Stimulation Methods 1:

Pulsed Stimulations for μSR Experiments

Stephen Cottrell
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Outline of talk

- **Introduction**
  Pulsed stimulations;
  Stimulation experiments and muon sources;
  What do stimulation experiments bring to μSR?

- **Overview of stimulation experiments**
  Example stimulation experiments;

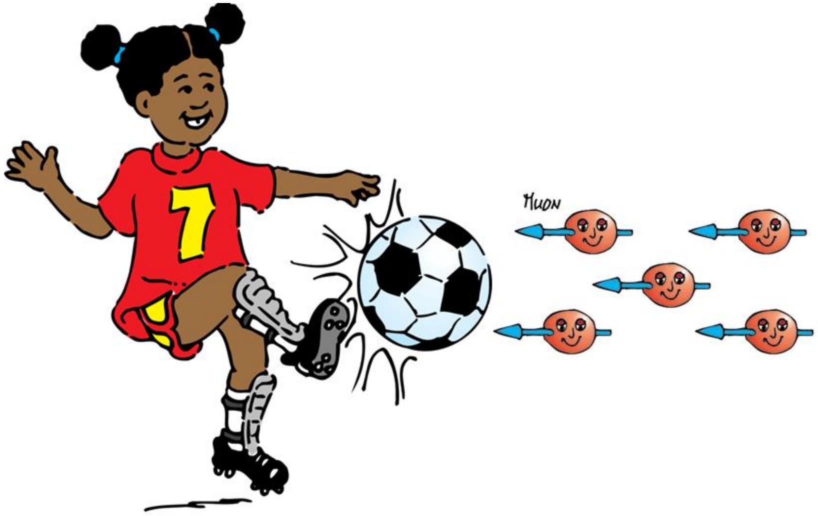
- **Radio-Frequency excitation**
  RF experiments;
  Potential applications of pulsed RF techniques;
  Final state spectroscopy;

- **Combining stimuli**
  RF+E-field experiments;

Introduction

Stimulation?

Apply an external stimulation while probing the sample with muons …



Then hopefully we learn something new about the ~~football~~ sample …

Possible Stimulations …

- Temperature
- Magnetic Field
- Pressure
- Electric Field
- Illumination
- Humidity
- Sound
- Flames
- …

Many possible!

Some are naturally **continuous for experiment**

Others may be applied only for a brief period as muons are implanted – i.e. **pulsed** (like kicking the ball!)

Pulsed Stimulations...

Many possible, but these are typical …

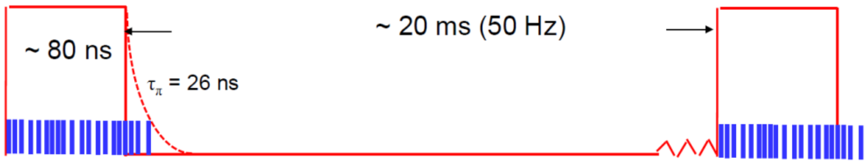
- Radio-frequency (RF) resonance
- Currents
- Magnetic fields
- Electric fields
- Acoustic resonance
- Illumination (flash lamp and laser)
- …

Why use pulsed stimuli?

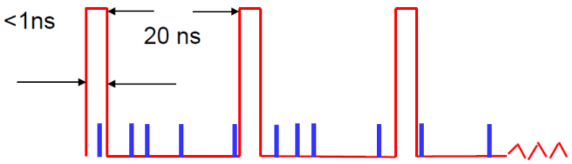
- Direct effect of time-varying environment
  - e.g. μSR signal following RF or laser pulse
- Observe slow formation of final muon states
  - e.g. delayed states that are a product of a reaction
- Measure recovery time of sample after a pulse
  - e.g. charge carrier recombination
- Avoid problems with steady state conditions
  - e.g. charge accumulation due to electric field

How to use pulsed stimuli?

- Muon sources come in two ‘flavours’ …
  - ‘pulsed beam’ (ISIS), all muons in short bunches



  - ‘continuous beam’ (PSI), muons arrive randomly



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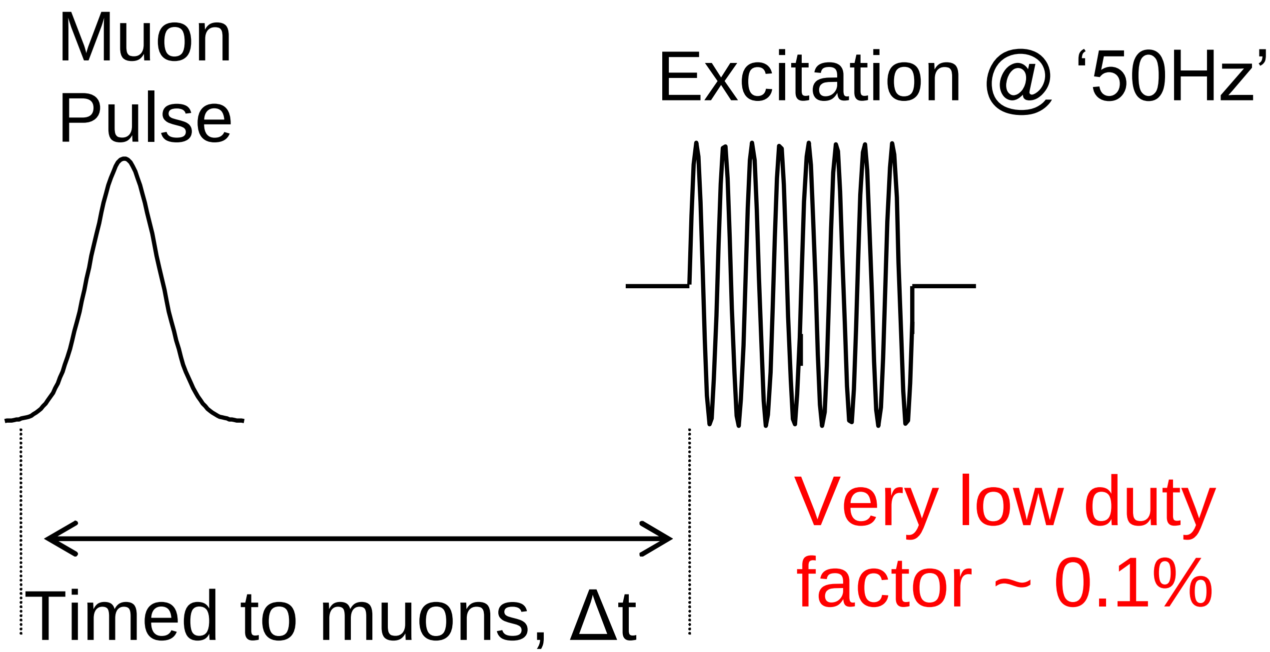
ISIS

At a pulsed source …

Low duty factor …

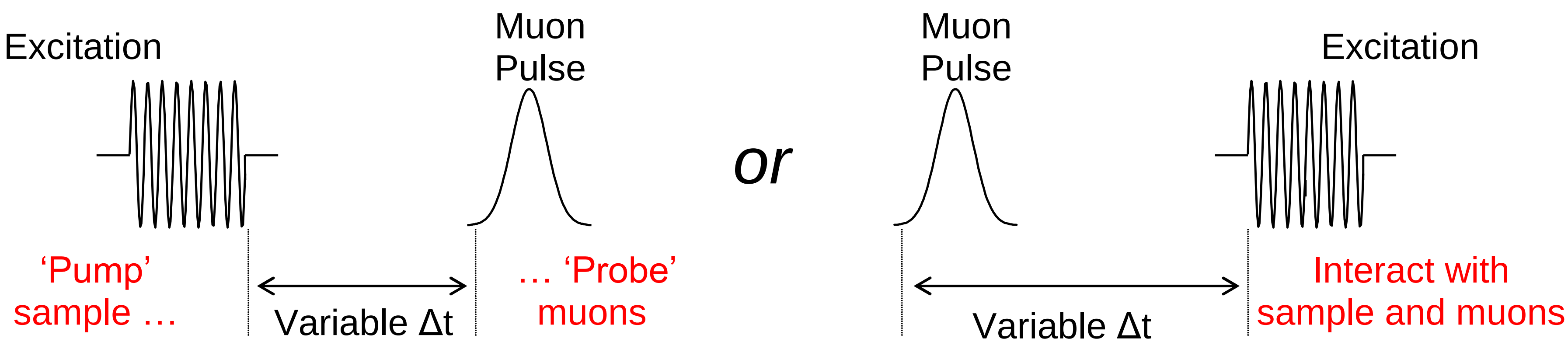


Very low duty factor ~ 0.1%

ISIS / J-PARC

Time relative to muons …

Before … *Coincident or after …*

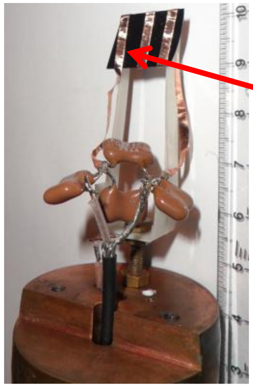


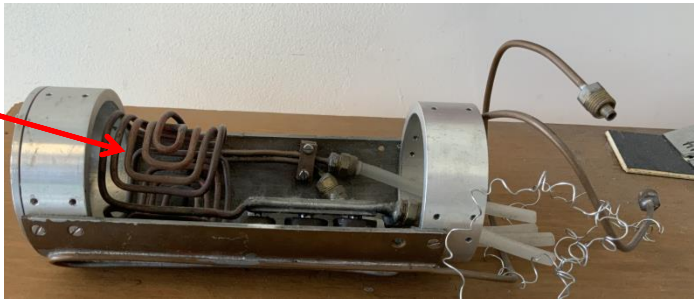
Change Δt,
study relaxation of sample

Change Δt,
study muon state kinetics

At a continuous source?

