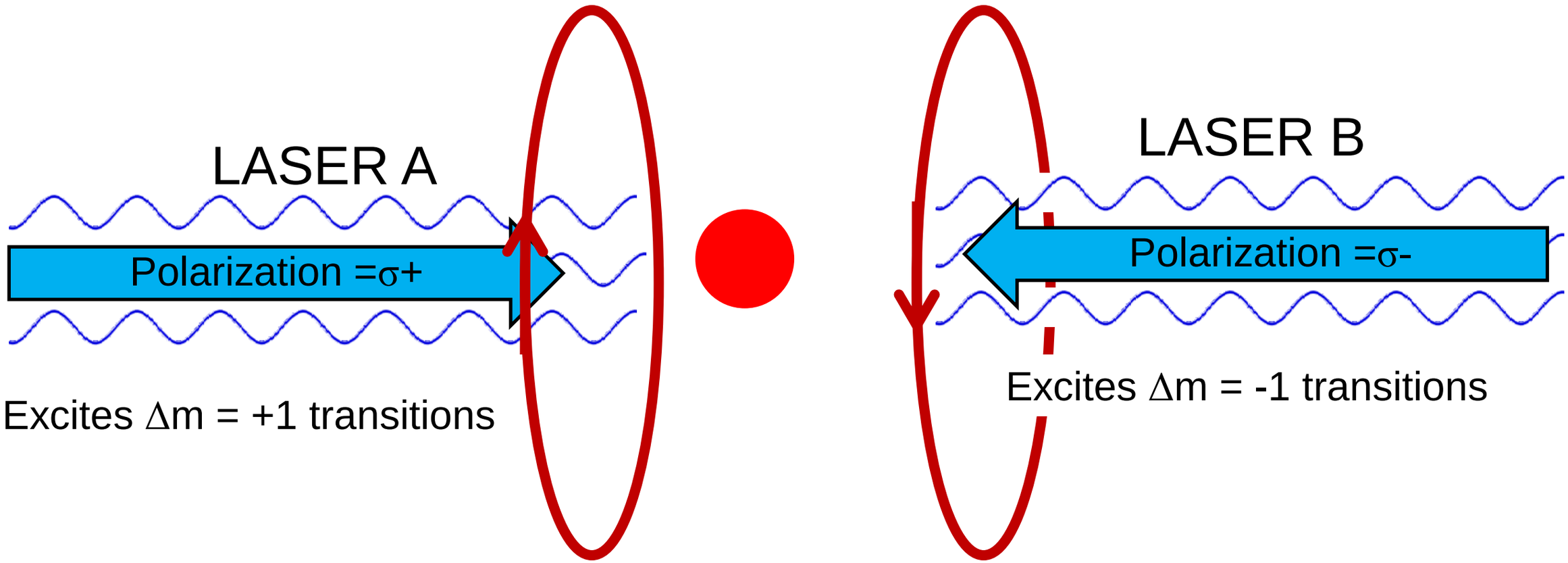
$\nu_{laser} = \nu_0 - 50$ MHz

If atom velocity = 0 it (weakly) absorbs photons from both beams (no net force)

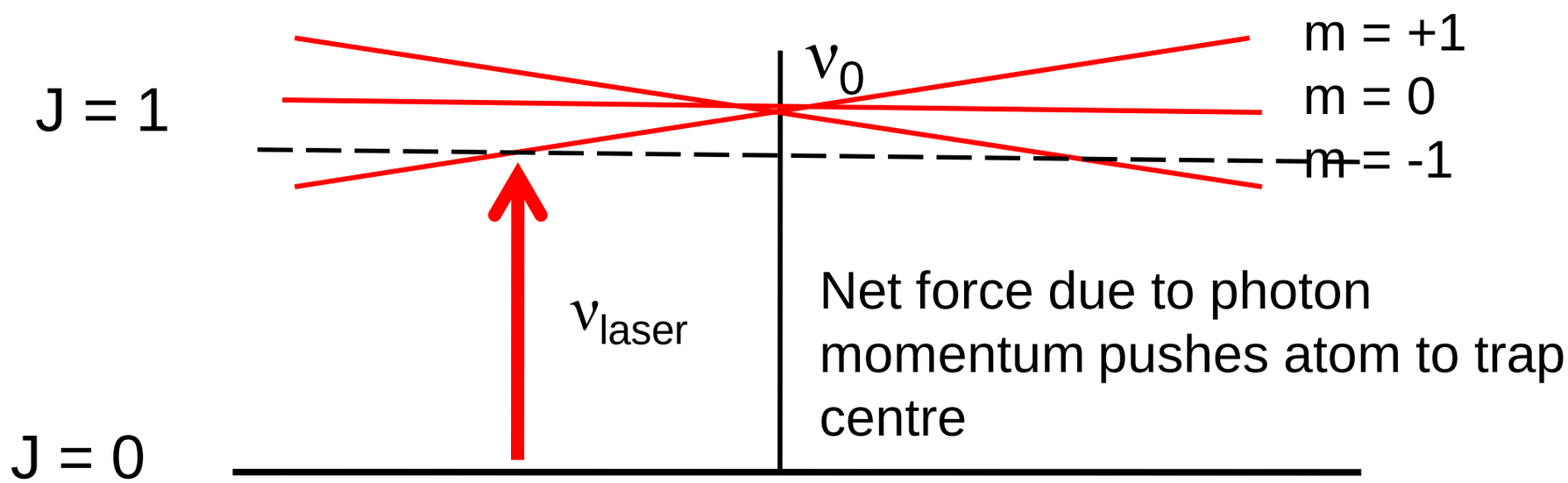
If atom velocity = + 30 m/s it absorbs photons from laser B
(Doppler shift: $(1 + v/c)\ \nu_{laser} = \nu_0$)

