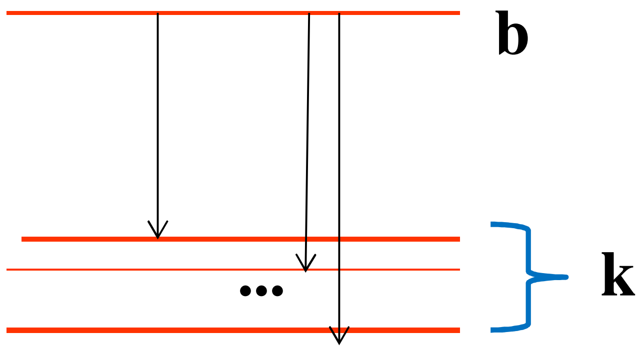
4(b) - Atomic lifetimes

Let us consider N(t) atoms in an excited state b at a particular time t.



The rate of change of N(t) is

$$\frac{dN(t)}{dt} = -N(t)\sum_k W^s_{kb}$$

Sum over all states k to which decay is allowed

Decay rates from b to k

Integrating:

$$N(t) = N(t=0)\exp\left(-t/\tau_b\right)$$

where $\tau_b^{-1} = \sum_k W^s_{kb}$ is the lifetime of level b

Please note:

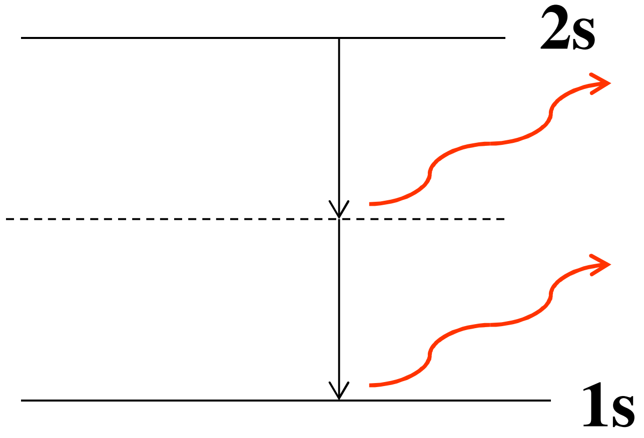
- In the absence of external fields, the lifetime of an atom cannot depend on the magnetic quantum number m of the level b (atoms = spherically symmetric systems)

- The lifetimes of hydrogenic ions are shorter than those in Hydrogen. They are given by the scaling law

$$\tau(Z) = Z^{-4}\,\tau(Z=1)$$

- In the dipole approximation the lifetime of 2s in H is infinite
- In practice, $\tau_{(2s)} \sim \frac{1}{7}\ \mathrm{s}$

(two-photon emission)



5- Measurements (radiative lifetimes)

- Importance of oscillator strengths (f values) of spectral lines: computations of gas discharges, plasmas, stellar atmosphere, etc
- In general, theoretical predictions are not accurate: experimental measurements are necessary

Direct measurements: f can be determined through the intensity of the spontaneous emission of a given spectral line.

$$I_{ki} \propto \underbrace{A_{ki}}_{\text{transition probability}} \; \underbrace{\dot{N}}_{\text{density}} \; \underbrace{V}_{\text{volume}} \; \hbar\omega_{ki}$$