- Timing more complex – muons arrive randomly!
- The advantage of the low duty factor is lost:
  - sample/equipment heating can be a problem
  - engineering is more difficult
  - compare RF coils at ISIS and TRIUMF!





PSI / TRIUMF

Overview of Pulsed Stimulation Experiments

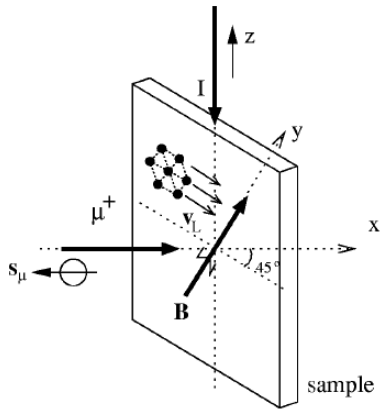
Pulsed Stimulations...

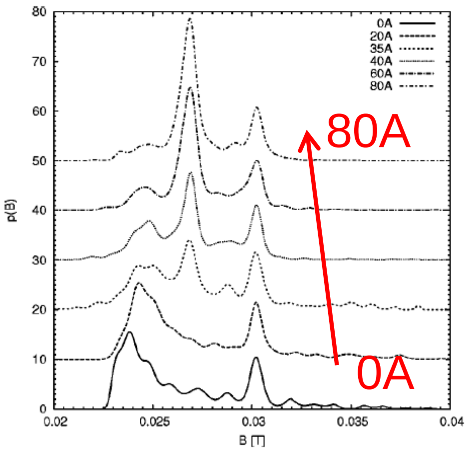
Some examples for …

- Currents
- Magnetic fields
- Electric fields
- Illumination

Pulsed Currents

- Current flow in a type II superconductor can lead to flux line motion
- Current measurements allow the study of the moving vortex lattice





PbIn, 30 mT, TF

- Motional narrowing of the μSR line shape observed
- Comparison to simulations to understand motion

- Pulsed (ISIS), AC (PSI) both contribute to study (higher B-fields at PSI allow smaller currents)

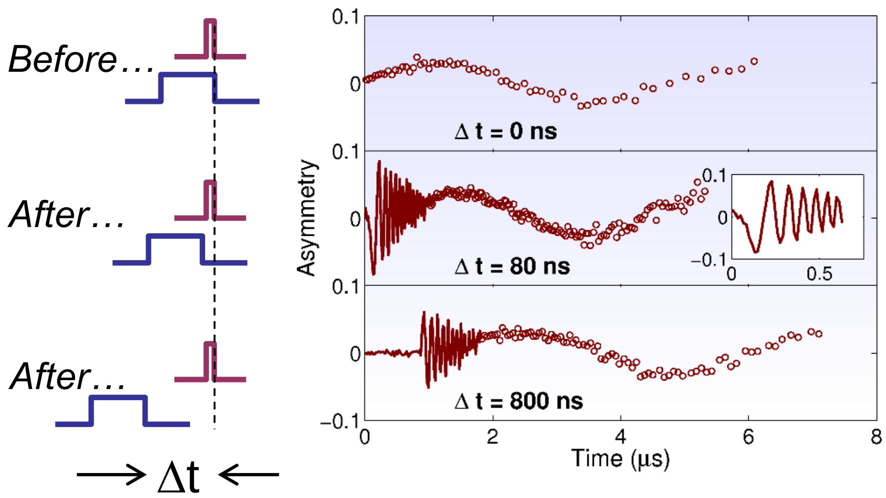
D. Charalambous, et al.,
Phys. Rev. B 66 (2002) 054506
Phys. Rev. B 73 (2006) 104514

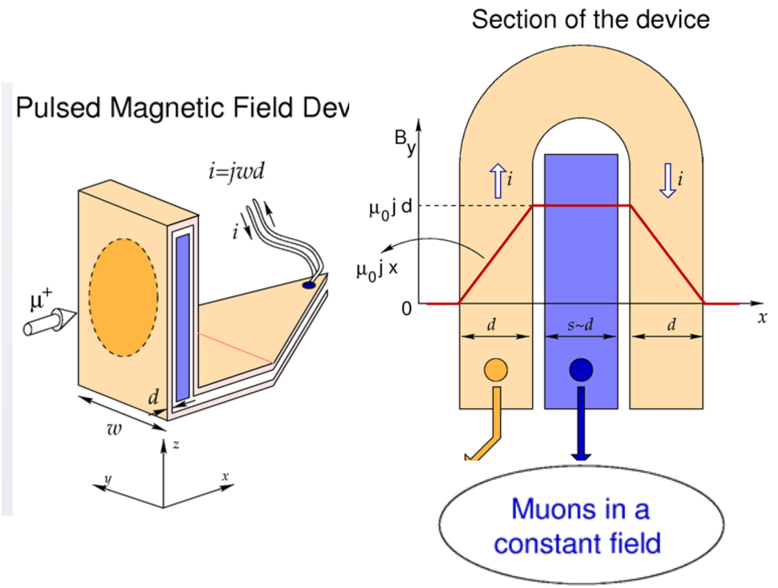
Science & Technology Facilities Council ISIS

Pulsed Transverse Fields

- Removes restriction of finite muon pulse width
- Allows study of slowly formed final muon states

Pulsed transverse field (15G) is applied …
(measuring Mu formed in Quartz)



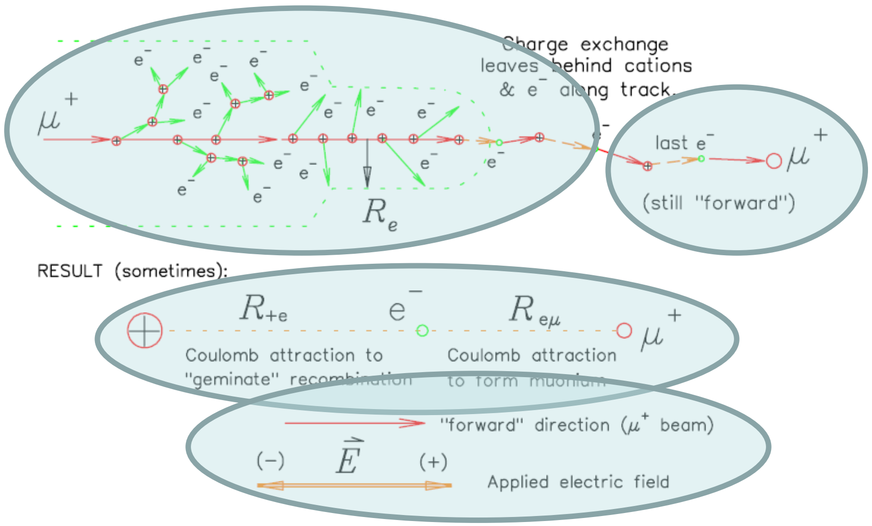


T. Shiroka et al., Phys. Rev. Lett. 83 (1999) 4405

Electric Fields

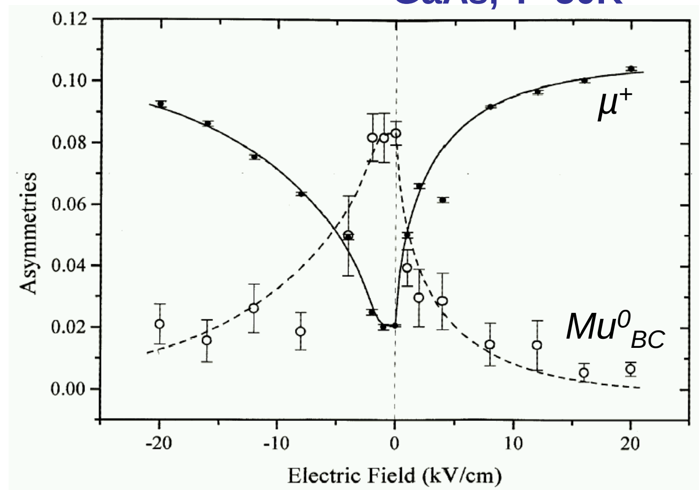
- Understanding electron transport in e.g. Rare Gas Solids
- Understanding Mu formation and Mu-H analogy in semiconductors

High energy muons create a track …



E-Fields transport excess charge …
μ+/Mu fractions modified

GaAs, T=50K



D.G. Eshchenko et al., Physics Lett. A 264 (1999) 226

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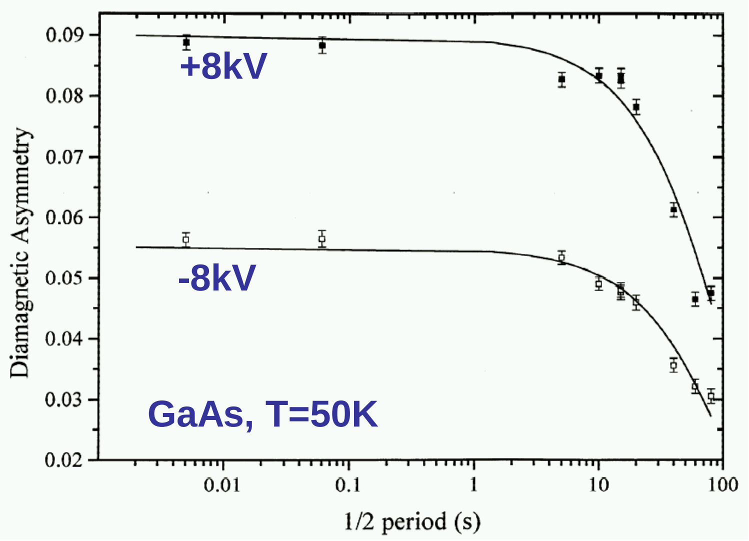
Switched or Static E-Fields?

