High Time Resolution Muon Spectroscopy

Exploiting a novel magnetic resonance technique

Muons provide a very sensitive probe of the atomic-level properties of materials. High Field muon techniques are being developed by the European facilities; the high time resolution available at the SμS enables unique phenomena to be investigated.

Examples of how high time resolution muon spectroscopy can be used for novel measurements include:

- Probing superconductivity; vortex states and length scales
- Studying magnetic systems such as spin liquids and low dimensional magnetism
- Characterising hydrogen impurities in semiconductors
- Investigating quantum fluctuations in high spin molecules.

A quick introduction to the muon technique

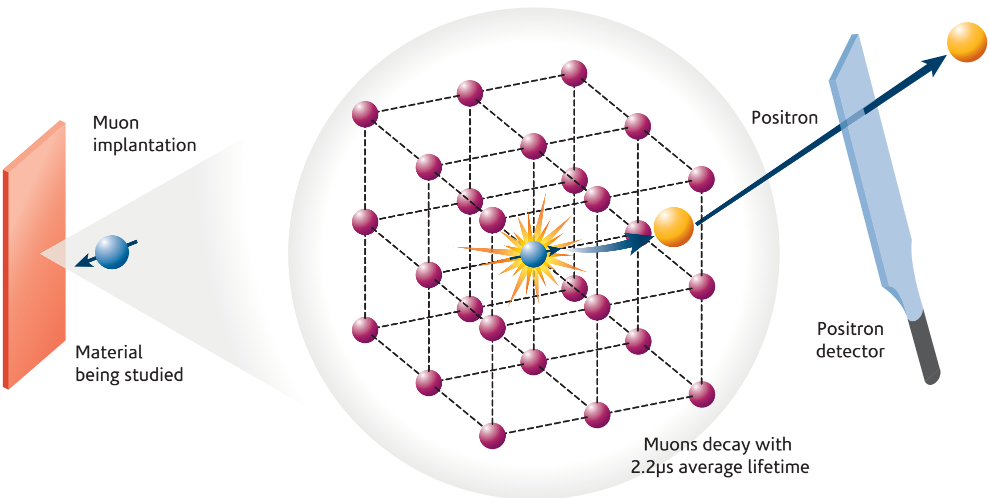
Muon spin resonance spectroscopy is less well known than other spin-spectroscopic techniques such as NMR and EPR, but it provides researchers with an important tool that can be used to study a wide range of problems in physics and chemistry.

The muon technique involves implanting spin-polarised positive muons into a material. Muons are short-lived particles, decaying after an average lifetime of 2.2μs to produce positrons. The decay positrons which emerge from a sample after muon implantation are detected revealing information about the muons' behaviour inside the material – particularly about how the muon polarisation changed within the sample. This, in turn, enables us to deduce information about the atomic-level properties of the material.

Muons are very sensitive probes of magnetic systems, often detecting effects that are too weak to be seen by other methods. They also have a wide variety of other applications – for example, in studies of superconductors, magnetic materials, molecular systems and chemical reactions, novel battery materials and a variety of organic systems. In some studies, the positive muon can be thought of as being like a light proton (muons have a mass of one ninth of the proton mass). Implanted muons will sometimes pick up an electron to form a light isotope of hydrogen called muonium (Mu). By following muon behaviour inside a material we can learn about proton and hydrogen behaviour. This is important in semiconducting materials, proton conductors and hydrogen storage materials and insulating materials.

References on the muon technique include:

- ***Muon spin rotation, relaxation and resonance: Applications to condensed matter*** *A Yaouanc, P Dalmas de Réotier, Oxford University Press (2010), ISBN 0199596476*
- ***Spin polarised muons in condensed matter physics*** *S J Blundell, Contemporary Physics, **40** (1999) 175*
- ***The Muon Method in Science****, V.P. Smilga, Yu.M. Belousov, Nova Science Publishers (1994) ISBN 9781560721611*



The muon technique – implantation of positive, spin-polarised muons into a sample, followed by detection of positrons emitted when the muons decay.