Problem: in many cases, N cannot be determined accurately

Solution: f is determined by measuring radiative lifetimes

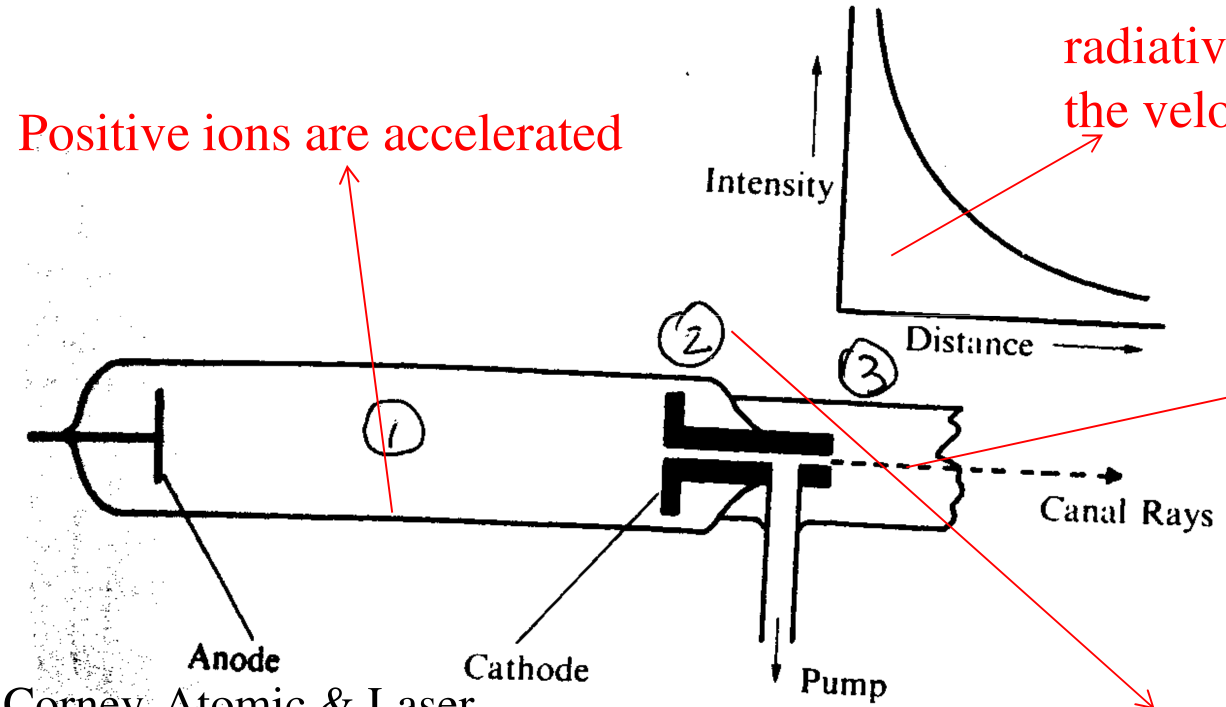
5(a) – The beam-foil method

Key idea: *"The time dependent exponential decay of excited atoms may be converted into a spatial variation of intensity by exciting a beam of fast-moving atoms at a given position".*

Positive ions are accelerated

The length of the glow allows the radiative lifetime to be measured if the velocity of the atoms is known

Moving atoms decay- a faint glow is emitted.