- Static E-Fields easily to implement and used in first experiments
- However, results erratic – likely connected with build up of charge at the sample electrode interface (electrode gap and charge mobility)
- Solution was periodic reversal of E-field polarity

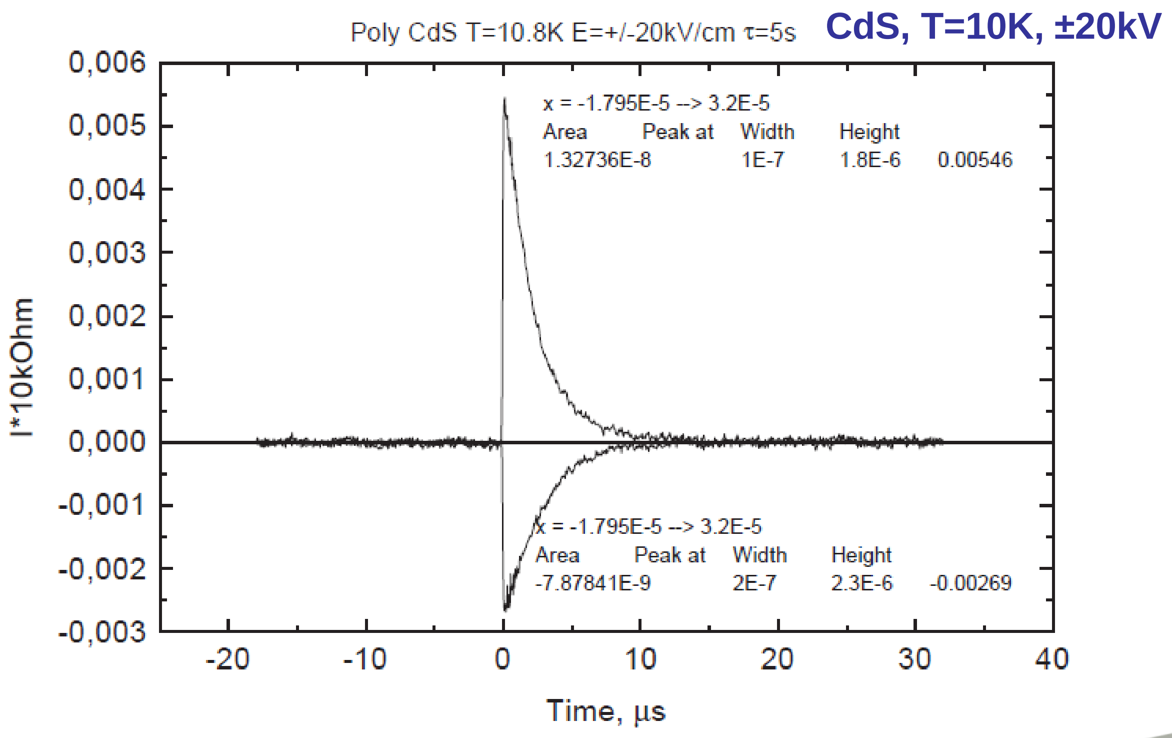
- Dependence of A_D on period of switching



D.G. Eshchenko et al., Physics Lett. A 264 (1999) 226

Track-induced Current

- Muon track-induced current – Bulk effect of microscopic process!



D.G. Eshchenko et al., Physica B 404 (2009) 880

Illumination

- Flash lamps and …

- See Koji's talk on Thursday!

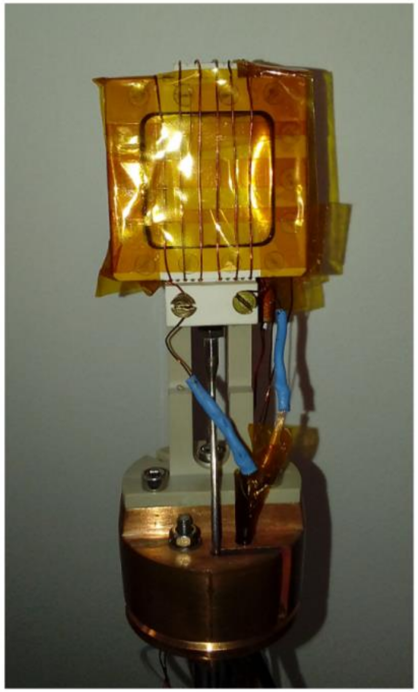
Radio-Frequency Stimulation

Resonance Experiment

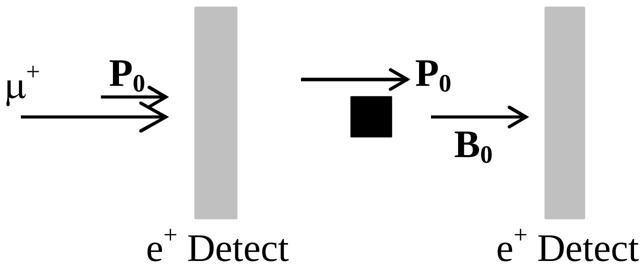
Apply an **RF Field, B_1**, perpendicular to a **static field B_0**;

Adjust the **RF frequency, ω_0**, to equal γB_0 (Larmor equation, γ = 13.5534 kHz/G for μ^+)

Just the same as doing an NMR experiment

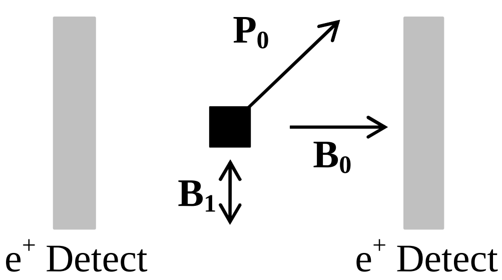


RF Resonance

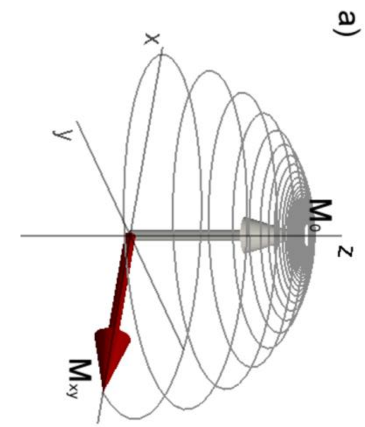


Implant muons: polarisation, P_0, parallel to static field B_0

Turn on RF Field, B_1 ...



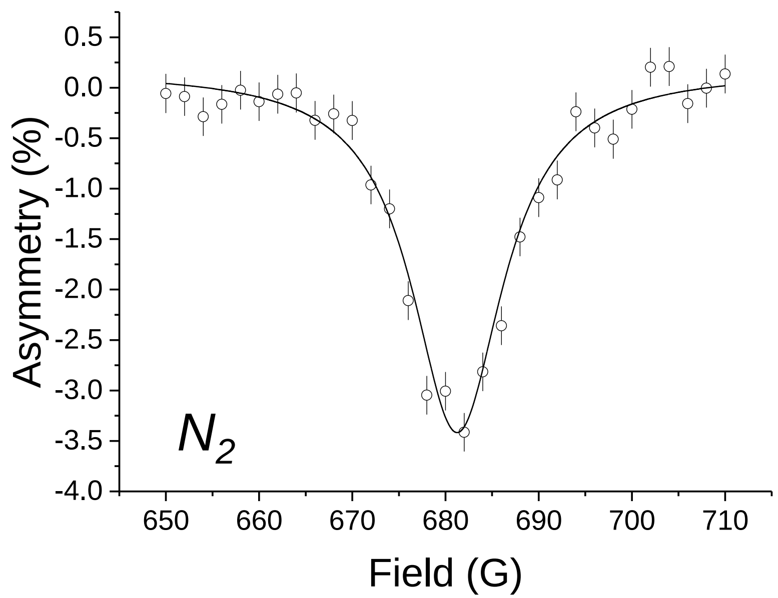
Muons couple to B_0 and B_1, complex motion



Resonance Experiment

Condition for resonance given by Larmor equation $\omega_0 = \gamma B_0$ …

Scan frequency or field to determine … but generally easier to scan field



Field scan (ω_0 = 9.2MHz)

Form of curve:

$$A_{\mathrm{RF}}(B) = A_{\mathrm{RF}}(B_0)\frac{\Delta B^2}{(B - B_0)^2 + \Delta B^2}$$