Science & Technology Facilities Council ISIS

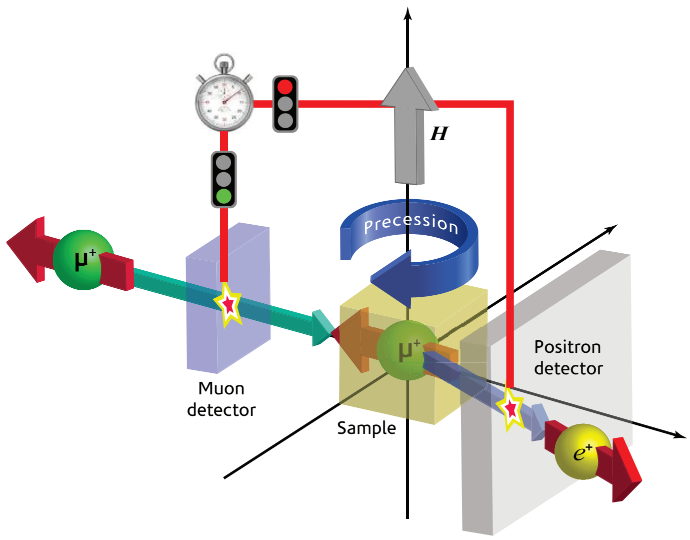
PAUL SCHERRER INSTITUT PSI

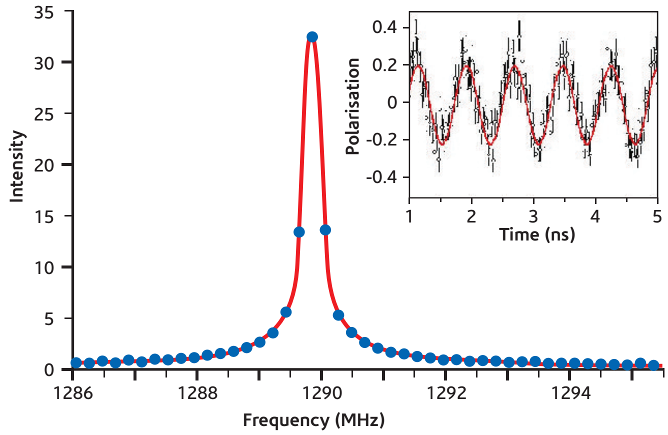
High resolution muon spectroscopy: the basic idea

Once implanted inside a material, muons interact with their local atomic environment. In some cases this interaction can be strong, and fast timing resolution is required to follow the evolution of the muon spin polarization. For example, in magnetic systems, large internal fields give rise to fast precession frequencies, while broad internal field distributions will lead to a rapid decay of the muon polarisation. In chemical systems, the measurement of detailed spectroscopic information requires large probing fields which, in turn, give rise to energy level splittings of the order of hundreds of megahertz.

The beam structure of the SμS, located at PSI, Switzerland is ideally suited to these types of measurement. Here, muons are implanted into the sample one by one, enabling accurate measurement of the time interval between the arrival of the muon and the detection of the decay positron. With careful instrument design and advanced detector technologies, time resolutions better than 80ps are possible.

This is illustrated by the transverse field experiment, where fast muon precession is measured in a large external magnetic field applied perpendicular to the initial muon spin polarisation. In this case a 9.5T field was applied to muons stopped in a silver plate and a precession frequency of ~1.3 GHz is measured. The decay of the muon response and the corresponding shape of the Fourier transform reflect the microscopic field distribution sensed by the muon.



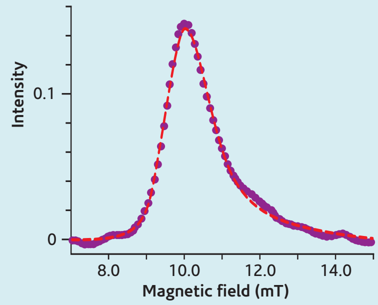


Precise measurement of the time interval between muon arrival and decay (top) enables high resolution spectra to be recorded.

Example Applications of High Resolutio

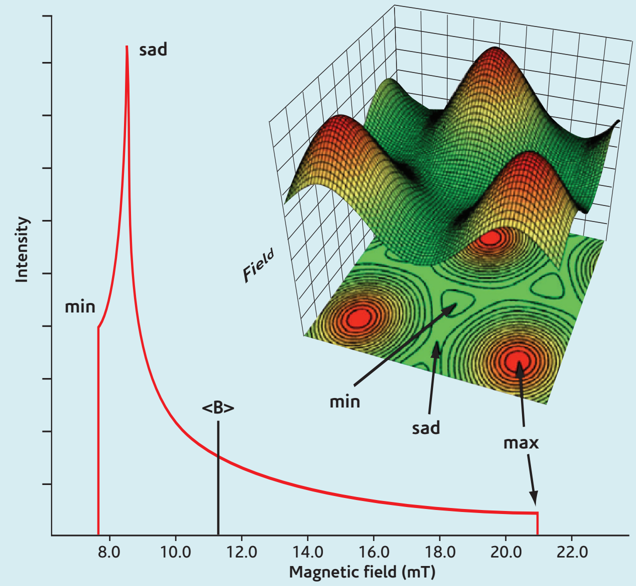
Superconductivity

The *vortex state* induced in a type-II superconductor when a strong magnetic field is applied can be studied using muons. The technique probes the magnetic field on a length scale much shorter than the inter vortex distance, enabling information about the internal vortex structure and interactions to be obtained.

