Neutrons and muons

NEUTRONS FOR SCIENCE

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Overview

- Properties of the muon
- Muon production and facilities
- Muon-matter interactions
- Applications in solid-state science
  - Magnetism
  - Superconductivity
  - Charge transport
  - Semiconductor defects
- Complementarity with neutrons

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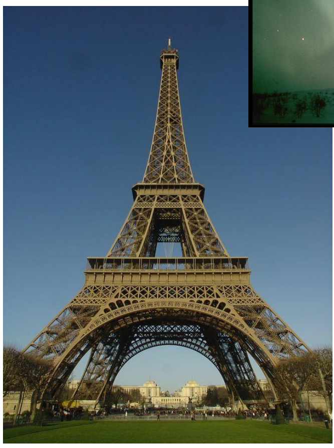
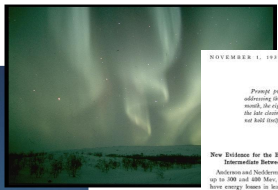
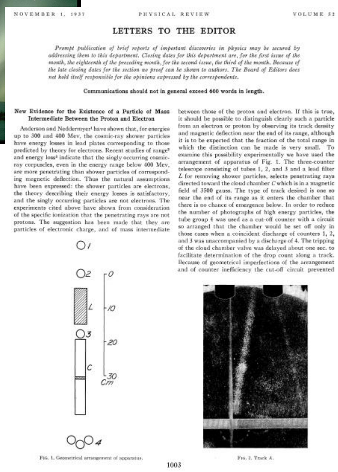
Muons: origins and properties

- Generated in upper atmosphere as H^+ in cosmic rays hits molecules
  - Muons survive to sea level (1 cm^{-2} min^{-1}) – identified in 1936

321m







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LETTERS TO THE EDITOR

Prompt publication of brief reports of important discoveries in physics may be secured by addressing them to this department. Closing dates for this department are, for the first issue of the month, the eighteenth of the preceding month, for the second issue, the third of the month. Because of the late closing dates for the section no proof can be shown to authors. The Board of Editors does not hold itself responsible for the opinions expressed by the correspondents.

Communications should not in general exceed 600 words in length.

New Evidence for the Existence of a Particle of Mass Intermediate Between the Proton and Electron

Anderson and Neddermeyer[1] have shown that, for energies up to 300 and 400 Mev, the cosmic-ray shower particles have energy losses in lead plates corresponding to those predicted by theory for electrons. Recent studies of range[2] and energy loss[3] indicate that the singly occurring cosmic-ray corpuscles, even in the energy range below 400 Mev, are more penetrating than shower particles of corresponding magnetic deflection. Thus the natural assumptions have been expressed: the shower particles are electrons, the theory describing their energy losses is satisfactory, and the singly occurring particles are not electrons. The experiments cited above have shown from consideration of the specific ionization that the penetrating rays are not protons. The suggestion has been made that they are particles of electronic charge, and of mass intermediate between those of the proton and electron. If this is true, it should be possible to distinguish clearly such a particle from an electron or proton by observing its track density and magnetic deflection near the end of its range, although it is to be expected that the fraction of the total range in which the distinction can be made is very small. To examine this possibility experimentally we have used the arrangement of apparatus of Fig. 1. The three-counter telescope consisting of tubes 1, 2, and 3 and a lead filter *L* for removing shower particles, selects penetrating rays directed toward the cloud chamber *C* which is in a magnetic field of 3500 gauss. The type of track desired is one so near the end of its range as it enters the chamber that there is no chance of emergence below. In order to reduce the number of photographs of high energy particles, the tube group 4 was used as a cut-off counter with a circuit so arranged that the chamber would be set off only in those cases when a coincident discharge of counters 1, 2, and 3 was unaccompanied by a discharge of 4. The tripping of the cloud chamber valve was delayed about one sec. to facilitate determination of the drop count along a track. Because of geometrical imperfections of the arrangement and of counter inefficiency the cut-off circuit prevented

FIG. 1. Geometrical arrangement of apparatus.

FIG. 2. Track *A*.

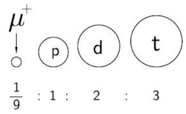
1003

Birth of the cloud chamber: C.T.R. Wilson

Muons: origin and properties

- Fundamental (indivisible) particle
  - charge +1 (μ^+) or -1 (μ^-)
  - mass ≈ 200 electron, 1/9 proton
  - Spin ½
  - Spontaneous decay: $\mu^+ \rightarrow e^+ + \upsilon_e + \bar{\upsilon}_\mu$ $\tau_{\mu+} \approx 2.2\ \mu s$

    $\mu^- \rightarrow e^- + \bar{\upsilon}_e + \upsilon_\mu$ $\tau_{\mu-}$ shorter due to nuclear capture
  - In practical terms it is the positive form that is important (lifetime) – but why?



	charge	spin	mass	moment	$\gamma\ /\ 2\pi$ (MHz T^{-1})	lifetime (μs)
e	$\pm$e	1/2	m_e = 0.51 MeV	657 μ_p	$28\text{x}10^3$	∞
μ	$\pm$e	1/2	207 m_e = 105.7 MeV	3.18 μ_p	135.5	2.19
p	$\pm$e	1/2	1836 m_e = 938 MeV	μ_p	42.6	∞

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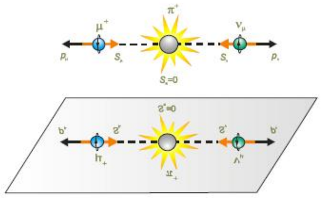
Muon production and polarisation

- Cosmic sources too feeble to be practical – use artificial sources
- Create from pions (π^+), in turn produced by firing high-energy protons (> 500 MeV) at target containing nuclei of intermediate mass (C,Be)

$$p + p \rightarrow \pi^+ + p + n$$

$$\pi^+ \rightarrow \mu^+ + \upsilon_\mu$$

  - Most of π^+ that lead to useable μ^+ are at rest in the *surface* of target
  - Conservation of spin (s) and momentum (p) for decay of π^+ at rest (s=0, p=0) leads to 100% polarisation of μ^+ spin opposite to momentum (parity violation means that mirror-image process doesn't occur and neutrino has spin antiparallel to its momentum)
    - Kinetic energy $\mu^+ \approx 4.1$ MeV
    - Half-life π^+ 26 ns



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  - So why is this useful?