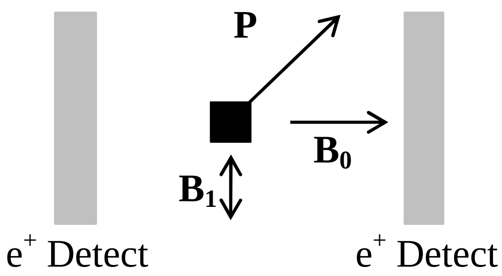
ΔB depends on:

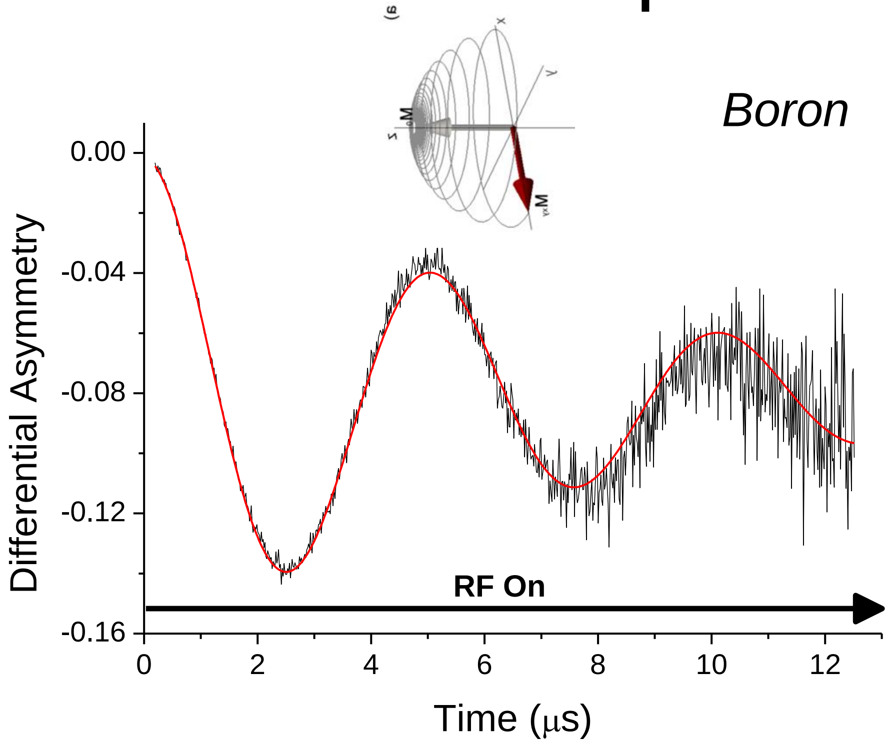
- RF power
- Depolarization
- Chemical reaction

CW NMR!

‘Longitudinal’ μSR signal – precession about B_1

At resonance …

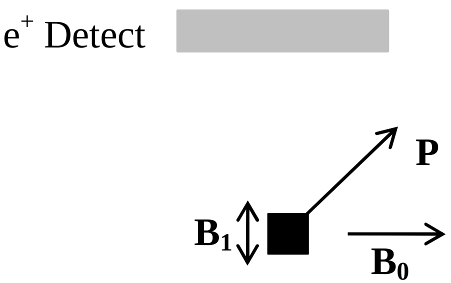




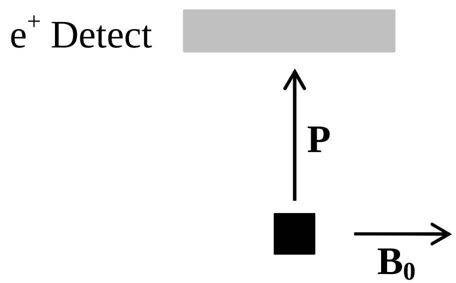
Precession about RF field, B_1, determined as ~15G from fit

‘Transverse’ μSR signal – precession about B_0

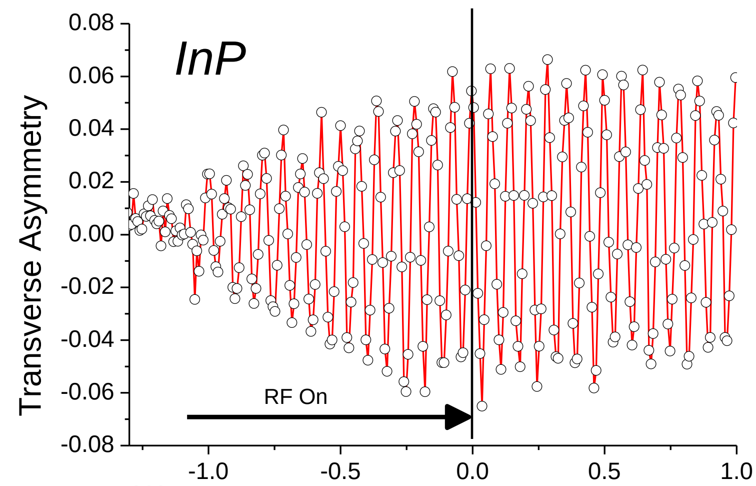
At resonance …

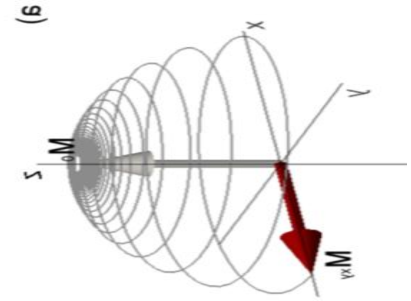


Amplitude of ‘transverse’ μSR signal grows



Turn off RF field after 90° rotation of P_0





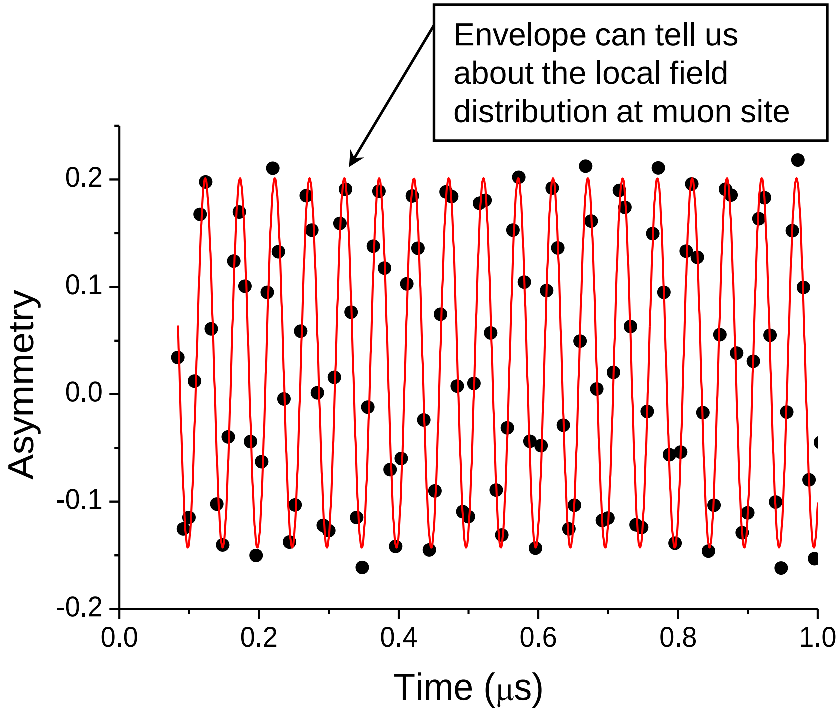
Muon precession at ~20MHz,

free precession signal following a 1.3μs RF pulse

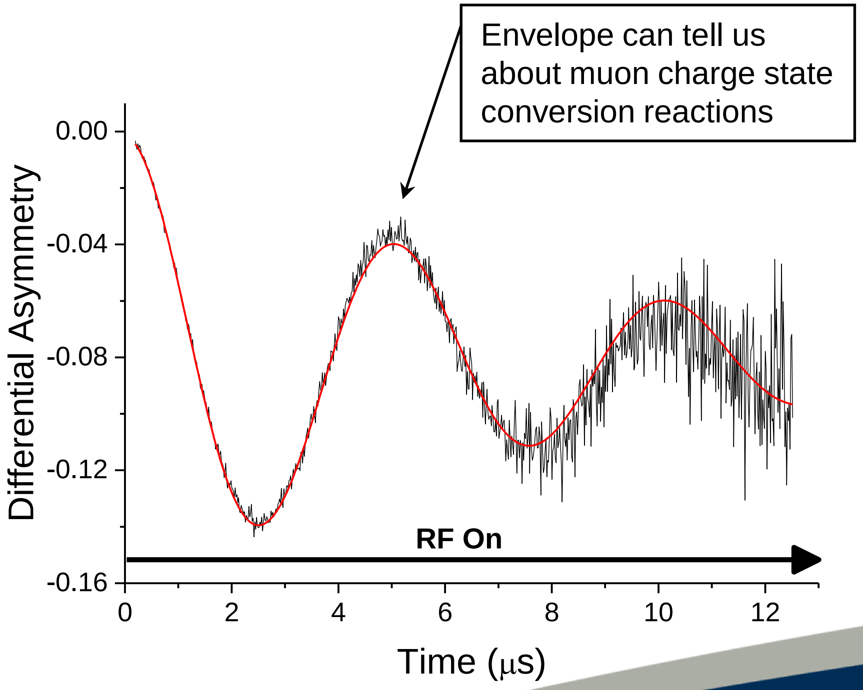
In NMR terms, this is a 90° pulse!