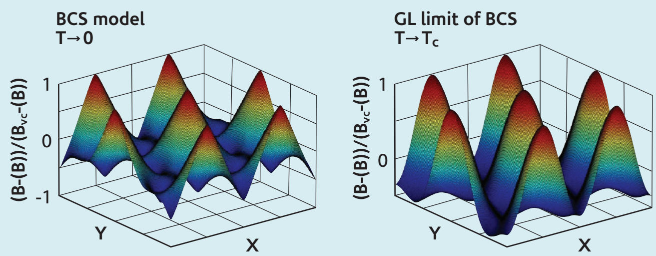


Field distribution (with model fit) of a vortex lattice formed in $SrFe_{1.75}Co_{0.25}As_2$ at 1.6K and an external field of 10mT.

Adapted from R. Khasanov et. al. Phys. Rev. Lett. 103, 067010 (2009).



Muons are now routinely used to determine *characteristic length scales*, such as the magnetic penetration depth and coherence length, and the muon technique enables vortex lattice topology to be investigated. Muons are also playing a key role in the search for experimental evidence for exotic vortex states. For example, a change in the spatial field distribution around the vortex cores has been predicted for clean superconductors at low temperatures and at fields close to the upper critical field. Spectrometers capable of extended temperature and field measurements promise to bring a new insight to these studies.



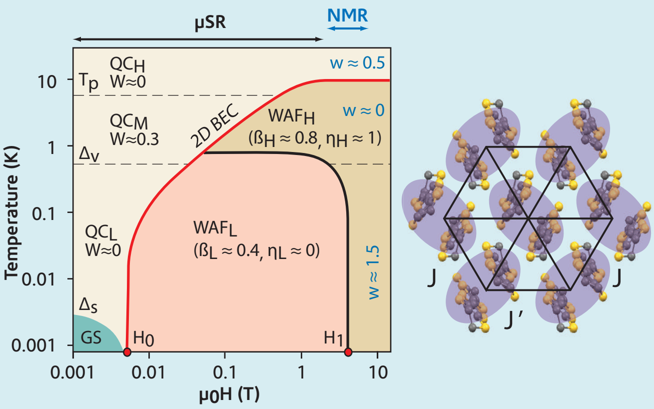
Models for conventional (left) and exotic (right) vortex states can be investigated using muon techniques.

A. Maisuradze et al., arXiu:1303.0209v1 (2013).

n Muon Spectroscopy in Condensed Matter Physics

Probing Spin Liquids

The *quantum spin-liquid phase* is an intriguing state where the usual magnetically ordered ground state is avoided even at the lowest temperatures for a system of strongly interacting magnetic spins. Spin liquid systems such as the Mott insulator χ-(BEDTTTF)$_2$Cu$_2$(CN)$_3$ display a complex phase diagram that can best be investigated using the extended field and temperature regimes offered by the latest muon spectrometers. Existing measurements have demonstrated no obvious signature for conventional magnetic ordering to 20mK.

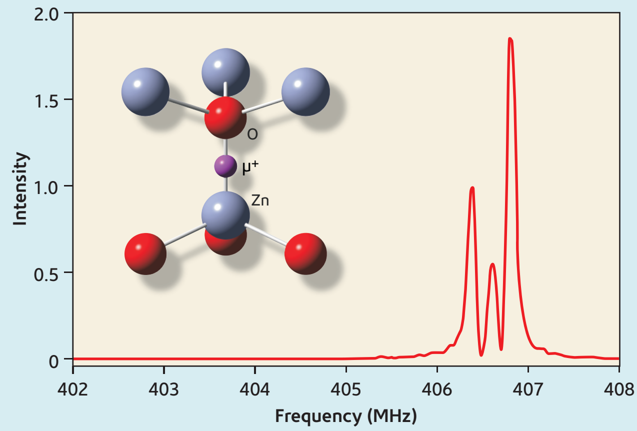


The rich phase diagram (left) of the Mott insulator χ-(BEDTTTF)$_2$ (right, grey + C and yellow = S) Cu$_2$(CN)$_3$ is readily explored using muon techniques. Compared to conventional resonance techniques, muon spectroscopy enables measurement over a greatly extended field range.

