Muon Spin Relaxation (μSR) Studies of Actinide Materials

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1. Scientific theme and description of the technique

The electronic properties of actinide materials are governed by their f-shell electrons, which can be itinerant or localized or sometimes both itinerant and localized in the same atom. This behavior produces a diversity of ground states, including charge density waves, superconductivity, and multipolar ordering of various kinds - ranging from ordinary magnetism to quadrupolar or octupolar ordering, for example. Competition from competing order parameters (such as antiferromagnetism and superconductivity) can be tuned to suppress a particular phase to zero temperature producing a 'quantum critical point' (QCP). Currently there is no complete theory which leads to a predictive understanding of these f-electron ground states. One reason for this is that the non-linear coupling of spin-charge and lattice degrees of freedom can produce intrinsic complexity on multiple length and time scales, allowing possible self-organization of new states of matter. This field of research, therefore, presents a major challenge for condensed matter physicists, chemists and materials scientists.

We propose to use muon spin rotation, relaxation or resonance (μSR) to study the magnetic and superconducting properties of primarily f-electron materials exhibiting novel ground states. μSR is a magnetic resonance technique which primarily utilizes 100% naturally spin polarized positive muons implanted at lattice interstitial sites. The μ^+ is a spin-1/2 particle with a lifetime of 2.2 microseconds decaying via the weak interaction into a positron and two undetected neutrinos. A μSR experiment generally consists of detecting both the muon's arrival and decay times and monitoring its spin polarization as a function of time by tracking the decay anisotropy of the emitted positrons. The local magnetic field is sensed through a combination of dipole and contact hyperfine coupling. The μSR technique is highly complementary to NMR, in that it requires no external magnetic field, can in principal be carried out in any material (single- or poly-crystalline, with or without nuclear moments) and has a large magnetic moment (≅3×proton moment) making it quite sensitive to internal fields as small as 1 Gauss. μSR is also complementary to neutron scattering in that it can detect magnetic field correlation times from 10^{-4} to 10^{-12} s. These times are generally slower than conventional neutron scattering techniques, which probe times ≤ 10^{-12}s. This synergism means that the ASRC (Advanced Science Research Center) μSR program will have strong in-house collaborative interactions with the Uranium NMR, Neutron and Materials Groups at JAERI.

2. Research Program

First, we spell out the differences which continuous and pulsed muon beams impose on planning a research program. The future J-PARC facility will provide the *most intense* muon beams in the world, with two 80 nanosecond long proton pulses separated by 600 ns at 25 Hz. These two 80 ns pulses will be kicked to different experimental spectrometers. In each spectrometer all muons arriving within the 80 ns time window are counted as being 'simultaneous.' Because individual muons are not detected, the intrinsic time resolution of the proton beam is 80ns. This will be reduced further by chopping the μ^+ beam, but only by about a factor of two or so with current technology. This should produce a background-free muon time spectrum (typically 15-20 μs long) with a time resolution of ≅ 30 ns, which is optimum for measuring slow muon relaxation in zero applied field (ZF), or in a field of any magnitude directed along the initial muon spin direction (longitudinal field, LF). Furthermore, because all muons in a given 30 ns pulse are accepted, very large counting rates (10^6 s^{-1}) can be utilized. The inherent pulse structure also allows efficient use of pulsed experimental environments, such external magnetic fields (dc or rf) or laser pulses, timed to coincide with the muon pulses in the sample.

An internal or applied field perpendicular to the muon's initial spin direction (transverse field, TF) causes muon precession, so that the 30 ns time resolution limits the TF field magnitude to about 700 Oe. This is a severe limitation, and means that most TF experiments (such as Knight shift or superconducting penetration depth measurements) need to be carried out at a continuous beam facility like TRIUMF, where individual muons can be counted due to the much lower instantaneous stopping rates. This allows much higher time resolution so that muon frequencies in fields up to 7 T can be measured. Allowing only one muon to be counted every 10 - 15 μs in continuous a beam limits the muon acceptance rate to about 3×10^4 s^{-1}, however.

We are planning to build our own μSR spectrometer at J-PARC which is particularly suitable for studying actinide materials. We will be able to access temperatures between 0.02 - 300 K in magnetic fields up to 3 T. Pressures up to 1 GPa will be usable in a different beam line. Furthermore, we plan strong collaborations with existing research efforts inside and outside Japan. Collaboration with world class sample providers will be of high priority.

A few of the general classes of experiments we will carry out include the following.

- **Plutonium and other radioactive actinide systems**

We have a strong collaboration with Los Alamos and Livermore National Labs in the U.S. to perform experiments on highly radioactive materials such as Pu and the newly discovered superconductor $PuCoGa_5$. These Pu-based materials are currently synthesized at Los Alamos or Livermore and measured at TRIUMF, but we envision that when the J-PARC facility is fully operational we will be able to combine the extraordinary materials synthesis capabilities within JAERI with the J-PARC muon beams, making a truly unique combination.

- **Spin lattice relaxation rate studies in novel magnetic materials**

Almost all magnetic materials can be effectively studied using the background free pulsed beam facilities at J-PARC. This would include, as examples, critical phenomena in ferro- and antiferromagnets, disordered magnets, octupolar ordering and non-Fermi liquid materials. Ultra high-precision experiments can be carried out due to the extraordinarily large anticipated counting rates. (Imagine being able to collect a million events in a few seconds!)

- **Measurements of small magnetic fields.**

An example of small moment magnetism occurs in superconductors possessing time-reversal-violating order parameters. Here the spin or orbital moments associated with the superconductivity produce magnetic fields of only a few Oe, ideal for μSR studies. Also, many heavy fermion materials exhibit incomplete Kondo screening of the local f-electron moments at low temperatures, resulting in frozen spin magnetism with moments as small as 0.001 μ_B. μSR is the most effective (and nearly the only) method for directly studying such systems. In addition to studying these Kondo-like systems themselves, it is important to establish the absence of small-moment magnetism, for example, because its presence could cause anomalous transport or thermodynamic properties which can masquerade as a presumed QCP.

- Knight shift studies

In ideal cases the Knight shift K_μ yields both the local spin susceptibility and the hyperfine/dipolar coupling. The muon Knight shift spectrum is usually quite simple because of its spin ½, generally consisting of a single line for each muon stopping site. The temperature dependence of the local spin susceptibility can be helpful in specifying the symmetry of a superconducting order parameter (singlet or triplet). Generally K_μ tracks the bulk susceptibility; cases where it may not indicate fundamental changes in the local electronic structure. Thus, Knight shift studies will be an important component in our program, necessitating travel to continuous beam facilities like TRIUMF for use of strong magnetic fields.

To summarize, the ability to study the basic properties of novel actinide materials using a highly complementary technique like μSR with unprecedented precision should make the μSR capabilities at JAERI/J-PARC truly world class..