RF μSR – What are we Learning?

'transverse' μSR signal – free precession

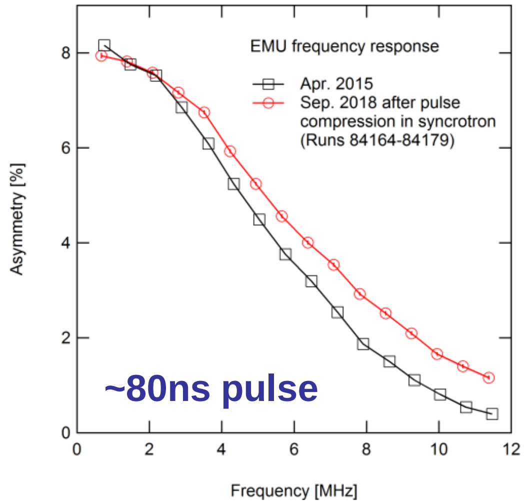


'longitudinal' μSR signal



Muon Precession at 20MHz!?

For *Pulsed Muon Sources*, finite muon pulse width limits maximum useable frequency …



~80ns ISIS pulse corresponds to a ~6MHz frequency bandpass

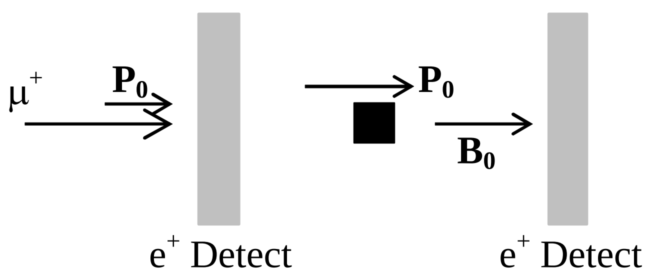
So how come we're seeing muon precession signals at ~20MHz?

90˚: ‘Beating the Pulse Width’

For **RF experiments**:

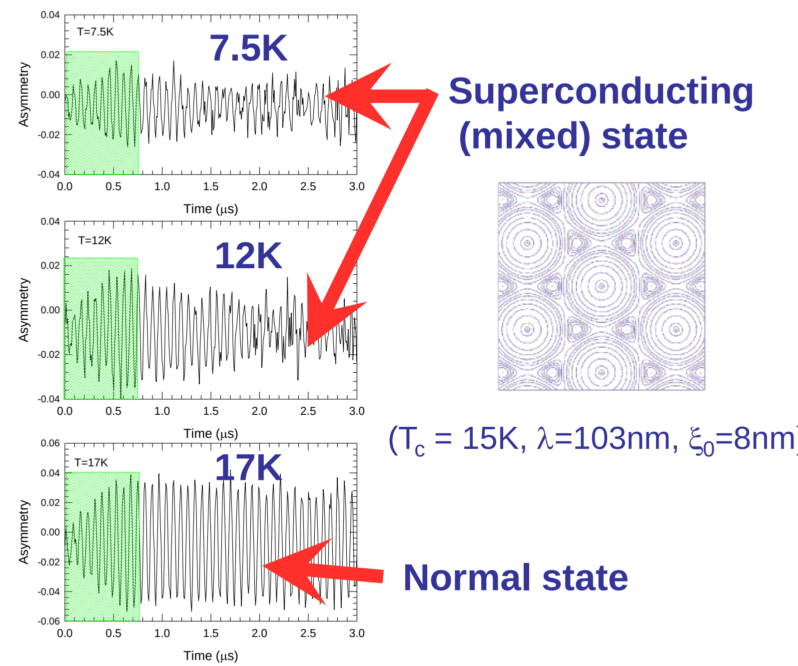
- muons implant in a large longitudinal field
- time structure of muon pulse removed

Frequency limit extended!



Example taken from study of YNi_2B_2C …

(A.D. Hillier et al., Physica B 326 (2003) 275)



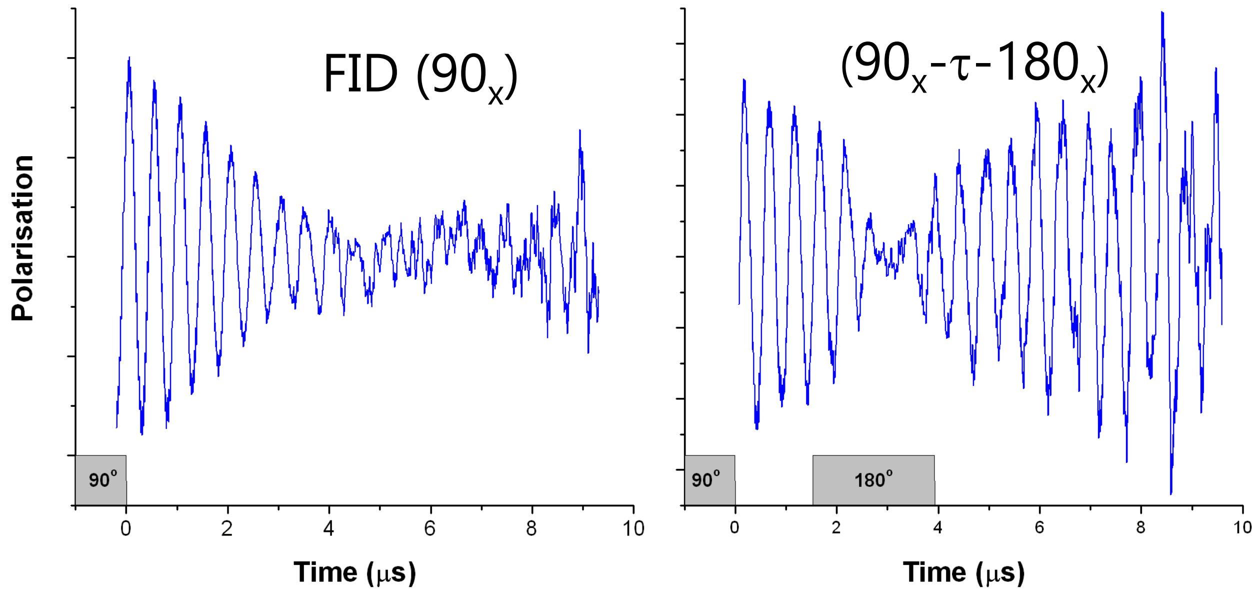
B_0=1034G (13.6MHz)

Applications

RF μSR – Benefits / Limitations?

- Many RF techniques from NMR/EPR might be applied to benefit for new science in μSR
- Uniquely, polarisation can be directly measured during RF pulses (compare to NMR)
- However, lifetime of muon can make implementing methods challenging – require short, high power RF pulses!

NMR-style pulse sequences

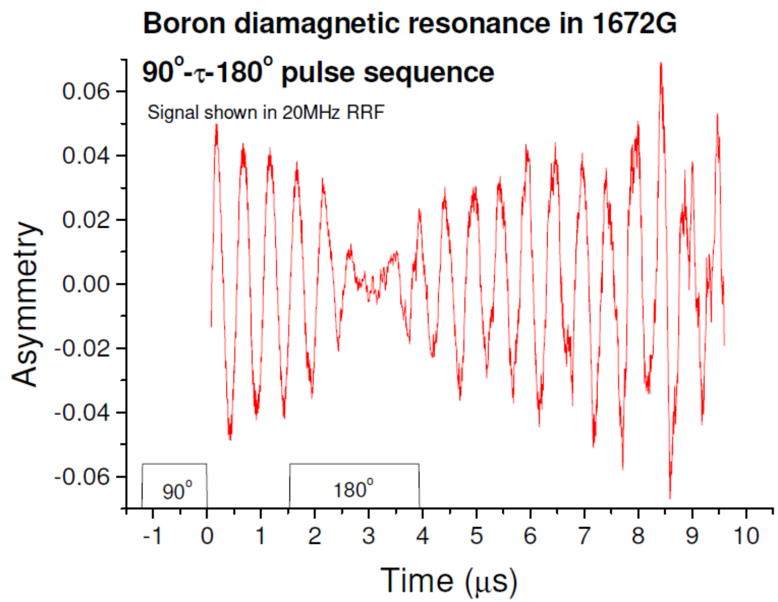


Examples: RF-μSR of diamagnetic muons in boron

S.P. Cottrell et al,
Appl. Magn. Reson. 15 (1998) 469

180°: Echoes and Hop Rates

- Precession damped by random nuclear fields
- Re-focus spins with 180° pulse at time τ, echo at 2τ
- Potential for studying muon hop rates



S.R. Kreitzman / RF resonance techniques for continuous muon beams 1067

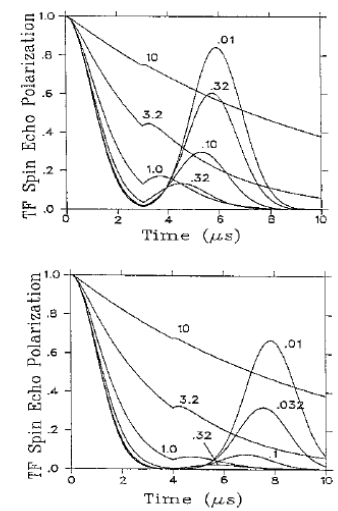


Fig. 6. Theoretical spin echo response for various hop rates. The value of Ω_2 is 1 μs and pulses are tacitly assumed to be *ideal*. The numerical captions denote hop rates in units of $1/t_\varphi$, which for fig. (a) is 3 μs and for fig. (b) is 4 μs. Note that even for reasonably rapid hop rates, the echo position can be distinguished.

