Historical overview of the RIKEN-RAL Muon Facility *from catalyzed fusion to materials/life science*

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Introduction

Construction of the RIKEN-RAL Muon Facility was proposed by RIKEN in 1989. It had been realized as the first large-scale scientific contribution from Japan to UK. Major research outputs achieved thus far and future prospects are briefly summarized here.

Birth of RIKEN-RAL Muon Facility

At the time of the cold fusion (proposed nuclear fusion phenomena of D_2 confined in special materials such as Pd) in 1989, muon-catalyzed fusion was recognized as a real nuclear fusion process without the need for high temperature. Immediately after the cold-fusion report, RIKEN proposed the construction of a muon facility at the world's strongest pulsed proton accelerator facility, the Rutherford Appleton Laboratory in UK, and the proposal was approved by the Japanese Government in 1990.[1)] A facility composed of a superconducting pion-decay section with 4 muon extraction lines was designed, constructed, and completed in 1994, as shown in Fig. 1, and various experimental programs were started.[2)]

Muon Catalyzed Fusion (μCF)

Among the various muon sciences, μCF, which is nuclear fusion catalyzed by the muon, is the most important as a new possible energy source. Fascinating features of the μCF can be summarized as follows: a) fusion phenomena within a small muonic atom/molecule and its successive reactions, b) no need for high temperatures, c) no radioactive waste production, d) no fuel problem, e) no critical phenomena for free-running, and f) already realized in a laboratory scale and only a few efforts are required for a break-even achievement.[3)]

μCF consists of the following processes: 1) the formation of a small muonic molecule and subsequent intra-molecular fusion reaction, and 2) the mediation of a chain of fusion reactions by a single μ^- (Fig. 2), up to more than 100 cycles. The numbers such as rates are quoted from a standard textbook.

After μ^- injection and stopping in a D-T mixture, either a (dμ) or a (tμ) atom is formed. Because of the difference in the binding energies, the μ^- initially in the (dμ) undergoes a transfer reaction to the (tμ), which reacts with D_2 or DT to form a muonic molecule (dtμ) at a rate of $\lambda_{dt\mu}$, where the formation of a specific state of (dtμd2e$^-$) through a resonant molecular formation (RMF) takes place. The (dtμ) molecule, after a rapid cascade transition, causes a fusion reaction since the distance between d and t is sufficiently close to allow fusion to occur, producing a 14.1-MeV n and a 3.5-MeV α.

After the fusion inside the (dtμ) molecule, most of the μ^- is liberated to participate in the second μCF. Some small fraction is captured by the emitted α^{++} (called the initial sticking probability ω_S^0). Once the $(\alpha\mu)^+$ is formed, because of kinetic energy, the μ^- can be stripped from the $(\alpha\mu)^+$ ion and liberated again

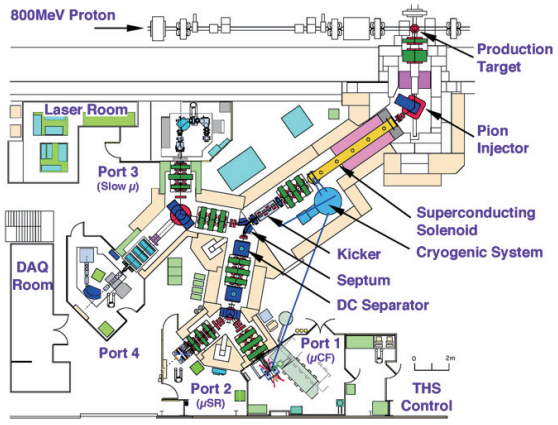

Fig. 1. Layout of the RIKEN-RAL Muon Facility in 2010.

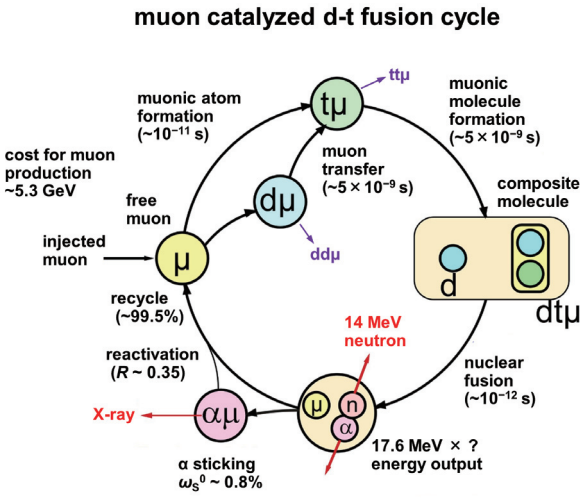

Fig. 2. Conceptual diagram of the D-T μCF cycle.

