

Subnuclear System Research Division RIKEN Facility Office at RAL

1. Abstract

Our core activities are based on the RIKEN-RAL Muon Facility located at the ISIS Neutron & Muon Source at the Rutherford Appleton Laboratory (UK), which provides intense pulsed-muon beams. The RIKEN-RAL Muon Facility is a significant and long-standing collaboration between RIKEN and RAL in muon science—with 2020 being the 30th years of continuous agreements between RIKEN and RAL. The Facility enables muon science throughout Japan and other field—it continues to attract proposals from a wide variety of Japanese universities and institutions (with over 80 groups having now used the facility), and including industrial users such as Toyota, and has been instrumental in establishing scientific links with other Asian universities.

Muons have their own spins with 100% polarization, and can detect local magnetic fields and their fluctuations at muon stopping sites very precisely. The method to study the characteristics of materials by observing time dependent changes of muon spin polarization is called “Muon Spin Rotation, Relaxation and Resonance” (μ SR method), and is applied to study electro-magnetic properties of insulating, metallic, magnetic and superconducting systems. Muons reveal static and dynamic properties of the electronic state of materials in the zero-field condition, which is the ideal magnetic condition for research into magnetism. For example, we have carried out μ SR investigations on a wide range of materials including frustrated pyrochlore systems, which have variety of exotic ground states of magnetic spins, so the magnetism study of this system using muon is quite unique.

The ultra-cold muon beam can be stopped in thin foil, multi-layered materials and artificial lattices, which enables us to apply the aSR techniques to surface and interface science. The development of an ultra-cold muon beam is also very important as a source of pencil-like small emittance muon beam for muon $g - 2$ /EDM measurement. We have been developing muonium generators to create more muonium atoms in vacuum even at room temperature to improve beam quality compared with the conventional hot-tungsten muonium generator. We have demonstrated a strong increase in the muonium emission efficiency by fabricating fine laser drill-holes on the surface of silica aerogel. We are also developing a high power Lyman-alpha laser in collaboration with the Advanced Photonics group at RIKEN. The new laser will ionize muoniums 100 times more efficiently for slow muon beam generation.

Over the past 2–3 years, a significant development activity in muon elemental analysis has taken place, proton radius experiments have continued and been developed, and chip irradiation experiments have also continued.

2. Major Research Subjects

- (1) Materials science by muon-spin-relaxation method and muon site calculation
- (2) Development of elemental analysis using pulsed negative muons
- (3) Nuclear and particle physics studies via muonic atoms and ultra-cold muon beam
- (4) Other muon applications

3. Summary of Research Activity

(1) Materials science at the RIKEN-RAL muon facility

Muons have their own spins with 100% polarization, and can detect local magnetic fields and their fluctuations at muon stopping sites very precisely. The μ SR method is applied to studies of newly fabricated materials. Muons enable us to conduct (1) material studies under external zero-field condition, (2) magnetism studies with samples without nuclear spins, and (3) measurements of muon spin relaxation changes at wide temperature range with same detection sensitivity. The detection time range of local field fluctuations by μ SR is 10^{-6} to 10^{-11} second, which is an intermediate region between neutron scattering method (10^{-10} – 10^{-12} second) and Nuclear Magnetic Resonance (NMR) (longer than 10^{-6} second). At Port-2 and 4 of the RIKEN-RAL Muon Facility, we have been performing μ SR researches on strong correlated-electron systems, organic molecules, energy related materials and biological samples to study electron structures, superconductivity, magnetism, molecular structures and crystal structures.

Among our scientific activities on μ SR studies from year up to 2022, following subjects of materials sciences are most important achievements at the RIKEN-RAL muon facility:

- (1) Quasi magnetic monopole state in the Ru-based pyrochlore systems, $R_2\text{Ru}_2\text{O}_7$;
- (2) Novel superconducting gap structure in the quasi two-dimensional organic superconductor, λ -(BETS) $_2$ GaCl $_4$;
- (3) Determination of muon positions estimated from density functional theory (DFT) and dipole-field calculations;
- (4) Chemical muonic states in DNA molecules.

Result-1) Quasi magnetic monopole state in pyrochlore systems gave us new interpretations to understand exotic phenomena, like the quantum spin liquid and spin fragmentation states. Result-2) A novel superconducting gap structure in the quasi two-dimensional organic superconductor, λ -(BETS) $_2$ GaCl $_4$. An intermediate one-band superconducting gap-structure was confirmed. Result-3) Well known and deeply investigated La $_2$ CuO $_4$ has opened a new scheme of the Cu spin. Taking into account quantum effects to expand the Cu-spin orbital and muon positions, we have succeeded to explain newly found muon sites and hyperfine fields at those sites. Result-4) Chemical states of the muon which attaches to DNA molecules were investigated by the avoided level-crossing muon-spin resonance experiments. In conjunction with DFT calculations, we are trying to reveal the electron motion through the DNA molecule.