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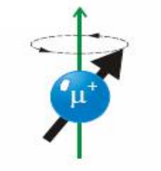
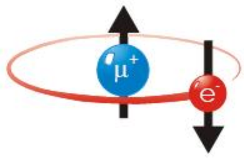
Muon implantation (*not* scattering!)

- μ^+ behaves like a light proton in terms of implantation in solids
  - 4.1 MeV kinetic energy rapidly lost by ionisation and e^- scattering → keV (ns)
  - Final stages of energy loss involve e^- loss and capture → 100s eV (ps)
    - Can end up in state with e^- captured (muonium - $\mu^+ e^-$)
    - If ends up positive, comes to rest at site favoured by charge – *e.g.* near O (*c.f.* O-H)
  - Thermalisation does not degrade spin polarisation appreciably





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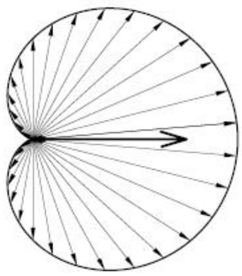
Muon decay

- The muon never emerges from solid – decays, $\tau_{\mu+} \approx 2.2\ \mu s$
  - Of the decay products, it is the positrons (e^+) that can be detected directly
  - Angular distribution of positrons reflects muon spin polarisation at point of decay (parity violation again, plus momentum distribution of decay products)

$$\mu^+ \rightarrow e^+ + \upsilon_e + \bar{\upsilon}_\mu$$



  - So…?

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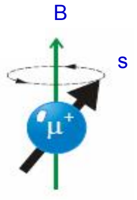
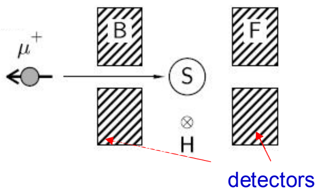
Muon as local probe of internal fields

- Implanted μ^+ (Larmor) precesses about any component of field (B) transverse to the direction of implantation
  - Angular frequency $\omega_\mu = \gamma_\mu B$ ($\gamma_\mu = ge/2\,m_\mu$) – between e^- (esr) and H^+ (nmr)
  - Typically falls in μs region (MHz)
  - μ^+ polarisation rotates between forwards and backwards direction (relative to direction of implantation) and distribution of detected positrons will reflect this





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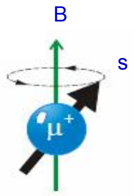
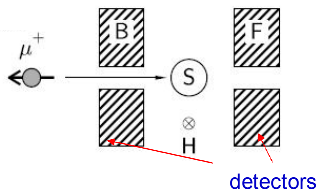
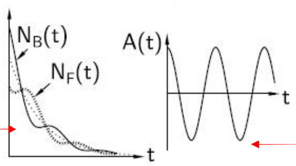




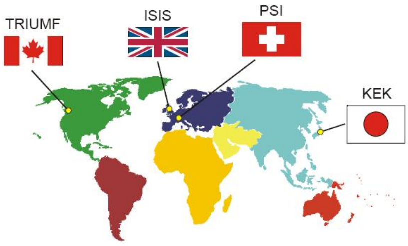
Positron signal in forward and backward detector – decaying with $\tau_{\mu+} \approx 2.2\ \mu s$

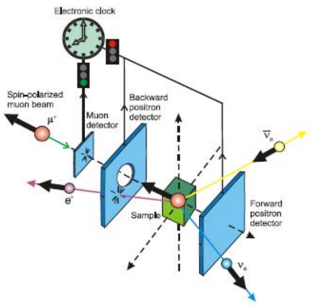


Asymmetry function A(t):
$A(t)=(N_B(t)-N_F(t))/(N_B(t)+N_F(t))$
Commonly normalize to A_{max} ~ 0.25:
$G(t)=A(t)/A_{max}$

The muon experiment: facilities

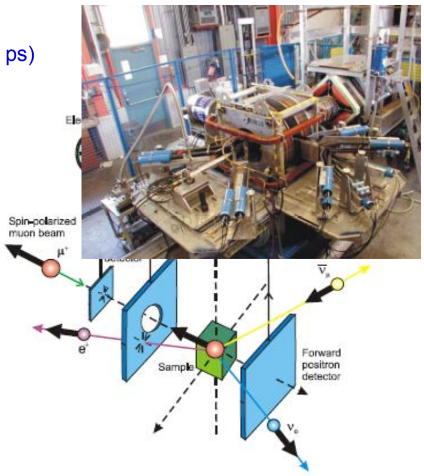
- Principal distinction: continuous or pulsed source
  - Continuous sources at the Paul Scherrer Institute (PSI) (Villigen, Switzerland) and the TRIUMF Meson Facility (Vancouver, Canada)
  - Pulsed sources at the KEK Meson Science Laboratory (Japan), and the ISIS Facility of the Rutherford Appleton Laboratory (UK)
- Most common instrument configuration: longitudinal
  - Detectors forward and backward with respect to initial muon polarisation
  - Any magnetic field applied along the same direction