Corney, Atomic & Laser spectroscopy, p. 145

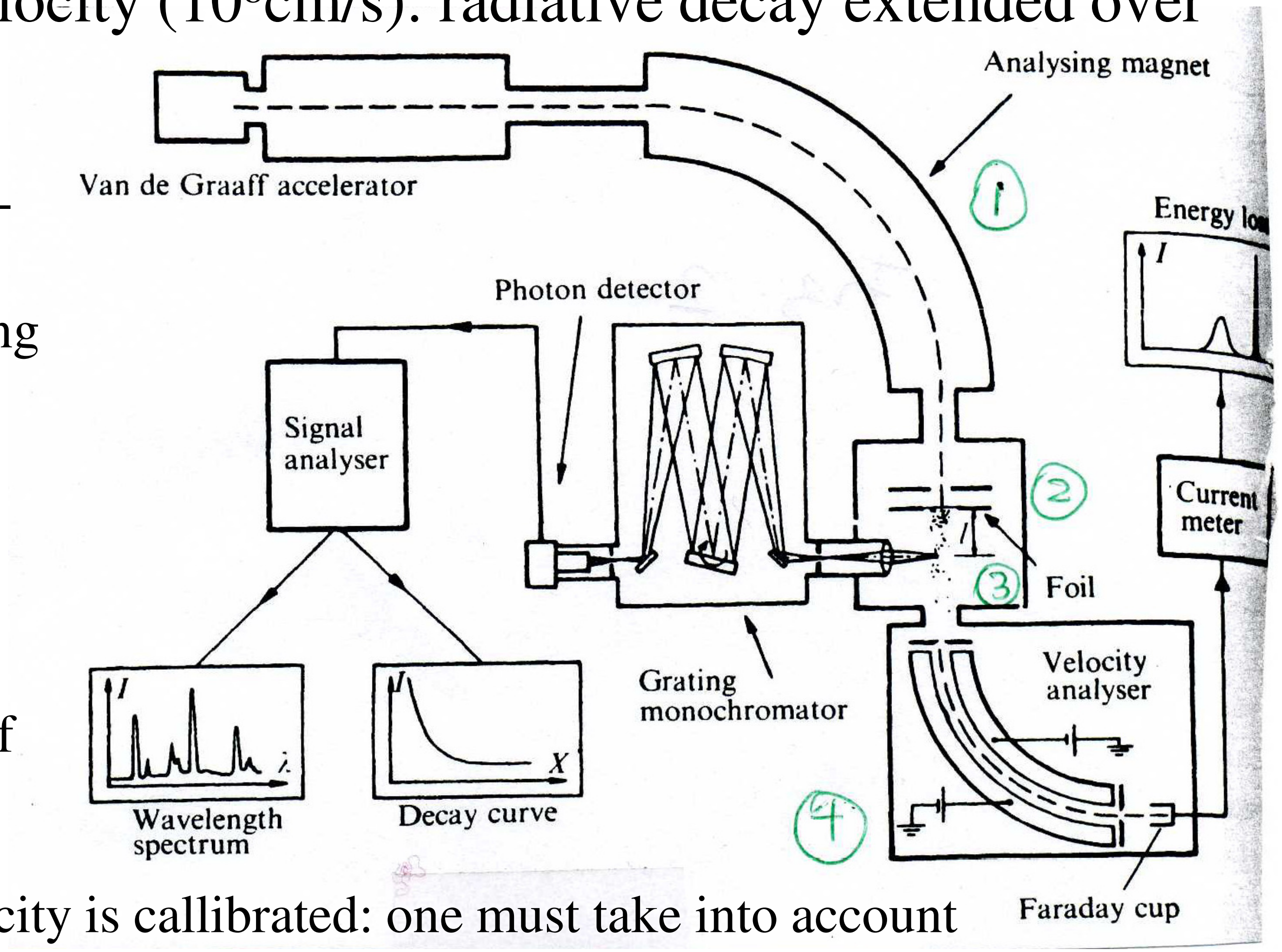
Excitation/excited atoms capture e^-s

Problem: repopulation of excited states by radiative decay from higher levels: too long lifetimes (overcome by Koenig+Ellett(1932) using optical excitation) and low velocity of the beam ($t > 10^{-6}$ s)

5(a) – The beam-foil method

Modern technique (Kay, 1963; Bashkin, 1964):

- Based on the fact that ions in the beam of a van der Graaf accelerator can be strongly excited by passing through a thin carbon foil
- Advantage: high velocity (10^8cm/s): radiative decay extended over several cms



1. Acceleration of ions- initial v spread determined by analyzing magnet

2. Ions collide with the foil: excitation

3. Ions decay: intensity/wavelength of emitted radiation is detected

4. Beam velocity is callibrated: one must take into account the energy loss of the ions passing through the foil

5(a) – The beam-foil method

Calculation of lifetimes

$$\underbrace{N_k(l)}_{\text{Density of ions at position } l} = \underbrace{N_k(0)}_{\text{Density of ions at the downstream face of the foil}} \exp(-t/\tau_k) \quad t = l/v$$