We have been continuing to develop muon-science activities in Asian countries. We enhanced international collaborations to organize new μ SR experimental groups and to develop muon-site calculation groups using computational method. We are creating new collaborations with new teams in different countries and also continuing collaborations in μ SR experiments on strongly correlated systems with researchers from Taiwan, Indonesia, China, Thailand and Malaysia including graduate students. We are starting to

collaborate with the new Chinese muon group who are developing the Chinese Muon Facility and trying to develop more muon activities in the Asian area.

(2) Development of elemental analysis using pulsed negative muons

There has been significant development of elemental analysis using negative muons on Port 4 and Port 1 over the past couple of years. Currently, elemental analysis commonly uses X-ray and electron beams, which accurately measure surfaces. However, a significant advantage of muonic X-rays over those of electronic X-rays is their higher energy due to the mass of the muon. These high energy muonic X-rays are emitted from the bulk of the samples without significant photon self-absorption. The penetration depth of the muons can be varied by controlling the muon momentum, providing data from a thin slice of sample at a given depth. This can be over a centimeter in iron, silver and gold or over 4 cm in less dense materials such as carbon.

Some techniques for elemental analysis are destructive or require the material under investigation to undergo significant treatment and some of the techniques are only sensitive to the surface. Therefore, negative muons offer a unique service in which they can measure inside, beyond the surface layer and completely non-destructively.

The areas of science that have used negative muons for elemental analysis have been very diverse. The largest area is the cultural heritage community as the non-destructive ability is particularly important and will become more so. This community have investigated swords from different eras, coins (Roman gold and silver, Islamic silver and from the Tudor Warship Mary Rose), miniature boats from Sardinia, reliefs on Baptist church gate, Bronze Age tools and cannon balls. In addition, energy materials (Li composition for hydrogen storage), bio-materials (search for iron to potentially help understand Alzheimer's), engineering alloys (manufacturing processes for new materials for jet engines), and functional materials (surface effects in piezo electrics) have also been investigated. The study was extended to see the difference by isotopes of silver and lead, which may give hint on the source of the material.

(3) Ultra-cold (low energy) muon beam generation and applications

Positive muon beam with thermal energy has been produced by laser ionization of muonium (bound system of μSR^+ and electron) emitted from a hot tungsten surface with stopping surface muon beam at Port-3. The method generates a positive muon beam with acceleration energy from several 100 eV to several 10 keV, small beam size (a few mm) and good time resolution (less than 8 nsec). By stopping the ultra-cold muon beam in thin foil, multi-layered materials and artificial lattices, we can precisely measure local magnetic field in the materials, and apply the μSR techniques to surface and interface science. In addition, the ultra-cold muon is very important as the source of pencil-like small emittance muon beam for muon $g - 2/\text{EDM}$ measurement. It is essential to increase the slow muon beam production efficiency by 100 times for these applications. There are three key techniques in ultra-cold muon generation: production of thermal muonium, high intensity Lyman-alpha laser and the ultra-cold muon beam line.

A high-power Lyman-alpha laser was developed in collaboration with the Advanced Photonics group at RIKEN. The new laser system is used at J-PARC U-line and, upon completion, will ionize muoniums 100 times more efficiently for slow muon beam generation. In this development, we succeeded to synthesize novel ceramic-based Nd:YAG and Nd:YAG crystals, which realized a highly efficient and stable laser system. We are working hard to improve the homogeneity of large size crystals in order to achieve full design power.

We also succeeded in developing an efficient muonium generator, laser ablated silica aerogel, which emits more muoniums into vacuum even at room temperature. Study has been done at TRIUMF utilizing positron tracking method of muon decay position. We demonstrated in 2013 at least 10 times increase of the muonium emission efficiency by fabricating fine laser drill-holes on the surface of silica aerogel. Further study was carried out in 2017 to find the optimum fabrication that will maximize the muonium emission. From the analysis, we found the emission has large positive correlation with the laser ablated area rather than with any other parameters. We also confirmed the muon polarization in vacuum. An alternative detection method for muonium emission using muonium spin rotation, which will be sensitive even to muoniums near the surface, was tested at RIKEN-RAL in 2018 and was found successful. The study was further applied to the measurement of the temperature dependence.

A new ultra-cold muon beam line for muon $g - 2/\text{EDM}$ was constructed based on our various experience. The new beamline has larger aperture for the beam, easier magnetic field monitoring, active field cancelling, and fine beam control for the injection to RFQ accelerator. The beamline is being tested at J-PARC S2 line.

