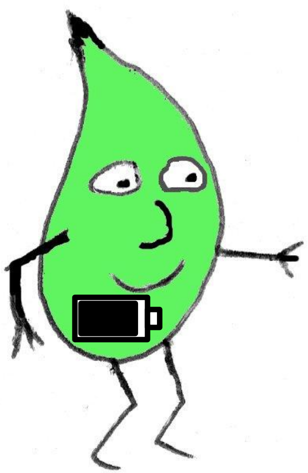
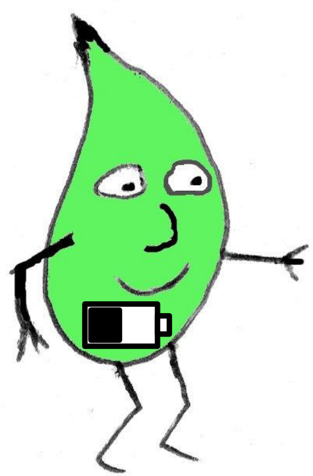
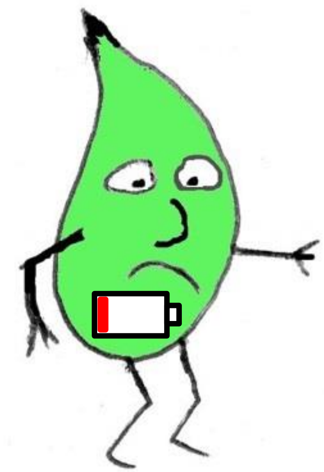
Science & Technology Facilities Council
ISIS Neutron and Muon Source

Charge states of hydrogen (and the muon)

James Lord

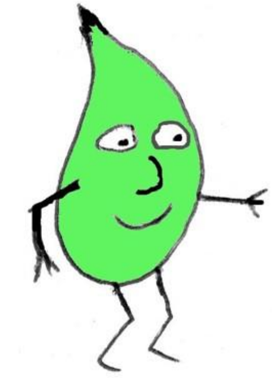


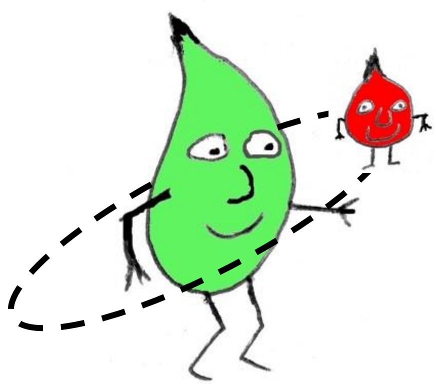


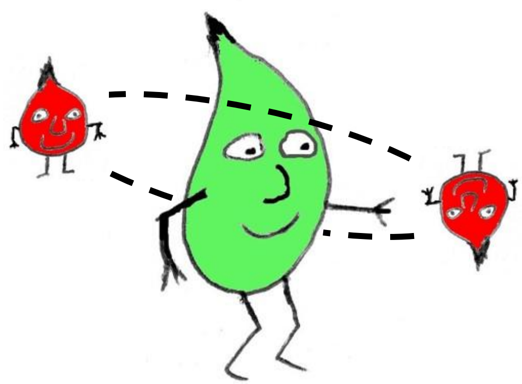


The positive muon

- Charge +e
- Point-like
- Mass >> m_e
- Looks like a hydrogen nucleus
- Can bind 1 or 2 electrons to form Mu^0 or Mu^-







Science & Technology Facilities Council
ISIS Neutron and Muon Source

Mu^+ (muon without electrons)

- In vacuum or gas: just Larmor precession
- In simple solids: finds interstitial space. Dipolar coupling with nuclei
- In oxides or fluorides: finds a location with high electron density (lone pairs) to form O-Mu or F-Mu-F
- In some materials inserts into an existing bond (e.g. Si)
- Electron wavefunctions stretched to screen the muon, but still overall positively charged defect