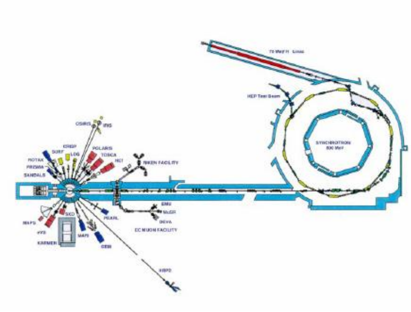
The muon experiment: facilities

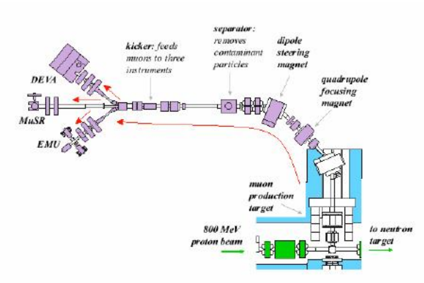
- Continuous sources
  - Each incoming muon detected – and clock started
  - Stop clock when corresponding positron detected (meanwhile reject other μ^+)
  - Repeat to accumulate statistics: A(t), G(t)
  - Advantages:
    - Can detect events at very short time (~ 100 ps)
  - Disadvantages
    - Relatively low intensities/weak signal
    - Often not extended to long times



The muon experiment: facilities

- Pulsed sources
  - Pulsed proton beam from synchrotron or linac directed on target
  - Pulses of muons produced with width set by proton pulse (~ 10ns) and π^+ lifetime
  - Repetition period must be much longer than muon lifetime (typically 20 ms)
  - Accumulate statistics over many pulses: A(t), G(t)





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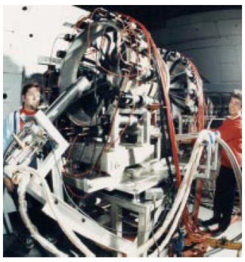
  - Accumulate statistics over many pulses: A(t), G(t)
  - Advantages:
    - Use all of muons in pulse – relatively intense
    - Background very low
  - Disadvantages
    - Cannot observe at shorter times than the pulse width



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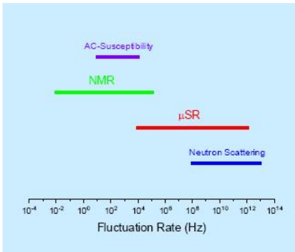
Science with implanted muons

- Muons implanted as μ^+ or muonium ($\mu^+ e^-$) after electron capture
- Muons highly sensitive to static and dynamic magnetic fields
  - Particular applications to systems with very small or dilute moments
  - Local probe – particularly good at sensing short-range effects
  - Works well in zero field – less perturbation of the system
  - Time-window $10^4 – 10^{12}$ Hz complements other techniques
  - Not element specific (nmr nuclei, neutron absorbers)
  - No spatial information – applicable to crystals, powders, films

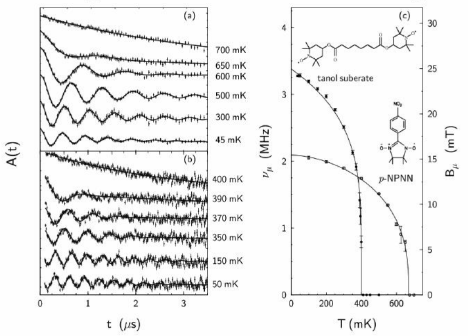
Average picture provided by susceptibility – can be misleading

Muons more likely to reveal nature of *local* magnetic environment and tell these two apart



Probing magnetism with muons

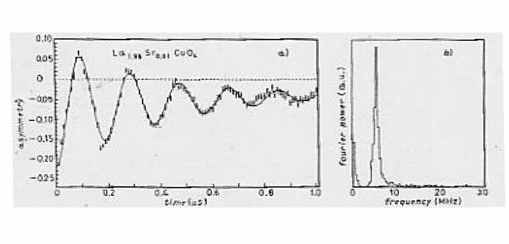
- Start with case of 'simple' ordered magnet
- Implanted muons 'feel' static internal field component and precess
  - This is μSR – muon spin *rotation*
  - $\omega_\mu = \gamma_\mu B$; $\gamma_\mu / 2\pi = 135$ MHz T^{-1}
  - With longitudinal geometry and no applied field, G(t) oscillates
  - Frequency gets smaller as magnet warmed to ordering temperature
  - Typically able to measure to 10^{-5} T; moment unknown unless muon site known

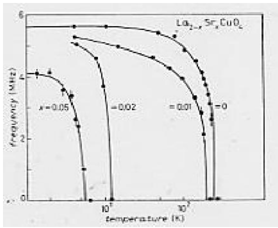


Blundell *et al, Europhys. Lett.* **31** (1995) 573

Probing magnetism with muons

- Muons particularly good at detecting *weak* moments (c.f. neutrons)
  - Detection of spin-freezing in high-T_c materials $La_{1-x}Sr_xCuO_4$ (s = ½)
    - Budnick *et al*, *Europhys. Lett.* **5** (1988) 651





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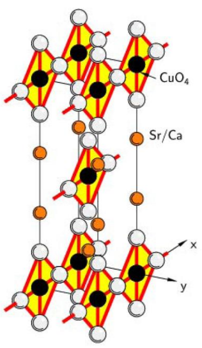
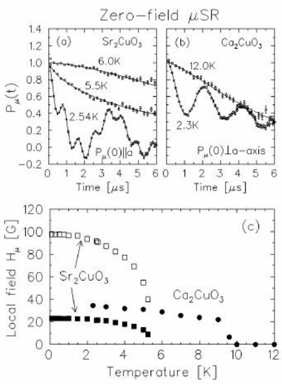
Probing magnetism with muons

- Exploration of effects of lower-dimensionality in cuprates
  - huge activity on cuprate chain and ladder materials e.g. Sr_2CuO_3 and Ca_2CuO_3
  - -Cu-O-Cu-O-Cu-O- chains well separated - J'/J small
  - Moment below T_N scales with J'/J – does it disappear as J'/J → 0?
    - Kojima *et al*, *PRL* **78** (1997) 1787
  - See what happens when chains pushed further apart