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(regeneration fraction R). Thus, μ^- of either a non-stuck μ^- or a regenerated one can participate in the second μCF cycle, while the thermalized $(\alpha\mu)^+$ is left out from the μCF cycle with an effective sticking $\omega_S = (1-R)\omega_S^0$. More than 100 fusion processes are known to take place with a single μ^-.

The phenomena of sticking and regeneration were deeply investigated at RIKEN-RAL by measuring both the X-ray from stuck $(\alpha\mu)^+$ representing initial sticking and the yield of fusion neutrons representing resultant phenomena including regeneration. Here, the strong pulsed muon was essentially helpful to remove the background related to T β-decay. The temperature-dependent measurement was conducted for solid/liquid D-T.[5] Although the X-ray yield remains constant, the loss rate decreases at higher temperatures; the regeneration seems to increase in the high-temperature solid (Fig. 3).

At RIKEN-RAL, the muon molecular formation rate in D_2 μCF ($\lambda_{dd\mu}$) was investigated by controlling ortho- and para-states to study the energy matching in RMF. Contrary to theoretical expectation, the opposite tendency was discovered in solid and liquid D_2.[6,7] Although the result is preliminary, a similar opposite tendency was discovered at RIKEN-RAL for the μCF in liquid D-T by using ortho- and para-D_2 with T_2 in a non-equilibrium liquid mixture.[8] The observed ortho-para dependence of the fusion neutron yield seems to be opposite to the theoretical expectation; a greater fusion neutron yield was observed for the ortho case in experiment. All of these observations clearly demonstrate the importance of the molecular correlation effect in RMF in D_2 and D-T μCF. The molecular correlation may change the energy-level diagram in D_2 and D-T in a scale of meV at an energy spacing of the order of eV.

Based on these results, several remarks can be made on possible further increase in the energy-production capability. By using ortho-D_2 rich and high-temperature (30 K) solid D-T μCF towards economical break-even, the following can be achieved: a) ortho-para optimized D_2 can be used to increase $\lambda_{dt\mu}$ to 800 μs^{-1} b) and in order to decrease ω_S (increase R), use of high-temperature solid D-T at 30 K may cause a decrease of ω_S to 0.1% (increase of R to 0.9%). Then, the fusion neutron occurrence is $1200/\mu^-$, which corresponds to an energy production capability of 21 GeV/μ^-, well exceeding the cost of muon production of 5.0 GeV/μ^-.

Energy production in solid D-T (30 K) is not realistic. Another idea is has been proposed: strongly correlated μCF. Let us consider the spatial structure of successive D-T μCF chain, where α^{++} ions are emitted in a space of within $(120\ \mu m)^3$ from a confined spot (within $(0.2\ \mu m)^3$ towards a larger $(\alpha\mu)^+$ ion space (within $(480\ \mu m)^3$). Therefore, if we have strong pulsed muons with stopping region and μCF region overlapping each other, because of collisions between the $(\alpha\mu)^+$ ions and α^{++} ions from nearby μCF reactions, exotic regeneration of $(\alpha\mu)^+$ stripping through high-temperature plasma caused by α^{++} may occur in the high-density μCF of D-T mixtures at moderately high temperature. There, $\omega_S = 0.1\%$ can be expected, leading to the same energy production capability, which is beyond the break-even energy balance.

Condensed Matter Physics

By using the capability of long-time-range measurement of the μ^+SR time spectrum, various condensed matter studies have been proposed and conducted, mainly by external users amounting to more than 200 scientists. Topics covered a) magnetic properties of high-T_C superconductors, b) dynamic character of exotic magnetic materials, c) diffusion of μ^+ in various materials d) reactions of $\mu^+$and muonium in chemical materials, and e) light-element diffusion in battery materials. A distinguished example is presented below.