Mu^0 (muon and one electron)

- “Hydrogen atom”
- In vacuum or gas: hyperfine coupling A=4463.302872… MHz
  - Repolarisation in LF
  - “Triplet” precession in low TF, 1.4 MHz/Gauss
- Liquid (solvated) or solid (interstitial): some spin density spreads onto neighbouring atoms (or sometimes squeezed). A=1500-4500 MHz, nearly isotropic

Mu^0 continued

- Reactions with host:
- Small molecules: attacks double bonds or rings. Forms radical.
- Lattices and polymers: attaches to bond centres or reactive atoms/bonds.
- Typical A_{Mu}=10-500 MHz, strongly anisotropic (but looks isotropic in a liquid or gas)
- Also hyperfine coupling with other nuclei in the molecule (e.g. H)

Mu^- (with two electrons)

- Less common than Mu^+ or Mu^0
  - Requires source of extra electrons
- Two electrons in 1s orbital, spins paired up/down so S=0
- In vacuum or gases: spin behaves as bare muon
- In liquids or solids: finds a large interstitial space or solvation cage. Dipolar coupling to nuclei.

Initial charge state

- High energy muon can capture an electron from the lattice, forming Mu^0
- Fast Mu^0 then has the electron stripped off
  - Rapid charge cycling
- Certain probability of Mu^0 when the muon comes to rest
- Depends on electron density, work function, implantation energy
- Usually independent of T

Charge state conversion

- Ionisation and carrier capture (e^- and h^+)
- Often linked to site changes
- Rapid conversion to equilibrium charge fractions at high temperature
- One-way conversion on ns timescale gives "missing fraction" and TF phase shifts
- Relaxation from subsequent charge cycling (semiconductors)
- Abstraction reactions in gases/liquids

$$Mu^0 + R\text{-}X \Rightarrow Mu\text{-}X + R\cdot$$

Metals

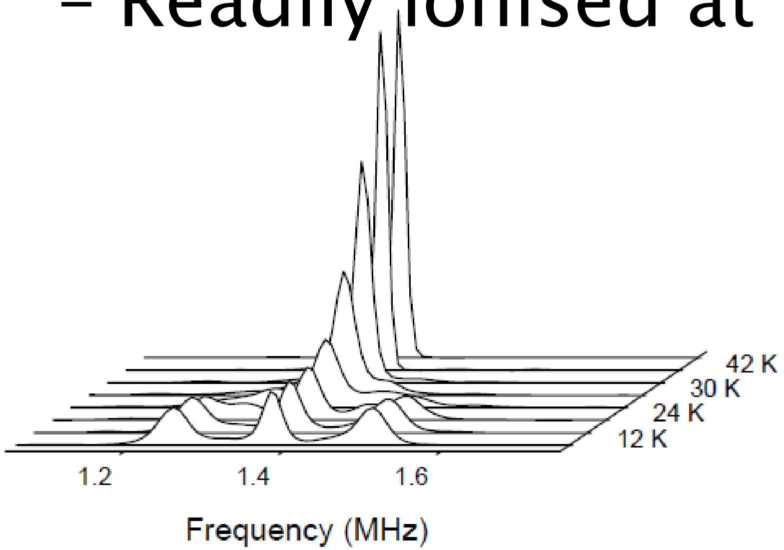
- Never see muonium in a metal – why?
- Muon 1s wavefunction overlaps conduction band
- Muonium electron(s) shared with all conduction electrons in sample
- Hyperfine coupling mostly averaged away, except in poor metals where:
  - Knight shift in TF
  - Korringa relaxation

Semiconductors

- Equilibrium charge state depends on the Fermi energy
- N-type sample: may favour Mu^-
- P-type sample: favours Mu^+
- Intrinsic or compensated: favours Mu^0
- High carrier density (high T or optically excited): rapid charge exchange