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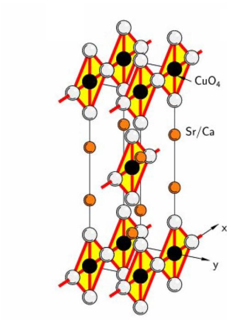
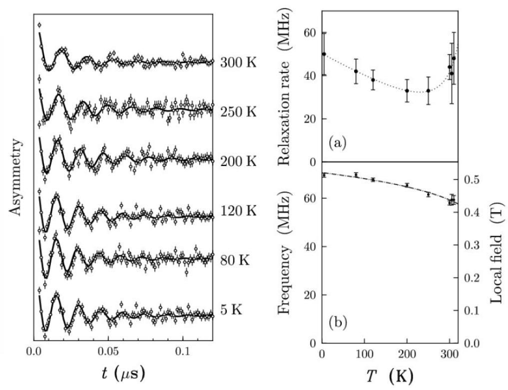
Probing magnetism with muons

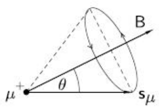
- Attempt to make $LaSrCoO_3$ by reduction of $LaSrCoO_4$ with CaH_2
  - Obtain target material with Sr_2CuO_3 structure – and chains are further apart.
  - However, internal field and ordering T *very* high (>300 K rather than 5 -10 K)
  - Closer analysis reveals $LaSrCoO_3H_{0.7}$ – H between chains – J' very strong
    - Hayward *et al, Science* **295** (200) 1882

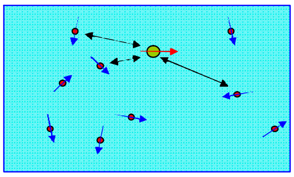




μSR with less than perfect order

- What happens when the field is not entirely uniform?
  - *e.g.* array of frozen, randomly oriented *nuclear* moments (in materials that have nuclear moments)
  - *e.g.* array of frozen, randomly oriented *electronic* moments – in spin glass
    - Uemura *et al, PRB* **31** (1985) 546

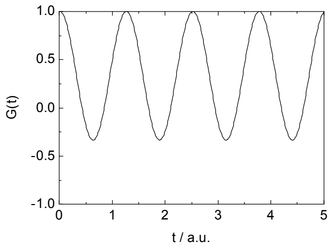


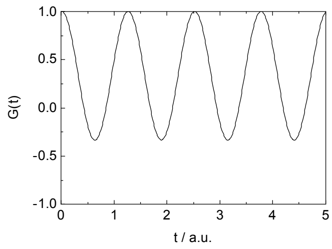


- For one component of the field: $G(t) = \cos^2\theta + \sin^2\theta\cos(\gamma_\mu Bt)$
- Average over completely random orientations: $G(t) = \frac{1}{3} + \frac{2}{3}\cos(\gamma_\mu Bt)$

Adding up all the components

- Add up the contributions from different field strengths: 1…



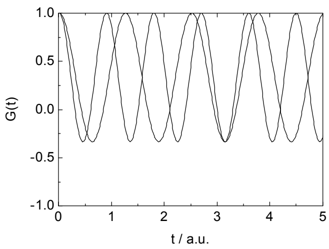


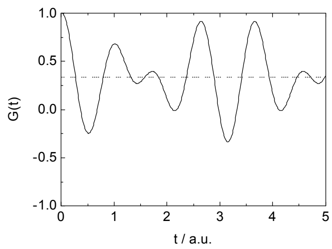
individual component

sum

Adding up all the components

- Add up the contributions from different field strengths : 1,2…



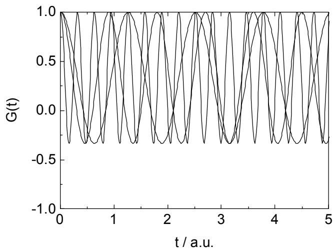


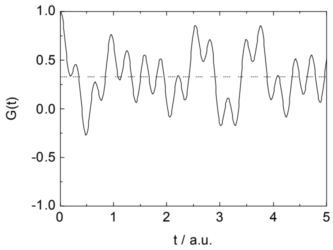
individual component

sum

Adding up all the components

- Add up the contributions from different field strengths: 1,2,3…

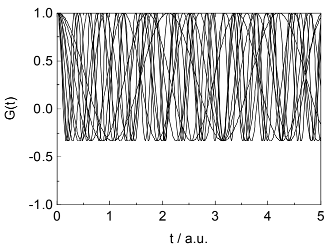


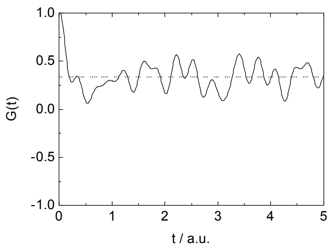


individual component sum

Adding up all the components

- Add up the contributions from different field strengths : 1,2,3…many



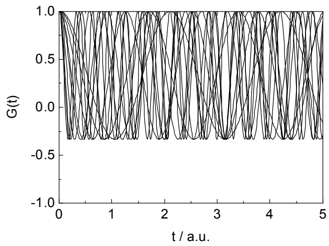


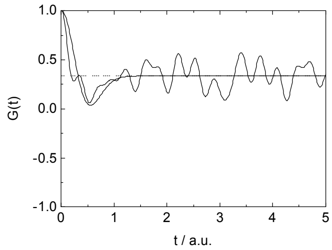
individual component sum

Adding up all the components

- Add up the contributions from different field strengths : 1,2,3…many
  - introduce a continuous Gaussian* distribution of width Δ/γ_μ :

$$G(t) = \frac{1}{3} + \frac{2}{3} e^{\frac{-\Delta^2 t^2}{2}} (1 - \Delta^2 t^2)$$





individual component | sum

* appropriate for *concentrated* collection of dipolar fields – for *dilute* systems *e.g.* some spin-glasses, this is Lorentzian: Walstedt and Walker, *PRB* **9** (1974) 4857; Crook and Cywinski, *JPCM* **9** (1997) 1149