Atom repeatedly absorbs (and emits in all directions) photons which slows it down until it is back to zero velocity

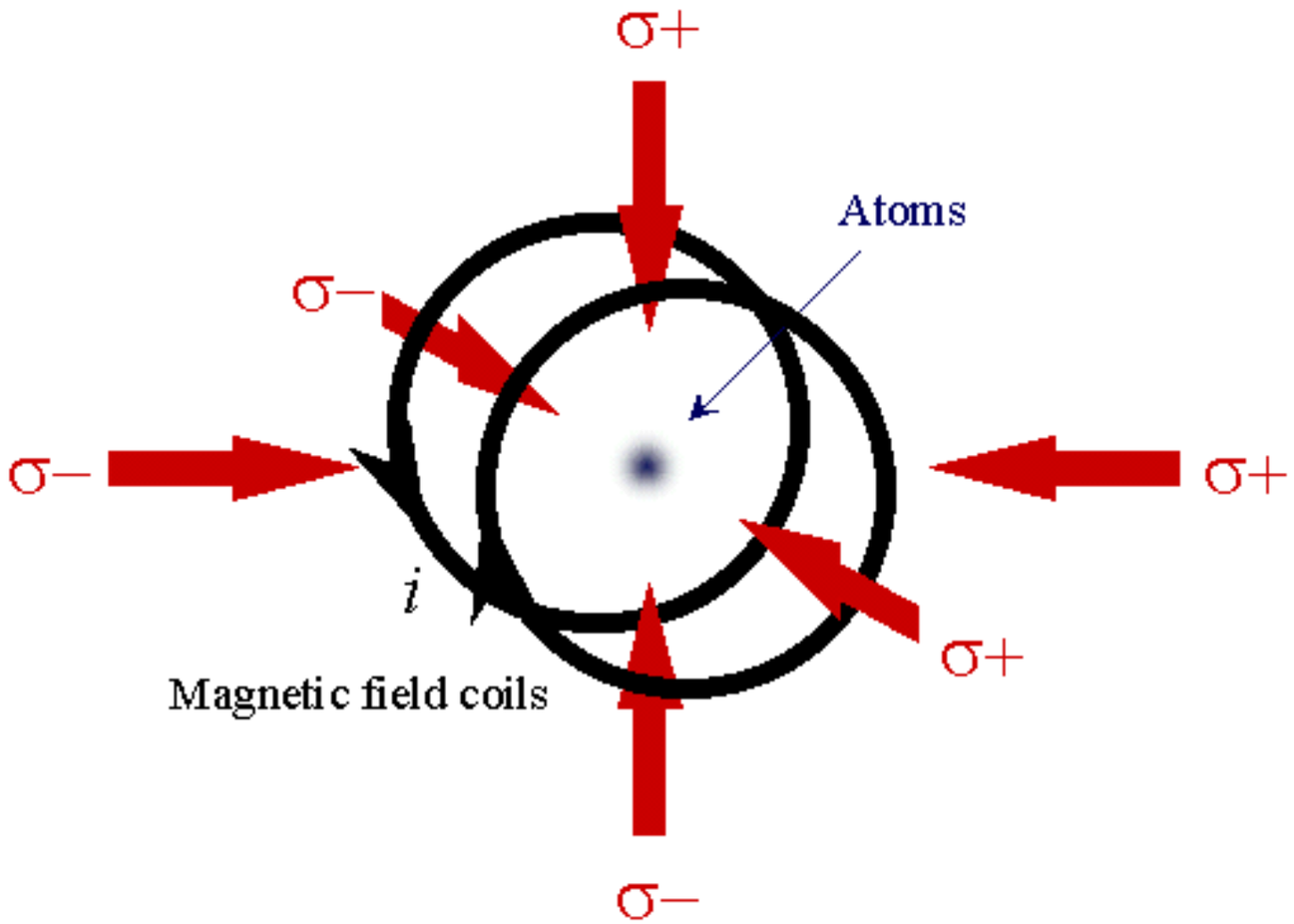
Position-dependent force

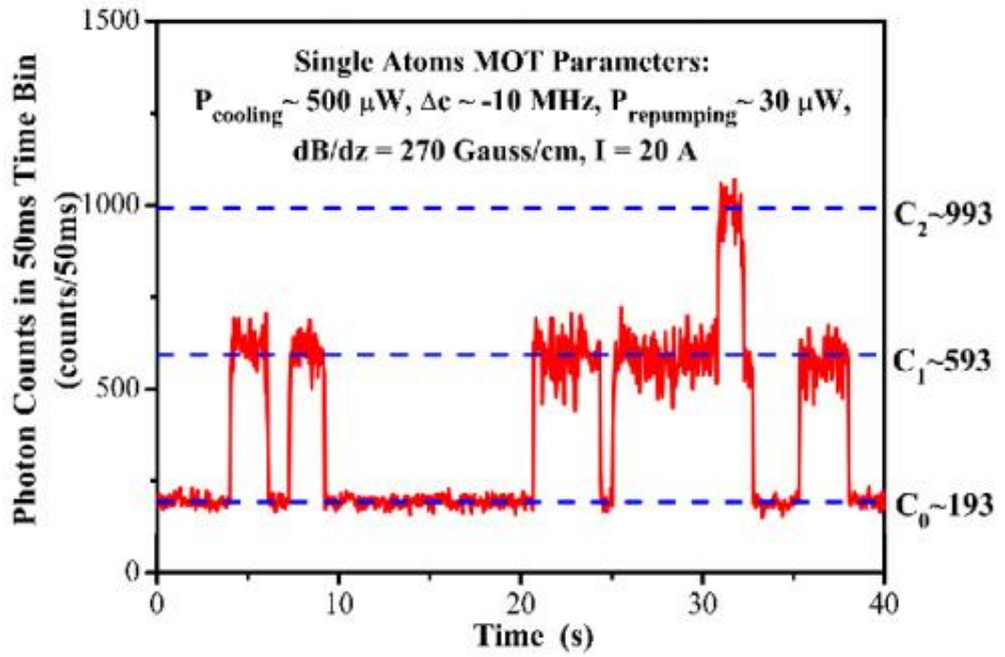


Anti-Helmholtz coils produce null magnetic field at trap centre, but increasing field away from centre which Zeeman-splits the atomic state