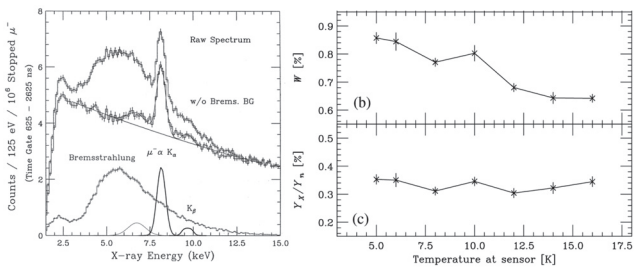

Fig. 3. Observed X-ray spectrum[4] (left) and temperature dependence of muon loss probability (ω_S), and the ratio of X-ray neutron yield for D-T μCF[5] (right).

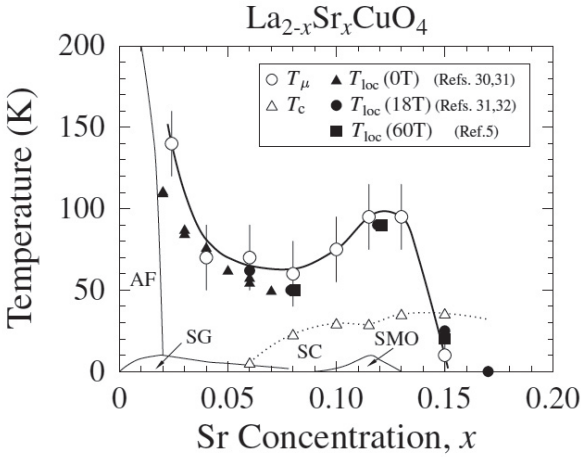

Fig. 4. Observed phase diagram of LSCO by a change of relaxation rate of muon (open circle) compared with electron resistivity (closed symbols).

Careful measurements of zero- and longitudinal-field muon-spin relaxation have been performed in order to investigate the dynamics of the electronic and magnetic states in the normal region of $La_{2-x}Sr_xCuO_4$ over a wide range of hole concentrations from 0.024 to 0.15.[9] It has been found that the dynamic depolarization rate of muon spins in zero field starts to increase monotonically from approximately 0.02 μs^{-1} with decreasing temperature at a high temperature of approximately 100 K. This effect is enhanced near a hole concentration of 1/8 per Cu, suggesting that the dynamics of fluctuating internal fields at the muon site in the normal state of $La_{2-x}Sr_xCuO_4$ changes at a high temperature of approximately 100 K and is correlated with the mobility of holes (Fig. 4). The result should be related to the origin of high T_C superconductivity.

For the μ^+SR studies of magnetism at surfaces and interfaces, the production of ultra-slow μ^+ is in high demand. By coupling pulsed lasers with pulsed muons, a production method of ultra-slow μ^+ by the laser resonant ionization of thermal muonium was developed at KEK. Advancements have been achieved at RIKEN-RAL by placing the system at the experimental port of a surface muon beam line[10] (Fig. 5). Energy was proved to be tunable from 0.1 keV to 20 keV. Space-time structure advancement was confirmed. The project is now under further development at J-PARC.

Life Sciences

By utilizing the unique and strongest pulsed muons, long-time range (more than 10 times the muon lifetime) became easily measured; consequently, the delicate muon spin response in biological environments is easily detectable.

The electron-transfer process plays highly important roles in biological activities. Two representative examples are the photosynthetic electron transfer chain and the respiratory chain in mitochondria. Most elementary molecules have been isolated, and their complicated structures have been determined one by one through crystallographic analysis. The remaining problem is the elucidation of the electron transfer mechanism.

Cytochrome *c* is a small hemoprotein and one of the members of the respiratory chain in mitochondria. It transfers an electron from protein complex III to IV, coupled with redox reactions of Fe located at the center of a porphyrin ring.