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Adding up all the components

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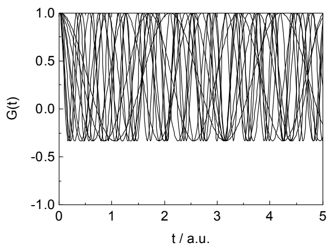

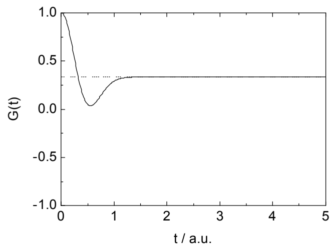

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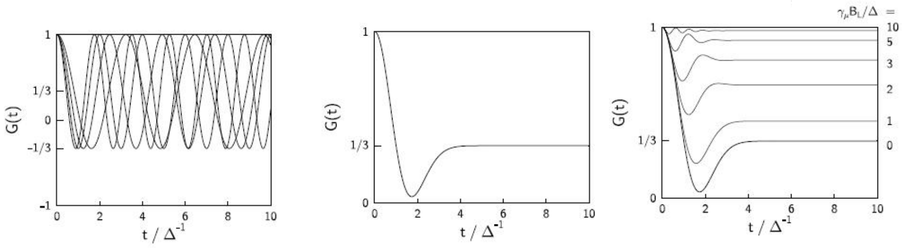
‘R’ is also for ‘Relaxation’

- Kubo-Toyabe relaxation function for frozen static moments in zero field and longitudinal geometry
  i.e. a form of muon spin *relaxation*

$$G(t) = \frac{1}{3} + \frac{2}{3} e^{\frac{-\Delta^2 t^2}{2}} (1 - \Delta^2 t^2)$$



  – At long times, G(t) recovers to 1/3 initial value – reflects 1/3 net component of random moments along longitudinal direction – no contribution to relaxation
  – An applied field increases the value of this field and hence the ‘1/3 tail’

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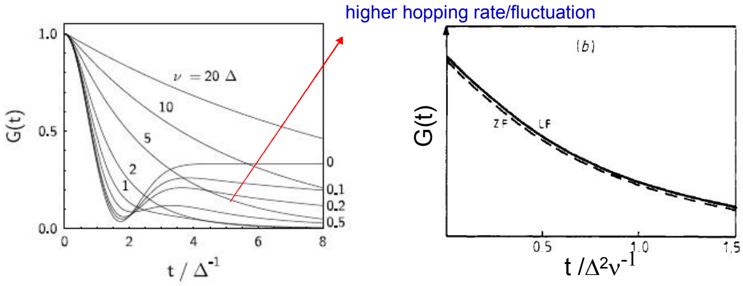
Fluctuating moments

- The muon spin often experiences fluctuations in the field – either because the field moves (*e.g.* in a paramagnet) or the muon hops
  - Hayano *et al*, *PRB* **20** (1979) 850
  – Assume rate of change of direction p(t)=exp(-νt)
  – Field orientation moves randomly at this rate within distribution $P(B_i)$
    - For *fast* relaxation rates: $G(t) = \exp(-\lambda_z t)$; $\lambda_z = 2\Delta^2/\nu$ (‘nmr’ - motional narrowing – ν)
    - For *slow* relaxation rates: G(t) = ⅓ exp(- ⅔ νt) – recover 1/3 tail
    - Full behaviour can either be simulated or approximated by analytic function (dynamical KT function) - Keren *PRB* **50** (1994) 10039
  – Applied field doesn’t make much difference to signal from paramagnet



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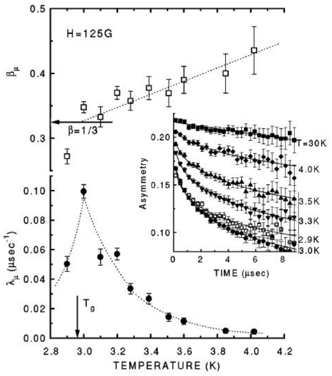
Probing the energy landscape

- Spin-glasses provide insights into glassy dynamics in general
  - Range of environments leads to distribution of relaxation times
  - No longer simple exponential decay for G(t) (implies one relaxation time)
  - Observe stretched exponential for many SGs (universal?): $G(t) \approx \exp(-\lambda_\mu t)^\beta$
  - e.g. 0.5 at% Mn in Ag (Ag has weak nuclear moment – passive matrix)
    - Campbell *et al, PRL* **72** (1994) 1291; Keren *et al PRL* **77** (1996) 1386

Stretched exponential exponent *vs* T

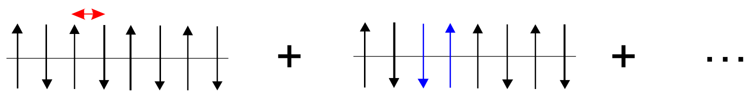
Relaxation rate λ vs T



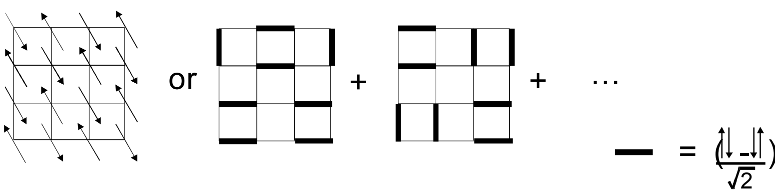
(Unnormalised) asymmetry A(t) vs T

Opening a very old can of worms

- Conventional (Néel) ground state may not be correct



- = RVB (resonating valence bond) or spin fluid ground state
- Similarly for layered magnets

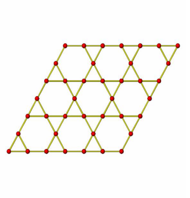


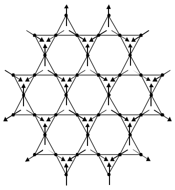
Pushing the boundaries of magnetism

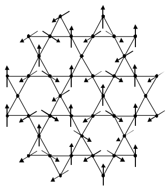
- Classical model works well most of the time – Néel order on cooling

- Quantum fluctuations more significant for
  - Small spin (S= ½)
  - Fewer neighbours - chains and planes
  - Frustrated interactions

- S= ½ kagome antiferromagnet brings all these together
  - Any good examples out there?