F. L. Pratt et al., Nature 471, 612 (2011).

Semiconductors and Oxides

Muonium, a bound μ$^+$e$^-$ system, can be considered as a light hydrogen isotope, and is frequently used to obtain information about *isolated hydrogen in semiconducting systems.* High transverse field muon spin rotation measurements enable high resolution spectroscopy of the different electronic states this hydrogen-like isotope can take, with the hyperfine coupling providing information about the local environment. Spin dynamics and state transitions can also be probed by investigating the field dependence of polarised muonium at different temperatures.

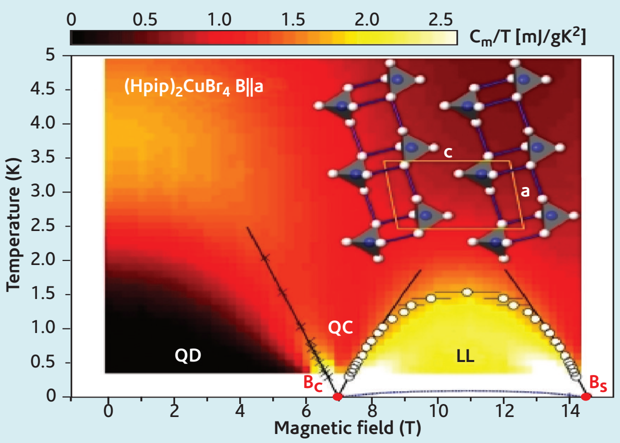


Spectroscopy of a Mu(H) centre formed in ZnO (inset). Line intensities reflect electron polarization.

Adapted from K. Shimomura et al., PSI Annual Report 2005.

Low Dimensional Magnetism

Low dimensional quantum spin systems are remarkable for their diverse and fascinating physical behaviour, and the muon technique provides an ideal probe of their properties. For example, quantum phase transitions between gapped and gapless phases of two-leg spin-ladder systems such as (Hpip)$_2$CuBr$_4$ can be driven using a large external magnetic field. The Luttinger Liquid regime, where the spectrum is gapless with no finite order parameter, can also be investigated from the high-temperature classical regime through a crossover.

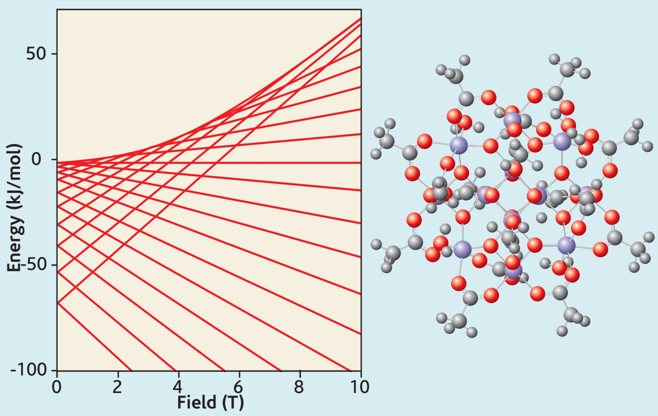


Phase diagram of the spin-ladder (Hpip)$_2$CuBr (inset). Transitions between the quantum disordered (QD), quantum critical (QC) and Luttinger-Liquid (LL) phases can readily be explored using muon techniques.

Adapted from Ch. Rüegg et al, Phys Rev Lett 101 247202 (2008).

High Spin Molecules

Molecular clusters containing as few as 10 to 100 atoms might be used as magnetic memory units, potentially offering greatly increased storage density. These systems are high spin molecules, comprising a small network of magnetic ions such that the net spin of the cluster is large (e.g. 10, 27/2 or 39/2). Unfortunately, molecular spin relaxation rates currently limit the application of these systems, and high field muon spectroscopy provides an ideal method for determining the quantum fluctuations and tunnelling driving this spin relaxation.