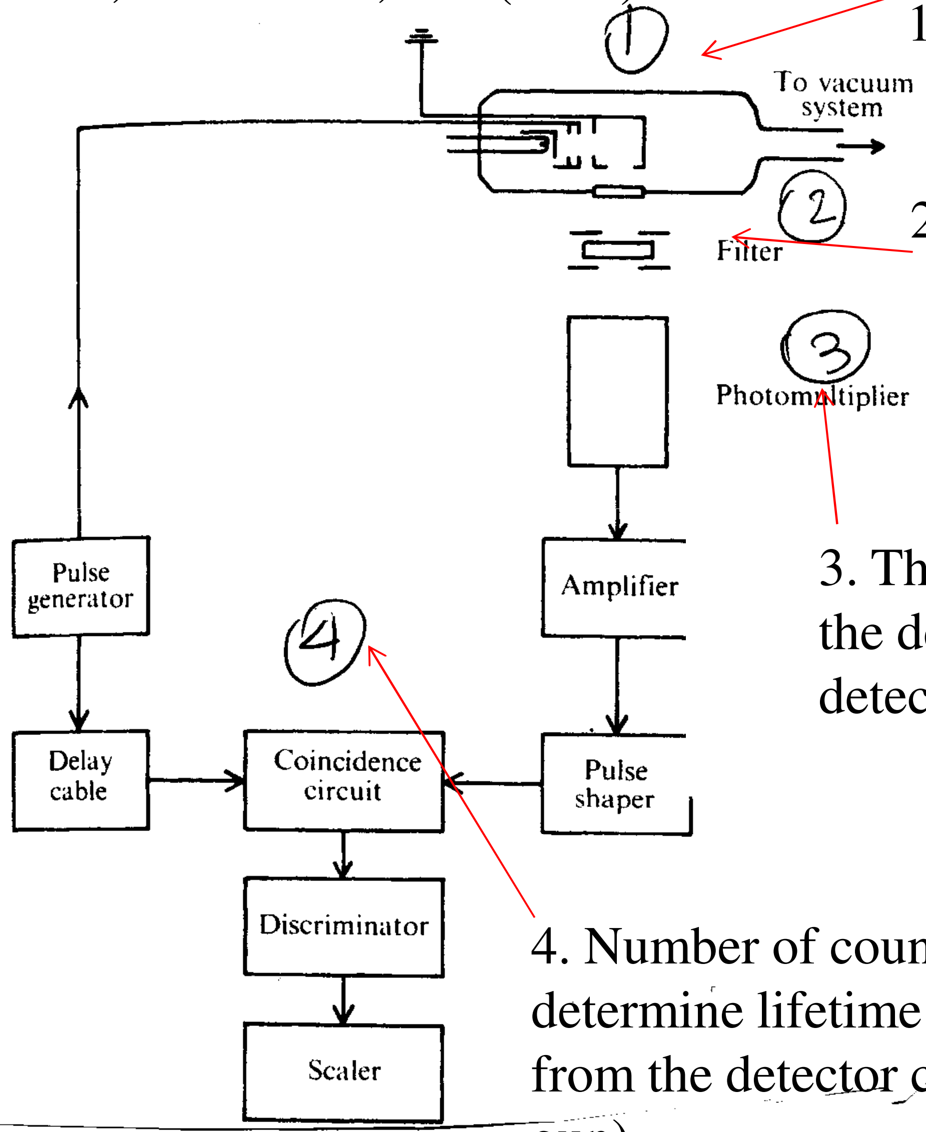
$\ln N_k(l)$ as a function of *l*: slope gives τ_k

Problem: radiative cascade: the plot shows a pronounced curvature due to the repopulation of the level being studied by the decay from higher-lying levels

- Remember: the ions are excited by collisions and this is an unselective process
- The radiative cascade can be avoided by replacing the foil by an intense laser: strict selection rules

5.(b) – The delayed coincidence method

Heron et al, Nature 174, 564 (1954)