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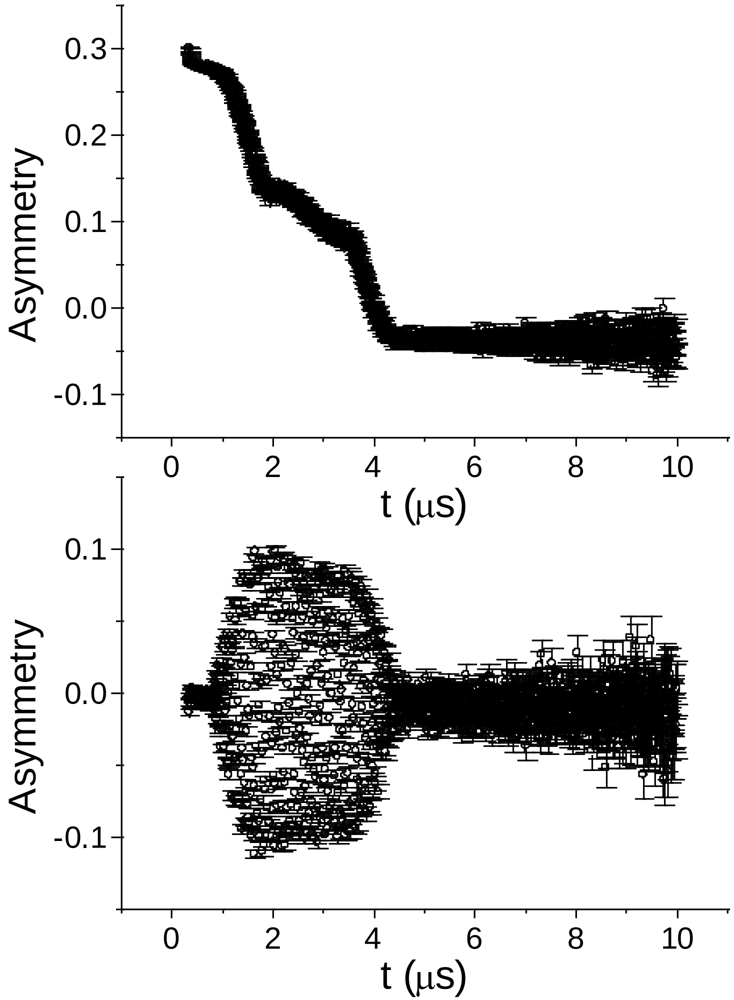
Composite Pulses



Muon RF coils frequently poorly shaped – inhomogeneous fields

Overcome imperfect inversion of 180° pulse using composite sequence $90^o_x180^o_y90^o_x$

Z component shows more complete inversion

Bonus! With μSR we can see the operation of each component of the composite pulse

N.J. Clayden et al,
J. Magn. Reson. 214 (2012) 144

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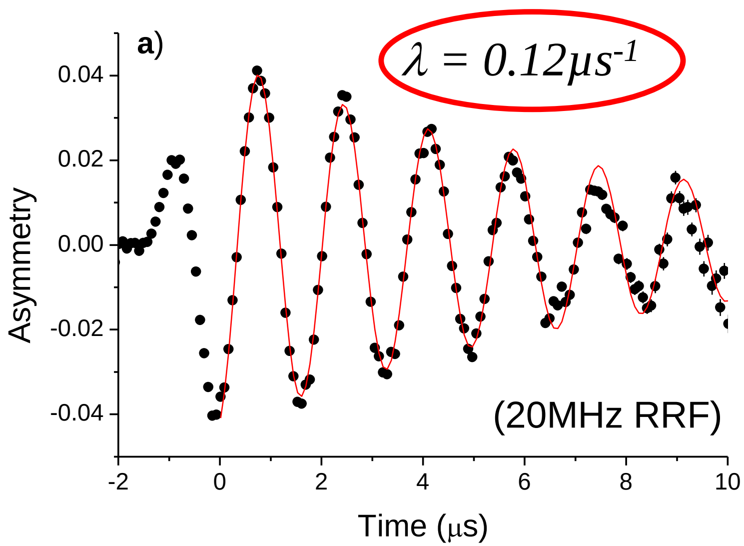
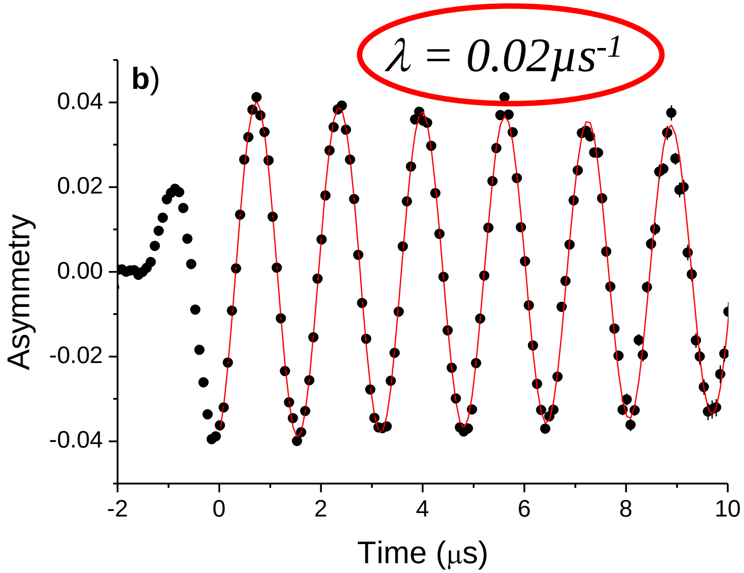
RF Decoupling

Demonstration: $Ca(OH)_2$

Continuous Wave RF Decoupling – μ^+-H dipolar coupling in $Ca(OH)_2$





Free precession of μ^+ following 90º pulse
proton coupling causes depolarisation

40 G decoupling pulse at proton resonance
'stir' nuclear spins / measure muon signal
proton coupling removed

S.P. Cottrell et al,
Physica B 289 (2000) 693

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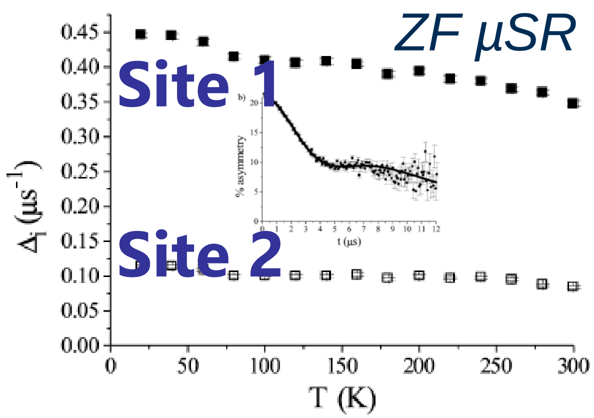
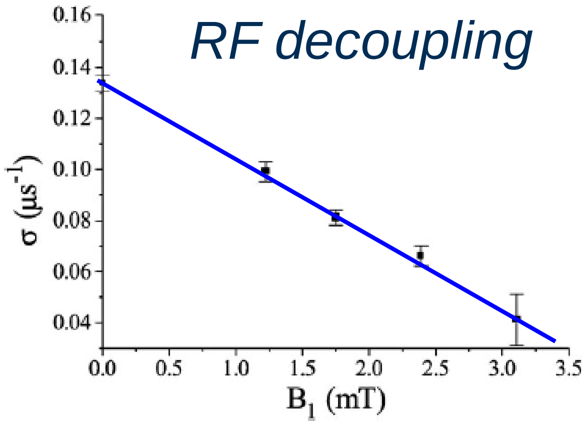
Muon Site Determination

Determining muon's site is an important step in understanding mobility data for proton conductor $Zr(H_2PO_4)(PO_4){\bullet}2H_2O$ …



and



- Two component ZF relaxation implies two muon stopping sites
- Site 1 confirmed by ^1H RF decoupling - large ^1H coupling consistent with muon incorporated into water (HMuO) and ZF relaxation for site 1
- Site 2 weaker coupling consistent with formation of P-O-Mu, μ^+ far from ^1H spins