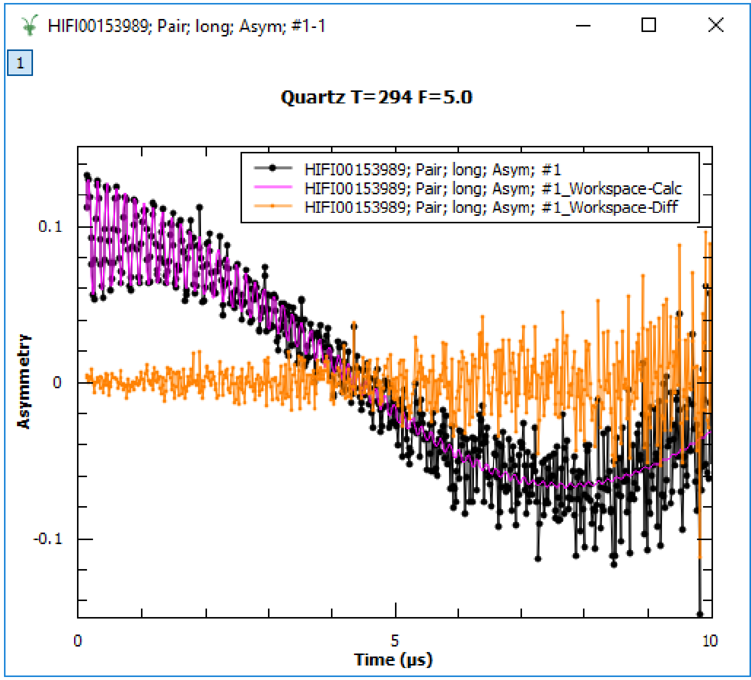
Semiconductors

- Mu^+ or Mu^- is a charged defect – dopant
- Sometimes weakly binds a conduction band electron or hole
  - Shallow Donor (or shallow acceptor)
  - Low hyperfine coupling <1MHz
  - Readily ionised at T>10-30K



Identifying charge states

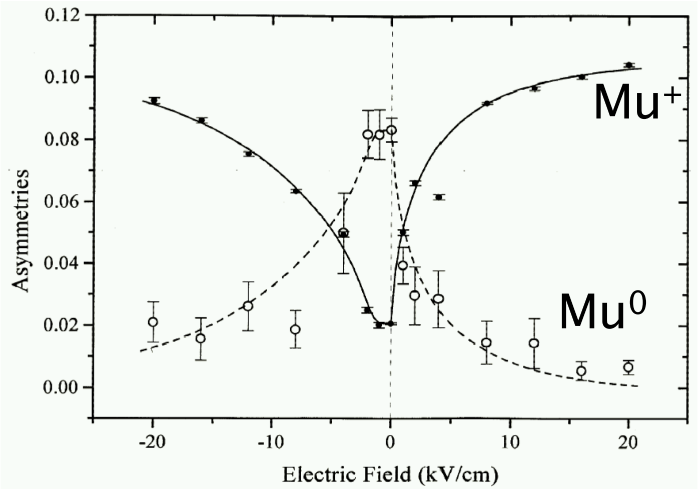
- TF20 – initial amplitude
- LF Repolarisation
- TF2 – Muonium triplet signal
  - Only where there are no nuclear moments
- TF Phase shifts with temperature
  - and amplitude varying with B



Influencing charge states

Initial charge state fractions normally fixed based on the material

- E-fields: sweep radiolytic electrons/holes away from the muon
- Light: provide more electrons/holes
- Doping: n-type or p-type sample
- Solutions: change the solvent
- Low energy muons: few radiolytic electrons formed



Conclusions

- Charge states can be useful
  - Fermi energy
  - Band structure
  - Carrier dynamics
  - Accessing different dynamic time scales
- Or can be inconvenient
  - Different response of Mu^0 and Mu^+ to magnetism of sample
  - Low radical fraction for ALCs
  - Low diamagnetic fraction for ionic motion