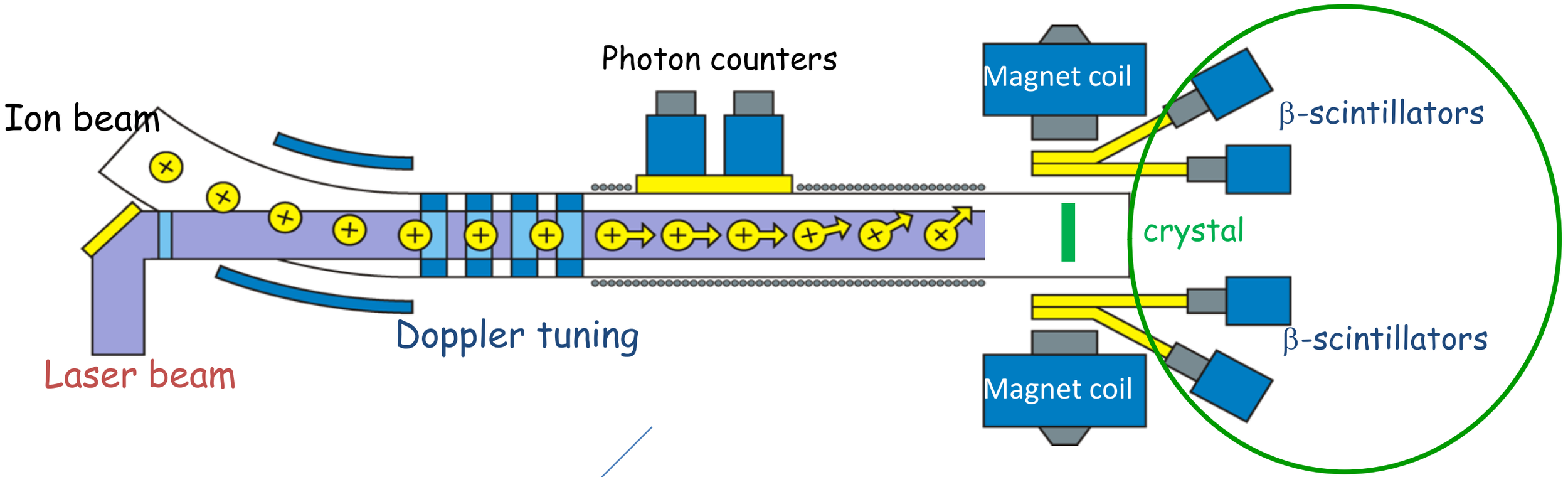


- Trapping works in 3-dimensions
- Laser spectroscopy possible with single atom sensitivity
- *But* it needs a cycling transition that does not pump out

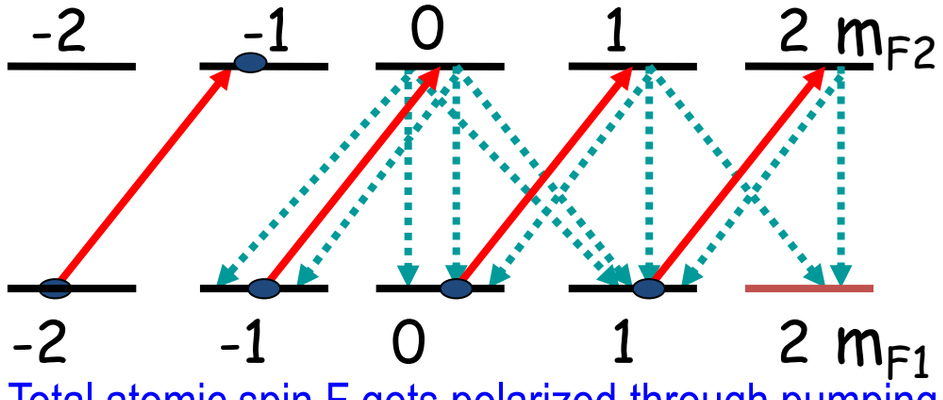




Laser polarization in collinear beams geometry

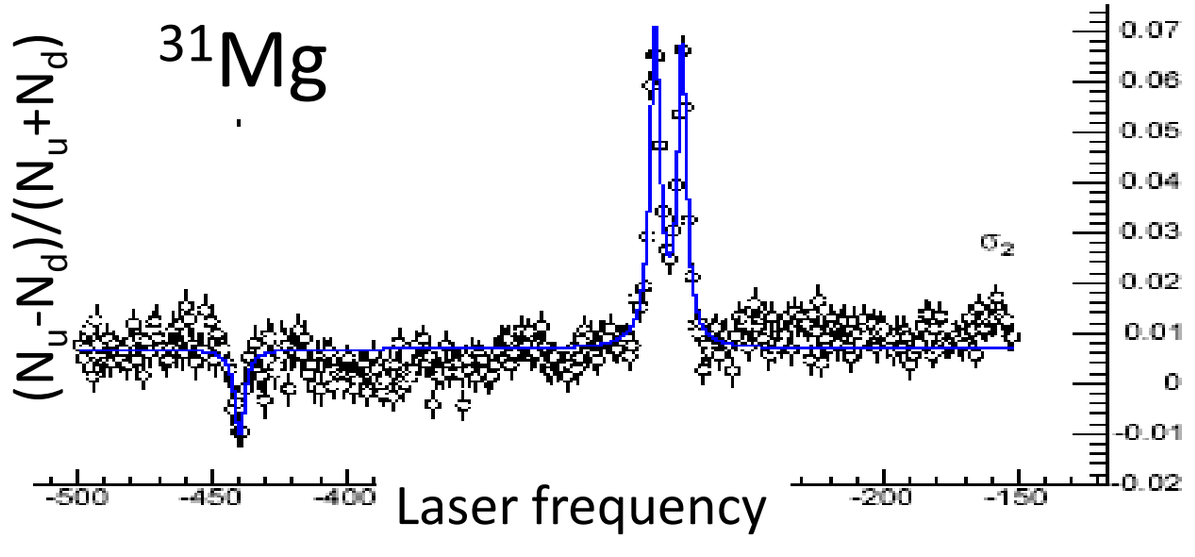


Resonant optical pumping with circularly polarized laser light to polarize the atoms and nuclei

