Stimulation Methods 1:

Pulsed Stimulations for $\mu$SR Experiments

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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. $\mu$SR 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
    - \( \approx 80 \text{ ns} \)
    - \( \tau = 26 \text{ ns} \)
    - \( \approx 20 \text{ ms (50 Hz)} \)

- ‘continuous beam’ (PSI), muons arrive randomly
  - \( <1 \text{ ns} \)
  - \( 20 \text{ ns} \)
At a pulsed source ...
Low duty factor ...

Muon Pulse

Timed to muons, Δt

Excitation @ ‘50Hz’

Very low duty factor ~ 0.1%

ISIS / J-PARC
Time relative to muons …

Before …

Excitation

‘Pump’ sample …

Variable $\Delta t$

Muon Pulse

… ‘Probe’ muons

Coincident or after …

Excitation

Muon Pulse

… ‘Probe’ muons

Variable $\Delta t$

Interact with sample and muons

or

Change $\Delta t$

study relaxation of sample

Change $\Delta t$

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

- 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)

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)

Before…

After…

After…

$\Delta t \leftarrow$

$\Delta t = 0 \text{ ns}$

$\Delta t = 80 \text{ ns}$

$\Delta t = 800 \text{ ns}$

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

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

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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**, $\omega_0$, to equal $\gamma B_0$ (Larmor equation, $\gamma = 13.5534$ kHz/G for $\mu^+$)

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

Form of curve:

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

$\Delta B$ depends on:
- RF power
- Depolarization
- Chemical reaction

Field scan ($\omega_0 = 9.2$MHz)

CW NMR!
‘Longitudinal’ $\mu$SR signal – precession about $B_1$.

At resonance …

Precession about RF field, $B_1$, determined as $\sim 15$G from fit.
‘Transverse’ $\mu$SR signal – precession about $B_0$

At resonance …

Amplitude of ‘transverse’ $\mu$SR signal grows

Turn off RF field after 90° rotation of $P_0$

Muon precession at $\sim$20MHz,
free precession signal following a 1.3\,$\mu$s RF pulse

In NMR terms, this is a 90° pulse!
RF $\mu$SR – What are we Learning?

‘transverse’ $\mu$SR signal – free precession

Envelope can tell us about the local field distribution at muon site

‘longitudinal’ $\mu$SR signal

Envelope can tell us about muon charge state conversion reactions

Asymmetry

Time (µs)

Differential Asymmetry

Time (µs)
Muon Precession at 20MHz!? 

For *Pulsed Muon Sources*, finite muon pulse width limits maximum usable 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$_2$B$_2$C …

\( T = 12K \)

\( T = 17K \)

\( T = 7.5K \)

\( B_0 = 1034G (13.6MHz) \)

\( \mu^+ \rightarrow P_0 \rightarrow e^+ \text{ Detect} \)

\( \mu^+ \rightarrow P_0 \rightarrow B_0 \rightarrow e^+ \text{ Detect} \)

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-\(\mu\)SR of diamagnetic muons in boron

\[ \text{FID (90}_\text{x} \text{)} \]

\[ (90}_\text{x} - \text{\(\tau\) - 180\(}_\text{x} \text{)} \]

\[ \text{Time (\(\mu\)s)} \]

\[ \text{Polarisation} \]

\[ \text{S.P. Cottrell et al,} \]
\[ \text{Appl. Magn. Reson. 15 (1998) 469} \]
180°: Echoes and Hop Rates

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

Muon RF coils frequently poorly shaped – inhomogeneous fields

Overcome imperfect inversion of 180° pulse using composite sequence $90^\circ_x 180^\circ_y 90^\circ_x$

Z component shows more complete inversion

Bonus! With $\mu$SR we can see the operation of each component of the composite pulse

RF Decoupling
Demonstration: Ca(OH)$_2$

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

Free precession of $\mu^+$ following 90$^\circ$ 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
Muon Site Determination

Determining muon’s site is an important step in understanding mobility data for proton conductor Zr(H₂PO₄)(PO₄)•2H₂O …

- Two component ZF relaxation implies two muon stopping sites
- Site 1 confirmed by $^1$H RF decoupling - large $^1$H 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, $\mu^+$ far from $^1$H spins

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

- Easily applied to spectroscopy of muoniated radicals (method for directly measuring $A_\mu$ 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:
\[ A_{Mu} = 514.25(1) \text{ MHz at } 298\text{K} \quad (514.4 \text{ MHz [1]}) \]
\[ A_H = 128.5(3) \text{ MHz ‘ipso’ proton} \quad (126.04 \text{ MHz [1]}) \]

I. McKenzie et al,

Dynamic Processes
Final State Spectroscopy

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

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

$RF + E\text{-Field}$
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

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

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

At resonance with $\mu^+$, (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 $h^+$ …

  $\mu_T^0 \rightarrow \mu^+ \text{ or } \mu^-$ 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 $h^+$*
Lots can be done with pulsed techniques ... and there’s plenty of interest!

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