At RIKEN-RAL, μSR spectra of cytochrome *c* and myoglobin were measured.[11] μSR data were analyzed by the Risch-Kehr (R-K) theory, which was developed in order to analyze the μSR data of conducting polymers. The principle of the electron transfer in biological macromolecules studies by μSR is as follows (Fig. 6). During the slowing-down process, the injected μ^+ has a chance to pick up electron(s) to form a neutral muonium. Then, the muonium becomes thermalized, stops at an electronegative site, and forms a chemical bond to a molecule. Then, depending on the nature of the molecule, the electron brought in by the μ^+ becomes localized to form a muonic radical state and/or diffuses rapidly along either intra-molecular chains or inter-molecular paths. These behaviors of the electron caused by the μ^+ can sensitively be detected by measuring the spin-relaxation process of the μ^+ by using the μSR method. The discrimination of intra- and inter-molecular electron transfer can be discussed on the basis of the magnetic-field dependence of relaxation rates. Thus, the spin relaxation of the μ^+ due to magnetic interaction with the moving electrons near muons was derived as a characteristic function (R-K function), $G(t) = \exp(\Gamma t)\mathrm{erfc}(\Gamma t)^{1/2}$, for a rapid electron spin flip rate with the relaxation rate Γ, which

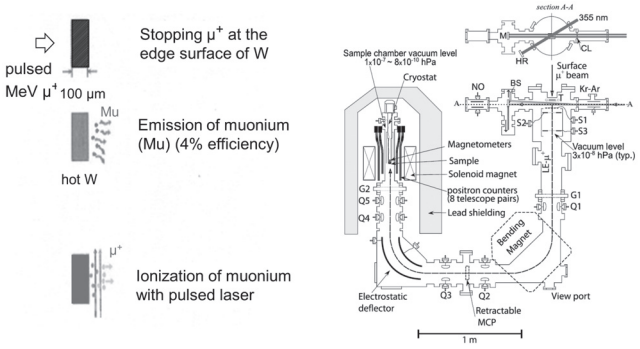

Fig. 5. Conceptual diagram showing the generation of an ultra-slow muon from an MeV muon by the laser ionization of thermal muonium (left). Layout of the dedicated experimental facility installed at port 3[10] (right).

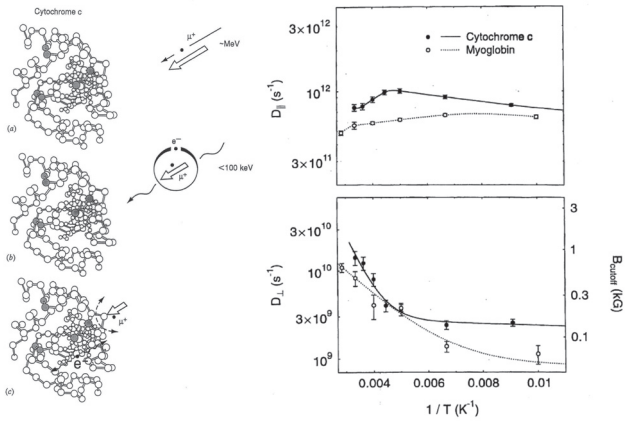

Fig. 6. Conceptual diagram showing the measurement of electron transfer in macromolecules by the μ^+SR method (left), and the measured temperature dependence of longitudinal (right-up) and transverse (right-down) diffusion rate.

has a characteristic dependence on the applied longitudinal field (B). In the case of one-dimensional intramolecular electron motion, Γ is proportional to $1/B$.

This experiment and several succeeding experiments on proteins and DNAs mainly performed at RIKEN-RAL have revealed the following facts: a) electron diffusion along the chain is temperature independent, while inter-chain diffusion has T-dependence reflecting glass transition at 100 K (Fig. 6); b) the hydration effect causes enhanced diffusion; c) the stopping position of the μ^+ in proteins seems to be away from Fe containing porphyrin but close to the part of the main poly-peptide chain.

Conclusion and future of RIKEN-RAL

As an extension of the pulsed muon facility, a facility to produce stronger pulsed muons has been constructed in Japan, the J-PARC MUSE, by KEK and JAEA. However, it is still important to keep RIKEN-RAL, since various experimental instrumentations are more easily equipped at RIKEN-RAL, promoting the increase of proposals and users. The author is grateful for the long-time collaboration of the following persons who contributed to the development of the RIKEN-RAL Muon Facility: Drs. M. Iwasaki, T. Matsuzaki, K. Ishida, I. Watanabe, R. Kadono, Y. Matsuda, S.N. Nakamura, P. Bakule, and F. Pratt.

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