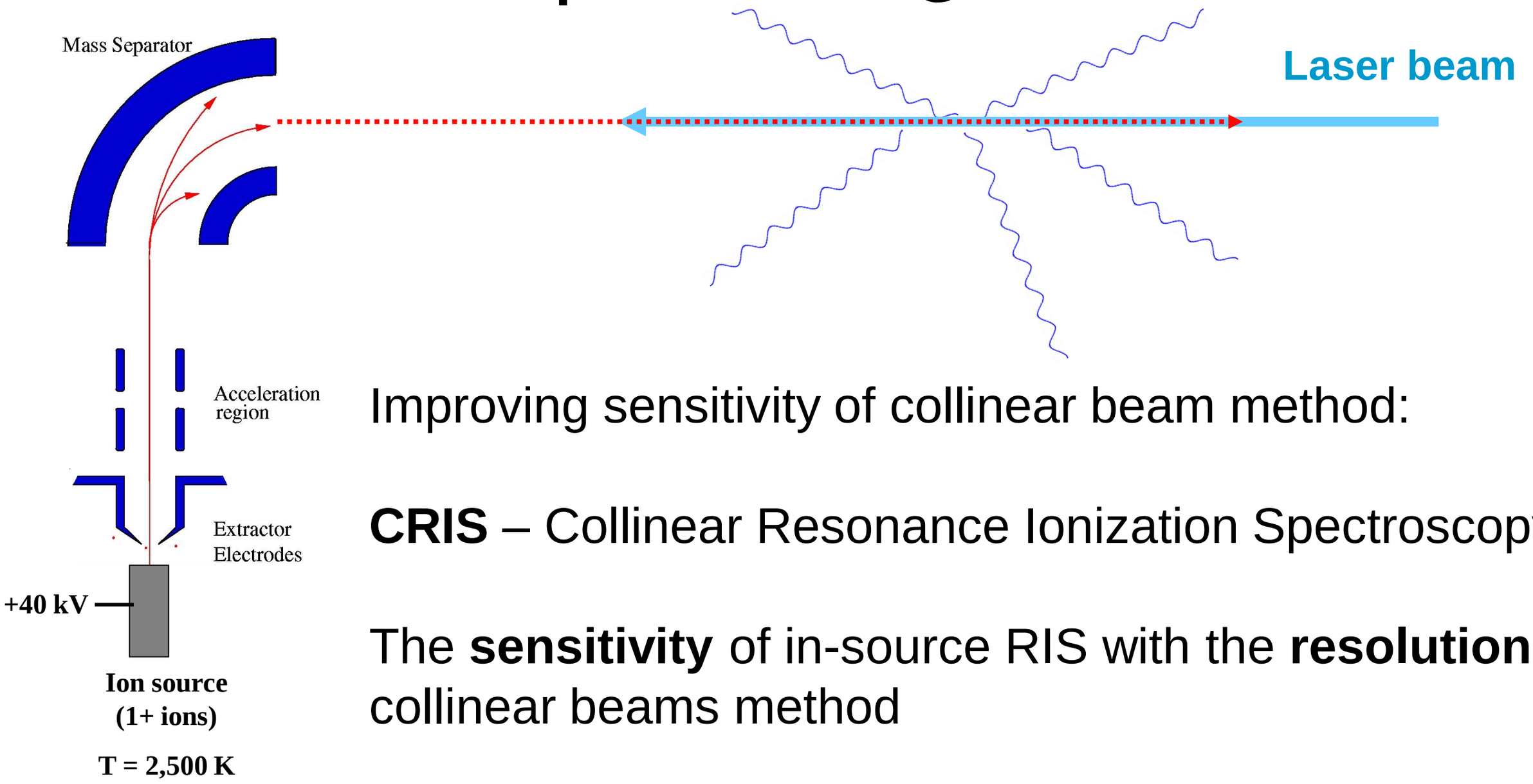


Total atomic spin F gets polarized through pumping !