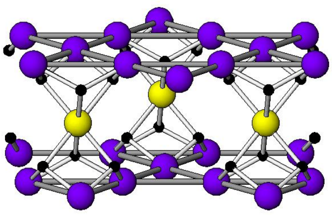


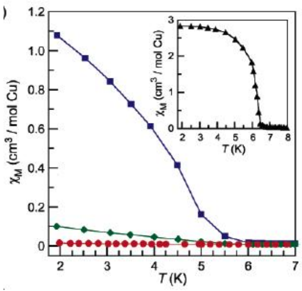


Herbertsmithite - a perfect S = ½ kagome afm?

- Parent compound: γ-$Cu_4(OH)_6Cl_2$ - dope with Zn: $Zn_xCu_{4-x}(OH)_6Cl_2$
  - Parent compound has pyrochlore structure
  - Zn selects sites between kagome layers (no JT distortion)
  - For Cu_3Zn compound, yields undistorted kagome layers separated by Zn
  - Zn severs weak FM component of Cu-Cu exchange (|θ|↑as %Zn ↑)
    - Intra-plane Cu-O-Cu 119º; inter-plane Cu-O-Cu 97º
      - Shores et al, JACS 127 (2005) 13462

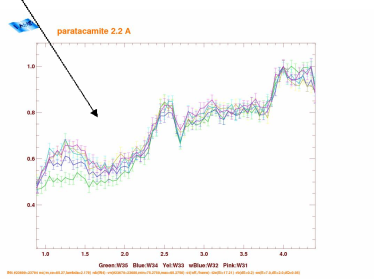
x = 0, 0.5, 0.66, 1.0





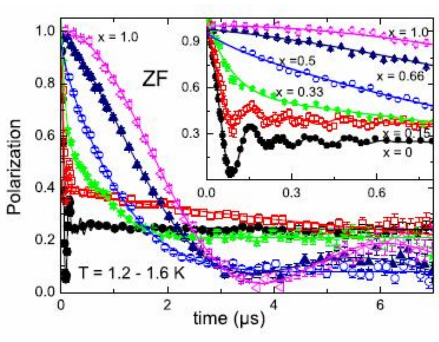
Neutron probe of correlations…

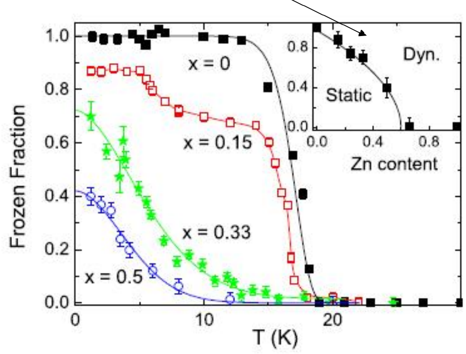
- No long-range order to 20 mK in x = 1 (pure kagome)
- Traces of excitation around 7 meV (but only for some of the spins)
  - Short-range correlation visible at 2 K but not 60 K
  - Scan along Q for data in energy range 7 – 8 meV; T = 2 to 60 K
  - Is this the energy required to break up spin singlets?



Spin freezing in paratacamite series - $Cu_{4-x}Zn_x$

- Track magnetic behaviour with x using μSR (Mendels *et al*, PRL **98** (2007) 077204)
  - For x ≤ 0.15 distinct oscillations plus paramagnetic term
  - For x = 0.33, x = 0.5 freezing transition broader and at lower T
  - Higher values of x only dynamic down to lowest T (50 mK – cf 300 K for θ)



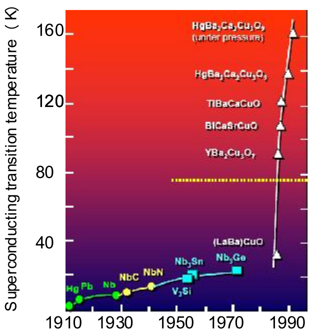


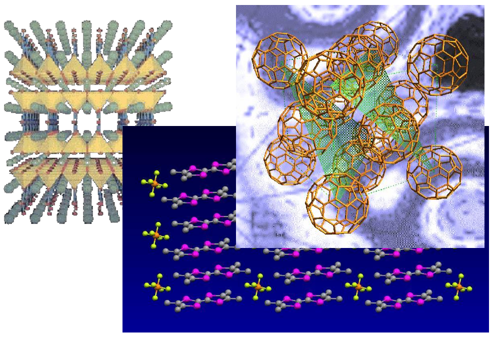
Superconductors

- Superconductors – why the fuss?
  - ‘superconductivity is perhaps the most remarkable physical property in the universe’ *David Pines*
  - It’s also one of the most useful – really and potentially

Superconductors

- Striking leaps in T_c in the past decades – but why?

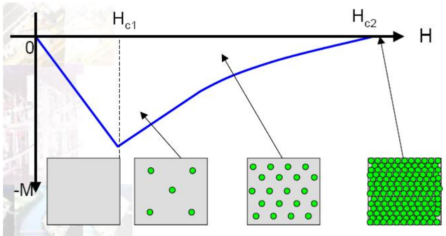




- Tremendous range of materials that are now known to superconduct
  - metals and alloys
  - oxides, especially cuprates
  - fullerenes
  - molecular solids
  - where next?

Superconductors

- What to look for? What provides clues about the underlying physics?
- Two characteristic length scales:
  - Coherence length (ξ) – scale for variation of sc wavefunction
  - Penetration depth (λ) – controls ability of sc to screen magnetic fields



- If λ is much greater than ξ ($\lambda > \xi/\sqrt{2}$) flux can penetrate entire sample
  - Does so as quantized flux lines (h/2e) called vortices which may form lattice
  - Behaviour of flux lattice a good test for theories of superconductivity
  - How to study?

