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 at 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" (μSR 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 μSR 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) Material 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 μSR 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 μSR 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 μSR 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 μSR studies from year 2017 to 2020, following subjects of material sciences are most important achievements at the RIKEN-RAL muon facility:

(1) Multi magnetic transitions in the Ru-based pyrochlore systems, $R_2Ru_2O_7$.
(2) Novel magnetic and superconducting properties of nano-size La-based high-T_c superconducting cuprates.
(3) Determination of muon positions estimated from density functional theory (DFT) and dipole-field calculations.
(4) Chemical muonic states in DNA molecules.

Result-1) Doped hole effects on the magnetic properties of corner-shared magnetic moments on pyrochlore systems gave us new interpretations to understand exotic phenomena, like the quantum criticality of magnetic moments and a quasi-magnetic monopole state. Result-2) The same nano-size effect was examined on the La-based high-T_c superconducting oxide changing the electronic state from insulating to superconducting. We confirmed the reduction in the magnetic interaction and the disappearance of the superconducting state leading the increase in the ferromagnetic interaction within the wide-range of the hole concentration. Result-3) Well known and deeply investigated La_2CuO_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 though the DNA molecule.

We have been continuing to develop muon-science activities in Asian countries. We enhanced international collaborations to organize new μSR 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 μSR 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 centimetre 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 μ^+ 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 $g-2$/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:YGAG crystal, which realized a highly efficient and stable laser system. However, larger size crystal than presently available is needed for full design power. We are working hard to improve the crystal homogeneity including the option of using material with slightly different composition.

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.

In RIKEN-RAL Port 3, the ultra-cold muon beam line, which had been designed with hot tungsten, was completely rebuilt to use advantage of the new room temperature silica aerogel target. The equipment was tested with surface muon beam and basic data such as muon stopping in aerogel were taken. A similar target design will be adopted in the ultimate cold muon source planned for muon $g-2$/EDM at J-PARC.

(4) Other fundamental physics studies

A measurement of the proton radius using $2S$-$2P$ transition of muonic hydrogen at PSI revealed that the proton charge radius is surprisingly smaller than the radius measured using normal hydrogen spectroscopy and e-p scattering by more than 5 times their experimental precision. The muonic atom has larger sensitivity to the proton radius because the negative muon orbits closer to the proton, although there is no reason why these measurements can yield inconsistent results if there exists no exotic physics or unidentified phenomenon behind. The cause of the discrepancy is not understood yet, thus a new measurement with independent method is much anticipated.

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) Other topics

RIKEN and ISIS have signed a new collaboration agreement for the period 2018–2023. This is the fourth in a continuous series of agreements, the first being signed in 1990, resulting in a partnership which will have lasted over 30 years. Under the new agreement, ownership and operation of the facility was passed to ISIS, a refurbishment programme of the facility has started, a user programme for Japanese scientists continued under the partnership between RIKEN and ISIS. The RIKEN-RAL collaboration is regularly highlighted as a good example of UK-Japanese science partnership at the UK-Japan Joint Committee on Science and Technology (chaired by the UK Chief Scientific Advisor to Government and a counterpart from Japan)—for example, Dr. King and Dr. Watanabe presented RIKEN-RAL at the November 2016 meeting of the Committee. The RIKEN-RAL collaboration has also enabled the development of collaborative activity between RIKEN and other Asian universities, *e.g.* through several MoUs with Indonesian and Malaysian universities.

It was very unfortunate that the research activity at RIKEN-RAL had severe restriction in FY2020 due to the COVID-19 pandemic. Almost all the access to RIKEN-RAL by external users had to be stopped. Even though, we managed to carry out several μSR experiments with mailed samples, where the ISIS staff mounted the samples and the external users controlled the experimental sequence remotely.

Members

Director

Philip KING

Senior Research Scientist

Isao WATANABE

Contract Researcher

Katsuhiko ISHIDA

List of Publications & Presentations

Publications

[Original Papers]

R. Kitamura, S. Bae, S. Choi, Y. Fukao, H. Iinuma, K. Ishida, N. Kawamura, B. Kim, Y. Kondo, T. Mibe, Y. Miyake, M. Otani, G. P. Razuvaev, N. Saito, K. Shimomura, and P. Strasser, "Development of negative muonium ion source for muon acceleration," Phys. Rev. Accel. Beams **24**, 033403 (2021).

S. Kanda, Y. Fukao, Y. Ikedo, K. Ishida, M. Iwasaki, D. Kawall, N. Kawamura, K. M. Kojima, N. Kurosawa, Y. Matsuda, T. Mibe, Y. Miyake, S. Nishimura, N. Saito, Y. Sato, S. Seo, K. Shimomura, P. Strasser, K. S. Tanaka, T. Tanaka, H. A. Torii, A. Toyoda, and Y. Ueno, "New precise spectroscopy of the hyperfine structure in muonium with a high-intensity pulsed muon beam," Phys. Lett. B **815**, 136154 (2021).

A. D. Pant, K. Ishida, N. Kawamura, S. Matoba, A. Koda, S. Nishimura, and K. Shimomura, "Study of muonium behavior in n-type silicon for generation of ultra cold muonium in vacuum," Physica B **613**, 412997 (2021).