Detection of HFS via asymmetric nuclear β-decay



Future plans – CRIS@ISOLDE

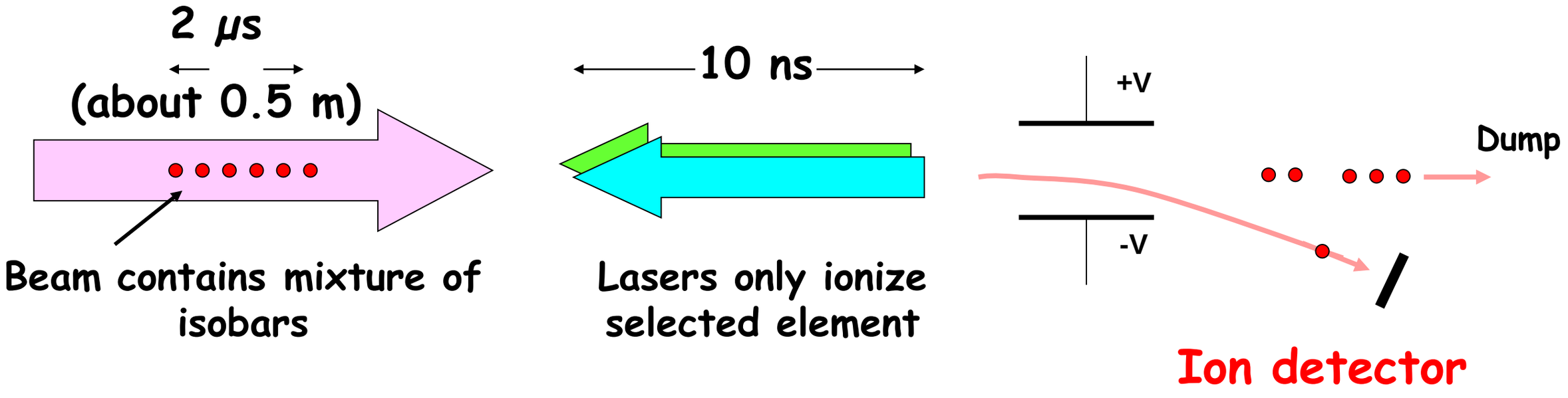


Improving sensitivity of collinear beam method:

CRIS – Collinear Resonance Ionization Spectroscopy

The **sensitivity** of in-source RIS with the **resolution** of collinear beams method

CRIS method: synchronize ion bunch with laser pulse



- Measure ion rate as function of laser frequency to find resonance

- Works well with even low repetition rate lasers – 10 Hz.