N. J. Clayden et al, PCCP 8 (2006) 8 3094

Science & Technology Facilities Council
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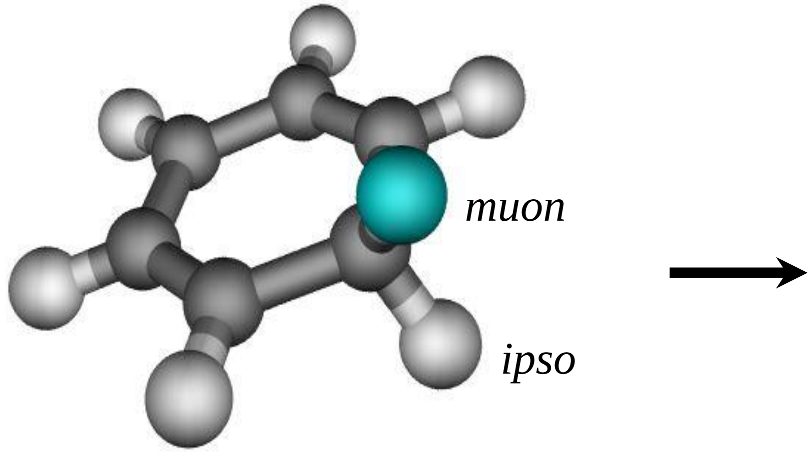
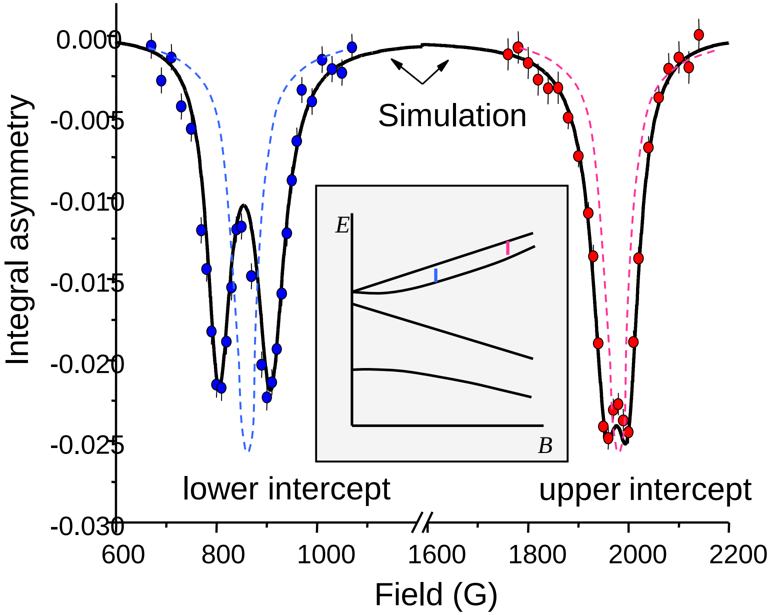
Muonium Chemistry

- Easily applied to spectroscopy of muoniated radicals (method for directly measuring A_μ at a pulsed source)
- Particularly useful for studying slowly formed species or dilute systems when high TF measurements aren't possible

Cyclohexadienyl Radical



Looking at the promptly formed radical state at 218 MHz



From fitting (Quantum), Isotropic hyperfine coupling:

A_{Mu} = 514.25(1) MHz at 298K (514.4 MHz [1])

A_H = 128.5(3) MHz ‘ipso’ proton (126.04 MHz [1])

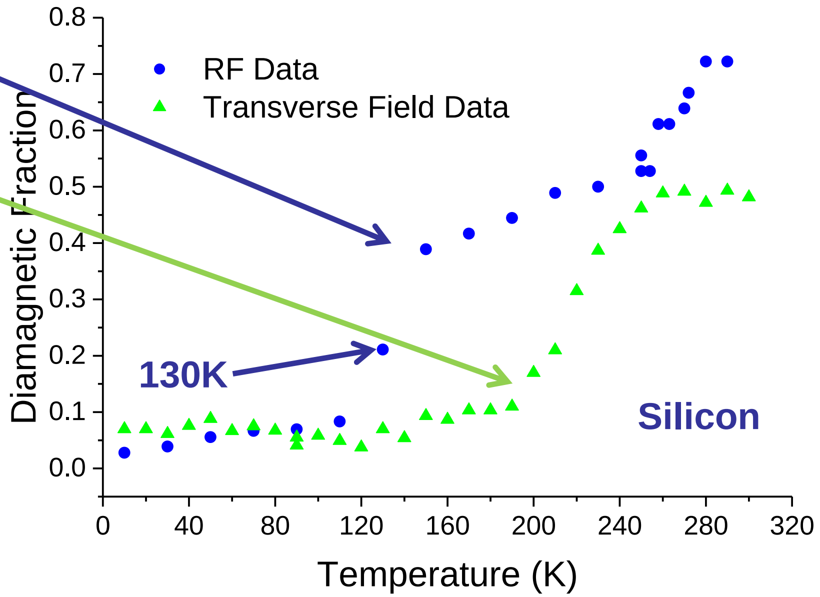
[1] Dake Y.U. et al, Chemical Physics 142 (1990) 229 – LCR data.

I. McKenzie et al,
J. Phys. Chem. B 117 (2013) 13614

Dynamic Processes

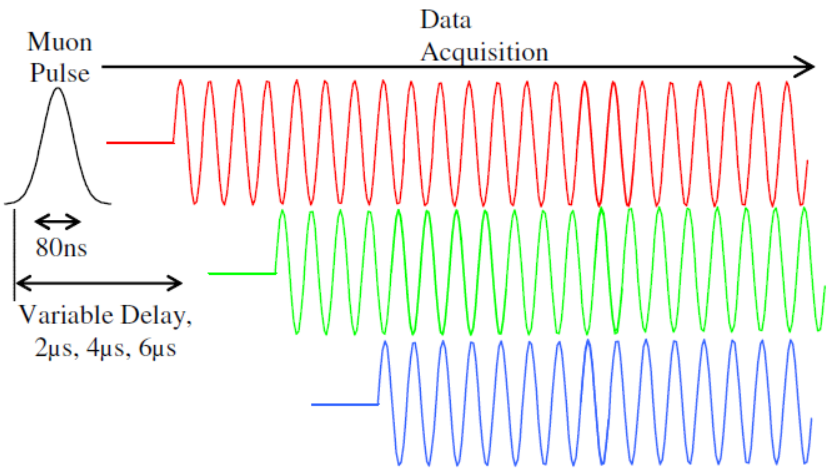
Final State Spectroscopy

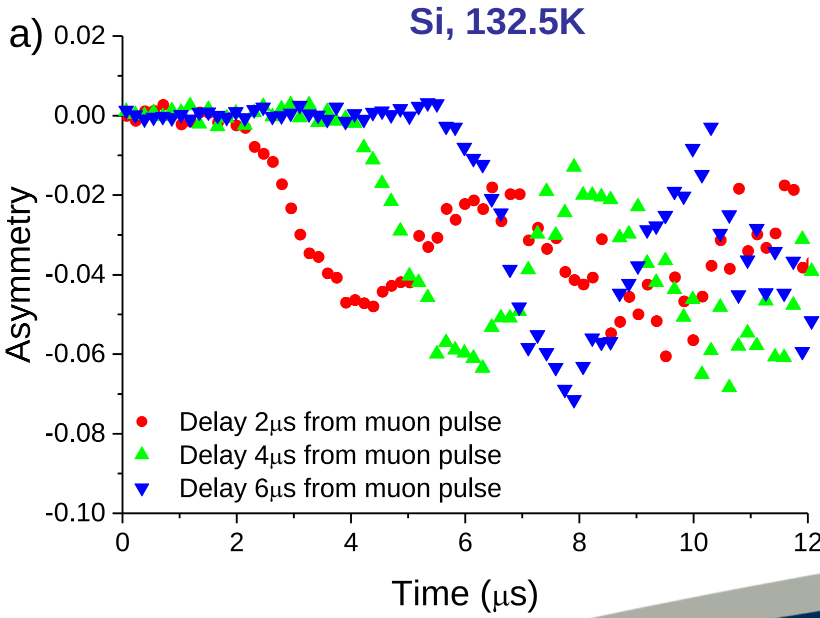
- RF shows appearance of diamagnetic species when TF Mu^{BC} signal disappears
- No diamagnetic signal in TF until much higher temperature
- In TF muons spend a short time as Mu^{BC} and dephase
- In the RF experiment, muon spins are ‘locked’ along high LF before the RF pulse



State Kinetics

- For favourable rates (μs^{-1}), can directly follow conversion of muon charge state (ionization of Mu^{BC} in this case, for Si)
- Follow the build-up of the final diamagnetic state using delayed RF excitation



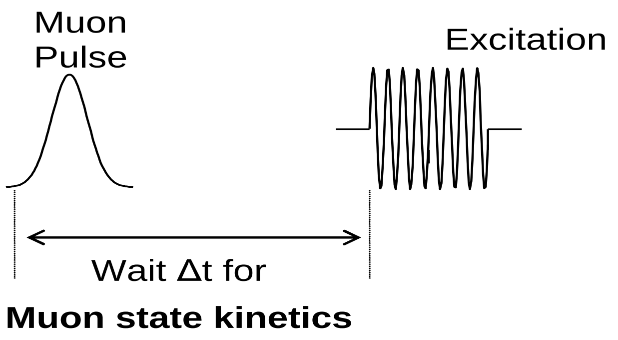


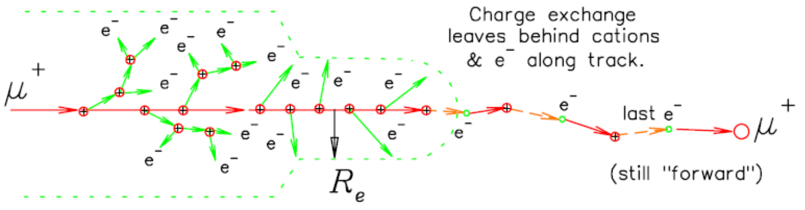
Combining Stimuli
RF + E-Field

D.G. Eshchenko et al., Physica B 404 (2009) 876

Why?