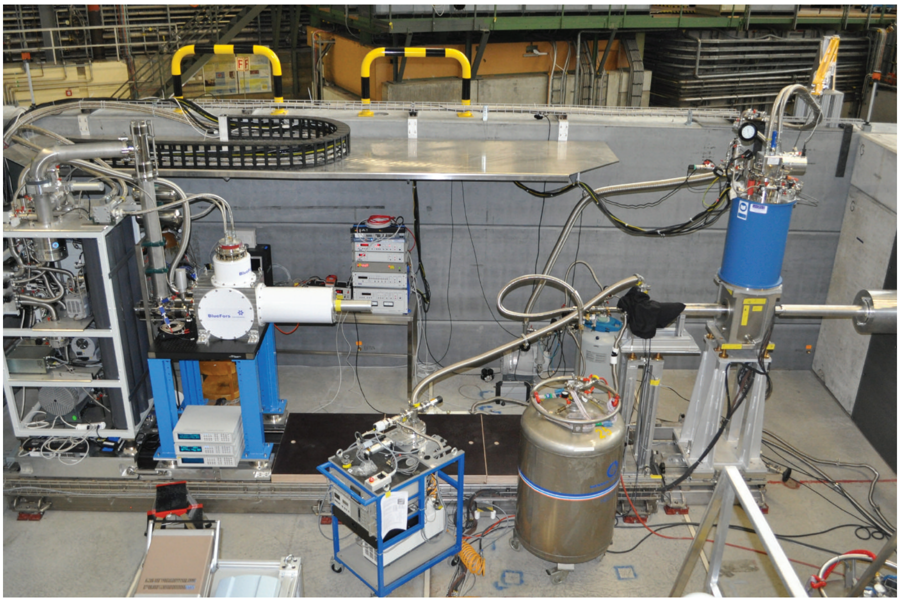


Energy levels for Mn$_{12}$-acetate$_{16}$ (S=10) (grey = C, white = H, red = O and purple = Mn). Fluctuations and tunneling rates can be probed using muon techniques.

G. Chaboussant et al., Phys. Rev. B70, 104422 (2004).

Facilities for Muon Spectroscopy

Europe is fortunate in having two muon sources that are complementary. The beam structure of the SμS, located at the PSI in Switzerland, makes it ideally suited for applications where high timing resolution is essential, such as following fast muon precession or rapid spin depolarisation. In contrast, the pulsed muon beam operated by the STFC in the UK, allows low background time differential data to be captured at high data rates. It also enables the effect of beam synchronous stimuli (such as Radio Frequency or laser radiation) to be investigated. Together, these facilities provide beams of muons for a wide variety of atomic-level studies in condensed matter, molecular, chemical, biological, geological and engineering materials. Further details of the various instruments and sample environment equipment can be found on the facility web sites.

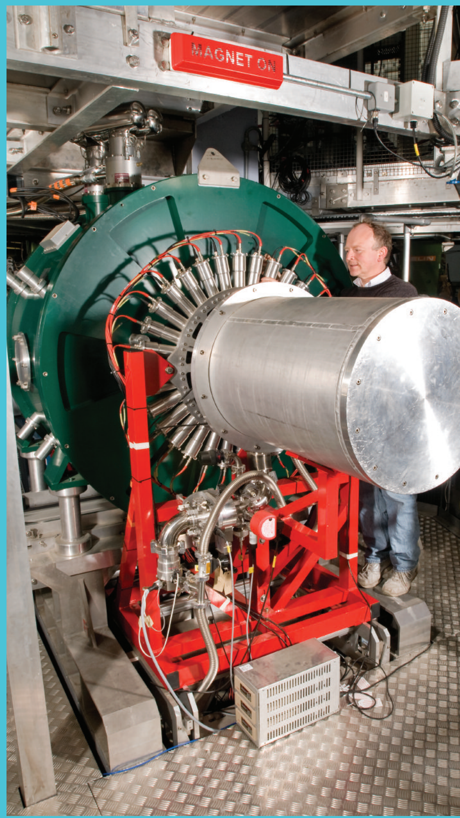


At both facilities a number of spectrometers are available with specialist sample environment equipment to enable a broad range of condensed matter and molecular studies on solid, liquid and gaseous samples. Temperature studies can extend from millikelvin temperatures to 1500 K and solid-sample pressures up to 2.5 GPa can be applied. Both facilities have recently completed major instrument upgrades to provide high magnetic fields; at ISIS fields of 5 T parallel to the muon spin are possible, while PSI provides a 9.5 T spectrometer optimised for spin rotation measurements.

Using the Facilities

Both facilities welcome experiment proposals from scientists of all disciplines. Calls for proposals occur twice a year: deadlines at ISIS are 16 April and 16 October, while at PSI deadlines are 10 December and 11 June. Proposals can be made using the online systems available through the respective web pages – typically a two-page science case is required.

Members of both groups are available to give advice on all aspects of muon science and running muon experiments. They can be contacted to discuss ideas for experiments, for technical and practical information on the muon instruments and to offer advice on draft proposals.



Above: *High field muon spectrometer at ISIS, UK.*

Left: *The new instrument for high field muon spectroscopy at PSI. The 9.5 T magnet is on the right side. On the left is the dilution refrigerator with the gas handling system visible, which can cool samples to temperatures below 20 mK.*

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