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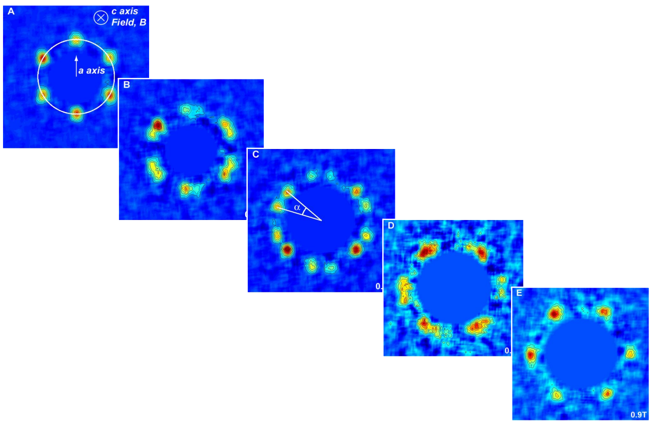
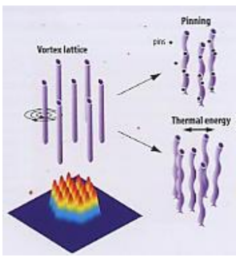
SANS

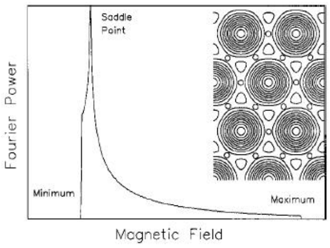
- Flux lines can scatter neutrons just as moments do
- Typical spacing between vortices puts scattering in SANS territory
  - E.g. MgB_2 (note – 98 mg Xtal – 0.75 mm² x 30 mm, 95% ^{11}B enriched
    - Cubitt and Dewhurst, Phys. Rev. Lett. 91 (2003) 47002

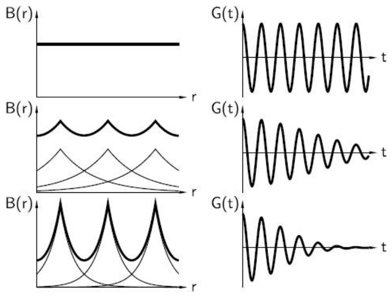




Muons and superconductors

- Muons can probe internal field distribution
  - apply transverse B to sample
    - In normal (non-sc) state, uniform field and simple muon precession signal
    - If vortices form, field felt by muon depends on implantation site relative to vortex
    - For larger λ, internal field variation less so relaxation rate (σ) less: $\sigma \approx 1/\lambda^2$
    - Generally difficult to get bulk measure of λ (don't need good x'tals as with SANS)





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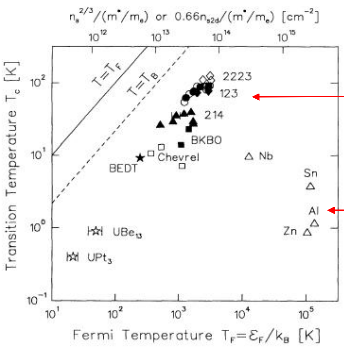
Muons and superconductors

- λ for clean sample at T= 0 can provide estimate for Fermi temperature (T_F)
- Plot T_c against T_F to provide clear distinction between ordinary and 'exotic' sc
- Common physics for exotic superconductors? But what?
  - Uemura *et al, PRL* **66** (1991) 2665



$$\frac{1}{100} < \frac{T_c}{T_F} < \frac{1}{10}$$

$$\frac{T_c}{T_F} \le \frac{1}{3}$$

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Probe of diffusion in solids

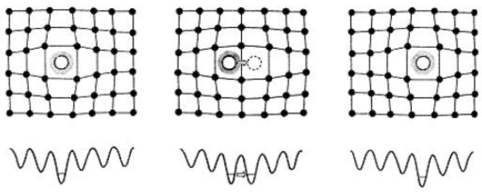
- Transport in solids – ions, atoms, electrons – in key technologies
  - Batteries, fuel cells, sensors, catalysts, conducting polymers
- Muons can probe such motion in several ways
  - relaxation of mobile μ^+ to study motion of light particles e.g. mimic of H^+
  - relaxation of static μ^+ to study motion of other species

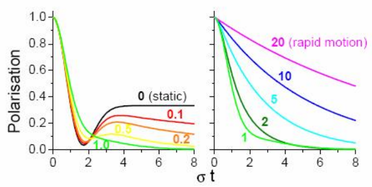
Illustrative figs of technologies etc

NEUTRONS FOR SCIENCE

Relaxation of diffusing muons

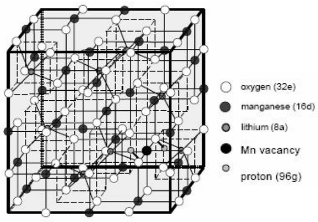
- μ^+ implanted in inorganic solid causes local lattice distortion
  - Muon spin senses local nuclear moments – KT relaxation
  - Hopping leads to relaxation of KT function (relaxation of ‘1/3 tail’)
  - Thermal assistance of motion includes phonons: overall $\nu = \nu_0 \exp(-E_a/kT)$
  - Quantum diffusion (tunnelling) at very low temperature
    - Storchak and Prokof’ev, *Rev. Mod. Phys*. **70** (1998) 929