- **RF** to probe final muon state
- **E-Field** to investigate track processes
- Prompt and final states can be compared by combined RF and TF measurements
(as for Si)



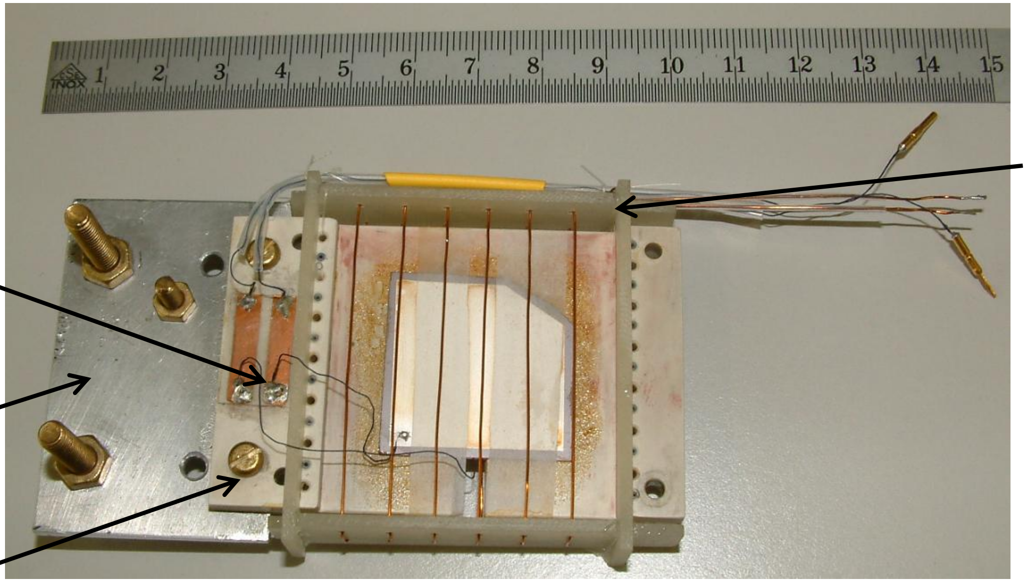


Equipment

- Kit gets progressively more complex as stimuli are combined
- Requires care with design … and sometimes perseverance to get everything working together!

E-field terminal, with fine wires linking front and back electrodes

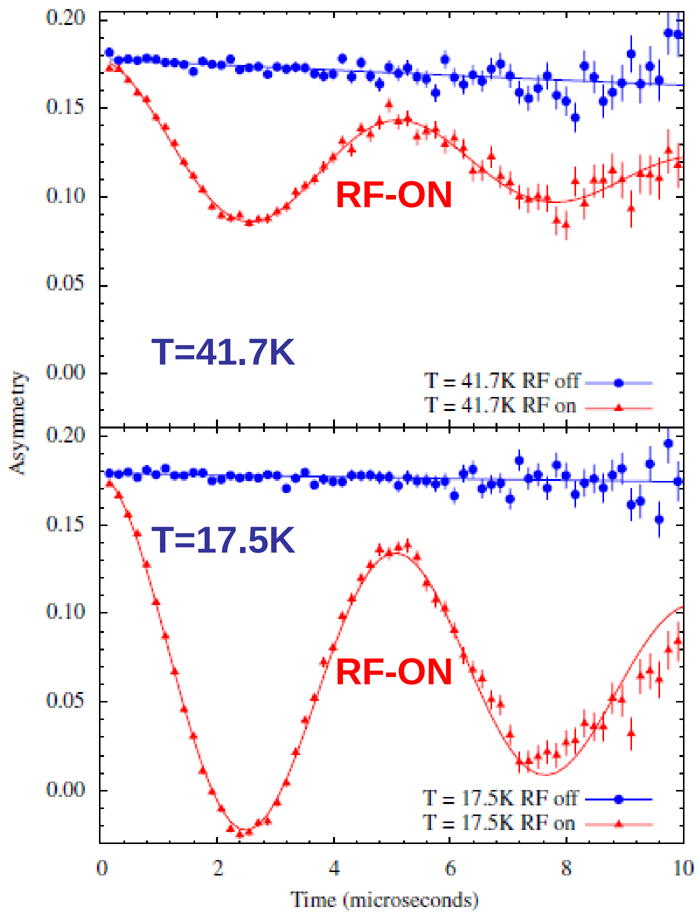
Using CCR, so cool by conduction through plate



RF coil wound on a carrier that can be removed for sample access

Sample mounted on ceramic plate for electrical isolation, Shapal chosen since also a 'good' thermal conductor

Measurements for GaP

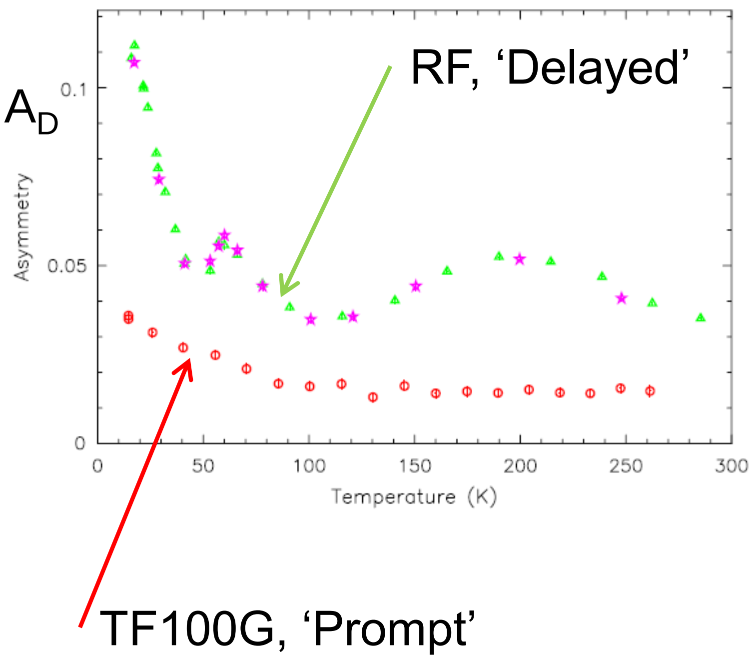


At resonance with μ^+, (20.3MHz, ~1498G)

- Clear precession about the RF field (red curve),
- Amplitude of RF signal increases at low temperature

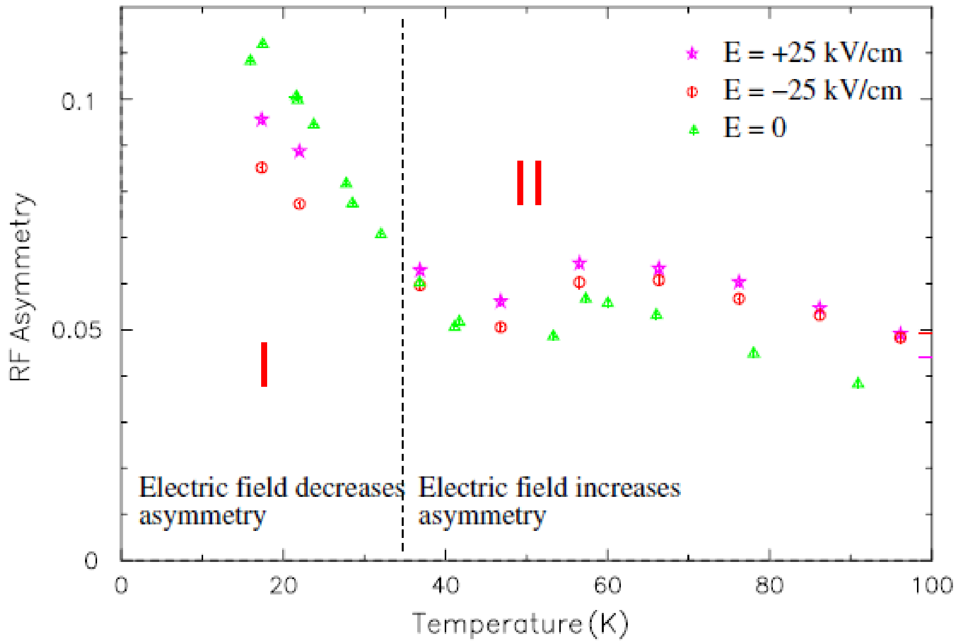
What was learnt for GaP?

- Dramatic increase in RF asymmetry, A_D, at T<40K
- Explained by considering interaction of Mu_T^0 with track e^- or h^+ …
  $Mu_T^0 \rightarrow Mu^+$ or Mu^- conversion
- Conversion time can be estimated by
  - analysing the RF phase shift
  - delayed RF measurements
  Conversion estimated <800ns



What was learnt for GaP?

- E-Field *reduces* RF asymmetry in region I, and *increases* it in region II
- Explanation for increase in region II with E-field: *Mu_{BC}^0 ionisation (as for GaAs)*
- Explanation for decrease in region I with E-field: *Reduction in the cross section for Mu_T^0 capture or e^- or h^+*



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Lots can be done with pulsed techniques ... and there's plenty of interest!

2005 pulsed techniques workshop held in Oxford (following μSR 2005)

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