(4) Other fundamental physics studies

We proposed the measurement of the proton radius by using the hyperfine splitting of the muonic hydrogen ground state. This hyperfine splitting is sensitive to the Zemach radius, which is a convolution of charge and magnetic-dipole distributions inside proton. We are planning to re-polarize the muonic hydrogen by a circularly polarized excitation laser (excites one of the $F = 1$ states and regenerates the muon spin polarization), and detect the recovery of the muon decay-asymmetry along the laser.

Preparation using muon beam is in progress. We measured the muon stopping distribution in low-density hydrogen-gas cell, which gave us consistent results with beam simulation. Another key is the lifetime of the upper hyperfine state of the muonic hydrogen that will contribute the polarization. We successfully observed the clear muon spin precession of muonic deuterium atom in 2018 for the first time in the world. The measurement with muonic protium was carried out in 2019 and the data is being analyzed.

(5) Facility operation and refurbishment

The research activity at RIKEN-RAL had severe restriction in FY2021 also since the COVID-19 pandemic continued. It was impossible for users from outside RAL to carry out the experiment. Even though, we managed to carry out several μSR experiments with mailed samples. Since July 2021, RIKEN-RAL muon facility had been undergoing major refurbishment, first time since its operation in 1994. The work covered many important components, such as the cooling water circuit, vacuum system, radiation shielding, magnet

power supplies and power cables, detectors and beamline control system. Most of the works were completed by December 2022. We had some test beam at the end of December and confirmed that most of the beamline components are working fine. As the ISIS beam delivery in early 2023 was very limited, we are planning the full beam test in middle 2023.

Members

Director

Philip KING

Senior Research Scientist

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List of Publications & Presentations

Publications

[Original Papers]

- M. D. Umar, K. Ishida, R. Murayama, D. P. Sari, U. Widyaiswari, M. Fronzi, I. Watanabe, and M. Iwasaki, "Muon-spin motion at the crossover regime between gaussian and lorentzian distribution of magnetic fields," *Prog. Theor. Exp. Phys.* **2021**, 083101 (2021).
- W. N. Zaharim, S. N. A. Ahmad, S. Sulaiman, H. Rozak, D. F. H. Baseri, N. A. M. Rosli, S. S. Mohd-Tajudin, L. S. Ang, and I. Watanabe, "Density functional theory study of 12 mer single-strand guanine oligomer and associated muon hyperfine interaction," *ACS Omega* **6**, 29641 (2021).
- A. Jamaludin, W. N. Zaharim, S. Sulaiman, H. Rozak, A. L. Sin, and I. Watanabe, "Density functional theory investigation of muon hyperfine interaction in guanine-cytosine double-strand DNA," *J. Phys. Soc. Jpn.* **91**, 024301 (2022).
- S. Yoon, W. Lee, S. Lee, J. Park, H. Lee, Y. S. Choi, S. -H. Do, W. -J. Choi, W. -T. Chen, F. Chou, D. I. Gorbunov, Y. Oshima, A. Ali, Y. Singh, A. Berlie, I. Watanabe, and K. -Y. Choi, "Quantum disordered state in the J_1 - J_2 square-lattice antiferromagnet $\text{Sr}_2\text{Cu}(\text{Te}_{0.95}\text{W}_{0.05})\text{O}_6$," *Phys. Rev. Res.* **5**, 014411 (2021).
- Ce Zhang, T. Hiraki, K. Ishida, S. Kamal, S. Kamioka, T. Mibe, A. Olin, N. Saito, K. Suzuki, S. Uetake, and Y. Mao, "Modeling the diffusion of muonium in silica aerogel and its application to a novel design of multi-layer target for thermal muon generation," *Nucl. Instrum. Methods Phys. Res. A* **1042**, 167443G (2022).
- A. Green, K. Ishida, K. Domoney, T. Agoro, and A. D. Hillier, "Negative muons reveal the economic chaos of Rome's AD 68/9 Civil Wars," *Archaeol. Anthropol. Sci.* **14**, 165 (2022).

Presentations

[International Conference/Workshop]

- K. Ishida on behalf of the J-PARC muon $g - 2$ /EDM collaboration (E34), "Muon $g - 2$ /EDM experiment at J-PARC," Fifth Plenary Workshop of the Muon $g - 2$ Theory Initiative, UK (The University of Edinburgh), September 5–9, 2022.

[Domestic Conferences/Workshops]

- 石田勝彦, 「ラザフォードアップルトン研究所ミュオン施設の紹介」, 第7回文理融合シンポジウム 量子ビームで歴史を探る—加速器が紡ぐ文理融合の地平—, つくば市 (KEK), 2022年11月3日.
- 石田勝彦, 「ミュオン触媒核融合の利用」, 中間子科学の将来討論会, 和光市 (理研), 2022年11月9–11日.
- 石田勝彦, 「理研 RAL 施設と原子・原子核研究」, 第13回 muon 科学と加速器研究研究会, 和光市 (理研), 2023年1月10–11日.