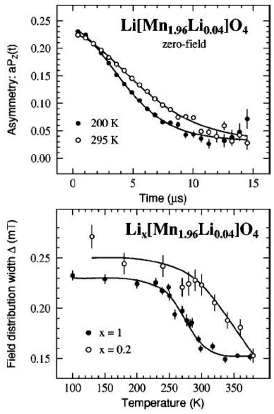


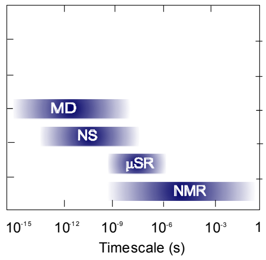


Relaxation by diffusing ions

- $Li_yx[Mn_{2-y}Li_y]O_4$ could be a key component (cathode) for Li batteries
  - Function depends on Li^+ flow in the spinel structure – optimise wrt x,y
  - μ^+ implants near O; observe dynamic KT form
  - Width of field (Δ) decreases above 230 K with x=1, y=0.04 – Li^+ motion
  - Li^+ motion only becomes significant above 300 K for x=0.2, y=0.04
    - Kaiser *et al, PRB* **62** (2000) R9236







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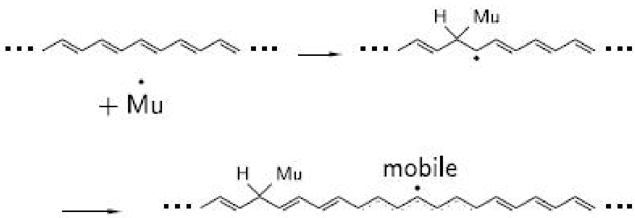
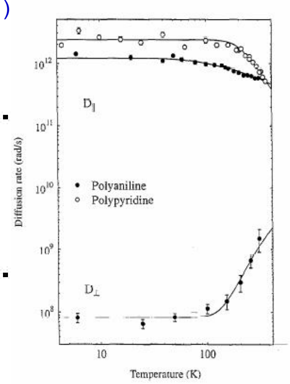
Electron motion in polymers

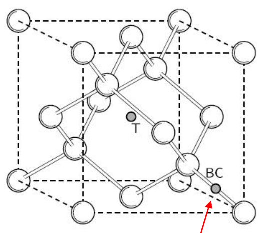
- μ^+ implanted in conducting polymer generally forms muonium (μ^+e^-) which reacts with the polymer to produce mobile spin (soliton)
  - Soliton moves up and down chain but cannot pass defect
  - Muon polarisation relaxes with each visit (μ^+- e^- hyperfine coupling)
  - Hence probe mechanism of charge transport
    - Nagamine et al, PRL 53 (1984) 1763; Pratt *et al PRL* **79** (1997) 2855; Pratt *et al, Syn. Met.* **101** (1999) 323; Blundell *et al, Syn Met.* **119** (2001)



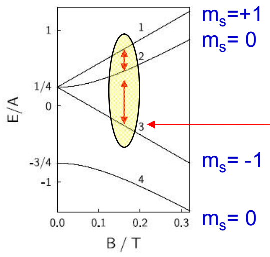


Semiconductor defects

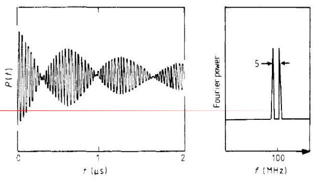
- Defects dominate much of the useful physics of semiconductors
  - H is a particularly important defect; mimic with muonium (H, H^+, H^-: μ^+, μ^+e^- μ^+2e^-)
  - Muonium studies most insightful for *individual* H defects – *very* sensitive
    - In low magnetic field see precession – transitions within the triplet
    - In higher field, measurement of precession signal yields A – sensitive to site
    - Patterson, *Rev. Mod. Phys.* **60** (1998) 69



Si: most tightly bound site for μ^+e^-



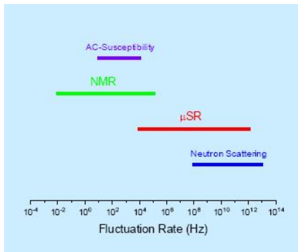
Breit-Rabi diagram for μ^+e^-
(isotropic A; A_{vac}=4.463..GHz)

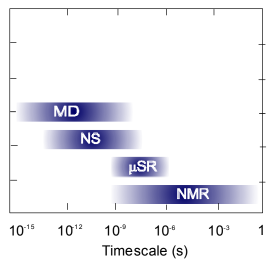


Intermediate field precession and Fourier transform – extract A

Summary

- What muons can do
  - Very sensitive probe of internal magnetic fields
    - Ordering temperature
    - Type of order – 'regular' or glassy
    - Fluctuations in paramagnets and glassy systems
    - Complements other techniques in dynamics
      - time range $10^4 – 10^{12}$ Hz
  - Superconductors
    - Characterisation of flux lattices – measure of penetration depth
  - Diffusion in solids
    - Mimic of light particles and diffusion mechanisms
    - Probe of diffusion of ions in solids, electrons in conducting polymers
  - Defects in semiconductors
    - Probe of nature of defect sites (H) in semiconductors





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Further reading

- Books and reviews
  - J. Chappert in ;Muons and Pions in materials research', eds J.Chappert and R.I. Grynszpan…
  - A. Schenck, 'Muon spin rotation spectroscopy' (1985) (Bristol, Hilger)
  - S.F.J. Cox, 'Implanted muon studies in condensed matter science', J.Phys.C:Solid State Phys. 20 (1987) 3187
  - S.J. Blundell, 'Spin polarised muons in condensed matter physics', Contemp. Phys (arXiv:cond-mat/0207699v)
  - P. Dalmas de Roetier and A. Yaouanc, 'Muon spin rotation and relaxation in magnetic materials', J.Phys.: Cond. Mat. 9 (1997) 9113
  - Patterson, Semiconductor defects, *Rev. Mod. Phys.* **60** (1998) 69
  - Storchak and Prokof'ev, Quantum diffusion, *Rev. Mod. Phys*. **70** (1998) 929

- Web resources
  - ISIS web site – esp. http://www.isis.rl.ac.uk/muons/trainingcourse/index.htm
  - TRIUMF web site: http://www.triumf.ca
  - PSI web site: http://www.psi.ch

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