Radio Frequency Muon Spin Resonance

User Guide


Version 0.2
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1 Introduction

This manual describes the radio frequency muon spin resonance (RFμSR) technique and the equipment used on the DEVA surface muon beam located at the ISIS facility at the Rutherford Appleton Laboratory (UK). The pulsed nature of the ISIS beam is particularly advantageous for RF measurements as the excitation pulse can be synchronised with the muon arrival. The low RF duty cycle (approximately 0.1%) afforded by such an arrangement ensures that RF heating of the target is avoided. In addition, data collection is inherently time differential with no experimental time penalty, making it easy to perform time-delayed measurements.
2 Experimental details

Muons are produced 100% spin polarised from pion decays, with a half-life of 2.197 µs. Following implantation, the muon spin evolves according to the local magnetic environment, whether internal or externally applied. The time evolution of the polarisation is followed by means of detectors arranged forward (upstream) and backward relative to the initial muon spin polarisation. Signals from these detectors measure the spatial distribution of the decay-positrons, which are emitted preferentially along the direction of the muon spin. Histograms are formed for each detector with the number of positrons detected during each 16 ns time-step being recorded. Data is typically collected for 32 µs following muon implantation.

In a conventional muon spin rotation experiment, a magnetic field $\mathbf{B}$ is applied transverse (TF) to the muon polarisation causing the muon spins to precess. If the system contains both muonium and diamagnetic species, the asymmetry $A(t)$ is given by:

$$A(t) = A_{\mu,h} e^{-\lambda_{\mu,h} t} \cos(\omega_{\mu,h} t + \phi_{\mu,h}) + A_{D} e^{-\lambda_{D} t} \cos(\omega_{D} t + \phi_{D})$$

(1)

where $A_{\mu,D}, \lambda_{\mu,D}, \omega_{\mu,D}$ and $\phi_{\mu,D}$ are the initial muon asymmetry, depolarisation rate, larmor precession frequencies and initial phases for the paramagnetic and diamagnetic environments respectively. For the diamagnetic species, the precession is at the Larmor frequency and is simply the product of the muon gyromagnetic ratio and field strength. For muonium, however, it is necessary to compute the energy levels in the Breit-Rabi diagram to obtain the precession frequency. In a field of 2 G, for example, $\omega_{D}$ is $\sim 0.027$ MHz and $\omega_{\mu}$ is $\sim 2.8$ MHz. Increasing the applied field to 20 G results in the muonium precession frequency being raised above the ISIS pass-band (defined by the 80 ns width of the muon pulse to be $\sim 6$ MHz). Consequently, the signal can no longer seen in the data.

In an alternative experiment, the magnetic field can be applied co-linear with the muon spin polarisation. In this longitudinal field (LF) environment there is no precession. Consequently, the paramagnetic and diamagnetic environments cannot be so easily distinguished.

These environments can be investigated, however, by the application of RF resonance methods similar to those first pioneered by Coffin et al. [1]. Such techniques were later developed by the Tokyo group [2, 3] as a tool for reaction studies using the pulsed beam of the KEK facility. More recently, RF methods have been utilised with both DC beams at TRIUMF [4, 5] and pulsed beams at ISIS [6] for studies in condensed-matter physics.

The RF technique combines both the TF and LF experiments by having a static field, $\mathbf{B}_0$, applied co-linear with the muon spin polarisation, and an oscillating $\mathbf{B}_1$ field applied transverse to it. As in NMR, adjusting the static field strength and RF frequency to meet the resonance condition for the system causes the probe spin to experience a torque due to the $\mathbf{B}_1$ field and precesses about that field for its duration. In practice, $\mu$SR data is collected both with and without the RF field to clearly...
identify its effect on the system, the experiment alternating between the two states, typically every 10 s, to reduce the effects of equipment instability.