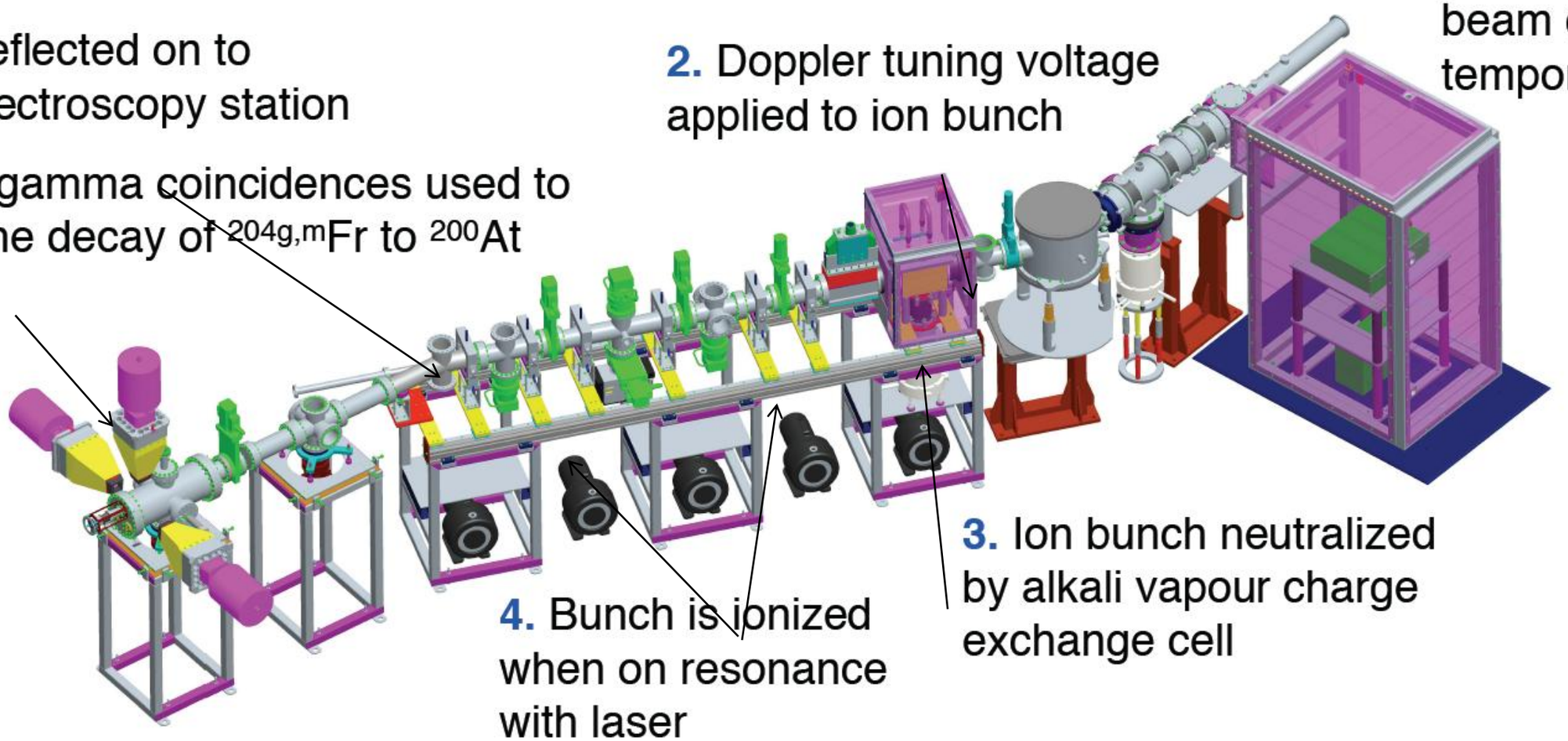
MANCHESTER 1824

The University of Manchester

Collinear Resonance Ionisation Spectroscopy CRIS@ISOLDE

1. Bunched ion beam of ~1μs temporal width

2. Doppler tuning voltage applied to ion bunch



3. Ion bunch neutralized by alkali vapour charge exchange cell

4. Bunch is ionized when on resonance with laser

(two-step resonance ionisation)