J. Sugiyama, O. K. Forslund, E. Nocerino, N. Matsubar, K. Papadopoulos, Y. Sassa, S. P. Cottrell, A. D. Hillier, K. Ishida, M. Mansson, and J. H. Brewer, "Lithium diffusion in $LiMnPO_4$ detected with $\mu^{\pm}$SR," Phys. Rev. Res. **2**, 033161 (2020).

M. Aramini, C. Milanese, A. D. Hillier, A. Girella, C. Horstmann, T. Klassen, K. Ishida, M. Dornheim, and C. Pistidda, "Spectroscopic tool for the investigation of the local chemistry of elements," Nanomaterials **2020**, 10, 1260 (2020).

E. Mocchiutti, A. Adamczak, D. Bakalov, G. Baldazzi, R. Benocci, R. Bertoni, M. Bonesini, V. Bonvicini, H. CabreraMorales, F. Chignoli, M. Clemenza, L. Colace, M. Danailov, P. Danev, A. de Bari, C. DeVecchi, M. DeVincenzi, E. Furlanetto, F. Fuschinod, K. S. Gadedjisso-Tossoua, D. Guffantia, K. Ishida, C. Labanti, V. Maggi, R. Mazza, A. Menegolli, G. Morgante, M. Nastasi, J. Niemela, C. Pizzolotto, A. Pullia, R. Ramponi, L. P. Rignanese, M. Rossella, N. Rossi, M. Stoilov, L. Stoychev, L. Tortora, E. Vallazza, G. Zampa, and A. Vacchi, "First measurement of the temperature dependence of muon transfer rate from muonic hydrogen atoms to oxygen," Phys. Lett. A **384**, 126667(2020).

C. Pizzolotto, A. Adamczak, D. Bakalov, G. Baldazzi, M. Baruzzo, R. Benocci, R. Bertoni, M. Bonesini, V. Bonvicini, H. Cabrera, D. Cirrincione, M. Citossi, F. Chignoli, M. Clemenza, L. Colace, M. Danailov, P. Danev, A. de Bari, C. De Vecchi, M. de Vincenzi, E. Fasci, E. Furlanetto, F. Fuschino, K. S. Gadedjisso-Tossou, L. Gianfran, D. Guffanti, A. D. Hillier, K. Ishida, P. J. C. King, C. Labanti, V. Maggi, R. Mazza, A. Menegolli, E. Mocchiutti, L. Moretti, G. Morgante, J. Niemela, B. Patrizi, A. Pirri, A. Pullia, R. Ramponi, L. P. Rignanese, E. Roman, M. Rossella, R. Sarkar, A. Sbrizzi, M. Stoilov, L. Stoychev, J. J. Suárez-Vargas, G. Toci, L. Tortora, E. Vallazza, M. Vannini, C. Xiao, G. Zampa, and A. Vacchi, "The FAMU experiment: muonic hydrogen high precision spectroscopy studies," Eur. Phys. J. A **56**, 185 (2020).

J. Beare, G. Beer, J. H. Brewer, T. Iijima, K. Ishida, M. Iwasaki, S. Kamal, K. Kanamori, N. Kawamura, R. Kitamura, S. Li, G. M. Luke, G. M. Marshall, T. Mibe, Y. Miyake, Y. Oishi, K. Olchanski, A. Olin, M. Otani, M. A. Rehman, N. Saito, Y. Sato, K. Shimomura,

K. Suzuki, M. Tabata, and H. Yasuda, “Study of muonium emission from laser-ablated silica aerogel,” Prog. Theor. Exp. Phys. **2020**, 123C01 (2020).

H. Yamauchi, D. P. Sari, I. Watanabe, Y. Yasui, L. -J. Chang, K. Kondo, T. U. Ito, M. Ishikado, M. Hahara, M. D. Frontzk, C. Chi, J. A. Fernandez-Bac, J. S. Lord, A. Berlie, A. Kotani, S. Mori, and S. Shamoto, “High-temperature short-range order in Mn_3RhSi,” Commun. Matter. **1**, 42 (2020).

M. Fujita, K. M. Suzuki, S. Asano, H. Okabe, A. Koda, R. Kadono, and I. Watanabe, “Magnetic behavior of T'-type Eu_2CuO_4 revealed by muon spin rotation and relaxation measurements,” Phys. Rev. B **102**, 045116 (2020).

Presentations

[Domestic Conferences/Workshops]

石田勝彦, 「理研 RAL での非破壊元素分析」, 第 4 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—, オンライン, 2021 年 1 月 28–29 日.

石田勝彦 (招待講演), 「理研 RAL と J-PARC」, パルス中性子ミュオン発生 40 周年記念シンポジウム, オンライン, 2020 年 12 月 23 日.

石田勝彦, 「英国理研 RAL ミュオン施設におけるミュオン利用分析研究の進展」, 第 3 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—, オンライン, 2020 年 9 月 25–26 日.

Press Releases

正負のミュオンで捉えたイオンの動き 2020 年 8 月 7 日—Li イオンの動きを, 負ミュオンで確認, 正ミュオンで詳細観察— 一般財団法人総合科学研究機構（CROSS）中性子科学センター・サイエンスコーディネータ 杉山 純国立研究開発法人理化学研究所（理研）仁科加速器科学研究センター・協力研究員石田 勝彦