Data can be treated in two distinct ways. Firstly, an experiment akin to continuous wave magnetic resonance can be carried out by sweeping either the $B_0$ field or RF frequency through the resonance condition. In this case, a time-integral RF asymmetry can be defined [7] as follows:

$$A_{\text{RF}} = \frac{N_{RF} - N_0}{N_{RF} + N_0} - \frac{N_{RF} - N_0}{N_{RF} + N_0}$$

where $N_{RF}$ is the time-integral positron rate with the RF excitation on, $N_0$ with it turned off and the subscript $i$ referring to the relevant group of detectors. A particular advantage of collecting data at a pulsed muon source is that the window of integration can be chosen after the experiment, according to the nature of the signal. It can be shown [7] that a Lorentzian line shape, $A_{RF}(\omega)$, is obtained:

$$A_{\text{RF}}(\omega) = A_{\text{RF}}(\omega_0) \frac{\Delta\omega^2}{(\omega - \omega_0)^2 + \Delta\omega^2}$$

where $A_{RF}(\omega_0)$ is the resonance amplitude, $\omega_0$ is the resonance frequency and $\Delta\omega$ is the half-width at half-maximum of the resonance line. Both $A_{RF}(\omega_0)$ and $\Delta\omega$ depend upon RF power, depolarisation or chemical reaction of the measured species and the muon lifetime [7].

Secondly, a time-differential signal can be measured on-resonance, displaying the spin precession around the RF field, $B_1$. This technique is particularly important for revealing slowly formed muon species, which may be directly measured simply by delaying the RF excitation relative to muon implantation. In the case where there is a paramagnetic precursor to a final diamagnetic state, the TF-µSR signal of the diamagnetic species can only be observed if the build-up of the final state is rapid. The timescale of this build-up being defined by the quantity $1/\omega_{\text{HF}}$ (where $\omega_{\text{HF}}$ is the hyperfine coupling of the paramagnetic state). The use of the RF resonance technique allows for the onset of the excitation pulse to be delayed relative to muon implantation and, by varying this delay, the build-up of the final muon state may be displayed explicitly [8]. A timing diagram for such an experiment is shown in Figure 1.
Figure 1 Timing diagram for the delayed RF excitation experiment.

An expression for the asymmetry $A(t)$ under these conditions follows from the work presented in reference [9] and involves three terms: the original prompt fraction, a fraction formed due to conversion during the delay, and a delayed fraction formed in the presence of the RF field:

$$A(t) = A_0 + A_d(t) + \int_{\delta_i}^{t} \frac{dA_d(t - \delta)}{dt} P(t - t' - \delta) dt' \quad (t > \delta_i) \quad (4)$$

where $A_0$ is the prompt diamagnetic asymmetry, the time dependence of the RF field is given by $P(t) = e^{-\alpha t} \cos(\omega t)$, $\alpha$ is depolarisation due to field inhomogeneities, and $\delta_i$ is the RF delay. The diamagnetic asymmetry resulting from state conversion, $A_D(t)$, is given by the expression:

$$A_d(t) = A_{Ma}^0 \left[ \frac{1 + 2x^2}{2(1 + x^2 + k_{Ma})} \right] \left[ 1 - e^{-k_{Ma}t} \right] \quad (5)$$

where $A_{Ma}^0$ is the prompt muonium asymmetry, $k_{Ma}$ a pseudo first-order rate constant for the conversion process and $x = B/B_{HF}$ with $B_{HF}$ the hyperfine field.

A similar expression to equation (4) above can be written for the muonium amplitude where, in this case, any delayed fraction could be due to charge neutralisation processes of either bare muons or molecular ions, $\text{Mu}^+$. 

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3 Getting started

3.1 Layout of the RF\(\mu\)SR system

The equipment required for a RF\(\mu\)SR experiment can be divided into three main components. The first comprises the electronics required to provide suitable timing signals such that the radio frequency cavity is energised only when muons are present in the sample. Secondly, a RF signal generator and amplifier are used to produce a signal of suitable frequency and power to excite the transition we wish to observe. Finally, a RF cavity into which the target material, be it a solid, liquid or gas, is contained within a non-ferromagnetic, non-conducting target cell.

The main features of the RF\(\mu\)SR system are shown in Figure 2.

<Figure 2 RF equipment photo about here>

The rack system available to the user provides all the electronics required for a RF\(\mu\)SR experiment; the three separate components, timing, signal generation and amplification are outlined below.