5. Ions deflected on to decay spectroscopy station

6. Alpha-gamma coincidences used to identify the decay of ^{204g,m}Fr to ^{200}At

ISOLDE

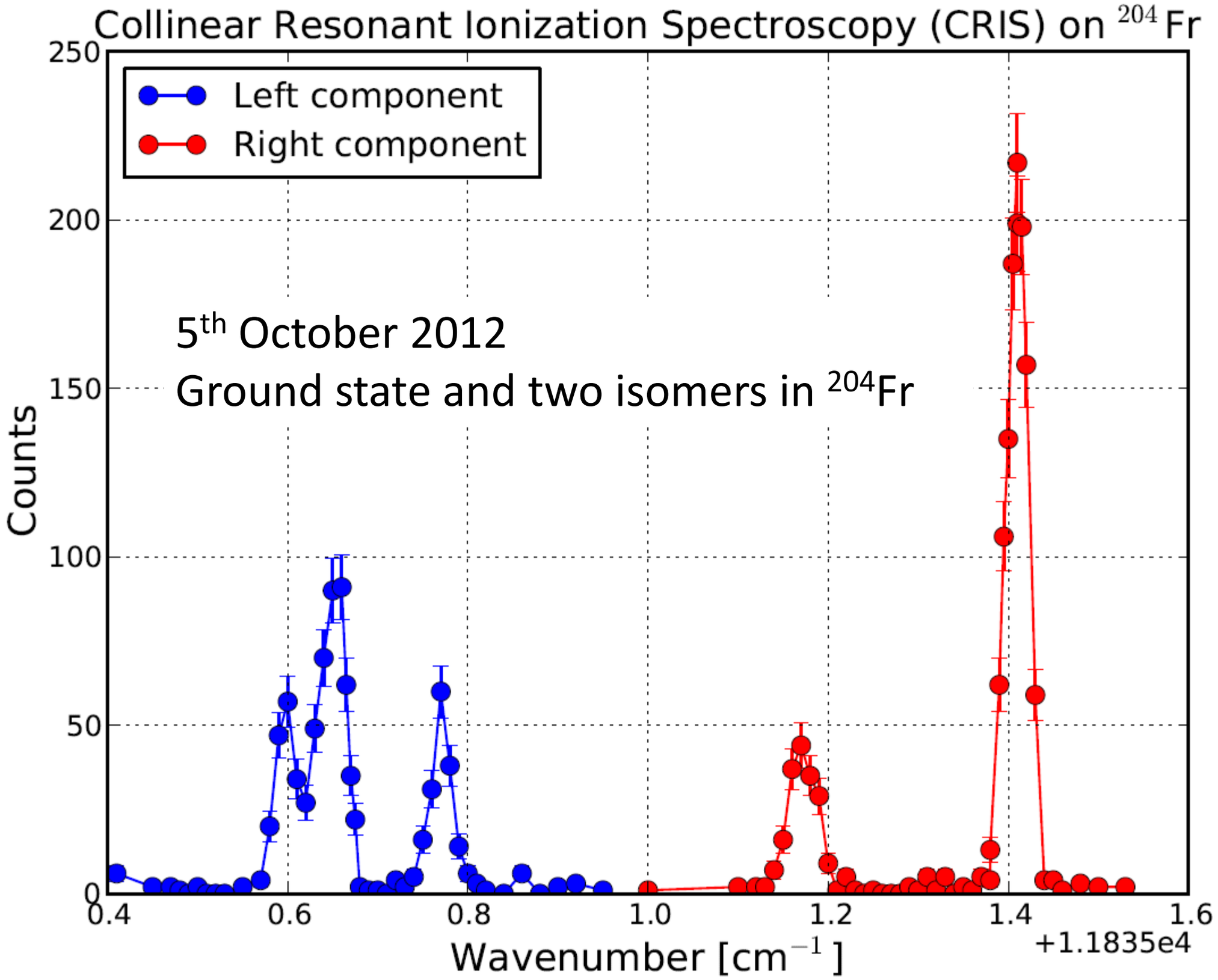
MANCHESTER 1824

KATHOLIEKE UNIVERSITEIT LEUVEN

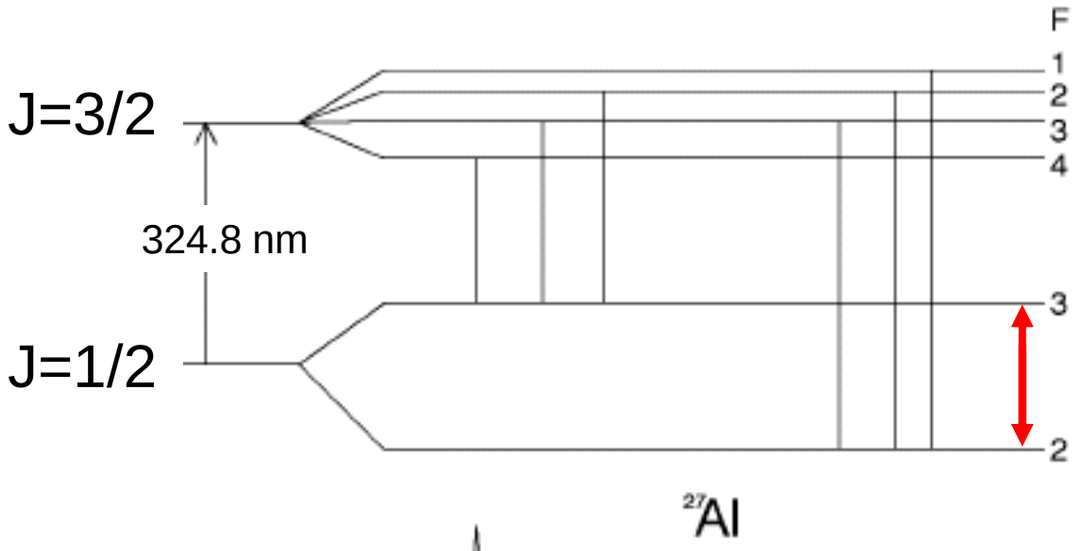
From K. Lynch, ARIS-2011 poster

MANCHESTER 1824

The University of Manchester



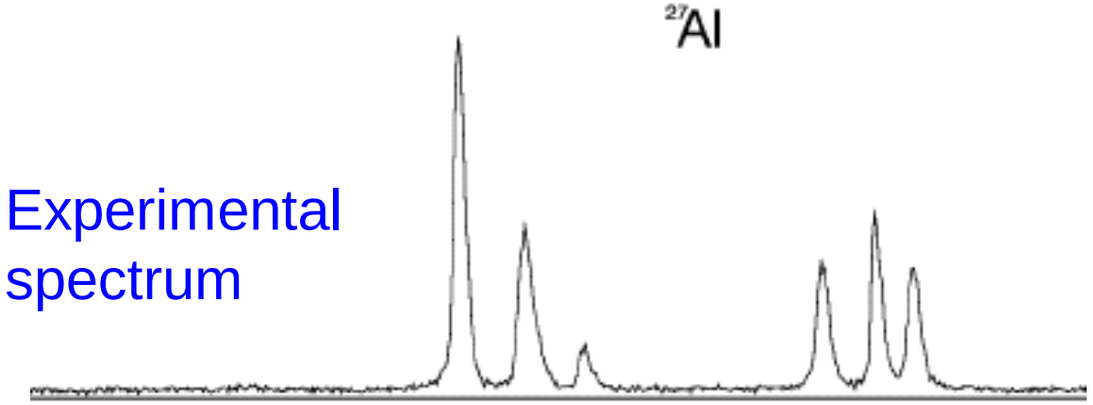
Determining the nuclear spin, I



Intervals depend on A_{upper} , B_{upper}, and I, J, F

Interval depends on A_{lower}, and I, J, F

Ratio A_{upper}/A_{lower} is independent of nuclear moment (ie same for all isotopes)



If the wrong value of I is used to fit the hyperfine structure then:

- May be impossible to fit structure (position or number of peaks)
- Deduced ratio A_{upper}/A_{lower} is wrong
- Deduced relative peak intensities are wrong (Racah coefficients)