1. A sample is excited to some level k

2. The excitation is cutoff instaneosly

3. The emitted photons produce a pulse in the detector – the number of pulses is detected

4. Number of counts vs delay time enables to determine lifetime (coincidence measurement: pulses from the detector compared to pulses from an electron gun)

5.(c) – The time of flight method

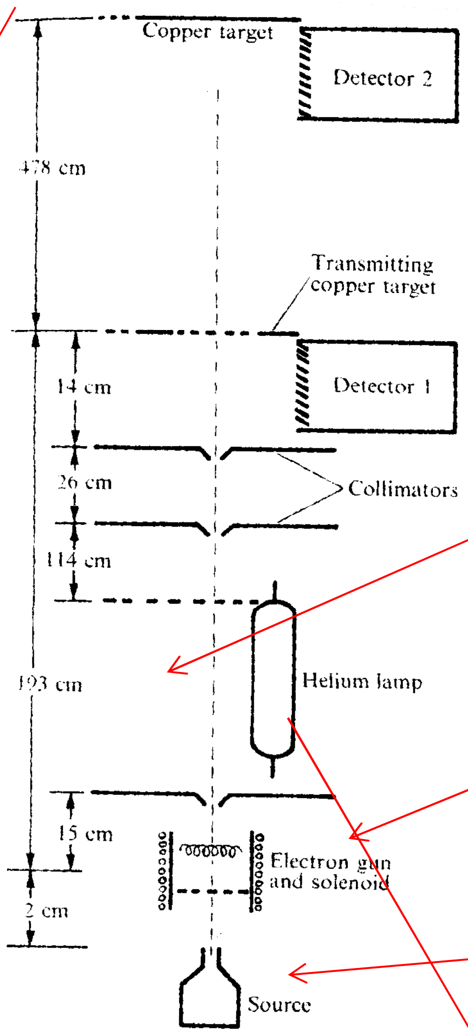
van Dyck et al, Phys. Rev. Lett. 25, 1403 (1970)

- Importance: this method is widely used for determining the lifetimes of metastable states (typically $>10^{-3}$s)

- Problems with beam-foil/delayed coincidence method:
  - Beam foil method: no decay would be observed for an apparatus of realistic size
  - Delayed coincidence method: the time necessary for obtaining sufficient data is unrealistic

Example: metastable states in He, He^+

5.(c) – The time of flight method



The gas collides with 2 targets:
(a) A 60% transparent copper mesh
(b) A copper plate
Collisions: electron multipliers detect electrons ejected from the target.

Excited atoms/molecules enter a low pressure region

Electron guns excite them to the required metastable level

Atoms/molecules pass through a cooled source chamber/effuse through a slot in the pumping region

Output pulses are recorded as functions of time after excitation: velocity distributions are determined

Lamp: if there are 2 close metastable levels, it excites the atom to a close, non metastable level

5.(c) – The time of flight method

Why "time of flight"?

One may determine the number of atoms which decay between both detectors by comparing the number of metastable atoms arriving at the detectors within specific velocity intervals

$$\underbrace{N_i(v)}_{\substack{\text{number}\\ \text{of atoms}\\ \text{at detector}\\ i}} = \int_{\text{surface}} \overbrace{\varepsilon_i}^{\text{surface efficiency of}} \underbrace{N_0(v)}_{\substack{\text{initial}\\ \text{number of}\\ \text{atoms}}} \exp(-t_i/\tau) \underbrace{ds}_{\substack{\text{area of}\\ \text{the detector}}}$$

If the initial velocity distribution is uniform across the beam

$$N_i(v) = \underbrace{\boxed{\int_{\text{surface}} \varepsilon_i \, ds}}_{\textbf{const.}} \; N_0(v) \exp(-t_i/\tau)$$

5.(c) – The time of flight method

Ratio (numbers of atoms at both detectors)

$$R = \frac{N_b(v)}{N_a(v)} = \frac{C_b}{C_a} \exp(-t/\tau)$$

$$t = t_b - t_a = \text{"time of flight"}.$$