3.2 Timing modules

The advantage of a pulsed muon source like ISIS, with respect to an RF experiment, is that RF power does not need to be maintained when no muons are present. Consequently, the duty cycle is reduced to a fraction of that required at a continuous muon source with a concomitant reduction in sample heating. The timing of the RF excitation pulse is provided by two C.A.E.N. dual timers as shown in Figure 3.

A detailed timing diagram for the RF\(\mu\)SR experiment is shown in Figure 4. Approximately 2.5\(\mu\)s prior to the arrival of the muons, an ‘Extract Trigger’ signal is received from the ISIS synchrotron starting the first CAEN dual timer. The length of the delay, shown as 1, determines the time before onset of amplifier noise blanking. At the end if the delay, the ‘End Marker’ is used to start a second delay, shown as 2, before the RF is gated. The timing is arranged such that the muons accumulate for the duration of the ISIS pulse (80ns FWHM) before the RF is applied. This removes any time uncertainty arising from the finite width of the muon pulse. After a delay of approximately 30\(\mu\)s, shown as 3, data acquisition is turned off along with the RF. The amplifier noise blanking remains switched on well after the RF has ended and this timing is determined by delay 4.
Figure 3 The picture shows the LeCroy level adapter along with the two C.A.E.N. dual timers making up the timing circuit for the RF experiment.

Figure 4 Timing diagram.
The wiring of the electronics will normally be carried out by the local contact and is shown in diagrammatic form in Figure 5.

![Diagram of the wiring of the two CAEN timers for a typical RFμSR experiment.](image)

**Figure 5** A diagrammatic view of the wiring of the two CAEN timers for a typical RFμSR experiment.

### 3.3 RF Signal generation

The RF rack contains a signal generator

![Signal generator showing (a) the left menu, (b) the right menu, (c) the numeric keypad and (d) the units keys.](image)

**Figure 6** The signal generator showing (a) the left menu, (b) the right menu, (c) the numeric keypad and (d) the units keys.

To set the RF frequency, press the top button of the right-hand menu (Figure 6b) and enter the value using the numeric keypad. The units are select using the far right
buttons labelled (d) in Figure 6. The RF level is set using the second button of the right-hand menu along with the numeric keys. The symbol, db, is entered as the fourth of the units buttons.

Each RF rack has a power amplifier capable of providing a maximum of 1 kW in the frequency range 20 kHz to 220 MHz.

At the back of the amplifier are three ports into which are placed cables carrying the blanking pulse signal, the low-power RF signal from the generator and the thick high-power output cable respectively.

### 3.4 RF cavity

The target cell comprises a container of the material under investigation around which is wound a suitable RF cavity. The direction of the $B_1$ field generated by this cavity must be arranged such that it is perpendicular to the applied longitudinal field $B_0$. In order for amplifier to deliver maximum power to the coil, it should be matched to 50 Ω. Details of coil design and the matching procedure are given in the following section.
4 RF Coils

There are three basic coil designs and the choice of which one to use will depend upon the sample orientation with respect to the muon beam.

4.1 Solenoid

This is the simplest coil design and an example of a solenoid coil is shown in Figure 8 along with the direction of the magnetic field produced.

< Figure 8 Picture here >

This coil consists of a number of turns of wire wrapped around the sample or sample cell.

4.2 Saddle

In a situation where a simple solenoid coil will produce a magnetic field in the wrong orientation with respect to the muon beam, a saddle coil can be used, Figure 9.

Figure 9 Diagram of a saddle coil showing the direction of the field it produces.

The saddle coil consists of a pair of counter-wound coils forming a cavity through which the magnetic field passes.

4.3 Birdcage

A more complicated coil, but one that produces a highly homogeneous magnetic field over a large region, is a birdcage coil, Figure 10.
This coil consists of two hoops of copper wire joined together by a number of capacitors.

4.4 Coil Tuning

For efficient transfer of RF power from the amplifier into the sample, the coil needs to be impedance matched to 50 $\Omega$. This matching is achieved by the use of a “matching box” containing a pair of capacitors.
5 Example experiments

5.1 Single resonance experiment

5.2 Double resonance experiment
References